

COMPREHENSIVE INVESTIGATION OF PLASMA BEHAVIOR IN DIVERSE
GAS DISCHARGE PHENOMENA: FLUID MODELING, CHAOTIC
DYNAMICS, AND SYNCHRONIZATION

A THESIS SUBMITTED TO
THE GRADUATE SCHOOL OF NATURAL AND APPLIED SCIENCES
OF
MIDDLE EAST TECHNICAL UNIVERSITY

BY

CİHAN YEŞİL

IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR
THE DEGREE OF DOCTOR OF PHILOSOPHY
IN
PHYSICS

SEPTEMBER 2023

Approval of the thesis:

**COMPREHENSIVE INVESTIGATION OF PLASMA BEHAVIOR IN
DIVERSE GAS DISCHARGE PHENOMENA: FLUID MODELING,
CHAOTIC DYNAMICS, AND SYNCHRONIZATION**

submitted by **CİHAN YEŞİL** in partial fulfillment of the requirements for the degree of **Doctor of Philosophy in Physics** Department, **Middle East Technical University** by,

Prof. Dr. Halil Kalıpçılar
Dean, Graduate School of **Natural and Applied Sciences**

Prof. Dr. Seçkin Kürkcüoğlu
Head of Department, **Physics**

Prof. Dr. İsmail Rafatov
Supervisor, **Physics, METU**

Examining Committee Members:

Prof. Dr. İsmet Yurduşen
Mathematics, Hacettepe University

Prof. Dr. İsmail Rafatov
Physics, METU

Prof. Dr. Seçkin Kürkcüoğlu
Physics, METU

Prof. Dr. Nevsan Şengil
Astronautical Engineering, TAA University

Assoc. Prof. Dr. Hande Toffoli
Physics, METU

Date:22.09.2023



I hereby declare that all information in this document has been obtained and presented in accordance with academic rules and ethical conduct. I also declare that, as required by these rules and conduct, I have fully cited and referenced all material and results that are not original to this work.

Name, Surname: CİHAN YEŞİL

Signature :

ABSTRACT

COMPREHENSIVE INVESTIGATION OF PLASMA BEHAVIOR IN DIVERSE GAS DISCHARGE PHENOMENA: FLUID MODELING, CHAOTIC DYNAMICS, AND SYNCHRONIZATION

YEŞİL, CİHAN

Ph.D., Department of Physics

Supervisor: Prof. Dr. İsmail Rafatov

September 2023, 188 pages

The work thoroughly examines various aspects of gas discharge phenomena. The foundational exploration begins with derivations of fluid equations from the kinetic Boltzmann equation. Plasma fluid equations are the foundation for numerical models, including a drift-diffusion approximation of particle fluxes. Self-consistent one-dimensional fluid models, the simple and extended fluid models, are developed and compared in addressing fundamental glow discharge requirements. Subsequently, the focus is on the transition from regular to chaotic oscillations in a DC-driven gas discharge–semiconductor system (GDSS) in nitrogen. This aims to understand the transition dynamics by revealing complex relationships among the control parameters. Further, the unpredictability and Delta Synchronization of Chaos (DSC) techniques are studied within the chaotic regime using detailed GDSS fluid models. These techniques validate chaotic solutions and chaos synchronization within the coupled drive and response systems. Furthermore, a comprehensive investigation of boundary conditions is conducted for fluid models of an RF argon discharge. Various boundary conditions are evaluated, emphasizing their impact on discharge behavior and proposing a revision of boundary conditions in commercial computational packages

to enhance accuracy and reliability. Finally, the electron Boltzmann solver is developed, enabling the precise calculation of electron transport and rate coefficients for fluid models. The solver is validated against conventional methods. Involving these coefficients in a CO_2 dielectric barrier discharge system concludes the necessity for advanced modeling and incorporation of gas heating equations.

Keywords: Gas discharge, kinetic Boltzmann equation, chaotic oscillations, gas discharge–semiconductor system, chaos synchronization



ÖZ

ÇEŞİTLİ GAZ DEŞARJ OLAYLARINDA PLAZMA DAVRANIŞLARININ KAPSAMLI İNCELEMESİ: AKIŞKAN MODELLEME, KAOTİK DİNAMİKLER VE SENKRONİZASYON

YEŞİL, CİHAN

Doktora, Fizik Bölümü

Tez Yöneticisi: Prof. Dr. İsmail Rafatov

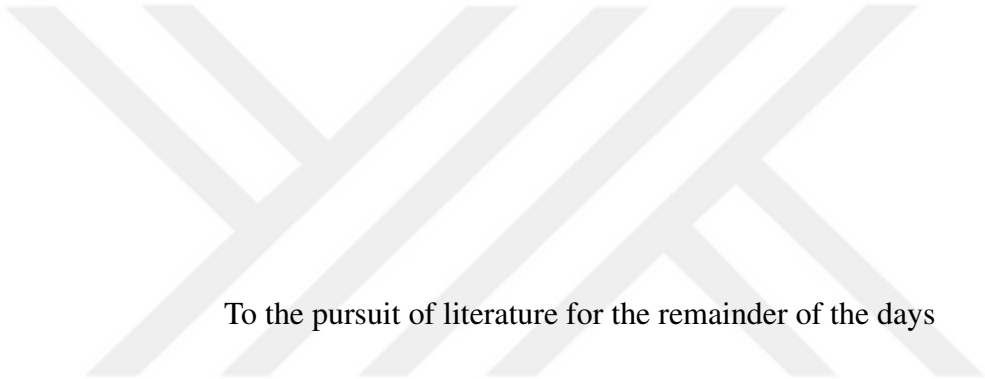
Eylül 2023 , 188 sayfa

Bu çalışma, gazdeşarj fenomeninin çeşitli yönlerini kapsamlı bir şekilde incelemektedir. Temel araştırma, kinetik Boltzmann denkleminin akış denklemlerine türetilmesiyle başlamaktadır. Sayısal modeller, parçacık akımları için sürüklenme-diffüzyon yaklaşımıyla plazma akış denklemlerine dayanmaktadır. Özgün bir boyutlu akış modelleri olan basit ve genişletilmiş akış modelleri, temel ısıldamadeşarj gereksinimlerini ele almak için geliştirilir ve karşılaştırılır. Ardından, odak noktası nitrojen içeren bir DC tahrikli gazdeşarj-yarıiletken sisteminde (GDSS) düzenli salınımlardan kaotik salınımlara geçişe yöneliktir. Bu, geçiş dinamiklerini anlamayı, kontrol parametreleri arasındaki karmaşık ilişkileri açığa çıkararak amaçlar. Dahası, öngörülemez ve Kaostaki Delta Senkronizasyon (DSC) teknikleri, detaylı GDSS akış modellerini kullanarak kaotik rejimde incelenir. Bu teknikler, bağlı sürücü ve tepki sistemleri içinde kaos senkronizasyonu ve kaotik çözümleri onaylar. Ayrıca, RF argondeşarjı için akış modelleri için kapsamlı bir sınama gerçekleştirilir. Farklı sınır koşulları değerlendirilir,deşarj davranışına etkileri vurgulanır ve ticari hesaplama paketlerindeki sınır

koşullarının doğruluğu ve güvenilirliğini artırmak için sınır koşullarının gözden geçirilmesi önerilir. Son olarak, elektron Boltzmann çözücüsü geliştirilir ve akış modelleri için elektron taşıma ve hız katsayılarının kesin hesaplamasını mümkün kılar. Çözücü, geleneksel yöntemlere karşı doğrulanır. Bu katsayıları CO_2 dielektrik bariyer deşarj sisteminde kullanmak, ileri seviye modellemeye ve gaz ısıtma denklemlerinin dahil edilmesine olan gerekliliği vurgular.

Anahtar Kelimeler: Gaz deşarj, kinetic Boltzmann denklemi, kaotik salınımlar, gaz deşarj-yarıiletken sistemleri, kaos senkronizasyonu





To the pursuit of literature for the remainder of the days

ACKNOWLEDGMENTS

I would like to commence by expressing my heartfelt appreciation to my thesis advisor, Professor Dr. İsmail Rafatov. His unwavering guidance and prompt responses to my inquiries have been a cornerstone of my research journey, providing me with invaluable insights and support.

I am also deeply grateful to my family for their unwavering encouragement and support, which has been a continuous wellspring of motivation throughout my life.

In conclusion, I would like to convey my profound admiration for Kağan Başkan, whose profound influence has shaped my thinking and provided me with strength during challenging times.

I also want to share my recently adopted personal motto: "As conditions shift, people and their pledges drift."

TABLE OF CONTENTS

ABSTRACT	v
ÖZ	vii
ACKNOWLEDGMENTS	x
TABLE OF CONTENTS	xi
LIST OF TABLES	xvi
LIST OF FIGURES	xviii
LIST OF ALGORITHMS	xxv
LIST OF ABBREVIATIONS	xxvi

CHAPTERS

1 INTRODUCTION	1
1.1 Low-temperature plasma	1
1.2 Gas discharge plasma	4
1.2.1 Gas discharge modeling	6
1.2.1.1 Subproblems of GDP modeling	7
1.2.1.2 Basic modeling approaches	9
1.2.2 Pattern formation and self-organization in planar gas discharges	14
1.2.2.1 Patterns in GDSS	15

1.2.2.2	Temporal oscillations in GDSS	16
1.2.2.3	Chaos phenomenon in GDSS	17
1.2.3	Synchronization in GDSS	20
1.2.3.1	Synchronization in chaotic systems	20
1.2.3.2	Chaos synchronization in GDSS	24
1.2.3.3	"Unpredictability" analysis and DSC method	25
1.2.4	Boundary Conditions for fluid modeling of RF discharge system	29
1.2.4.1	Motivation for universal BC	29
1.2.4.2	Existing literature	30
1.2.4.3	Properties of Radio-frequency (RF) discharges	31
1.2.4.4	BC investigations in RF plasmas	32
1.2.5	Input parameters of a DBD fluid model in CO_2	33
1.2.5.1	Basic configurations and properties of DBDs	33
1.2.5.2	Importance of CO_2 DBD system	38
1.2.5.3	Modeling of CO_2 DBD system	40
1.2.5.4	Boltzmann solvers and input parameters	41
2	GAS DISCHARGE MODELLING: FLUID EQUATIONS	45
2.1	Fluid description of plasma	45
2.2	Two-fluid model equations	49
2.3	"Drift-diffusion" approximation	52
2.4	"Simple" and "Extended" Fluid Models	54
2.5	Limitations and applicability	56

3	TRANSITION FROM PERIODIC TO CHAOTIC OSCILLATIONS IN A PLANAR SEMICONDUCTOR–GAS DISCHARGE SYSTEM	61
3.1	The statements of the problem	61
3.2	Description of Gas Discharge Model	65
3.2.1	Governing equations	65
3.2.2	Boundary conditions	68
3.2.3	"Simple fluid model"	69
3.2.4	Coupling of a semiconductor cathode layer	70
3.2.5	Parameter regime	70
3.3	Results and discussions	71
3.4	Conclusions to this chapter	79
4	THE DELTA UNPREDICTABLE SYNCHRONIZATION ANALYSIS OF CHAOS IN GAS DISCHARGE-SEMICONDUCTOR SYSTEMS	81
4.1	The statements of the problem	82
4.2	Preliminaries	86
4.3	The algorithms of the "unpredictability" and DSC methods	88
4.4	The "extended fluid model"	91
4.4.1	The stable and unstable stationary states	92
4.4.2	The dimensionless bifurcation diagram	94
4.5	Delta synchronization in the "extended fluid model"	95
4.5.1	The "unpredictability" analysis of drive and response systems	96
4.5.2	Delta synchronization between the drive and response systems	101
4.5.3	The generalized synchronization in the coupled system	103
4.6	Conclusions to this chapter	105

5	ANALYSIS OF BOUNDARY CONDITIONS FOR FLUID MODELS OF GAS DISCHARGES: IMPLEMENTATION OF THE ION BOUNDARY FLUX	107
5.1	The statements of the problem	107
5.2	The description of gas discharge model	109
5.2.1	Governing equations	109
5.2.2	Reaction rate and transport coefficients	110
5.2.3	Boundary conditions	113
5.3	Results and discussion	114
5.3.1	Effects of the different forms for the ion boundary flux	114
5.3.2	Effect of approximation for T_i	116
5.3.3	Effect of low and high electric field regions	118
5.3.4	Effect of different BCs	121
5.3.5	Effect of secondary electron emission coefficient	121
5.3.6	Comparison to PIC/MCC simulation results	123
5.4	Conclusions to this chapter	126
6	ANALYSIS OF INPUT PARAMETERS FOR A DIELECTRIC BARRIER DISCHARGE IN CO_2 FLOW AT ATMOSPHERIC PRESSURE	129
6.1	The statements of the problem	130
6.2	The Boltzmann equation solver	132
6.2.1	Model definition	132
6.2.2	Fluid model coefficients	134
6.2.3	Comparison of the coefficients	137
6.3	Gas discharge model	140

6.3.1	Governing equations	140
6.3.2	Parameter regime	141
6.3.3	Plasma-chemical model	142
6.3.4	Boundary conditions	143
6.4	Validation and verification of the models	144
6.4.1	Integral characteristics of CO_2 DBD	144
6.4.2	Temporal evolution of discharge variables	146
6.5	Conclusions to this chapter	148
7	CONCLUSION	151
	REFERENCES	155
	CURRICULUM VITAE	185

LIST OF TABLES

TABLES

Table 3.1	Set of plasma chemical reactions used in the "extended fluid model".	67
Table 3.2	Boundary conditions used in the "extended fluid model".	69
Table 4.1	The values of the control parameters (U_t , R_s) and the intersection points of the load line with the CVC curve (U , J) for (a), (b), (c) and (d) points in figure 4.3.	93
Table 4.2	Boundary conditions for the drive GDSS.	96
Table 4.3	Boundary conditions for the response GDSS.	97
Table 4.4	The elements of $\{t_n\}$, $\{s_n\}$ and $\{\delta_n\}$ sequences of the drive system.	99
Table 4.5	The elements of $\{t_n\}$, $\{s_n\}$ and $\{\delta_n\}$ sequences of the response system.	101
Table 4.6	Common dimensionless u_n , v_n , and δ_n values for the coupled system.	103
Table 4.7	Boundary conditions for the auxiliary GDSS.	104
Table 5.1	Set of plasma chemical reactions.	111
Table 5.2	The set of boundary conditions.	113
Table 5.3	Effect of boundary conditions on the maximum bulk plasma density. The pressure is $p = 3$ torr, the applied voltage amplitude and frequency are $V_0 = 300$ V and $f = 0.85$ MHz, the discharge gap is $d = 1$ cm. . . .	123

Table 6.1 Set of plasma chemical reactions used in DBD model for CO_2 gas at atmospheric pressure.	136
Table 6.2 The geometric properties of different coaxial DBD cells. The inner electrode diameter (I.E.D.), inner dielectric diameter (I.D.D.), outer di- electric diameter (O.D.D.), and outer electrode diameter (O.E.D.) values are defined for three different configurations.	142
Table 6.3 Boundary conditions at the walls of the discharge gap.	144



LIST OF FIGURES

FIGURES

Figure 1.1	The graphical representation of the parameter ranges appropriate for applying kinetic, hybrid, and fluid models can be found in [1]. . . .	13
Figure 1.2	The schematic of a planar DC gas discharge system with high ohmic cathode [2]	14
Figure 1.3	Some of the experimentally observed patterns [2]	16
Figure 1.4	The plasma variables develop into temporal homogeneous oscillations after transient solutions [3].	17
Figure 1.5	The phase portrait of the drive system of a GDSS [4]. Red marked points correspond to the instants t_n when the trajectory of the phase point passes through the closest point to the initial point where calculations have started.	26
Figure 1.6	The phase portraits of the drive (blue) and response (red) systems of a GDSS [4]. Green (drive) and yellow (response) data points represent the instants, when phase points pass through the closest place of corresponding initial points of the drive and response systems. . . .	27
Figure 1.7	The schematic illustration of typical CCP and planar-coil type ICP reactors [5].	31
Figure 1.8	Basic planar configurations of volume DBDs: (a1) symmetric, (a2) asymmetric, (a3) floated dielectric [6].	34

Figure 1.9	DBD reactor cross-sectional illustrations for the treatment of gases: coaxial volume DBD with an outer electrode arrangement (a) and an inner electrode arrangement (b) [6].	36
Figure 1.10	The illustrations of Filamentary and Diffuse DBDs.	37
Figure 3.1	The schematic illustration of a planar gas discharge coupled with a semiconductor layer.	62
Figure 3.2	Electron cross sections for the first six reactions in table 3.1: (1) elastic, (2) ionization, (3), (4), (5), and (6) excitation collisions in nitrogen used in the model. Numbering corresponds to positions of corresponding processes in table 3.1.	68
Figure 3.3	The first two rows show the time oscillations of the discharge current J and the voltage U , and the last row represents the corresponding phase portraits of these oscillations. The calculations are carried out at the values of the total applied voltage $U_t = 665$ V (a), 708 V (b), 725 V (c), and 827 V (d), and the semiconductor conductivity $\sigma_s = (2.6 \times 10^5 \Omega \text{ cm})^{-1}$. The two additional lines are the load line $U = U_t - R_s J$ and the current-voltage characteristics (CVC) of the gas discharge $U = U(J)$	72
Figure 3.4	Current-voltage characteristics (CVCs) curves derived from the "simple" and "extended" fluid models.	73
Figure 3.5	Profile of the ion density $n_i(x, t)$ as functions of time and spatial position at the total applied voltages $U_t = 665$ V and 827 V and the semiconductor conductivity $\sigma_s = (2.6 \times 10^5 \Omega \text{ cm})^{-1}$. These results correspond to (a) and (d) regimes in figure 3.3.	74
Figure 3.6	The period-doubling bifurcation diagram in the space of the discharge current density J and total applied voltage U_t . The regimes (a), (b), and (c) correspond to the regimes specified with the same labels in figure 3.3.	75

Figure 3.7	Lorenz map, corresponding to the regimes (a)–(d) with total applied voltages $U_t = 665, 708, 725, \text{ and } 827 \text{ V}$ from figure 3.3, depicts the data points of the $(n + 1)$ th maximum of the discharge current $J(t)$ versus its n th maximum. This figure also includes the maps derived from the LMEA model with the Maxwellian EEDF and the "simple fluid model".	76
Figure 3.8	The bifurcation diagram is defined in the parameter space of U_t and $1/R_s$. The curves depict the onset of regular (periodic) oscillations via Hopf bifurcation. The markers, representing the transition from periodic to chaotic oscillations, indicate the onset of irregular oscillations. The computations are performed using the "simple" fluid (LFA) and "extended" fluid (LMEA) models. The black asterisk marker * data points indicate the bifurcation curve obtained from the instability development time [7].	77
Figure 4.1	The schematic illustration of a Poisson stability.	86
Figure 4.2	The schematic illustration of a Delta unpredictability.	87
Figure 4.3	The CVC curve: $U = U(J)$ and the load line: $U = U_t - R_s J$ of the "extended fluid model". The intersection point (A, B) shows the equilibrium point at the regime $U_t = 20.23$ and $R_s = 30709$. The points (a), (b), (c), and (d) denote stationary solutions with $U_t = 23.2, 24.7, 25.3 \text{ and } 28.9$ at $R_s = 30709$. Note that the load lines for (a), (b), (c), and (d) are not depicted for clarity of demonstration.	92
Figure 4.4	Phase trajectories initiated at $(A_0, B_0) = (17.81, 8.22 \times 10^{-5})$ spiral inwards towards the stationary point $(A, B) = (17.92, 7.55 \times 10^{-5})$ in the dimensionless (U, J) plane.	93
Figure 4.5	The phase portraits include the trajectories of the system oscillations, the CVC curve $U = U(J)$ and the load line $U = U_t - R_s J$ for the corresponding regimes (a), (b), (c) and (d), indicated in figure 4.3.	94

Figure 4.6 Bifurcation diagram of the LMEA model with onsets of periodic and chaotic oscillations in the dimensionless parameter space. Points denoted by (a), (b), (c) and (d) denote the regimes with $U_t = 23.2$, 24.7, 25.3 and 28.9 at $R_s = 30709$ 95

Figure 4.7 Phase portrait of the drive system at $U_t = 28.9$ and $R_s = 30709$ in the dimensionless (U, J) plane. The points denoted by red markers represent the corresponding U and J values at the time values of the sequence of convergence $\{t_n\}$ 98

Figure 4.8 Phase portrait of the response system at $V_t = 28.9$ and $R_s = 30709$ in the dimensionless (V, J) plane. The points denoted by red markers represent the corresponding V and J values at the time values of the sequence of convergence $\{t_n\}$ 100

Figure 4.9 Phase portraits of the drive (blue) and response (red) systems at $V_t = 28.9$ and $R_s = 30709$ in the dimensionless $((U, V), J)$ plane. The points denoted by green and yellow markers represent the corresponding U, V and J values at common time values u_n of the sequence of convergence for the drive and response systems, respectively. 102

Figure 4.10 Implementation of the auxiliary system approach at $W_t = 28.9$, $R_s = 30709$ 105

Figure 5.1 Electron cross sections for (1) elastic, (2) direct ionization, (3) excitation, (4) excitation, and (5) stepwise ionization reactions in argon, included in the numerical model. The numbering on the curves corresponds to the indices of reactions in table 5.1. 112

- Figure 5.2 Spatial profiles of the time-averaged ion density, n_i , obtained from three formulations for the boundary ion flux Γ_i ("universal", "purely drift", "purely thermal"). Inset displays the same profiles in the immediate vicinity of the left boundary of the domain. Note that only half of the whole domain is plotted because the other side shows the same behavior due to symmetry. Discharge conditions are pressure $p = 3$ torr, the total applied voltage amplitude and frequency are $V_0 = 300$ V and $f = 0.85$ MHz, and the discharge gap is $d = 1$ cm. 115
- Figure 5.3 Spatial profiles of the time-averaged ion density, n_i , obtained from the models based on three formulations of ion boundary flux Γ_i ("universal", "purely drift", "purely thermal") with defined T_i approximations (indicated as case (i), (ii), and (iii)). Discharge conditions are the same as in figure 5.2. 116
- Figure 5.4 Spatial profiles of the time-averaged electron and ion temperatures T_e and T_i , derived from different ion temperature approximations (case (i), (ii), and (iii)). The discharge conditions are the same as in figure 5.2. 117
- Figure 5.5 The dependence of three dimensionless formulations of ion boundary $\tilde{\Gamma}_i$ ("universal", "purely drift", and "purely thermal") on the dimensionless drift velocity $U_{d,i}$ 119
- Figure 5.6 Spatial profiles of the time-averaged (a) electron n_e (solid lines) and ion n_i (dashed lines) number densities, and (b) of the ion flux density Γ_i (solid lines) and electron temperature T_e (dash-dot lines), obtained from the models implementing BCs from table 5.2. The vertical lines in panel (a) indicate the sheath layers. The conditions are the same as in figure 5.2. 120
- Figure 5.7 Spatial profiles of the time-averaged (a) ion n_i (red colored lines) and electron n_e (black colored lines) number densities, and (b) electron temperature T_e , obtained from the model of BC-3 with different values of γ . The conditions are the same as in figure 5.2. 122

Figure 5.8	Spatial profiles of the time-averaged, normalized (a) electron and (b) ion number densities n_e and n_i . The conditions are the same as in figure 5.2.	124
Figure 5.9	(a) Spatial profiles of the time-averaged ion density n_i , (b) enlarged form of the panel (a), and (c) the time-averaged profiles of the potential for different modeling approaches and boundary conditions. Discharge conditions are pressure of the argon gas $p = 0.1$ torr, the total applied voltage amplitude and frequency $V_0 = 270$ V and $f = 13.56$ MHz, the discharge gap $d = 2$ cm, and the background gas temperature $T_g = 0.03$ eV	125
Figure 6.1	Electron cross sections for the reactions in table 6.1: (1) elastic, (2) ionization, (3), (4), (5), and (6) elastic collisions in figure 6.1a; (8), (12) dissociative attachments, (13), (14), (15), (16), and (17) three body attachments in figure 6.1b, used in the hand-made Boltzmann solver model. Numbering corresponds to positions of corresponding processes in table 6.1	135
Figure 6.2	Mobility and diffusion coefficients for electron and electron energy transport as functions of mean electron energy. In 6.2a and 6.2b, the Einstein relation and the relation defined in [8,9] are also considered.	138
Figure 6.3	The rate coefficients vs. mean electron energy for different EEDFs derived from different Boltzmann solvers. The number indices of the rate coefficients correspond to the reaction numbers in table 6.1. .	139
Figure 6.4	The schematic illustration of a coaxial DBD tube. Geometrical features for three different coaxial discharge tube configurations are given in table 6.2.	141

Figure 6.5 Computed and measured RMS of the discharge current vs. RMS of the applied potential. The numerical and experimental results of three different discharge gaps, namely figure 6.5a, 6.5b, and 6.5c are 1.9 mm, 3.5 mm, and 5.5 mm, respectively. The configuration parameters are based on table 6.2. 145

Figure 6.6 Temporal evolution of discharge current and applied voltage in the regimes indicated by point *A* in the figures 6.5a, 6.5b, and 6.5c. The excitation frequency and applied voltage amplitudes are (a) $f = 106$ kHz and $V_0 = 7$ kV, (b) $f = 111$ kHz and $V_0 = 8.5$ kV, (c) $f = 104$ kHz and $V_0 = 9$ kV. 147



LIST OF ALGORITHMS

ALGORITHMS

Algorithm 1	Algorithm for Unpredictability	89
Algorithm 2	Algorithm for Delta (Complete) Synchronization of Chaos . . .	90



LIST OF ABBREVIATIONS

GDSS	Gas Discharge Semiconductor System
DC	Direct Current
DSC	Delta Synchronization of Chaos
DBD	Dielectric Barrier Discharge
PIC	Particle in Cell
MCC	Monte Carlo Collisions
GDP	Gas Discharge Plasmas
RF	Radio Frequency
BC	Boundary Condition
LMEA	Local Mean Energy Approximation
CCP	Capacitively Coupled Plasma
ICP	Inductively Coupled Plasma
CVC	Current Voltage Characteristics
LFA	Local Field Approximation
RMS	Root Mean Square
AC	Alternating Current
EEDF	Electron Energy Distribution Function
EDF	Energy Distribution Function
ITO	Indium Tin Oxide
CCD	Charge Coupled Device
NDR	Negative Differential Resistivity
CS	Complete Synchronization
PS	Phase Synchronization
LS	Lag Synchronization

GS	Generalized Synchronization
ILS	Intermittent Lag Synchronization
AS	Almost Synchronization
IPS	Imperfect Phase Synchronization
LTTA	Lorentz Two Term Approximation
SEE	Secondary Electron Emission
SEI	Specific Energy Input
BE	Boltzmann Equation
LTE	Local Thermodynamic Equilibrium
SFM	Simple Fluid Model
PECVD	Plasma Enhanced Chemical Vapor Deposition



CHAPTER 1

INTRODUCTION

1.1 Low-temperature plasma

Plasma, consisting of electrons, ions, and neutrals, are generated when an adequate amount of energy is injected into a gas, resulting in partial or complete ionization. This energy can be supplied through chemical reactions, electrical fields, or thermal ways. Within a plasma, this energy is stored among plasma species. Langmuir [10] introduced the term "plasma" to describe a dynamic combination of electrons, ions, and neutrals, which marks the beginning of plasma physics as a distinct field of investigation. Over time, plasma science has rapidly developed and become influential in various industrial domains, encompassing microelectronics, ozone production, modifying surface properties, pollution reduction, stimulating CO lasers, flat plasma panel technology, excimer lamps, converting methane, automotive innovation, packaging solutions, decontamination methods, and advancements in medical applications [11–15]. Plasmas' non-equilibrium nature is an important characteristic, with different species exhibiting distinct temperatures. This non-equilibrium state gives rise to a wide range of reaction pathways that are not achievable through conventional chemical methods.

Plasmas are non-equilibrium due to the various degrees of freedom observed in different plasma species. This is particularly evident in the case of electrons, which possess significantly higher kinetic energy than the excitation energy of heavier species within the plasma. The concept of temperature can be defined in plasma once thermal equilibrium is attained across all species. Under certain cases, individual constituents such as electrons or ions can have well-defined temperatures if thermalization oc-

curs among these constituents. Plasmas are generally classified into two temperature regimes: low and high, with the electron temperature serving as a reference. High-temperature plasmas, characterized by temperatures in the millions of Kelvin, are employed for the process of nuclear fusion involving hydrogen isotopes to yield energy and solar plasmas [16, 17]. However, in the scope of this thesis, our focus is on low-temperature gas discharge plasmas, where electron temperatures typically fall within the electronvolt range. This temperature range is sufficient to effectively initiate dissociation and ionization reactions, while the temperature of heavier species often tends to remain at room temperature.

The primary distinguishing feature of a low-temperature plasma is its ability to maintain a non-equilibrium state between electrons and heavier species, which forms the basis for a broad scope of applications. One critical application involves utilizing a reactive plasma initiated by energetic electrons in combination with a cold surface. This unique combination finds extensive use in a range of critical technologies:

Plasma-based surface treatment: Precise and controlled modifications of a cold surface is provided using a reactive plasma driven by hot electrons. This technology is widely employed in semiconductor manufacturing, materials processing, and surface engineering for tasks like surface cleaning, etching, deposition, and activation [18].

Plasma-enhanced material synthesis: Low-temperature plasmas enable precise and controlled growth of advanced materials by interacting highly energetic charged particles with a cold surface. This process facilitates the growth of thin films, nanoparticles, and nanostructures, which finds diverse applications in nanotechnology, catalysis, and energy storage, where the ability to engineer materials at the nanoscale contributes significant advantages [19].

Plasma medicine: The distinctive properties of low-temperature plasmas make them highly promising for a wide range of biomedical applications. Plasma jets produced by energetic electrons of gas discharge systems interacting with a cold surface are used for sterilization, wound healing, cancer treatment, and tissue engineering [11, 20]. The plasma species also exhibit antimicrobial properties and can select target cells, which makes them appealing for addressing healthcare challenges.

Plasma-enhanced combustion: Low-temperature plasmas are employed to enhance combustion processes by introducing reactive species into the combustion zone. This technique enables improved fuel ignition, more complete combustion, and reduced emissions. Therefore, this technology finds applications in various areas, such as energy production, automotive engines, and environmental remediation [21].

Gas conversion: Plasma offers an advanced technique for converting electrical energy into chemical energy. They serve as a highly efficient technique to transform hydrocarbons into valuable substances like acetylene. Moreover, plasmas can convert harmful chemicals into environmentally friendly end-products, making them essential in addressing environmental issues [22].

Light generation: Plasma lamps can produce intense light with heightened energy efficiency. It is worth noting that the energy-efficient lamps we use today are, in fact, plasma lamps. Their efficient light generation capabilities make them popular in various applications requiring intense illumination.

The selection of a suitable plasma type relies on the specific application and the desired pressure range. Generally, plasmas are heated using electric fields. These fields can be either a constant (DC), an alternating (AC), (RF), or even an electromagnetic wave at GHz frequencies [23]. Plasma composition varies depending on the electric field frequency because of the dissimilar resistance to the motion by electrons and ions. Moreover, magnetic fields can be employed to manipulate the charged particles within a plasma, enabling the confinement of electrons and ions along the field lines. Four distinct methods for generating plasma can be categorized according to the frequency of the electric fields employed in the process.

DC plasmas: DC plasmas involve the generation of a plasma between pair of electrodes, with a current moving in the system, maintained by the charged particles in the plasma. By utilizing magnetic fields, the plasma density can be improved by separating the charged particles from the plasma boundaries. An application of this is magnetron plasmas, where magnetic fields are employed to improve the density and confinement of the plasma [15].

AC plasma (kHz): Within AC plasmas, the electric field operates at a sufficiently low

frequency, enabling charged particles to obey the oscillations of the field. Electrons and ions actively participate in the current flow. This kind of plasmas are commonly employed in applications that require high power and large scale.

RF plasmas (MHz): The electric field in an RF plasma operates at considerably high radio wave frequencies. Within this frequency range, solely electrons are able to pursue the oscillating electric field while the ions remain relatively stationary. In RF plasmas, the current is predominantly driven by "displacement current" rather than "conduction current". This feature enables the formation of insulating layers on the electrodes, a critical element in treating insulating materials such as silicon oxide [15]. Plasma heating in RF plasmas occurs through Ohmic current losses within the plasma bulk. This phenomenon arises when the plasma bulk's electrons interact with the oscillating sheaths at the plasma boundary.

Wave heated plasmas (GHz): In a plasma operating within the GHz range, the electric field's frequency reaches a level that electrons are only directed by the electric field. However, the electromagnetic wave itself is unable to penetrate the plasma deeply because of inherent limitation of skin depth associated with electromagnetic waves in a conductive medium.

In contrast to low-pressure conditions, creating these four plasma categories is also possible at atmospheric pressure levels. As the pressure increases, the collision rate significantly increases, resulting in thermal equilibrium of plasmas. Consequently, the electron and ion temperatures in these plasmas become comparable.

1.2 Gas discharge plasma

Gas discharge plasma, the well-known variant of low-temperature plasma, has extensive practical applications in diverse scientific and technological fields. Electrical discharges are primarily utilized to produce plasmas. The process of initiating a discharge, also known as production, ignition, or breakdown, includes the creation of charge carriers within an insulating medium. This occurs under the influence of an external electric field. During breakdown, there is a rivalry between the generation and depletion of free-charge carriers, which happens when a threshold, characterized

by a breakdown voltage (V_b), is exceeded. Below this critical point, discharge initiation does not happen because the ionization process fails to counterbalance the loss of free-charges. The threshold value is influenced by factors such as the medium's chemical composition, density (or gas pressure), the frequency of the applied electric field, and additional factors such as electrode geometry and material.

The fundamental discharge types can be classified as follows:

DC discharges: In a DC discharge, the cathode and anode electrodes of the plasma device are connected to a DC power supply using a ballast resistor. When a breakdown occurs, an electron avalanche creates a negatively charged region (negative glow) near the cathode, effectively shielding the external electric field. The conditions for plasma formation are met in the adjacent region, known as the positive column. The discharge is sustained by a small ambipolar field that drives the current only in the radial direction. A plasma sheath appears near the anode to balance the varying mobilities of electrons and ions. The mentioned discharge form is commonly known as a "glow discharge", typically operates at currents ranging from a few milliamps to several hundred milliamps. In order to maintain the discharge, ion-induced secondary electrons continuously supply the necessary seed electrons at the cathode. However, at reduced current values, the contribution of secondary electrons may be inadequate, requiring volume ionization through background radiation to maintain the discharge. In this regime, one may observe dark, Townsend, or corona discharge phenomena [15]. The electrodes can become heated at high currents, producing electrons through thermionic emission. This process shows significant efficiency, requiring simply low voltage inputs to achieve currents within the ampere range, resulting in the formation of a thermal arc.

RF discharges: RF discharges include generating plasmas by applying oscillating electric fields at radio frequencies. This approach provides several advantages, including the dominance of displacement currents and the ability to use insulating electrodes. There are two main types of RF discharges:

CCP (Capacitively coupled plasma): In CCP, the plasma is contained between the electrodes, effectively forming a capacitor within the electrical configuration of the system [24]. Typically, one electrode is grounded, while an RF generator drives the

other electrode at several hundred volts. A matching unit, often comprised of an LC network, connects RF generator and electrode.

ICP (Inductively coupled plasma): In ICP, the plasma is subjected to electric fields through induction via an alternating magnetic field. An oscillating electric field is generated within the plasma volume by a coil tied to the RF generator, driven by the alternating magnetic field. This process leads to the formation of plasma current [15]. The plasma performs to a secondary coil within a transformer configuration. The primary external coil may consist of a few windings surrounding a quartz tube, which confines the plasma.

Microwave discharges: Microwave plasmas are produced at operating frequencies typically within the GHz range or wavelengths in centimeters. Achieving this requires using waveguides, which pose significant technical challenges compared to RF plasmas. It is important to note that the skin depth limits the extent to which microwave waves penetrate the plasma. This limitation necessitates the use of specific antenna configurations to efficiently couple power into larger plasma volumes. Such configurations include resonant reactor geometries or employing a series of microwave windows for power coupling. Another approach involves surface waves, which can propagate along the vessel's surface, maintaining plasma confinement over extended distances. This principle is utilized in surfatrons or plasmalines [15].

1.2.1 Gas discharge modeling

The development of gas discharge physics has demonstrated an extensive range of possibilities for designing gas discharge devices. This is mainly due to the remarkable ability to implement numerous diverse modes and regimes of gas discharge plasmas (GDP). Considerable processes, including interactions among charged, and neutral particles, their interconversion, interactions with electromagnetic fields, transport mechanisms, and additional factors [25], contribute to the inherent versatility. These sophisticated interactions collectively govern gas discharge plasmas' properties, making their behavior highly versatile and adaptable to various applications and conditions.

These processes involve the behavior of electrons, whose dynamics are governed by the "Electron Energy Distribution Function" (EEDF) shape. A multitude of elastic and inelastic collisions between electrons and atoms or molecules in a discharge medium influence the formation of the EEDF under specific conditions [25]. The significant role is also attributed to the transportation of electrons, which is driven by spatial gradients and external fields. The EEDF in glow discharge plasmas is consistently found in a non-Maxwellian state [25]. Consequently, while studying the characteristics of gas discharge plasmas, determining the EEDF becomes one of the fundamental challenges.

The interactions, including positive and negative atomic and molecular ions, play a vital role in the overall behavior of gas discharge plasmas. These interactions are responsible for regulating the loss of charges through several bulk recombination mechanisms [25]. Charge loss is further influenced by attachment and detachment processes. Moreover, the transport of ions is significantly influenced by ambipolar drift and diffusion [25], making a substantial contribution to the overall plasma dynamics. The collective impact of these complex ion-related processes directly affects the plasma's stability, density, and overall behavior.

The possible diverse modes and regimes in gas discharge plasmas give them a distinctive character. One effective approach is to use numerical modeling to acquire a comprehensive knowledge of these complex processes. Numerical simulations offer valuable insights into the behavior and dynamics of gas discharge plasmas, allowing researchers to explore a wide range of conditions, parameters, and interactions that might be challenging to study experimentally. Furthermore, scientists can further enhance their understanding of plasma characteristics and optimize the design and operation of gas discharge devices for diverse applications.

1.2.1.1 Subproblems of GDP modeling

As suggested in [25], the numerical analysis of gas discharge plasmas (GDP) is a complex and multifaceted problem, which involves divisions of the problem into several interconnected subproblems [25]. The solution to each subproblem depends on the insights and knowledge gained from related subproblems. This self-consistent na-

ture of numerical analysis emphasizes the sophisticated interdependencies and complexities in gas discharge plasma phenomena. As a result, researchers must adopt a comprehensive perspective to approach the numerical modeling of GDP. Indeed, the solutions to individual subproblems must be compatible and coherent within the larger context of the plasma system. It makes accurate and reliable numerical simulations of gas discharge plasmas achievable. Subproblems can be categorized in the following manner:

Calculation of electromagnetic field: In gas discharge plasmas, the electromagnetic field has a dual purpose. Firstly, it shapes the "Electron Energy Distribution Function" (EEDF) and generates ionizing electrons through Joule heating, thereby maintaining the discharge [25]. Furthermore, within an inhomogeneous discharge plasma, electromagnetic fields give rise to the movement of electrons and ions, establishing quasi-neutrality [25]. In DC discharges, the electric field fulfills both. However, the high-frequency electric field in RF discharges primarily induces Joule heating, whereas the quasi-stationary potential field maintains particle flows and quasi-neutrality [25]. The behavior of electromagnetic fields within the plasma is described by the Maxwell equations. The source terms include the charges and currents originating from external electrodes, antennas, and the plasma medium.

Calculation of EEDF: Once the electromagnetic fields are characterized, solving the Boltzmann equation determines the "Electron Energy Distribution Function" (EEDF). Indeed, the balance between the energy gained by electrons through Joule heating (energy diffusion) and the energy lost due to collisions influences the EEDF [25]. Momentum relaxation primarily occurs through elastic collisions with neutral particles, whereas energy loss is typically induced by inelastic collisions involving the excitation of atoms and molecules [25]. Consequently, the resultant EEDF, shaped by electron collisions, generally deviates seriously from the Maxwell distribution. The "tail" of the distribution generally shows an exponential decline with energy, and its shape is considerably susceptible to the probabilities of distinct elementary processes. This sensitivity is significant for processes with a high energy threshold. Electron impact ionization is an example of such a process, and it plays a vital role in sustaining the gas discharge by creating additional free electrons. A precise comprehension of the exact structure of the EEDF is essential for precisely determining

the rate constants of these processes. Using the standard Maxwell distribution can result in uncontrollable severe errors. Therefore, accurately characterizing the EEDF is crucial for understanding and predicting the behavior of gas discharge plasmas.

Calculation of the plasma concentration profile: Accurately determining charged particle concentrations in gas discharge plasmas requires solving a system of transport equations. These equations consider the production and destruction of charged particles directly linked to corresponding fluxes influenced by transportation (drift and diffusion) in a quasistationary field. Thus, knowledge of the EEDF is crucial as it determines rates of ionization as well as various plasma-chemical reactions, ultimately determining charged particles' birth and death [25]. Additionally, EEDF provides an understanding of the characteristics of electric and magnetic field profiles within the plasma, which is crucial for accurate analysis and interpretation of the plasma behavior.

As stated in [25], it is essential to recognize that none of the subproblems can be addressed in isolation because they are inherently interconnected. The Boltzmann equation alone cannot fully determine the EEDF due to the self-consistency of the electromagnetic field. The field is influenced by external elements like electrodes, antennas, and coils. Additionally, the field is affected by the charges and currents carried by the charged plasma particles. One must know these charges and current profiles to accurately determine the fields governed by the transport equations. However, solving these transport equations necessitates prior knowledge of the fields. Without considering the self-consistency, any approach to specific analytical or numerical problems, such as finding the EEDF under specified fields or deducing fields from a provided plasma configuration, leads to limited relevance to realistic plasma behavior. The interconnected nature of these aspects requires a holistic and self-consistent approach to analyze gas discharge plasma meaningfully.

1.2.1.2 Basic modeling approaches

After carefully considering the abovementioned factors, it becomes clear that a numerical solution to this self-consistent problem is unavoidable. The inherent complexity of contemporary plasma technologies further reinforces the need for exten-

sive use of computer simulation methods to study gas discharge plasmas (GDPs). Gas discharge modeling approaches can be classified into three main categories: fluid models, kinetic and particle models, and hybrid models [25]. Fluid models offer both advantages and drawbacks. On the positive side, these models are characterized by their relatively simple nature and computational efficiency. These models derive from the moments of the kinetic Boltzmann equation. As a result, the fluid model equations encompass continuity, momentum, and energy balance equations. These equations are associated with the Maxwell equations for the self-consistent fields [25–28].

When deriving fluid equations for gas discharge plasma, it is assumed that all plasma species' distribution functions closely approximate the Maxwellian distribution [25]. This assumption is subject to the criterion that the processes' characteristic temporal and spatial scales and those of the electric and magnetic fields surpass the temporal and spatial scales required for the distribution functions to relax towards the Maxwellian state [25, 26]. For elastic collisions involving particles of comparable masses (e.g., neutral, excited particles, and ions), the relaxation process happens rapidly, often within a single collision time. Consequently, Maxwellian distributions of all species, characterized by a single common temperature, are established in such cases. However, an exception arises with electrons, whose Energy Distribution Function (EDF) often remains in a non-equilibrium state due to their low mass.

As suggested in [25], two primary adaptations of the fluid model are commonly employed to accommodate the non-Maxwellian nature of the "Electron Energy Distribution Function" (EEDF) [25]. The first is the "simple fluid model", which assumes that the EEDF is shaped by local values of specific parameters, mainly the electric and magnetic fields, observed at a specific point within space and time [25]. This approach is also known as the "Local Field Approximation" (LFA), where rate coefficients for reactions initiated by electrons and transport coefficients of electrons are defined as dependent on the local electric field. However, in collisions between electrons and heavy particles, electron energy relaxation occurs much more slowly than momentum relaxation, and it takes place over a longer distance. As a result, the LFA is unsuitable for describing processes involving electrons in spatially inhomogeneous fields, such as those found in the near-electrode regions of discharges, and it can lead to substantial errors in the modeling and understanding of electron behavior in gas

discharge.

To enhance the fluid model by addressing the limitations of the "Local Field Approximation" (LFA) and incorporating, to some extent, non-local electron transport, an alternative approach called "extended fluid model" (also known as "the Local Mean Energy Approximation" (LMEA)) was proposed in [29]. Numerous studies have extensively utilized this approach in various modifications [8, 25, 29–36]. In the "extended fluid model", the transport and rate coefficients for electrons are defined as electron temperature functions rather than solely determined by the local electric field. The solution of the electron Boltzmann equation accomplishes this, considering the behavior and interactions of electrons in the plasma. Moreover, the energy balance equation for electrons considering both volume processes and heat transfer through heat conduction is simultaneously solved to determine the spatial distribution of the electron temperature.

The "extended fluid model" provides a more comprehensive and accurate representation of electron behavior, incorporating electron temperature as the main factor governing electron transport and reactions, particularly in regions where significant variations occur in the electric field. This makes the model particularly valuable for studying plasma phenomena in spatially non-uniform fields, such as those in the vicinity of electrode regions in gas discharges. The approach is crucial for various plasma applications and technologies, enabling a more accurate analysis and understanding of the complex relationship between electron dynamics and the non-uniform plasma environment.

An exact and comprehensive definition of electron behavior can be achieved by solving the Boltzmann equation [25, 37–41]. However, this mathematical approach becomes highly complicated, especially when dealing with multi-dimensional scenarios. Another method, referred to as PIC/MCC (Particle in cell/Monte Carlo collision) [25, 42, 43] is characterized by its computational demands and limited applicability in gas discharge modeling because this couples Monte Carlo simulations for electron and ion dynamics with the Poisson equation for the self-consistent electric field.

Additionally, the accuracy of detailed direct-method computations in plasma pro-

cesses is limited by the low reliability of cross-section data, especially regarding the angular dependence of scattering [25]. In self-consistent problems, the model's reliability is determined by its least accurately known part, often called the "bottleneck" (In modeling gas discharge systems, the focus is typically on the dynamics of electrons). Therefore, even if certain aspects of the numerical model are based on developed and reliable methods, the enhancement in result accuracy remains limited if other components are inadequately described. This highlights the importance of addressing all aspects of the plasma model with care and accuracy to ensure a reliable and meaningful analysis of gas discharge plasmas.

Considering the challenges in gas discharge modeling, aiming for equilibrium in the accuracy of different components within the numerical model is a sensible strategy. Utilizing equally precise methods for various aspects of the model guarantees that the limitations of any single part do not provide the overall reliability and precision of the results [25]. This balanced approach becomes crucial in obtaining accurate simulations and understanding the complex behavior of plasmas in practical gas discharge modeling situations. By carefully addressing all aspects of the model with an equal focus on accuracy, the potential to make more informed decisions and develop superior plasma-based technologies is heightened.

Hybrid models balance between fluid models that are computationally efficient yet approximative and particle and kinetic models that offer accuracy at the cost of time-consuming computations [25, 30, 42, 44–49]. These methods involve the division of the electron ensemble into two distinct behaving groups: "fast" and "slow" electrons. Energized electrons exceeding the threshold of inelastic collisions are categorized as "fast" electrons, while all other electrons with lower energies belong to the "slow" electron group. Slow electrons, characterized by relatively low average energies (temperatures not exceeding several electronvolts), play a vital role in maintaining equilibrium between density and temperature across electron ensemble and mainly account for the electron current. The EEDF closely approximates the standard Maxwellian distribution, making them well-suited for using the fluid model with a "drift-diffusion" approximation of flux density. Similarly, the fluid model also describes ions and excited (metastable) particles. On the other hand, the behavior of fast electrons, which play a crucial role in ionization and excitation processes, is typically

estimated by applying the Monte Carlo method [25]. The ionization and excitation sources derived from Monte Carlo simulations are then incorporated into the particle balance equations for slow electrons and ions, thus ensuring a self-consistent system. Due to the relatively low concentration of fast electrons compared to slow electrons, the effect of fast electrons on Poisson equation can be ignored. Hybrid models have become extensively utilized in numerical studies and are now considered a viable method for effectively modeling gas discharge plasmas. In summary, hybrid models provide a practical and accurate approach by combining the fluid modeling of ions and slow electrons with particle or kinetic definitions of fast electrons, offering a powerful tool for gas discharge modeling and understanding their complex behavior.

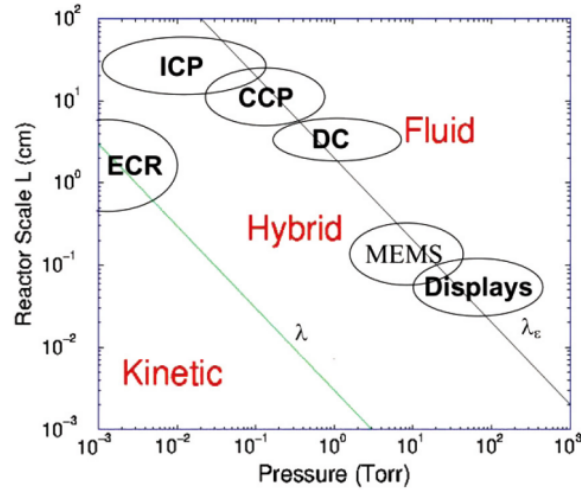


Figure 1.1: The graphical representation of the parameter ranges appropriate for applying kinetic, hybrid, and fluid models can be found in [1].

Figure 1.1 is a schematic diagram showing different parts in parameter space, defined by plasma size (L) and pressure (p), where kinetic, hybrid, and fluid models are applicable. As the figure shows, in regions where plasma size (L) is less than the energy relaxation length (λ_e), significant non-local kinetic effects arise. For instance, in the region where the product pL is less than the energy relaxation length (λ_e), the fluid model becomes unsuitable for describing the electronic component. It is important to note that, based on the analysis in [50], plasmas of atomic gases show non-local behavior in the $pL < 10 \text{ cmTorr}$ range. In such situations, the hybrid and kinetic models are more appropriate for providing accurate descriptions of the plasma behav-

ior, considering the influence of non-local kinetic effects on the system's dynamics. The diagram in figure 1.1 serves as a valuable guide, assisting in choosing appropriate modeling approaches based on the specific plasma conditions characterized by the system's characteristic size (L) and gas pressure (p). It emphasizes incorporating non-local kinetic effects in specific parameter regimes to achieve more accurate and insightful plasma simulations and analyses.

1.2.2 Pattern formation and self-organization in planar gas discharges

Investigating pattern formation in nonlinear systems represents one of the most challenging fields of modern science. Pattern formation includes a series of events spontaneously occurring in a self-organized manner, which leads to regarding of pattern formation and self-organization as a unified phenomenon in the scientific literature [51]. This phenomenon is observable in various natural and technological contexts involving biological, chemical, and physical systems [52].

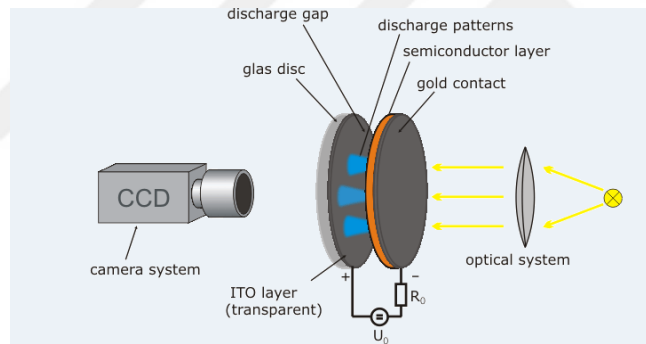


Figure 1.2: The schematic of a planar DC gas discharge system with high ohmic cathode [2]

Figure 1.2 shows a schematic representation of plasma planar systems with a high ohmic barrier, known as the gas discharge-semiconductor system (GDSS). Various research groups have conducted extensive investigations on this GDSS, notably the Purwins group in Munster, Germany, and the Astrov group in St. Petersburg, Russia. The GDSS exhibits a rich range of patterns in its behavior, making it an interesting and complex system for study. The observed patterns in this system demonstrate various forms of self-organization and pattern formation, providing valuable insights into these

phenomena in plasma and other nonlinear systems. The research conducted on pattern formation in the GDSS and similar systems contribute significantly to the advancement of our knowledge in complex systems and have implications for fundamental research and practical applications. In contrast to usual long discharge tubes, this experimental setup has a unique configuration with a small aspect ratio, which means that the width is much greater than the height, along with a high ohmic semiconductor cathode.

The discharge gap is designed in a layered structure resembling a sandwich, with a planar electrode at the anode side composed of a transparent indium tin oxide (ITO) layer coated onto a glass disc. The transparency of the anode ensures that the camera system records the patterns without any obstruction. Moreover, the planar electrode at the cathode side is a semiconductor layer, such as GaAs, attached with gold contact. Specifically, in the experimental study presented in [53], both the gas-discharge part and the semiconductor part (where the semiconductor serves as the cathode of the gas discharge) were found to be capable of triggering instabilities and providing a unique characteristic contributing to this system's diverse and rich observed patterns. To facilitate visualization, an optical system is specifically designed to enable camera-based observations. A DC voltage source powers the entire system, and the pattern formation processes are recorded using a CCD (charge-coupled device) camera system via the transparent electrode. This setup allows researchers to observe and study the complex patterns formed in the GDSS. Overall, this experimental setup presents a valuable platform for studying pattern formation and self-organization in planar plasma systems with high ohmic barriers.

1.2.2.1 Patterns in GDSS

When the GDSS is supplied with an adequate current, it forms spatiotemporal patterns in the lateral direction, which are observed in spatially and temporally inhomogeneous variations in the direction of the current flow in the discharge. These patterns can be visualized through the luminescence radiation emitted from the discharge gap. One can establish a direct relationship with the corresponding current density distribution in the gas by examining the "luminance density distribution" (the

spatial arrangement and variation of brightness across a surface) observed in the discharge. Therefore, the luminescence radiation provides valuable information about the spatial distribution and variations in the current density, enabling the observation and analysis of the spatiotemporal patterns in the GDSS.

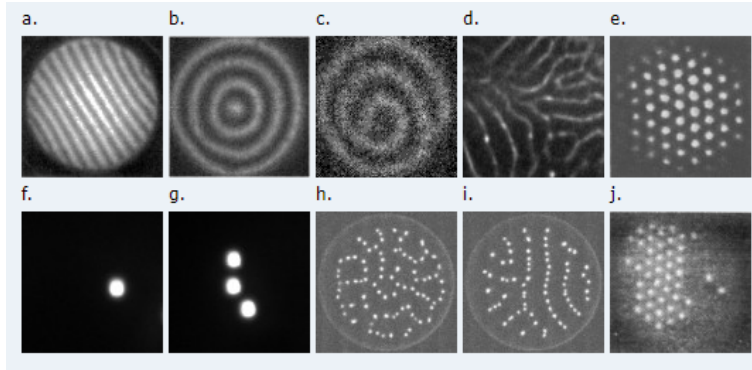


Figure 1.3: Some of the experimentally observed patterns [2]

Some of the experimentally observed structures from reference [2] are illustrated in figure 1.3. The displayed different patterns are as follows: a) Stripe pattern, b) Target pattern, c) Spiral pattern, d) Labyrinth pattern, e) Hexagonal pattern. Figure 1.3 also includes a structure (f) characterized by a localized bright spot. This particular pattern is relatively unaffected or undisturbed by fluctuations in the system. Due to this stability, the pattern exhibits particle-like properties and is referred to as "dissipative solitons" [54]. Dissipative solitons manifest as isolated structures and come together and interact with each other, showing particle-like properties to construct complex structures. Examples include in figure 1.3 molecule formation (g), nets (h), chains (i), and the combination of different phases in one domain (j). These complex patterns emphasize the self-organization and dynamic behavior of the GDSSs.

1.2.2.2 Temporal oscillations in GDSS

Apart from the structures mentioned above, GDSSs can display a wide range of temporal behaviors, such as homogeneous stationary and oscillating modes, as well as chaotic behavior [55, 56]. Experimental investigations have identified the presence of homogeneous oscillations in a GDSS containing nitrogen gas [57]. Representative numerical oscillations of homogeneous discharge current and voltage can be seen in

figure 1.4 [3]. Furthermore, a later study [58] shows the existence of chaotic oscillations of the same system used in Chapters 3 and 4 of this thesis. These diverse behaviors highlight the complex and dynamic nature that can emerge in GDSS.

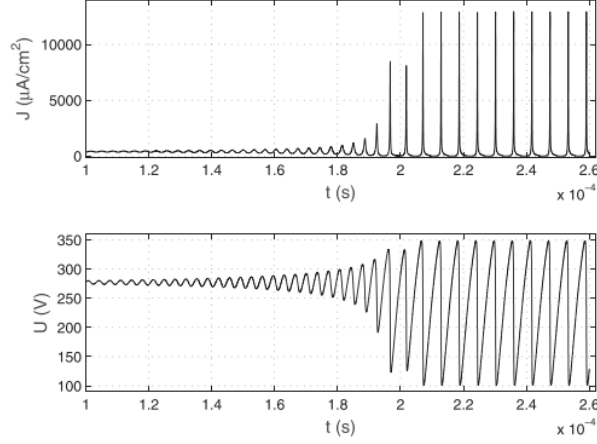


Figure 1.4: The plasma variables develop into temporal homogeneous oscillations after transient solutions [3].

In the figure 1.4 and the chaotic oscillations demonstrated in Chapters 3 and 4 of this thesis, the main emphasis is on investigating temporal oscillations of physical parameters, such as current and voltage, rather than pattern formation. Indeed, the transverse directions of the GDSS are assumed to be spatially homogeneous, resulting in only the spatial variations occurring perpendicular to the electrodes. This is significant for this thesis because the developed models are one-dimensional, and only temporal oscillations of plasma parameters are derivable. Our focus will be the chaotic oscillations of these parameters as a function of time. Therefore, understanding the fundamentals of chaos phenomena in plasma systems is essential to find these oscillations meaningful.

1.2.2.3 Chaos phenomenon in GDSS

In recent decades, there has been a significant increase in the exploration of chaos in laboratory plasmas. Chaotic behavior in dynamical systems can be categorized into two main types: temporal (or low-dimensional) chaos, which is often associated with pure oscillatory motion, and spatiotemporal chaos (or weakly developed turbulence),

characterized by wave-like patterns [59, 60]. In plasma devices, chaos, and turbulence are generally considered undesired features [60], particularly in applications that require crucial cycle-to-cycle reproducibility. Hence, understanding and controlling chaos in gas-discharge plasmas becomes essential to improve the reliability and performance of gas-discharge systems. Various methods based on discrete and continuous feedback and synchronization have been developed to achieve control over complex behavior in nonlinear dynamic systems, including in plasma devices [60]. These control techniques play a crucial role in managing instabilities and chaos, providing opportunities to enhance gas-discharge plasmas' stability and overall performance in real-world applications.

A well-known phenomenon in gas discharge systems is the emergence of instability within the homogeneous stationary state of an electric current-carrying medium. This occurs when the differential resistivity, defined by the relationship between the applied voltage (U) and discharge current (I), becomes negative [53, 61]. This phenomenon occurs when an increase in current is accompanied by a decrease in voltage across the discharge. Consequently, the current-voltage characteristic (CVC) shows a branch with a negative slope, dU/dI , indicating the presence of negative differential resistivity (NDR) within the system. This situation corresponds to the sub-normal glow regime between the Townsend discharge and the normal glow discharge phases in the classical glow discharge context. The negative differential resistivity (NDR) in this regime has significant importance in the emergence of instabilities and oscillations, which are frequently observed in gas discharges [31, 62–65]. These instabilities can lead to the formation of various patterns of structures within the gas discharge, emphasizing the complex and dynamic nature of the plasma behavior in this particular regime.

The instabilities induced by negative differential resistivity (NDR) in the gas discharge can disrupt spatial homogeneity in the transverse direction, particularly at higher current densities. The lack of homogeneity behavior leads to the formation of structures, which are spatially localized regions. These structures can be both stationary and dynamic, which play a crucial role in shaping the behavior of the gas discharge. The complex interaction between these structures is also responsible for the observed temporally and spatially chaotic current fluctuations in the GDSS [66]. The

presence of chaotic current oscillations adds further complexity and unpredictability to the system's behavior. As a result, investigating chaos in GDSS is crucial for effectively understanding and controlling these plasma devices' dynamics. Understanding the role of these instabilities is also essential in optimizing and improving the performance of gas-discharge systems used in various practical applications.

In this thesis, in chapters 3 and 4, the analysis primarily focuses on subnormal oscillations that occur in a spatially homogeneous mode. The models are one-dimensional, specifically limited to the direction perpendicular to the gas discharge and semiconductor layers. Subnormal oscillations in glow discharges have been extensively studied through experimental and theoretical approaches in numerous publications [31, 63–65]. However, the underlying chaotic behavior in these systems and the mechanisms responsible for the transition from regular periodicity to a chaotic regime have not yet been entirely understood. Despite previous research on subnormal oscillations, there is still a need to comprehensively understand the chaotic dynamics in these GDSSs to advance our knowledge and control over such behaviors.

One well-accepted pathway to temporal chaos is period-doubling bifurcation [55,67], observed in various gas discharge devices, including those with high-ohmic semiconductor electrodes [55, 68, 69]. The onset of period-doubling bifurcations was experimentally detected under conditions similar to those examined in this thesis. Previous numerical investigations have also identified and studied period-doubling bifurcations in GDSS [70,71]. These results stress the significance of period-doubling bifurcations as a mechanism leading to temporal chaos in GDSS.

The Lorenz map and bifurcation diagram were obtained in the extensive investigation of GDSS dynamics [71], illustrating the transition from a periodic oscillatory regime to a fully chaotic state via period-doubling bifurcations. The chaos transition observed in chapter 3 of this thesis exhibited a perfect period-doubling cascade, resembling simple nonlinear mappings like the logistic map [72].

In this thesis, the investigation of GDSS in nitrogen begins in Chapter 3. The focus is on understanding the occurrence of chaotic oscillations within the system. A one-dimensional fluid model based on the "Local Mean Energy Approximation" (LMEA) approach is utilized for this purpose. Moving on to Chapter 4, chaos synchronization

between master-slave chaotic systems of GDSSs is explored. Similar fluid models with corresponding chaotic regimes are employed for this study, aiming to understand the dynamics of chaos synchronization in GDSSs.

1.2.3 Synchronization in GDSS

1.2.3.1 Synchronization in chaotic systems

The word "synchronization" has its origins in a Greek root, which conveys the meaning of "to share the common time." This original meaning of synchronization has persisted throughout history and is still evident in everyday usage, referring to the agreement or correlation in time between various processes [73]. The investigation of synchronization phenomena in the evolution of dynamic systems has a rich history of investigation, dating back to the early days of physics. In the 17th century, Christiaan Huygens discovered that two weakly coupled pendulum clocks hanging on the same beam could synchronize in phase [74]. Other instances of synchronization were observed in various natural phenomena, such as the synchronized lighting of fireflies or the peculiar behavior of adjacent organ pipes, which can almost silence one another or produce sound in perfect unison. These early observations were a foundation for further investigation and understanding of synchronization in various systems across scientific disciplines.

In recent times, the study of synchronization has extended to chaotic systems. In this context, the emergence of collective dynamics, where multiple chaotic systems synchronize their behavior, is not a trivial process. Chaotic systems are known for their sensitivity to initial conditions, meaning that even a tiny difference in the starting states of two identical systems will lead to exponentially diverging trajectories over time. As a result, chaotic systems inherently resist synchronization, as their states would grow increasingly unsynchronized due to the exponential growth of differences. This inherent sensitivity to initial conditions has a significant challenge when attempting to achieve synchronization in chaotic systems. It makes the study of chaotic synchronization an exciting and complex area of research.

The sensitivity of initial conditions presents a practical challenge since experimental

initial conditions are never known with perfect precision. Therefore, achieving collective synchronized behavior in coupled chaotic systems becomes highly relevant and of great interest. It involves finding methods to couple and control chaotic systems to synchronize their motions, resulting in coherent and ordered dynamics. This pursuit of synchronization in chaotic systems has opened up new avenues of research and has significant implications in diverse fields, including physics, biology, engineering, and information processing.

As a preliminary definition, synchronization of chaos refers to a phenomenon where two or more chaotic systems, which may be equivalent or nonequivalent, adjust a specific property of their motion to exhibit a common behavior. This synchronization can arise due to coupling or forcing between the systems. The degree of synchronization can vary, from the complete agreement of trajectories to the locking of phases, where specific aspects of the chaotic systems become coordinated in time. Chaos synchronization has broad implications and can manifest in various forms, depending on the nature of the coupled chaotic systems and the type of coupling or forcing applied.

The first important distinction in achieving synchronized states lies in the difference between unidirectional and bidirectional coupling configurations. In the case of unidirectional coupling, two subsystems form a global system where one subsystem serves as the "drive" or "master" and the other functions as the "response" or "slave." The response system follows a function of the dynamics of the drive system, which acts as an external chaotic driver for the responder. This leads to the occurrence of external synchronization. Examples include communication with chaos, where information is transmitted through the chaotic signal. In contrast, bidirectional coupling involves both subsystems being coupled to each other. The coupling factor influences their rhythms, leading to a mutual synchronization behavior. This situation is frequently observed in fields such as physiology, where the cardiac and respiratory systems can exhibit mutual synchronization, or in nonlinear optics, where coupled laser systems with feedback can also demonstrate bidirectional synchronization. It is essential to emphasize that these two processes are fundamentally different philosophically and in their dynamics. There is currently no known way to reduce one process to another or formally establish a direct connection between these two cases. Their distinct nature makes them interesting and significant in different applications and scientific

contexts [75].

Various synchronization states have been extensively studied over the past decade in the realm of coupled chaotic elements. These include:

Complete (Identical) Synchronization (CS): In this state, two or more chaotic systems evolve in perfect agreement, meaning their trajectories coincide entirely. CS has been thoroughly investigated in the literature [76, 77].

Phase Synchronization (PS): PS occurs when the phases of coupled chaotic systems become synchronized, while the amplitudes may still differ. This type of synchronization has been a subject of significant interest [78, 79].

Lag Synchronization (LS): LS is characterized by synchronized chaotic trajectories with a constant time lag between the systems. It has been studied in various systems [80].

Generalized Synchronization (GS): GS goes beyond PS and LS, where the dynamics of one system can be mapped to another, allowing for a more flexible synchronization behavior [81, 82].

Intermittent Lag Synchronization (ILS): In ILS, the synchronization between chaotic systems is intermittent, meaning they alternate between synchronized and desynchronized [80, 83].

Imperfect Phase Synchronization (IPS): IPS refers to a state where the phases of the coupled systems are close to being synchronized but not precisely in phase with each other [84].

Almost Synchronization (AS): AS is a state where the coupled systems are not entirely synchronized but exhibit synchronization within a finite region in phase space [85].

CS (Complete Synchronization) was the first form of synchronization discovered and is considered the simplest in chaotic systems. It involves perfectly aligning the chaotic trajectories of two systems using coupling signals, ensuring they remain synchronized over time. The mechanism for CS was initially observed in unidirectionally coupled

identical chaotic systems, where the conditional Lyapunov exponents of the subsystem to be synchronized are all negative [77]. In this state, the coupled systems evolve in complete harmony, maintaining identical behavior and motion throughout their dynamics. CS is a fundamental synchronization state in the study of chaotic systems and has been extensively explored in various applications and disciplines. GS (Generalized Synchronization) surpasses CS and other more straightforward synchronization states by enabling the coupling of entirely different systems and linking them. In GS, the output of one system is related to a specified function of the output of the other system [81, 82]. An intermediate regime called Phase Synchronization (PS) can be achieved in coupled nonidentical oscillatory or rotatory systems. In PS, the phases of the coupled systems become locked or synchronized, while the amplitudes may still exhibit a weak correlation [78]. This phenomenon has been studied and characterized in the context of coupled oscillators, particularly the Rossler system [78]. Lag Synchronization (LS) serves as an intermediate state between Phase Synchronization (PS) and Complete Synchronization (CS). In LS, the outputs of two coupled systems become asymptotically bounded, meaning the difference between the output of one system at a time t and the output of the other system shifted in time by a lag time τ_{lag} remains limited [80]. While LS involves the locking of phases and amplitudes similar to PS and CS, it differs by the presence of a time lag between the synchronized outputs [80]. Intermittent Lag Synchronization (ILS) is a synchronization state where two coupled systems predominantly show Lag Synchronization (LS), but intermittent bursts of local nonsynchronous behavior can occur [80, 83]. These bursts of nonsynchronous behavior correspond to specific regions in the attractor where the local Lyapunov exponent along a globally contracting direction becomes positive [80, 83]. Analogously, Imperfect Phase Synchronization (IPS) refers to a situation where phase slips occur within a Phase Synchronization (PS) regime [84]. In IPS, the phases of the coupled systems remain close to being synchronized, but occasional phase slips or phase jumps may occur, causing temporary desynchronization before resuming synchronization. On the other hand, Almost Synchronization (AS) is a state where the difference between a subset of variables of one system and the corresponding subset of variables of the other system remains asymptotically bounded [85].

1.2.3.2 Chaos synchronization in GDSS

In the past, generalized synchronization was one of the most effective methods to detect synchronization in complex systems. Researchers utilize various instruments and methodologies to detect generalized synchronization in complex systems like GDSS. Three primary approaches are commonly employed for this purpose:

Analysis of Conditional Lyapunov Exponents: This method involves investigating the Lyapunov exponents, which quantify the sensitivity to initial conditions in a dynamical system. For generalized synchronization, the conditional Lyapunov exponents of coupled systems are analyzed to assess the degree of synchronization between them [81].

Auxiliary System Approach: The auxiliary system approach is another powerful method to detect generalized synchronization. It involves developing an auxiliary system that interacts with the original system and then examining the relationship between the auxiliary and original system variables to identify synchronization patterns [86].

Statistical Estimations of Predictability: This method analyzes the predictability properties of coupled systems. This approach employs mutual information, cross-correlation, and other statistical quantities to assess the degree of synchronization between the systems [82,87,88].

In some instances, applying the generalized synchronization method may have proven ineffective in detecting synchronization in the specific circumstances of the systems under investigation. As a result, researchers have sought alternative approaches to identify and analyze synchronization in these systems. These alternative approaches may involve using different synchronization measures, statistical estimations, or developing novel methods specifically tailored to the unique dynamics of the system. In the case of GDSS, the absence of generalized synchronization has been demonstrated in a study cited as Ref. [89], using the auxiliary system approach. By applying this method, it was concluded that generalized synchronization is not present in the GDSS system under investigation. However, it is essential to explore various techniques and adapt them to the specific characteristics of the studied system. In other words, the

absence of generalized synchronization does not rule out the possibility of other synchronization forms or cooperative behaviors in the system.

In [4], we propose a novel method to demonstrate chaos synchronization in the GDSS, called "Delta Synchronization of Chaos" (DSC), which depends on analyzing "unpredictability" in chaotic systems. By introducing this unique perspective, we gain new insights into the intricate dynamics and behaviors that emerge when chaotic systems are coupled. Through the examination of the unpredictability aspects of the coupled chaotic systems, we gain a deeper understanding of their synchronization patterns and the underlying mechanisms driving this synchronization. This new method contributes to the ongoing research efforts in chaos synchronization [4, 90, 91].

1.2.3.3 "Unpredictability" analysis and DSC method

The "unpredictability" analysis, extensively explored in references [92–95], is vital in confirming a system's chaotic behavior, particularly Poincaré chaos. This type of analysis focuses on trajectories that first converge to their initial points due to the Poisson stability phenomenon and then deviate from these initial paths after a specific time. These successive convergence and separation events are strong evidence of the system's unpredictable and essentially chaotic nature, commonly referred to as Poincaré chaos. One notable advantage of the "unpredictability" method lies in its ability to determine the values of convergence and separation times, referred to as the "sequence of convergence" and "sequence of separation," respectively. The time moments corresponding to Poisson stability represent the moments, at which the phase trajectory converges back to the initial point, while orbits originating from points within the unpredictable trajectory at times t_n and 0 diverge at specific moments s_n , thus establishing the time values for the "sequence of separation." Consequently, the "unpredictability" analysis is a valuable tool for confirming chaotic motions in dynamic systems and offers profound insights into the intricate behaviors inherent in chaotic systems. In the context of numerical "unpredictability" analysis of GDSSs, the process involves recording the moments of convergence to the initial point and the moments of separation from the initial orbit sections, thereby capturing the dynamics of "unpredictability" within the system. This analysis was carried out in the

study [4], where the chaotic regime of the GDSS system in nitrogen was examined using the "simple fluid model" (LFA) of Chapter 3 of this thesis. As depicted in Figure 1.5, the corresponding points are marked in red, representing the moments when the phase points converge to the initial point. As the simulation progresses, the radius of the disk, defined by these red phase points, progressively shrinks. In other words, each cycle of phase points moves closer to its initial position as the simulation time advances.

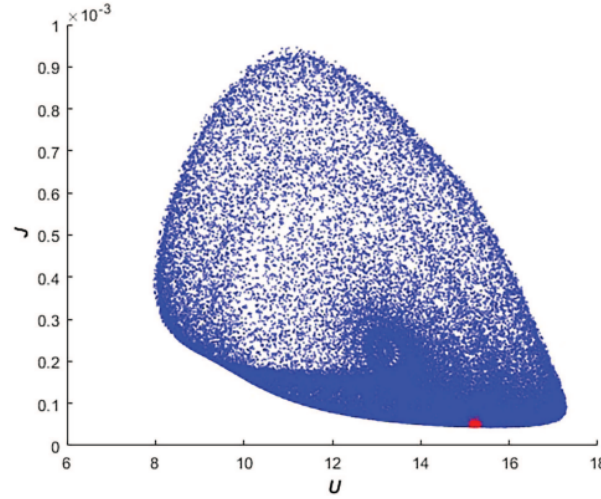


Figure 1.5: The phase portrait of the drive system of a GDSS [4]. Red marked points correspond to the instants t_n when the trajectory of the phase point passes through the closest point to the initial point where calculations have started.

Based on the "unpredictability" analysis, a new technique, the "Delta Synchronization of Chaos" (DSC) method, has been introduced and comprehensively explained in [4]. For successful implementation of the method, it is essential that the coupled systems (drive and response) must separately show unpredictable solutions and that the simulation time is sufficiently long. The DSC analysis investigates the occurrence of common finite convergence and separation sequences between the coupled drive and response systems. The presence of these shared sequences confirms the synchronization of chaos.

Suppose the drive and response systems, where the latter is developed using data obtained from the former, possess unpredictable trajectories. The objective is to observe the synchronization between these two dynamic systems. The DSC entails the

existence of shared sequences of moments for convergence and separation in unpredictable orbits. As the orbits belong to two different models, one cannot expect the emergence of a common time sequence in the unpredictability of both motions. However, in [4], utilizing the LFA modeling approach of GDSS in nitrogen, the common separation and convergence sequences are identified, providing evidence for the existence of the DSC in this coupled system.

It is important to note that the common convergence sequence indicates points on the trajectories that do not ultimately converge to the initial points but approach a finite non-zero distance. Hence, this method is referred to as delta synchronization of chaos. In simpler terms, a distance with a delta length is present in the phase portrait between the phase point and the initial point of the calculation at the last common time of both systems.

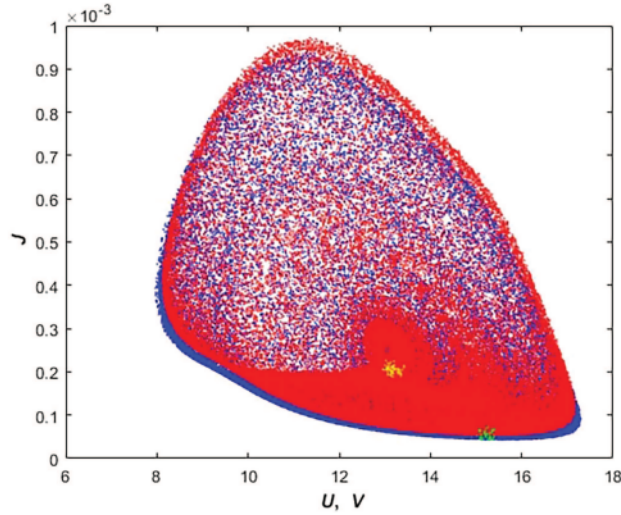


Figure 1.6: The phase portraits of the drive (blue) and response (red) systems of a GDSS [4]. Green (drive) and yellow (response) data points represent the instants, when phase points pass through the closest place of corresponding initial points of the drive and response systems.

Figure 1.6 illustrates the drive and response systems in corresponding phase portraits, where blue and red data points represent the phase points of drive and response systems, respectively. The green and yellow marked points correspond to the common moments of both systems, which gradually approach their respective initial points. Moreover, in [4, 89], it was demonstrated that the coupled system lacks generalized

synchronization. Consequently, the DSC method presents a more comprehensive approach to chaos synchronization by deriving common time moments for both convergence and separation scenarios.

The investigation of chaos synchronization in GDSSs has been limited in the existing literature. Recent studies in [4, 89] have shown that GDSSs lack generalized synchronization, and other chaos synchronization methods have not been explicitly explored. However, introducing the DSC method has demonstrated the existence of chaos synchronization in GDSSs. Chapter 4 of this thesis investigates the chaos synchronization on the subnormal discharge regime of GDSS in nitrogen, characterized by chaotic oscillations, which are based on the models developed in Chapter 3, using one-dimensional "extended fluid models" with "drift-diffusion" approximation. We will develop drive and response systems for the LMEA approach ("extended fluid model" of Chapter 3 of this thesis) of the same GDSS in [4] to investigate chaos synchronization in the coupled system. We will determine common time values for convergence and separation scenarios by implementing the DSC method, which means that the two models act in unison in the coupled system with a numerically determined common "sequence of finite convergence" and "sequence of separation". The results obtained from the synchronization analysis using the "simple" and "extended" fluid modeling approaches will be compared, and new numerical characteristics will be defined to facilitate the comparison of chaos synchronization degree. Using the "extended fluid model", which is more advanced than the "simple fluid model" for the mentioned reasons, enables us to draw more accurate conclusions in our analyses.

Chapter 5 focuses on the correct definition of boundary conditions in fluid modeling RF discharges. The need for more consensus in the literature on this matter is addressed, as boundary conditions substantially impact both the near-boundary walls and the plasma bulk region. In this context, the chapter introduces a universal boundary condition, explains the underlying physical principles of RF discharges, and emphasizes the significance of appropriate boundary conditions in the fluid modeling approach.

1.2.4 Boundary Conditions for fluid modeling of RF discharge system

1.2.4.1 Motivation for universal BC

Fluid equations with "drift-diffusion" approximation are commonly used to address low-temperature plasma physics problems. These equations encompass [96]

$$\frac{\partial n_k}{\partial t} + \nabla \cdot \mathbf{\Gamma}_k = S_k, \quad (1.2.1)$$

$$\mathbf{\Gamma}_k = \text{sgn}(q_k) \mu_k n_k \mathbf{E} - D_k \nabla n_k, \quad (1.2.2)$$

where n stands for particle number density, $\mathbf{\Gamma}$ denotes particle flux density, S indicates the source term, D and μ are diffusion and the mobility coefficients, $V_d = \text{sgn}(q_k) \mu_k \mathbf{E}$ represents drift velocity, q stands for particle charge, and E is the electric field. Chapter 2 explains the fluid definition of plasma particles by considering these equations.

In the "extended fluid approach", a self-consistent set of equations of plasma species is established by combining equations 1.2.1 for each charged segment with the electron energy component and the Poisson equation. By considering the moments of the Boltzmann equation, equation 1.2.1 can be derived for charged particles in a quasineutral plasma, where electric field is relatively weak, and distribution function is approximated using the "Lorentz two-term approximation" (LTTA) [8, 27, 97]. In the LTTA, it is assumed that the particle distribution in the movement path is nearly isotropic. However, when considerable electric fields are present, ion drift velocity usually surpasses thermal velocity, and the ion velocity distribution is no longer isotropic, requiring another approach (This is one of the reasons for the emphasis on the ion boundary flux in Chapter 5). Nevertheless, a "drift-diffusion" pair, similar to Eqs. 1.2.1, 1.2.2 with a nonlocal dependency of the average velocity on the field, can still hold validity under the influence of substantial fields.

When modeling gas discharges, formulating a boundary-value problem is essential to obtain a unique solution. This involves imposing boundary conditions at all boundaries of the discharge volume. Given the diverse conditions and interactions that can occur in different discharge scenarios, developing a universal formulation of the boundary conditions becomes necessary. This universal formulation enables to effec-

tively represent and account for all fields and particles when simulating discharges, allowing for a comprehensive analysis of plasma behavior and phenomena.

1.2.4.2 Existing literature

Various approaches to formulate boundary conditions (BCs) for plasma simulations, each varying in complexity and level of understanding, have been explored. The most straightforward and crudest approach involves setting particle density to zero at absorbing boundary, leading to a Dirichlet condition [98]. When the "wall" is a plane in which the field's perpendicular direction is considered, it acts as a surface that reflects particles, leading to a Neumann BC [99]. An alternative approach, referred to as the "primitive approach" [29], neglects the influence of the altered layer's thickness close to the absorbing wall and approximates the flux to the wall using a "half-Maxwellian" distribution, combining the flux density with the Maxwell distribution over half of the velocity space [29]. Because the flux density involves drift and diffusion terms, the Dirichlet condition is substituted with a more comprehensive condition known as the Robin condition.

The advanced understanding approach considers that the absorbing wall's presence significantly alters the distribution function's isotropy in its nearness, making the LTTA inappropriate. LTTA assumes a nearly isotropic distribution function, while near the wall, particles only move toward it, leading to nonzero values in that direction. The kinetic approach involves greater mathematical complexity. Some authors [100–103] avoided anisotropic kinetic Boltzmann equations and neglected the distortion, restricting their analysis to the Maxwell distribution, leading to an "intermediate" method that is an "effective BC" based on LTTA, although it is not fully comprehensive. This approach is known as the "two-stream approximation" [104, 105] and was initially developed for diffusion flux [100], with later attempts to extend it to the "drift-diffusion" model. However, the inclusion of drift in LTTA along with diffusion resulted in a nonphysical switch [101], whose physical meaning remains unclear [29].

Chapter 5 of this thesis introduces a universal formula that eliminates the need for switching mechanisms. Instead, it consistently applies the "two-stream" approach, treating drift and diffusion terms as equal constituents of the overall flux, instead of

regulating drift terms under a specific relation between the electric field's directions and the particles' motion. Additionally, the chapter demonstrates the extent to which improvement can be made in BC formulation without resorting to LTTA or solving kinetic equations in the vicinity of the boundary [96, 106].

1.2.4.3 Properties of Radio-frequency (RF) discharges

Radio-frequency (RF) plasmas have gained significant importance in hi-tech industries, particularly semiconductor manufacturing, where they serve as indispensable core devices for material etching and thin film deposition [106–108]. Two commonly used RF plasma sources in the semiconductor industry are the capacitively coupled plasmas (CCP) reactor and the inductively coupled plasmas (ICP) reactor, as shown in Fig. 1.7.

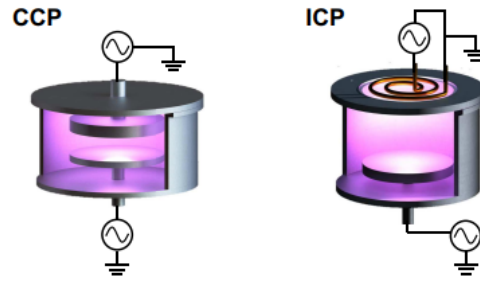


Figure 1.7: The schematic illustration of typical CCP and planar-coil type ICP reactors [5].

The CCP reactor, developed in the 1970s, consists of two parallel-plated electrodes spaced a few centimeters apart in a vacuum chamber. One electrode is driven by an RF power source with a typical power ranging from 100 W to 1000 W and a frequency between 13.56 MHz and 200 MHz. The plasma density in CCP is relatively low, typically in the 10^{15} m^{-3} range to 10^{16} m^{-3} . In contrast, the ICP reactor is driven by an RF power supply, utilizing either a planar coil or antenna positioned on top of a vacuum chamber or a helical coil surrounding a dielectric cylinder. The RF current in the coil generates electromagnetic fields that convey energy to electrons within the chamber. The magnitude of the RF current determines the plasma density in ICP, which is usually 1 – 2 orders of magnitude higher than that in CCP due to the higher

power efficiency of inductive coupling.

Understanding the fundamental processes in RF plasmas is crucial to improve the operational efficiency of plasma sources in real-world applications. One fundamental aspect is the electron heating or electron power absorption mode in time-varying fields, which determines the EEDF. High-energy electrons above certain thresholds are responsible for gas-phase chemical reactions such as ionization, excitation, and dissociative attachment processes, significantly influencing the concentrations and spatial distributions of charged species and neutral radicals. Furthermore, the energy distribution functions of various ionic species in the sheath region play a crucial role in the plasma's interaction with material surfaces, particularly in processes like atomic layer etching [109]. Secondary electron emission, induced by ion and atom bombardment and photon irradiation, contributes to the behavior of electrons in the plasma. Some interactions, such as ion sputtering and surface chemical reactions, are vital in material processing. Moreover, high-energy electrons play a more critical role than low-energy electrons in ionizing and exciting background atoms or molecules, impacting the macroscopic properties of the RF discharge. The discharge mode, represented by the spatiotemporal distributions of excitation/ionization rates caused by highly energetic electrons, has been extensively studied in recent experiments and simulations [110–112]. Understanding the discharge mode is essential for optimizing critical plasma parameters and improving independent control of ion flux and energy on the substrate.

1.2.4.4 BC investigations in RF plasmas

Modeling RF plasmas using continuum (hydrodynamic) approaches have been instrumental in basic and applied research [113–116]. However, defining appropriate boundary conditions (BCs) for the solid surfaces of RF plasmas remains unresolved in the literature. Traditional fluid models for RF plasmas often adopt the same BCs as those used for DC plasmas. While some studies have explored BCs for DC glow discharges in detail [101, 117], their suitability for RF plasmas has not been thoroughly investigated. Moreover, DC plasma BCs frequently consider secondary electron emission (SEE) due to ion impact on surfaces [117, 118], often neglected in RF

plasmas. As RF plasma fluid models commonly use the LMEA method, requiring the solution of the electron energy equation, Chapter 5 applies the LMEA modeling equations to analyze the effects of various BCs.

A fluid plasma model necessitates BCs for the number, energy densities, and electric potential. The surfaces are typically grounded or assigned a known voltage in terms of electric potential. However, they could also be dielectrics [29, 119], where the surface accumulates a non-uniform charge due to the electric current. In some studies, the number and energy densities are set to zero at the surface [120, 121], assuming the surface absorbs charged particles. Alternatively, the flux to the surface can be specified based on drift toward the boundary [119, 122] and thermal motion [29, 123]. SEE effects at boundaries are often disregarded in RF discharges, although occasionally included [124]. However, the rationale selection of BCs is rarely provided in the reviewed works, and the importance of this choice needs to be fully discussed and quantified. Chapter 5 of this thesis is driven by the need for a consensus on the exact form of BCs required for the fluid modeling of RF gas discharge systems. The chapter demonstrates that the choice of BC can significantly impact both the sheath and bulk plasma regions, where thermal and diffusion fluxes to the surface are crucial considerations.

Chapter 6 of this thesis compares electron transport and electron-induced rate coefficients obtained from different solvers. These input parameters are then implemented into a one-dimensional fluid model of the Dielectric Barrier Discharge (DBD) system in CO_2 to investigate the effects of different definitions of electron transport and rate coefficients on this system.

1.2.5 Input parameters of a DBD fluid model in CO_2

1.2.5.1 Basic configurations and properties of DBDs

"Dielectric Barrier Discharges" (DBDs) are autonomous electrical phenomena that occur within electrode arrangements, including an insulating material along the discharge setup. The presence of the dielectric barrier is essential for enabling self-pulsing plasma operation, leading to the formation of a nonequilibrium plasma at

atmospheric pressure and low temperatures in a reliable and cost-effective manner.

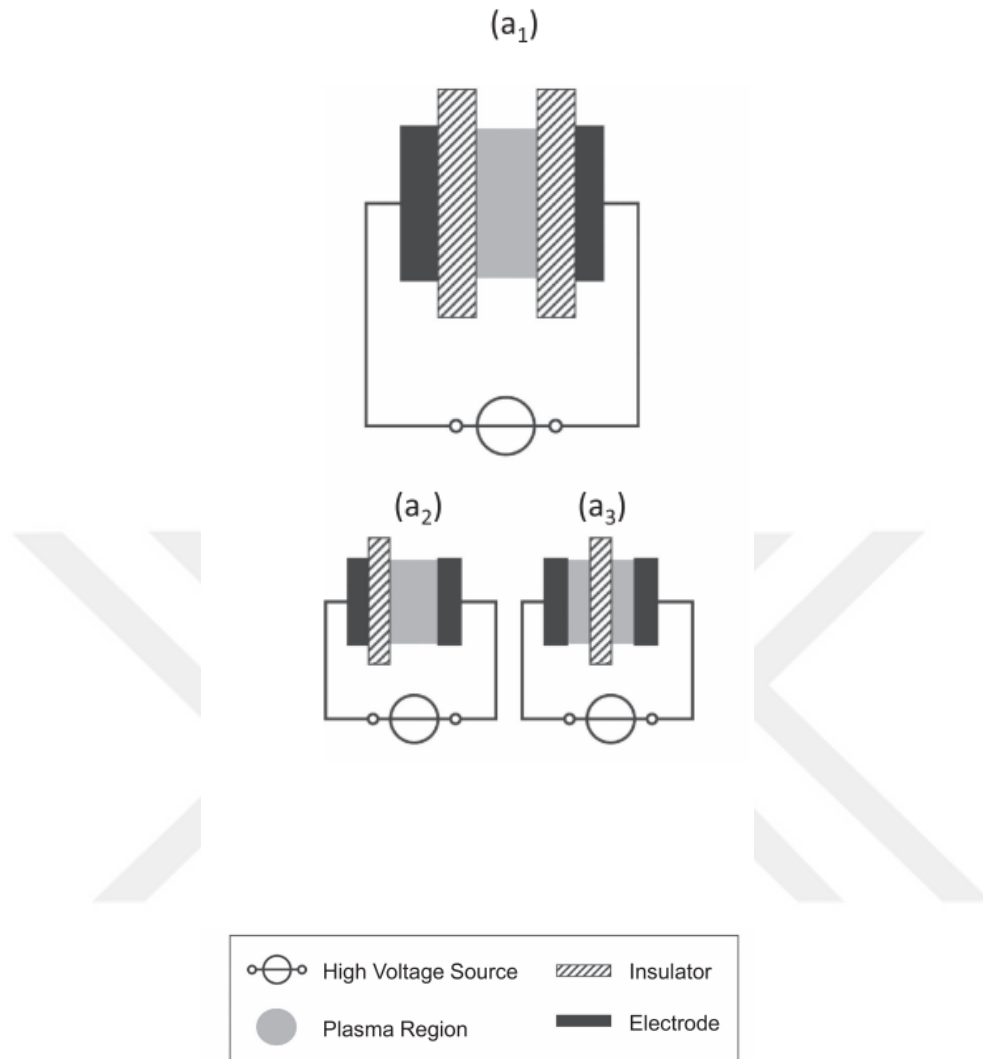


Figure 1.8: Basic planar configurations of volume DBDs: **(a1)** symmetric, **(a2)** asymmetric, **(a3)** floated dielectric [6].

Often referred to as "silent discharges," "barrier discharges," or "ozonizer discharges," DBDs have a rich history, first introduced for ozone generation in 1857 [125], and have since found diverse applications in surface treatment, degrading contaminant molecules in gaseous medium, activating gas laser, powering plasma display, and generating excimer radiation. DBDs serve as electric wind generators in aerodynamic systems and find use within innovative analytical detection devices. The designs and underlying principles of DBDs involve incorporating insulating materials in the discharge path. Common materials employed as dielectric include glass, quartz, ceram-

ics, enamel, mica, plastics, silicon rubber, or Teflon. Remarkably, barrier discharges can operate under a wide range of conditions, encompassing gas pressures from several tens of Torr to atmospheric pressure, gas gap widths varying from fractions of a millimeter to several millimeters, voltage amplitudes spanning from hundreds of volts to several kilovolts, and field frequencies ranging from tens to several hundreds of kilohertz. Based on this general characterization, various designs can be developed.

Figure 1.8 illustrates the primary fundamental configurations in a planar structure. In volume DBDs, there are variations in the electrode configurations concerning the placement and number of dielectric barriers. Specifically, one or both electrodes can be surrounded by barriers ((a1) and (a2)), or a barrier can separate the structure into two areas (a3) [126]. In arrangement (a1), both electrodes are shielded from the species that may come from the plasma bulk medium. On the other hand, configurations (a2) and (a3) provide interactions between one or both electrodes and plasma, resulting in erosion or corrosion. Nonetheless, operating the plasma at a lower high voltage amplitude is possible in (a2) and (a3) due to a more significant portion of the applied voltage distributed across the gas gap. In the case of (a3), the dielectric is floated, and while this does not impact the general operational principle, it presents an option for treating gases using DBDs. Several novel configurations have emerged as researchers and engineers have adapted DBDs to address various challenges.

In addition to the different planar configurations (parallel plate electrode configurations), Figure 1.9 shows cylindrical configurations (coaxial electrode configurations), which play a significant role in DBD investigations [127–129]. These cylindrical devices with coaxial electrode arrangements are commonly employed as conventional gas reactors, such as generating ozone from oxygen or air. In these setups, insulating tubes are utilized either as "outer electrode" (Figure 1.9(a)) or to surround "inner electrode" (Figure 1.9(b)). Instead of using a solid metal electrode, it is possible to employ a metallic film that is either deposited or stuck onto the inner or outer wall of the dielectric tube, which then functions as the inner or outer electrode. The utilization of cylindrical configurations broadens the scope of DBD applications and provides valuable insights into various gas treatment processes, making it an essential area of research and experimentation. In Chapter 6, we will also develop a one-dimensional

fluid model based on the "drift-diffusion" approximation for particle fluxes in a coaxial DBD operating at atmospheric pressure with CO_2 .

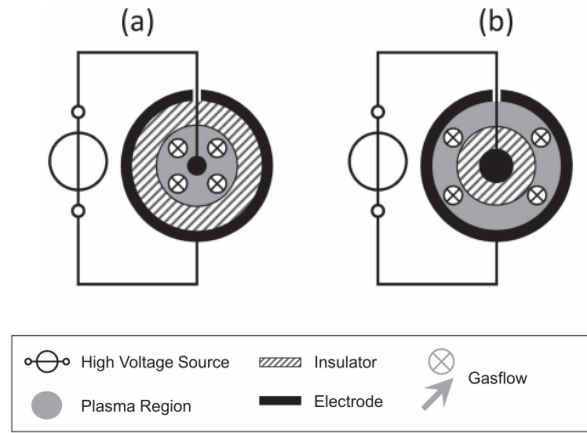
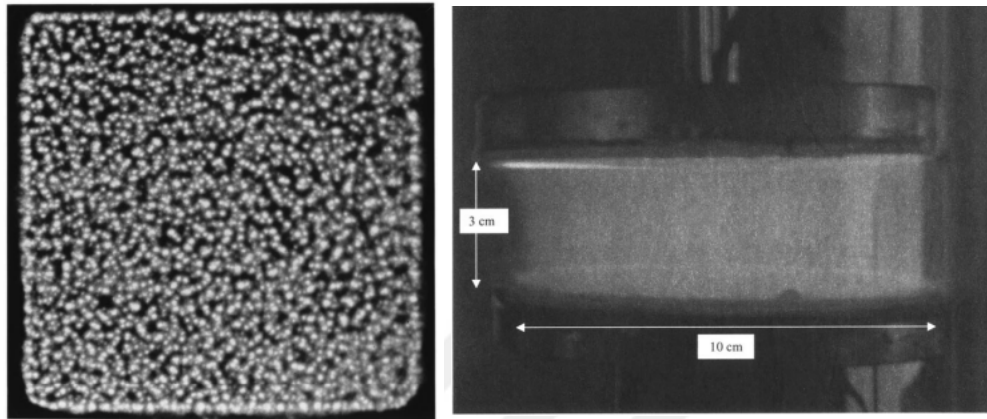


Figure 1.9: DBD reactor cross-sectional illustrations for the treatment of gases: coaxial volume DBD with an outer electrode arrangement **(a)** and an inner electrode arrangement **(b)** [6].

Depending on the gas mixture, dielectric surface properties, and operating conditions, various discharge modes can manifest in DBDs at atmospheric pressure, including diffuse, filamentary, and mixed modes [130–132]. The typical filamentary regime is characterized by many short-lived current filaments, commonly observed in most industrial applications of barrier discharges. In contrast, the plasma of a diffuse barrier discharge uniformly covers the entire electrode area [131]. Within the diffuse modes, two different types can occur: the atmospheric pressure Townsend discharge and the atmospheric pressure glow discharge [132–135]. The occurrence of these discharge modes depends on factors such as cell geometry, amplitude and frequency of the applied voltage, gas composition, and gas pressure [130–132]. Complex discharge dynamics can arise from the periodic or chaotic alternation between different discharge modes. Phenomena like period multiplication, mixed-mode oscillations, and transitions to chaotic states in experimental and theoretical investigations of electrical discharges in gases have been observed [136–141]. A detailed analysis of the discharge properties is often necessary to explain such behaviors. Moreover, DBDs can be categorized into two groups based on the breakdown mechanism: those resulting from a streamer breakdown and those from a Townsend breakdown. Streamer

breakdown typically leads to a filamentary discharge, while Townsend breakdown leads to a more homogeneous discharge. However, the precise connection between the breakdown mechanism and the discharge homogeneity still needs to be fully understood [142].



(a) Filamentary breakdown of a DBD system, which is obtained by a small parallel-plate ozonier with a 1 mm air gap [143]. (b) Diffuse barrier discharge operated in an atmospheric-pressure mixture of helium and air [143].

Figure 1.10: The illustrations of Filamentary and Diffuse DBDs.

Figure 1.10a provides visual evidence of filamentary breakdown in a small parallel-plate ozonizer with a 1 mm air gap. The image likely captures the characteristic appearance of numerous short-lived current filaments (microdischarges) during the filamentary discharge mode. On the other hand, Figure 1.10b depicts the operation of a diffuse barrier discharge in a mixture of helium and air at atmospheric pressure. Indeed, the plasma uniformly covers the entire electrode area, indicating a diffuse discharge mode. Thus, understanding the different discharge modes and underlying mechanisms is essential for optimizing DBD applications and advancing plasma science and technology. Further research and analysis are required to comprehensively understand these phenomena and harness their potential in various practical applications.

1.2.5.2 Importance of CO_2 DBD system

Many industries, particularly the automotive and aerospace sectors, still heavily rely on combustion methods using fossil fuels, leading to significant emissions of polluting and greenhouse gases [144]. Among these gases, carbon dioxide (CO_2) is one of the most problematic for the environment, mainly due to its contribution to global warming [145, 146]. Carbon capture and storage in geological reservoirs have been considered a potential solution to address the substantial emission of significant amounts of CO_2 into the atmosphere. However, this approach has various drawbacks, encompassing high expenses, environmental evaluations, and enduring risks. Additionally, this needs to align with the fundamental aspects of sustainable development because CO_2 continues to be classified as waste demanding long-term storage. An environmentally friendlier alternative has arisen: treating CO_2 as a valuable resource and converting it into products with improved value by combining it with hydrogen gas. Electrochemical and photochemical methods for reducing CO_2 have been effectively employed to acquire products like CO , formic acid, and methane [147–149]. Additionally, splitting pure CO_2 into CO and O_2 is noteworthy, as CO shows higher reactivity than CO_2 and can be employed in diverse catalytic processes, including the production of acetic acid (Monsanto process) and liquid hydrocarbons (Fischer-Tropsch process) [150–152].

In recent years, cold plasmas have emerged as a novel approach to valorizing CO_2 [153]. These plasmas work beyond thermodynamic equilibrium, where electrons mainly gain energy using applied electric power. This energy, in turn, triggers gas activation through processes like electron-induced reactions. This leads to the formation of novel reactive species that readily transform into different molecules while the gas maintains its temperature around room levels. Plasma, when coupled with heterogeneous catalysis, can also enable the synthesis of particular organic molecules [154, 155]. For example, $CO_2 - CH_4$ treatments using Ni catalysts have been used to produce syngas and slight quantities of organic compounds [156, 157]. Additionally, plasma converts peak currents from sustainable energy sources into new fuels due to its capacity for swift activation and deactivation. Moreover, capturing, transporting, and storing CO_2 may no longer be essential, as specialized plasma sources

could directly and immediately process emissions from industrial smokestacks. Diverse plasma sources have been utilized for the conversion of CO_2 , such as coronas, gliding arcs, and dielectric barrier discharges [158–171]. Unlike coronas and gliding arcs, DBDs offer the advantage of treating the entire gas flow because the whole gas flows through the region where the discharge is active. Additionally, DBD reactors possess a straightforward design that could be readily expanded for industrial purposes, as presented by ozone synthesis [172–174]. The use of flowing DBD setups has shown promise, achieving a sufficient balance between proper energy efficiency and heightened CO_2 conversion rates for low SEI [175]. Although DBDs alone might not offer an economically viable approach for CO_2 splitting, they hold promise when integrated with catalysts in a packed bed design. In such situations, DBDs have the capability to facilitate the targeted generation of value-added chemicals and improve the energy efficiency of CO_2 dissociation [169, 176–178].

Despite numerous experimental studies on CO_2 dissociation using various excitation frequencies, gas flow rates, and reactor configurations, there still needs to be more fundamental knowledge concerning the prevailing mechanisms in CO_2 Dielectric Barrier Discharges (DBDs) at atmospheric pressure. As a result, conversion rates and energy efficiencies often remain below 10%. Gaining a deeper understanding of the complex and highly transient plasmas that occur spatially and temporally is essential, making in-situ experimental studies highly desirable. However, conducting such studies takes much work. One particular aspect of interest is whether vibrational excitation, proposed in [179] for discharges using higher excitation frequencies, plays a dominant role in atmospheric pressure DBDs. Addressing this question can shed light on the underlying mechanisms that govern CO_2 dissociation and potentially pave the way for improved energy efficiencies and conversion rates in this process. Further research and experimental investigations are required to explore and elucidate the intricate processes occurring in CO_2 DBDs at atmospheric pressure. A comprehensive understanding of these mechanisms is crucial for optimizing and enhancing the efficiency of CO_2 conversion, thus contributing to developing more sustainable and environmentally friendly approaches to tackle CO_2 emissions.

1.2.5.3 Modeling of CO_2 DBD system

Numerical simulations are important in comprehensively understanding dissociation mechanisms and predicting optimal operational parameters. Various modelling approaches are employed to investigate gas discharge plasmas, including fluid models, fully kinetic treatments of charge carriers, and hybrid methods [180]. In the context of CO_2 discharges, global reaction kinetic models have been commonly utilized to analyze dissociation mechanisms. These models involve solving a set of rate equations for different species, allowing for the study of low-pressure RF discharges [181], DC glow discharges [182], and moderate-pressure microwave discharges [183,184] in pure CO_2 . Global modeling studies have also been conducted for CO_2 laser discharge plasmas [182,185–191] in different mixtures, and DC-supplied $He - CO_2$ glow discharges [192]. However, a notable gap exists in the reported works on modelling CO_2 dissociation in dielectric barrier discharges (DBD), with only a few earlier works addressing this topic [193]. Recently, there has been an emerging interest in this area [183,194–196], using a zero-dimensional fluid model that incorporates extensive reaction kinetics, considering the vibrational kinetics of CO_2 . In contrast, a study in [193] employed a time-dependent, spatially two-dimensional model. This model solves the continuity equation for charged particles and Poisson equation to calculate the local electric field. However, this study considered a simplified set of reactions and did not account for the vibrational relaxation processes of CO_2 . Given that a typical DBD consists of multiple randomly distributed filaments across the dielectric surface, a spatially two- or three-dimensional model, including a stochastic description of filament generation, is necessary for a comprehensive theoretical description of discharges. In a related work, [197], a time-dependent, spatially one-dimensional approach was used to model and analyze the spatiotemporal discharge dynamics of the active zone. This treatment is well-suited for analyzing planar DBDs operating in the homogeneous or glow mode [198] and has proven successful in studying single micro discharges (e.g., Refs. [199] and [200]).

In chapter 6 of this work, we will develop a one-dimensional fluid model for the CO_2 DBD system with a cylindrical geometry, where the plasma is only formed in the radial direction of a coaxial electrode configuration.

1.2.5.4 Boltzmann solvers and input parameters

In the DBD system using CO_2 gas, dissociating inert CO_2 into CO and O_2 requires energy. Low-temperature atmospheric plasma processes offer a convenient and innovative solution because electrons supply most energy, minimizing energy losses due to gas heating. In this plasma, CO_2 splitting is primarily driven by electron-induced dissociation, forming CO and O atoms [194]. For the direct dissociation of CO_2 , electrons must possess energies higher than the $C = O$ bond dissociation energy of the CO_2 molecule, which is 5.52 eV. Typically, only a small fraction of electrons in the tail of the EEDF meet this requirement, as most electrons have energies around 2 – 3 eV [195]. The energetic electrons then activate the gas molecules through excitation, ionization, and dissociation reactions, generating reactive species that can efficiently form new molecules. The relatively low gas temperature in the plasma prevents backward recombination reactions. It is important to note that each gas behaves differently in a discharge due to variations of reactions with electrons and their energy dependence [201–203]. Gas heating can significantly impact the EEDF by influencing the reduced electric field E/N or mean electron energy. Consequently, it affects electron transport, reaction rates, and ultimately the CO_2 conversion [204]. However, in Chapter 6, the influence of gas heating mechanisms on electron behavior is neglected by assuming a constant room temperature for the gas. Instead, this chapter focuses on the effects of various EEDFs on the transport and rate coefficients of electrons in DBD models of CO_2 . It derives input parameters from different solvers and common assumptions widely used in the literature.

Fluid models of gas discharges employ the Boltzmann equation (BE) moments to describe the transport of electrons, ions, and other reactive particle species. These moments consist of the continuity equation, the momentum equation, generally approximated by the "drift-diffusion" equation, and the energy equation (usually only for electrons) [8, 28, 205–207]. Transport and rate coefficients play a crucial role in these equations and serve as input data for the fluid model. These coefficients are specific to the discharge conditions, particularly concerning electrons, as they depend on the EEDF, which is often non-Maxwellian and varies significantly under different conditions. Calculating the EEDF and electron coefficients for a given discharge

condition involves solving the electron BE using fundamental collision cross-section data [8, 208]. A common approach is solving the BE with necessary approximations for various reduced electric field values, generating lookup tables of coefficients against the reduced field or mean electron energy. These tables are then used in the fluid model through interpolation to define the transport and rate coefficients.

BE solvers used to obtain electron-related input data for fluid models were developed in the 1970s and 1980s. These solvers had different objectives than fluid discharge modeling; they were initially designed to replicate specific experiments and achieve high numerical precision in computing physical quantities [37, 209–217]. The requirements for fluid modeling include the ability of the BE solver to work over a wide range of discharge conditions (reduced electric field, ionization degree, gas composition, and field frequency) rather than simulating a single experiment.

Several user-friendly BE solvers are used and cited in fluid discharge modeling, such as the commercial ELENDIF [213] and the freeware BOLSIG [8]. However, these solvers were initially designed with a different objective. They may be limited in their application to certain discharge conditions, inconvenient for generating coefficient tables, or inadequately documented (especially BOLSIG), making it difficult to evaluate their appropriateness for fluid modeling. To address these limitations, BOLSIG+ [218] was developed, capable of handling a broader range of discharge conditions, providing faster and easier usage, and paying more attention to consistent coefficient definitions. BOLSIG+ utilizes theoretical descriptions and approximations from [8]. In Chapter 6, an electron Boltzmann solver is explicitly developed for electron-induced reactions in CO_2 DBD. The resulting transport coefficients are compared with those derived from online electron Boltzmann solvers and common approximations in the literature.

In fluid models, certain approximations are often used for transport coefficients. For example, the Einstein relation between the diffusion coefficient (D) and mobility (μ) is commonly employed, given by $D_e = \mu_e k_B T_e / e$, which is exact for a Maxwellian EEDF or a constant momentum-transfer frequency but approximate for real discharge conditions. Additionally, fluid models frequently use an electron energy equation where the energy transport coefficients (μ_ε and D_ε) are approximated as $\mu_\varepsilon = 5/3 \mu_e$

and $D_e = 5/3D_e$ [9]. These relations are derived by assuming a Maxwellian EEDF, constant momentum-transfer frequency, and constant kinetic pressure [9]. Evaluating the accuracy of these approximations is important, and Chapter 6 of this thesis also examines some of them against the results of the BE solver to gain insight into their validity under various conditions.

To sum up, this thesis presents a comprehensive and in-depth exploration of the diverse aspects of gas discharges. Chapter 2 establishes the governing equations of plasma fluid theory and introduces the concepts of "simple" and "extended" fluid models. Periodic and irregular oscillations of GDSS in nitrogen are investigated and classified in Chapter 3. In Chapter 4, the synchronization phenomenon of chaotic regimes of Chapter 3 in the master-slave approach is detected using the DSC method. Chapter 5 presents a universal BC in RF gas discharge systems and compares it with the BCs used in the literature. Chapter 6 develops a Boltzmann solver to be used as the input parameters of the DBD system in CO_2 . The validness of the input parameters derived from different solvers and common approximations widely used in the literature are also tested.



CHAPTER 2

GAS DISCHARGE MODELLING: FLUID EQUATIONS

In this chapter, the fluid modeling equations for glow discharge are derived from the moments of the kinetic Boltzmann equation. These include continuity, momentum, and energy equations, which are crucial for describing the behavior of the "two-fluid" components: electrons and positive ions. Furthermore, under specific assumptions and reductions, the Maxwell equations are coupled to the "two-fluid" equations to capture the interaction between the plasma particles and electromagnetic fields. The "drift-diffusion" approximation is employed to derive particle flux densities, combined with the continuity equation. With the governing equations of plasma fluid theory established, the concepts of "simple" and "extended" fluid models are introduced. A comparison is made between these two modeling approaches to determine their strengths and limitations, aiming to identify which model is more suitable for fulfilling specific fundamental requirements.

2.1 Fluid description of plasma

Each plasma species can be described with a velocity distribution function, indicated by $f(\mathbf{r}, \mathbf{v}, t)$, the probability of finding particles in a six-dimensional $(\mathbf{r}, \mathbf{v})$ space at a specific position, velocity, and time [16, 25]. According to the kinetic theory of plasmas, the Boltzmann equation describes the time evolution of this distribution function under the influence of external forces. Solving the Boltzmann equation is computationally inefficient and a challenging task. Instead, both sides of this equation are multiplied with the powers of velocity and integrated along the velocity domain. Orders of velocity power separately yield moments of the Boltzmann equation and

functional integrals defining the macroscopic quantities such as the plasma particles' number density, velocity, and energy. As proposed in [25], the resulting equations are coupled with Maxwell equations for the foundation of self-consistent plasma fluid theory.

Fluid modeling equations of each plasma species are derived by taking the moments of the following kinetic Boltzmann equation,

$$\frac{\partial f_i}{\partial t} + \nabla_{\mathbf{r}} \cdot \mathbf{v} f_i + \frac{q_i}{m_i} \nabla_{\mathbf{v}} \cdot [(\mathbf{E} + \mathbf{v} \times \mathbf{B}) f_i] = \sum_k C_k f_k. \quad (2.1.1)$$

The solution $f_i(\mathbf{r}, \mathbf{v}, t)$ is a velocity distribution function such that the quantity $f_i d\mathbf{r} d\mathbf{v}$ defines the number of particles belonging to species i within the volume element $d\mathbf{r} d\mathbf{v} = dx dy dz dv_x dv_y dv_z$ in six-dimensional phase space at time t . $\mathbf{E}$ and $\mathbf{B}$ represent the local electric and magnetic fields. q and m are the particle's charge and mass. $\nabla_{\mathbf{r}}$ and $\nabla_{\mathbf{v}}$ denote the gradients in the coordinate and velocity spaces, respectively. The operator C_k describes both elastic and inelastic collisional processes experienced by the particles of species k , which leads to changes in the distribution function f_i .

The zeroth-order moment: The equation 2.1.1 is multiplied with $v^0 = 1$ on both sides and integrated over the velocity domain [25],

$$\begin{aligned} & \int \frac{\partial f_i}{\partial t} d\mathbf{v} + \int \nabla_{\mathbf{r}} \cdot \mathbf{v} f_i d\mathbf{v} \\ & + \frac{q_i}{m_i} \int \nabla_{\mathbf{v}} \cdot [(\mathbf{E} + \mathbf{v} \times \mathbf{B}) f_i] d\mathbf{v} = \int \left(\sum_k C_k f_k \right) d\mathbf{v}. \end{aligned} \quad (2.1.2)$$

Furthermore, as described in [25], the particle density and the fluid velocity (averaged over mass) are introduced through the following integrals:

$$n_i(\mathbf{r}, t) = \int f_i(\mathbf{r}, \mathbf{v}, t) d\mathbf{v} \quad (2.1.3)$$

and

$$\langle \mathbf{v} \rangle = u_i(\mathbf{r}, t) = \frac{1}{n_i(\mathbf{r}, t)} \int \mathbf{v} f_i(\mathbf{r}, \mathbf{v}, t) d\mathbf{v}. \quad (2.1.4)$$

Note that the third element in equation 2.1.1 vanishes due to the Gauss theorem. Similarly, when collisions do not alter the density, the integration of the right-hand side becomes zero, indicating the absence of particle creation or destruction. However, if

collisions do influence the density, it is $S_i = \sum_k n_k \langle C_k \rangle$. Then, the equation 2.1.1 becomes as suggested in [25],

$$\frac{\partial n_i}{\partial t} + \nabla \cdot (n_i \mathbf{u}_i) = S_i, \quad (2.1.5)$$

which is the continuity equation governing the conservation of particles. The term S_i is responsible for inelastic processes affecting species' populations. Furthermore, the charge and mass balance equations are able to be derived by multiplying 2.1.5 with the particle charge q_i and mass m_i , respectively.

The first-order moment: When the kinetic equation 2.1.1 is multiplied by velocity $\mathbf{v}$ and integrated over velocity domain, we obtain [25]

$$\begin{aligned} \int \mathbf{v} \frac{\partial f_i}{\partial t} d\mathbf{v} + \int \mathbf{v} \nabla_{\mathbf{r}} \cdot (\mathbf{v} f_i) d\mathbf{v} \\ + \frac{q_i}{m_i} \int \mathbf{v} \nabla_{\mathbf{v}} \cdot [(\mathbf{E} + \mathbf{v} \times \mathbf{B}) f_i] d\mathbf{v} = \int \mathbf{v} \left(\sum_k C_k f_k \right) d\mathbf{v}. \end{aligned} \quad (2.1.6)$$

The first component of this equation (see equation 2.1.4) is

$$\int \mathbf{v} \frac{\partial f_i}{\partial t} d\mathbf{v} = \frac{\partial}{\partial t} (n_i \mathbf{u}_i). \quad (2.1.7)$$

The following expression gives the second component

$$\begin{aligned} \int \mathbf{v} \nabla_{\mathbf{r}} \cdot (\mathbf{v} f_i) d\mathbf{v} &= \nabla_{\mathbf{r}} \cdot \int \mathbf{v} \mathbf{v} f_i d\mathbf{v} \\ &= \nabla_{\mathbf{r}} \cdot n_i \langle \mathbf{v} \mathbf{v} \rangle = \nabla \cdot \left(n_i \mathbf{u}_i \mathbf{u}_i + \frac{1}{m_i} \mathbf{P}_i \right). \end{aligned} \quad (2.1.8)$$

The pressure tensor is $P_i = m_i \int \langle \mathbf{w} \mathbf{w} \rangle f_i d\mathbf{w} = m_i n_i \langle \mathbf{w} \mathbf{w} \rangle$, which is usually represented as $\mathbf{P}_i = p_i \mathbf{1} + \pi_i$, where $\mathbf{1}$ is the identity tensor, $p_i = \frac{1}{3} m_i n_i \langle w^2 \rangle = \frac{1}{3} (P_{xx} + P_{yy} + P_{zz})$ is the partial pressure such that $\mathbf{w}$ indicates the thermal velocity with relations of $\mathbf{v} = \mathbf{u}_i + \mathbf{w}$ and $\int \mathbf{w} f_i d\mathbf{w} = \langle \mathbf{w} \rangle = 0$, and π_i is also a pressure tensor having the property of being traceless.

By using integration by parts and above inferred identities, the third term of the equation 2.1.6 is transformed to

$$\frac{q_i}{m_i} \int \mathbf{v} \nabla_{\mathbf{v}} \cdot [(\mathbf{E} + \mathbf{v} \times \mathbf{B}) f_i] d\mathbf{v} = -\frac{q_i n_i}{m_i} (\mathbf{E} + \mathbf{u}_i \times \mathbf{B}). \quad (2.1.9)$$

As proposed in [25], we finally obtain

$$\frac{\partial n_i \mathbf{u}_i}{\partial t} + \nabla \cdot (n_i \mathbf{u}_i \mathbf{u}_i) + \nabla \cdot \frac{1}{m_i} \mathbf{P}_i = \frac{q_i n_i}{m_i} (\mathbf{E} + \mathbf{u}_i \times \mathbf{B}) + \frac{1}{m_i} \mathbf{R}_i, \quad (2.1.10)$$

where the last term, expressed as $\mathbf{R}_i/m_i = \sum_k n_k \langle C_k \mathbf{v} \rangle$, represents the rate of momentum exchange through collisions among different species. Since

$$\frac{\partial(n_i \mathbf{u}_i)}{\partial t} = \mathbf{u}_i \frac{\partial n_i}{\partial t} + n_i \frac{\partial \mathbf{u}_i}{\partial t} \quad (2.1.11)$$

and

$$\nabla \cdot (n_i \mathbf{u}_i \mathbf{u}_i) = \mathbf{u}_i \nabla \cdot (n_i \mathbf{u}_i) + n_i \mathbf{u}_i \cdot \nabla \mathbf{u}_i, \quad (2.1.12)$$

the equation 2.1.10 becomes [25]

$$m_i n_i \frac{d\mathbf{u}_i}{dt} = q_i n_i (\mathbf{E} + \mathbf{u}_i \times \mathbf{B}) - \nabla p_i - \nabla \cdot \pi_i + \mathbf{R}_i, \quad (2.1.13)$$

which corresponds to the fundamental principle of momentum conservation.

The second-order moment: When the kinetic equation 2.1.1 is multiplied by v^2 and integrated over velocity space, we obtain

$$\begin{aligned} & \int v^2 \frac{\partial f_i}{\partial t} d\mathbf{v} + \int v^2 \nabla_{\mathbf{r}} \cdot (\mathbf{v} f_i) d\mathbf{v} \\ & + \frac{q_i}{m_i} \int v^2 \nabla_{\mathbf{v}} \cdot [(\mathbf{E} + \mathbf{v} \times \mathbf{B}) f_i] d\mathbf{v} = \int v^2 \left(\sum_k C_k f_k \right) d\mathbf{v}. \end{aligned} \quad (2.1.14)$$

This equation can be equivalently expressed as follows.

$$\begin{aligned} & \frac{\partial}{\partial t} \int v^2 f_i d\mathbf{v} + \nabla_{\mathbf{r}} \cdot \int v^2 \mathbf{v} f_i d\mathbf{v} + \frac{q_i}{m_i} \int \nabla_{\mathbf{v}} \cdot [v^2 (\mathbf{E} + \mathbf{v} \times \mathbf{B}) f_i] d\mathbf{v} \\ & - \frac{q_i}{m_i} \int 2\mathbf{v} \cdot [(\mathbf{E} + \mathbf{v} \times \mathbf{B}) f_i] d\mathbf{v} = \int v^2 \left(\sum_k C_k f_k \right) d\mathbf{v}. \end{aligned} \quad (2.1.15)$$

The first term of this equation

$$\int v^2 f_i d\mathbf{v} = n_i u_i^2 + n_i \langle w^2 \rangle, \quad (2.1.16)$$

the second term is

$$\begin{aligned} \int v^2 \mathbf{v} f_i d\mathbf{v} &= n_i u_i^2 \mathbf{u}_i + 2n_i \mathbf{u}_i \cdot \langle \mathbf{w} \mathbf{w} \rangle + n_i \mathbf{u}_i \langle w^2 \rangle + n_i \langle w^2 \mathbf{w} \rangle \\ &= n_i u_i^2 \mathbf{u}_i + \frac{2}{m_i} \mathbf{u}_i \cdot \mathbf{P}_i + \frac{3p_i}{m_i} \mathbf{u}_i + \frac{2}{m_i} \mathbf{q}_i, \end{aligned} \quad (2.1.17)$$

where the heat flow vector is defined as $\mathbf{q}_i = n_i \langle \frac{1}{2} m_i w^2 \mathbf{w} \rangle$.

The third component is zero due to complete derivation inside the integral, leading to the cancellation of this term at the boundaries, while the fourth term is

$$-2 \frac{q_i}{m_i} n_i \mathbf{u}_i \cdot \mathbf{E}. \quad (2.1.18)$$

By multiplying the equation 2.1.15 with $m_i/2$ and with a definition of $\varepsilon_i = \frac{1}{2}m_i w^2$, the right-hand side of equation obtains the form as described in [25]

$$\frac{1}{2}m_i \int v^2 \left(\sum_k C_k f_k \right) d\mathbf{v} = \mathbf{R}_i \cdot \mathbf{u}_i + \sum_k n_k \langle C_k \varepsilon_i \rangle = \mathbf{R}_i \cdot \mathbf{u}_i + Q_i, \quad (2.1.19)$$

which represents the rate of energy transfer per unit volume by all other species.

In the case of a separable distribution function in real and velocity space, $f_i(\mathbf{r}, \mathbf{v}, t) = n_i(\mathbf{r}, t)F(v)$, and considering a Maxwellian distribution function

$$F_i(v) = \left(\frac{m_i}{2\pi k_B T_i} \right)^{3/2} \exp\left(-\frac{m_i v^2}{2k_B T_i}\right), \quad (2.1.20)$$

with a definition of $k_B T_i = \frac{p_i}{n_i} = \frac{1}{3}m_i \langle w^2 \rangle$, the energy equation is as follows.

$$\begin{aligned} \frac{\partial}{\partial t} \left(\frac{1}{2}m_i n_i u_i^2 + \frac{3}{2}n_i k_B T_i \right) + \nabla \cdot \left[\frac{1}{2}m_i n_i u_i^2 \mathbf{u}_i + \frac{5}{2}n_i k_B T_i \mathbf{u}_i + \pi_i \cdot \mathbf{u}_i \right] \\ = -\nabla \cdot \mathbf{q}_i + q_i \mathbf{E} \cdot (n_i \mathbf{u}_i) + \mathbf{R}_i \cdot \mathbf{u}_i + Q_i \end{aligned} \quad (2.1.21)$$

where $p_i = n_i k_B T_i$ represents the partial pressure and k_B is the Boltzmann constant.

The first term inside the time derivative, $(\frac{1}{2}m_i n_i u_i^2 + \frac{3}{2}n_i k_B T_i)$, represents the total density of random thermal and kinetic energy. The Joule heating is attributed to the electric field on the right-hand side of this equation. The term Q_j contains elastic and inelastic collisions. By employing the equations 2.1.5 and 2.1.13, the energy balance equation is eventually transformed into its final form as described in [25]

$$\frac{3}{2}n_i \frac{d}{dt}(k_B T_i) + p_i \nabla \cdot \mathbf{u}_i = -\pi_i : \nabla \mathbf{u}_i - \nabla \cdot \mathbf{q}_i + Q_i. \quad (2.1.22)$$

The set of fluid equations that govern the behavior of species i in a plasma consists of the equations 2.1.5, 2.1.13, and 2.1.22. However, more than these equations are required to develop a complete set. Moreover, it is necessary to define the functions π_i , $\mathbf{R}_i$, $\mathbf{q}_i$, and Q_i because these functions play a vital role in closing the system and obtaining a solution for this system.

2.2 Two-fluid model equations

The fluid equations 2.1.5, 2.1.13, and 2.1.22 obtained in the earlier section are utilized to characterize the plasma composed of electrons and a single species of ions. These

equations will show the behavior of two coupled fluid components. When the viscosity effects are neglected, the fluid equations for the electron fraction can be expressed as follows.

$$\begin{aligned}\frac{\partial n_e}{\partial t} + \nabla \cdot (n_e \mathbf{u}_e) &= S_e, \\ m_e n_e \frac{d\mathbf{u}_e}{dt} &= q_e n_e (\mathbf{E} + \mathbf{u}_e \times \mathbf{B}) - \nabla p_e + \mathbf{R}_{e,i}, \\ \frac{3}{2} n_e \frac{d}{dt} (k_B T_e) + p_e \nabla \cdot \mathbf{u}_e &= -\nabla \cdot \mathbf{q}_e + Q_{e,i}.\end{aligned}\tag{2.2.1}$$

The fluid equations describing the ions

$$\begin{aligned}\frac{\partial n_i}{\partial t} + \nabla \cdot (n_i \mathbf{u}_i) &= S_i, \\ m_i n_i \frac{d\mathbf{u}_i}{dt} &= q_i n_i (\mathbf{E} + \mathbf{u}_i \times \mathbf{B}) - \nabla p_i + \mathbf{R}_{i,e}, \\ \frac{3}{2} n_i \frac{d}{dt} (k_B T_i) + p_i \nabla \cdot \mathbf{u}_i &= -\nabla \cdot \mathbf{q}_i + Q_{i,e}.\end{aligned}\tag{2.2.2}$$

In the given equations, n represents the particle number density, and S is the rate at which particles are created or destructed. $\mathbf{R}$ and Q indicate the momentum and energy exchange rates, respectively. m denotes the mass of the particles, and $p = nk_B T$ is the pressure. The electron and ion species are indicated by the subscripts e and i .

To govern the behavior of electric and magnetic fields, "two-fluid" equations must be complemented by Maxwell equations to describe plasma system comprehensively [25].

$$\begin{aligned}\frac{1}{\mu_0} \nabla \times \mathbf{B} &= \mathbf{j} + \varepsilon_0 \frac{\partial \mathbf{E}}{\partial t}, \\ \nabla \cdot \mathbf{B} &= 0, \\ \nabla \times \mathbf{E} &= -\frac{\partial \mathbf{B}}{\partial t}, \\ \varepsilon_0 \nabla \cdot \mathbf{E} &= \sigma,\end{aligned}\tag{2.2.3}$$

where the symbols σ and $\mathbf{j}$ represent the charge and current densities, respectively. These quantities play a vital role in the Maxwell equations because they provide the spatial distribution of electric charges and the flow of electric current within the plasma system. These two density variables are as follows [25].

$$\begin{aligned}\sigma &= n_i q_i + n_e q_e, \\ \mathbf{j} &= n_i q_i \mathbf{u}_i + n_e q_e \mathbf{u}_e.\end{aligned}\tag{2.2.4}$$

In the given equations, q_e and q_i represent the charge of electrons and ions, respectively. ε_0 and μ_0 correspond to permittivity and permeability of free space. By analyzing the effect of collisions, reasonable approximations for the friction forces $\mathbf{R}_{e,i}$ and the rate of heat transfer $Q_{e,i}$ in the momentum and energy equations, corresponding to the last two ones of the set of equations 2.2.1 and 2.2.2, can be derived. Due to the direct proportionality between momentum lost and relative velocity, the resultant force term could be expressed as suggested in [25].

$$\mathbf{R}_{e,i} = -\nu_{e,i} m_e n_e (\mathbf{u}_e - \mathbf{u}_i), \quad (2.2.5)$$

where $\nu_{e,i}$ is the collision frequency ($\tau = 1/\nu_{e,i}$ represents the mean free time between the collisions). Similarly, it is evident that $\mathbf{R}_{e,i} = -\mathbf{R}_{i,e}$. The heat transfer rate becomes [25]

$$Q_{e,i} = -Q_{i,e} = -3 \frac{m_e}{m_i} \nu_{e,i} n_e k_B (T_e - T_i). \quad (2.2.6)$$

Note that, under two different limits, the energy equations simplify. The first limit is that collisions occur so rapidly that $T_e \approx T_i$, and hence $Q_{e,i} = -Q_{i,e} \approx 0$. In the second limit, the collisions are significantly slower than a specific time-scale of interest τ ,

$$\frac{1}{\tau} \gg \delta \nu_{e,i}, \quad (2.2.7)$$

where the energy exchange factor is $\delta = 2m_e/m_i$, such that heat exchange can be ignored, $Q_{e,i} = -Q_{i,e} \approx 0$. In both cases, the energy equations can be integrated to derive the equation of state [25]

$$pn^{-5/3} = \text{Constant}, \quad (2.2.8)$$

This equation establishes a relation between p and n . It provides the closure of the system of fluid equations 2.2.1 and 2.2.2 into the particle and momentum conservation equations, which correspond to the first two components of them. In general, the equation of state can be expressed as $pn^{-\gamma} = \text{Constant}$ such that the corresponding energy equations for electrons and ions are simplified to

$$\begin{aligned} p_e n_e^{-\gamma_e} &= \text{Constant}, \\ p_i n_i^{-\gamma_i} &= \text{Constant}, \end{aligned} \quad (2.2.9)$$

where $\gamma = 1$ for the isothermal compression [25]. By indicating the number of degrees of freedom as N , the parameter γ becomes $(2 + N)/N$.

2.3 "Drift-diffusion" approximation

This section considers gas discharge plasmas that are weakly ionized and have low temperatures. The discharge is far from being in the state of local thermodynamic equilibrium (LTE), implying that the temperature of electrons is considerably higher compared to the temperatures of other species. In the relevant discharge models, it is commonly assumed that the temperature of all heavy particles is equal to the background gas temperature, and total pressure is represented by $p = Nk_B T$, where N denotes the density calculated by summing up n_i for all species i : $N = \sum_i n_i$. When the inertia terms on the left-hand side of equation 2.1.13 are neglected (and similarly in corresponding equations of 2.2.1 and 2.2.2) and set $B = 0$, the particle flux density for species i can be formulated as follow [25]:

$$\mathbf{\Gamma}_i = n_i \mathbf{u}_i = \text{sign}(q_i) \mu_i n_i \mathbf{E} - D_i \nabla n_i, \quad (2.3.1)$$

where the particle mobility and diffusion coefficients are defined as $\mu_i = |q_i| / m_i \nu_i$, and $D_i = k_B T_i \mu_i / q_i$ with collision frequency ν_i .

Suppose a plasma consisting of three species: electrons, positive ions, and neutral species, indicated by the subscripts e , i , and n , respectively. In the absence of a magnetic field, the corresponding momentum equations in the equation sets of 2.2.1 and 2.2.2 can be expressed in a more simplified form as proposed in [25]

$$\begin{aligned} \rho_e \frac{\partial \mathbf{u}_e}{\partial t} + \rho_e (\mathbf{u}_e \cdot \nabla) \mathbf{u}_e &= -\nabla p_e - e n_e \mathbf{E} \\ -m_e n_e \nu_{e,n} (\mathbf{u}_e - \mathbf{u}_n) - m_e n_e \nu_{e,i} (\mathbf{u}_e - \mathbf{u}_i), \end{aligned} \quad (2.3.2)$$

$$\begin{aligned} \rho_i \frac{\partial \mathbf{u}_i}{\partial t} + \rho_i (\mathbf{u}_i \cdot \nabla) \mathbf{u}_i &= -\nabla p_i - e n_i \mathbf{E} \\ -m_i n_i \nu_{i,e} (\mathbf{u}_i - \mathbf{u}_e) - m_i n_i \nu_{i,n} (\mathbf{u}_i - \mathbf{u}_n), \end{aligned} \quad (2.3.3)$$

where $\rho_{e,i}(\mathbf{r}, t) = m_{e,i} n_{e,i}(\mathbf{r}, t)$ represents the expression for the mass density, $\mathbf{u}_n$ is the mass-average velocity of the neutral gas atoms, and the electron and positive ion charges are taken as $q_e = -e$ and $q_i = e$.

When the condition $m_e \ll m_i$ is considered, it is reasonable to make the assumption that $\rho_e (\mathbf{u}_e \cdot \nabla) \mathbf{u}_e \ll \rho_i (\mathbf{u}_i \cdot \nabla) \mathbf{u}_i$. With this assumption, we can simplify the equation

2.3.2 as provided in [25] and [219],

$$-\nabla p_e - en_e \mathbf{E} - m_e n_e \nu_{e,n} (\mathbf{u}_e - \mathbf{u}_n) - m_e n_e \nu_{e,i} (\mathbf{u}_e - \mathbf{u}_i) = 0. \quad (2.3.4)$$

Given that $\mathbf{u}_e \gg \mathbf{u}_n, \mathbf{u}_i$, and $p_e = n_e k_B T_e$, this equation can be further simplified to

$$k_B T_e \nabla n_e + en_e \mathbf{E} + (m_e \nu_e) n_e \mathbf{u}_e = 0. \quad (2.3.5)$$

Also, it can be written as

$$\mathbf{\Gamma}_e = n_e \mathbf{u}_e = -\mu_e n_e \mathbf{E} - D_e \nabla n_e, \quad (2.3.6)$$

where the electron mobility and diffusion coefficients are defined as $\mu_e = e/(m_e \nu_e)$, and $D_e = k_B T_e \mu_e / e$, respectively, and the collision frequency of electrons is $\nu_e = \nu_{e,n} + \nu_{e,i}$.

When the momentum conservation for the ionic species is examined, the equation 2.3.3 can be written in the following form [25, 219]

$$-\nabla p_i - en_i \mathbf{E} - m_i n_i \nu_{i,e} (\mathbf{u}_i - \mathbf{u}_i) - m_i n_i \nu_{i,n} (\mathbf{u}_i - \mathbf{u}_n) = 0. \quad (2.3.7)$$

By neglecting the $\mathbf{u}_n$ and taking into account the relations $\nu_{i,e} m_i n_i (\mathbf{u}_i - \mathbf{u}_e) = -\nu_{e,i} m_i n_i (\mathbf{u}_i - \mathbf{u}_e)$, and $m_e \nu_e \ll m_i \nu_{i,n}$, the equation 2.3.7 becomes

$$\mathbf{\Gamma}_i = n_i \mathbf{u}_i = -\mu_i n_i \mathbf{E} - D_i \nabla n_i, \quad (2.3.8)$$

where $\mu_i = e/(m_i \nu_i)$, and $D_i = k_B T_i \mu_i / e$ are the mobility and diffusion coefficients of ions, respectively.

If the derived expressions from equations 2.3.6 and 2.3.8 for the flux densities, ($n_e \mathbf{u}_e$ and $n_i \mathbf{u}_i$), are substituted into corresponding particle balance equations, which are the first equations of 2.2.1 and 2.2.2, then "two-fluid" equations with "drift-diffusion" approximation are obtained as follows [25]:

$$\frac{\partial n_k}{\partial t} + \nabla \cdot (\text{sign}(q_k) \mu_k n_k \mathbf{E} - D_k \nabla n_k) = S_k. \quad (2.3.9)$$

Here, the subscript k denotes the species of particles such as electrons, ions, and neutral species. The term S_k represents the creation or destruction rate of species k through collision processes. To develop a self-consistent model, the continuity equations in the equation 2.3.9 are coupled with the Poisson equation for the electric field. Each chapter of this thesis employs the models developed from these equations and will be discussed in the following sections.

2.4 "Simple" and "Extended" Fluid Models

This section describes the "simple" and "extended" fluid models. As previously stated, the fluid modeling equations for glow discharge include the moments of the Boltzmann equation: continuity, momentum, and energy equations. These equations are coupled with the Poisson equation to derive a self-consistent electric field distribution. Moreover, fluid models' momentum equations are transformed into "drift-diffusion" equations for particle flux densities, as discussed in Section 2.3. These equations include the transport and rate coefficients, serving as input data for the fluid model [25, 103]. Since these coefficients account for the dynamics of the system through the collision of particles, determination of them plays a crucial role in the accuracy of the fluid model. Indeed, deriving electron transport and collisional reaction rates determines the reliability and accuracy of the fluid modeling since the electron dynamics are the least precisely known element in the model. "The Local Field Approximation" (LFA) and "Local Mean Energy Approximation" (LMEA) are commonly used techniques to describe the transport and rate coefficients of electrons in fluid modeling of gas discharge plasmas [25, 32]. In the LFA approach, the transport (mobility and diffusion) and rate coefficients are defined as a function of the local reduced electric field, E/p , where E is the electric field magnitude and p is the pressure. The fluid modeling based on this approximation of input parameters is known as the "simple fluid model". In addition, it is important to note that transport coefficients can be treated as constants by providing the Einstein relation, $D/\mu = k_B T/e$. (Chapter 3 of this thesis defines the transport coefficients of electrons as constants in the "simple fluid model", and Chapter 6 tests the accuracy and validity of this relation in treatments of electron dynamics.)

The equations for this modeling approach are [25]

$$\frac{\partial n_l}{\partial t} + \nabla \cdot \mathbf{\Gamma}_l = S_l, \quad (2.4.1)$$

$$\mathbf{\Gamma}_l = \text{sgn}(q_l) \mu_k n_l \mathbf{E} - D_l \nabla n_l, \quad (2.4.2)$$

$$\epsilon_0 \nabla \cdot \mathbf{E} = \sum_l q_l n_l, \quad \mathbf{E} = -\nabla \varphi, \quad (2.4.3)$$

where the subscript l indicates the l th species, such as electrons, ions, or metastable atoms. Γ_l represents the "drift-diffusion" approximation of particle flux density. For the electron and ion species, the source term can be defined as $S_e = S_i = \alpha |\Gamma_e| - \beta n_e n_i$, where $\alpha = \alpha(E)$ represents the first Townsend coefficient, or the rate coefficient of electron impact ionization, and β denotes the electron-ion recombination coefficient.

In fluid modeling of gas discharges, the "Simple fluid model" (LFA) is generally limited because of its inability to incorporate nonlocal ionization phenomena in the negative glow region. The "Local Field Approximation" (LFA) assumes that electron impact ionizations only occur in the cathode sheath region, where the electric field strength is high enough. However, there are ionizations in the regions, where the electric field strength is weak due to the presence of fast electrons from the cathode region. This phenomenon is known as "nonlocal-ionization" and is not considered within the LFA. Consequently, the source of electrons in the discharge tube is not only related with high electric field regions but also from the negative glow region due to nonlocal ionizations.

The "extended fluid model" approach, also referred to as the "Local Mean Energy Approximation" (LMEA) [32, 103] is developed to incorporate, to some extent, the nonlocal ionizations of electrons into fluid modeling. This approach extends the equations 2.4.1, 2.4.2, and 2.4.3 by adding an electron energy equation

$$\begin{aligned} \frac{\partial n_e}{\partial t} + \nabla \cdot \Gamma_e = & - e \Gamma_e \cdot E - \frac{3}{2} \frac{m_e}{m_g} \nu_{ea} n_e k_B (T_e - T_g) \\ & - \sum_j \Delta E_j R_j, \end{aligned} \quad (2.4.4)$$

where n_e represents the electron energy density, which is derived by $n_e = n_e \frac{3}{2} k_B T_e$, and Γ_e is the electron energy flux density, which is expressed as

$$\Gamma_e = -D_e \nabla n_e - \mu_e E n_e, \quad (2.4.5)$$

where the energy transport coefficients are related to the transport coefficients through the equations $D_e = (5/3)D_e$ and $\mu_e = (5/3)\mu_e$. Additionally, the right-hand side of equation 2.4.4 comprises source terms in various phenomena. These are Joule heating (or cooling) of electrons due to electric field and energy losses due to elastic and inelastic collisions. In this context, the electron-atomic elastic collision frequency is

denoted by ν_{ea} , R_j represents the reaction rate associated with energy loss (or gain) ΔE_j , m is the particle mass and T_g represents the background gas temperature.

The LMEA approach to fluid modeling defines electron transport and rate coefficients as functions of electron temperature (T_e) by solving the kinetic electron Boltzmann equation (Chapter 6 also develops an electron Boltzmann equation solver for a DBD system in CO_2). The lookup tables for these coefficients are constructed to store and interpolate through the fluid model as input parameters. Moreover, the spatial distributions of electron temperatures are derived by solving the electron energy equation 2.4.4. Unlike the LFA, which underestimates the ionizations occurring in regions with weak electric field strength, the LMEA defines input parameters by considering the mean electron energy and provides a more accurate representation of electron behavior in regions with varying electric field strengths.

2.5 Limitations and applicability

The comparison between the small mean free path (λ_i) and the characteristic system size (L) is the primary factor that determines the suitability of applying the fluid model for species i [25]. According to [26], the applicability condition is expressed as

$$L > \lambda_i. \quad (2.5.1)$$

It is important to note that the fluid model loses its applicability when considering distances from the boundaries shorter than the mean free path of the particles. Nevertheless, findings from the kinetic analysis reveal that it is possible to complete fluid model equations by formulating effective boundary conditions (Chapter 5 of this thesis investigates and analyzes the effects of BCs on fluid modeling of RF gas discharge systems in detail.).

Particles with similar mass, including neutral and excited atoms, molecules, and their respective ions, commonly show momentum and energy relaxations that happen typically within a single collision or a few collisions. Consequently, if the condition 2.5.1 is satisfied, these particles' Energy Distribution Functions (EDFs) closely approximate a Maxwellian distribution with a temperature set by the thermostat [25].

In contrast to neutral gas particles, charged particles experience the influence of an electric field that leads to heating of them (see the Joule heating term of the equation 2.4.4). Thus, these charged particles' temperatures or mean energies exceed the thermostat's temperature. In such scenarios, the EDFs of charged particles approximate a Maxwellian distribution only when the degree of ionization is sufficiently high (greater than 10^{-4} - 10^{-3}), where the relaxation of the particle energies is mainly defined through collisions between electrons and between ions [25].

In the case of ions subjected to a strongly applied electric field within a low ionized plasma (characterized by a ionization degree ranging from less than 10^{-4} to 10^{-3}), the EDF is non-Maxwellian, deviating from a Maxwellian EDF. Nevertheless, in the context of fluid equations, ion EDF's actual shape has a minimal impact on the determination of transport coefficients (mobility and diffusion) because these coefficients represent the collective properties of the entire ensemble of ions [25]. Therefore, the characteristics of ions can be approximated using a Maxwellian EDF, while accounting for the temperature determined through energy balance considering the heating effects caused by the electric field [25–27].

Electrons are highly mobile particles with a small mass compared to heavy particles; therefore, relaxation processes for momentum and energy demonstrate significant differences. When electrons collide with weighty particles, energy relaxation length (λ_e) and time (τ_e) are significantly larger than electron mean free path (λ) and corresponding time ($1/\nu_{ea}$), respectively. Specifically, in the context of elastic collisions, the conditions described below are applicable [25, 26]:

$$\lambda_e > 100\lambda, , \quad (2.5.2)$$

$$\tau_e > 100^2/\nu_{ea}. \quad (2.5.3)$$

In situations where the electric field is weak, the acquired energy of an electron across the distance of energy relaxation is negligible relative to the temperature of more massive particles. In this case, the EEDF resembles a Maxwellian distribution with a temperature equivalent to heavy particles. However, under the influence of strong electric fields, the characteristics of the EEDF are affected by the degree of ionization.

When the degree of ionization is relatively high, the EEDF approximates a Maxwellian distribution with the electron partial temperature. In this case, it is reasonable to introduce a system of three equations for electron density, momentum, and energy, which were explained in detail in previous sections of this chapter. This modeling approximation is known as the "standard fluid approach" [25,26].

For lower degrees of ionization, the situation becomes notably more intricate and depends on multiple factors, including the ionization degree, collision features, and electric field strength. When the ionization degree is low, the EEDF shows significant deviations from a standard Maxwellian distribution, mainly when strong electric fields are present. Under such conditions, a kinetic description for electrons should be used to accurately capture electron behavior [25].

Depending on the specific conditions, the energy distribution functions (EDFs) of fluid particles, mainly electrons, can show Maxwellian and non-Maxwellian behavior. In the following chapters, where fluid models are developed, both Maxwellian and non-Maxwellian EDFs will be employed for electrons to define transport and rate coefficients, allowing for comparing the obtained results and assessing their impact on the system's behavior. It is crucial to emphasize that accurately describing the electrons' dynamics, such as their nonlocal nature, often requires using kinetic models. While the "Local Mean Energy Approximation" (LMEA) approach provides some insights into the nonlocal dynamics of electrons when compared to the "Local Field Approximation" (LFA), it is essential to acknowledge its limitations. By incorporating kinetic models, a more comprehensive understanding of electron behavior can be achieved, enabling a more accurate representation of the interactions and contributions to the overall system dynamics.

In conclusion, this chapter has investigated the fluid modeling of glow discharge by deriving the fluid equations derived from the moments of the kinetic Boltzmann equation. The continuity, momentum, and energy equations have been obtained to understand the behavior of the "two-fluid" modeling of plasma. We have derived a self-consistent model for the electric field by integrating the Poisson equation with "two-fluid" equations. Utilizing the "drift-diffusion" approximation of particle flux densities, combining momentum and continuity equations has provided a single equation

used to define "simple" and "extended" fluid models. The strengths and limitations of these two fluid modeling approaches are compared with each other regarding meeting the glow discharge's fundamental needs. In the following chapters, these fluid model equations will form the backbone of the one-dimensional plasma discharge models.





CHAPTER 3

TRANSITION FROM PERIODIC TO CHAOTIC OSCILLATIONS IN A PLANAR SEMICONDUCTOR-GAS DISCHARGE SYSTEM

In this chapter, the transition from periodic to irregular chaotic oscillations in a dc-driven "barrier" discharge, comprised of planar gas discharge and a semiconductor layer, is investigated. The analysis is focused on oscillations that occur in the subnormal discharge regime of spatially homogeneous one-dimensional fluid models. The "drift-diffusion" approximation of gas discharges is employed in developing numerical models called the "simple fluid model" and the more detailed "extended fluid model". The applied total voltage and the semiconductor layer resistance are considered two control parameters. To characterize the transition from regular to irregular regimes through period-doubling bifurcations, two illustrative figures, namely, the bifurcation diagram and the Lorenz map, are derived. The effects of the two modeling approaches in the analysis are also addressed. Although the dynamical behavior of the system obtained by the "simple" and "extended" fluid models have qualitatively similar results, they have differences in quantitative perspectives as a function of the control parameters. We introduce the statements of the problem in section 3.1. The models and the parameter regime are described in section 3.2. The calculated results are discussed in section 3.3. Finally, conclusions to the problem are presented in section 3.4.

3.1 The statements of the problem

The appearance of chaos in laboratory plasmas draws the attention of researchers, and it has become a heavily active research area for the last couple of decades. Tem-

poral chaos, pure oscillations in low dimensional cases, and spatio-temporal chaos, perceived as weakly developed turbulence showing itself as a wave, are generally considered two kinds of chaotic behavior in dynamical systems [220,221]. In practical situations, such as plasma devices, chaos and turbulence states of the systems are usually seen as undesired features [221]. Indeed, in the applications where cycle-to-cycle replicas are necessary, chaotic behavior may be unwanted [222]. It shows us the importance of controlling and understanding chaos phenomena in gas discharge plasma systems. Several methods have been developed for controlling over complex behavior of nonlinear dynamical systems to treat instabilities and chaos within plasma devices [221,222].

In this chapter, we will investigate the transition from a periodical to chaotic oscillations in a laterally extended dc-driven system of planar gas discharge in nitrogen, coupled to a semiconductor layer (see figure 3.1). This coupled system is known as a gas discharge–semiconductor system (GDSS), extensively studied both numerically and experimentally (see, e.g., reviews [54,68,223]).

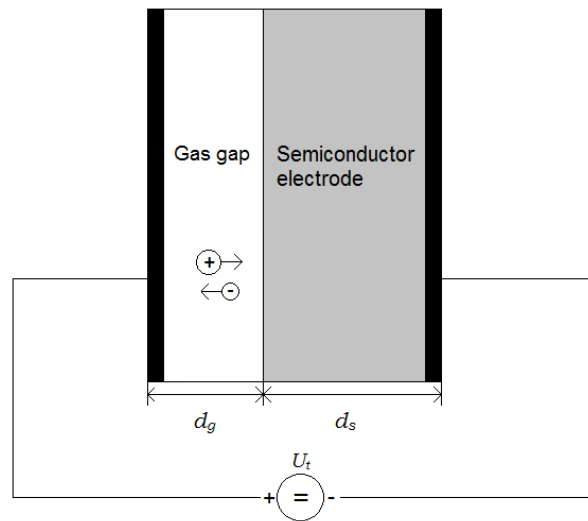


Figure 3.1: The schematic illustration of a planar gas discharge coupled with a semiconductor layer.

The research teams of Astrov (St. Petersburg, Russia) and Purwins (Münster, Ger-

many) are two well-known research groups performing experiments on GDSS. It was discovered that semiconductor–gas discharge system has the nature of showing complex dynamical behavior. These behavior emerge in the form of stationary mode, which is homogeneous in the transverse direction, oscillatory mode, as well as spatial and spatio-temporal patterns. The structures can be classified as stripes, hexagons, spirals, rings, etc., and also evolve as chaotic and solitary waves [54, 68, 223].

Underlying physical reasons for selforganization and pattern formation phenomena in the Townsend and glow discharges were discussed in [223] (see also [53, 62]). When the applied voltage is plotted as a function of the discharge current, and the slope representing the differential resistivity becomes negative, the homogeneous stationary state of a charge-carrying discharge medium may lose its stability and become unstable [53, 61]. The sustainability of this kind of discharge is observed with increasing the discharge current associated with decreasing discharge voltage. As a result, the negative differential resistivity (NDR) manifests itself with having a negative slope branch, $dU/dI < 0$, on the current-voltage characteristic (CVC). This region corresponds to a regime between the Townsend and normal glow discharge on CVC. It is known as a subnormal regime in the classical glow discharge. Based on the NDR principle, the oscillations and instabilities are usually observed in this regime (see, e.g., [31, 62–65]). The right branch of the Paschen curve is an account for the development of instabilities. The mechanism for the NDR and related instabilities occurring in the discharges are described with distortion of the spatial profile of electric field E , accompanied by a strong electric field increasing dependence of the ionization coefficient, α . These instabilities, occurring at adequately high current densities and filamentation, are responsible for emerging of stationary and dynamical structures by disturbing the homogeneity in the transversal direction. In addition, it is stated in [223] that the temporal and spatial chaotic current oscillations observed in GDSS are based on this mechanism.

A previous study has experimentally confirmed and demonstrated the emergence of instabilities within GDSS [53]. This experiment's considered system is similar to ours, including the gas-discharge gap and semiconductor cathode layer. In this study, a monotonic CVC of the semiconductor component, defined by the gas pressure p and discharge gap length d , has been derived, and the resulting instabilities were attributed

to appearing N-type NDR in the CVC [53]. If $pd > e \cdot (pd)_{\min}$, N-type NDR in the CVC exists, and if $pd < e \cdot (pd)_{\min}$, where $(pd)_{\min}$ corresponds to Paschen curve's minimum, then the CVC is monotonic [224].

We have a different scenario if the semiconductor electrode has a nonmonotonic CVC. More precisely, if the semiconductor component is in the regime of S-type NDR, a prominent instability emerges in a semiconductor domain, which may lead to dynamical patterns in the gas discharge part of the system [53].

The thermal destabilization mechanism also plays a role in the appearance of instabilities in the GDSS. Especially under cryogenic conditions, at which the current densities are too small to redistribute the electric field inside the discharge gap, the current filamentation emerges in the Townsend regime of the GDSS [223, 225, 226]. Moreover, the increasing dependency on the electric field value E at the cathode with secondary emission coefficient γ gives rise to another destabilization mechanism, which manifests as homogeneous self-sustained oscillations in the GDSSs [63–65, 223].

This chapter will focus on the subnormal oscillations obtained from a spatially homogeneous one-dimensional fluid model. The model only considers variations in one direction, perpendicular to planar electrode layers, and ignores the spatial changes parallel to the layers (transversal directions). Numerous investigations of subnormal oscillations in the glow discharges have been theoretically and experimentally made in many publications (see, e.g., [31, 63–65]). However, these systems' transitions from regular to irregular oscillations and chaotic behavior are still not precisely understood. The generally accepted view is that period-doubling bifurcation is a typical route to temporal chaos [67, 227, 228], and it is observable in various gas discharge systems [221, 222, 229–233]), including gas discharge–semiconductor systems [55, 68, 69]. It should be noted that experimental conditions to observe the onset of period-doubling bifurcations in [69] are similar to those used in our study. The period-doubling bifurcations in GDSS were numerically described in [234] and examined in detail in [71]. More precisely, in [71], the bifurcation diagram and Lorentz map were derived to characterize the transition of GDSS to chaos through period-doubling bifurcations. This chaos transition in [71] has displayed an excellent period-doubling cascade with a substantial similarity with simple nonlinear mappings, such

as the logistic map [72].

In [71, 234], the numerical models, based on the plasma fluid equations with a "drift-diffusion" approximation for particle fluxes, are called the "simple fluid model". In this model, only two species were considered, namely, electrons and positive ions. The ionization rate was determined as a function of the reduced electric field, E/p , by using the Townsend formula [224]. This approach achieves describing the basic discharge properties in a self-consistent way. However, this kind of numerical model ignores the nonlocal ionizations occurring in the discharge regions, where the electric field is weak [103]. In this chapter, the studies given in [71, 234] are extended by using a more detailed and realistic fluid model, called "extended fluid model", also known as "Local Mean Energy Approximation" (LMEA) [32]. Within this approach, the electron transport coefficients (mobility and diffusion coefficients) and electron-induced reaction rate coefficients are defined as functions of the mean electron energy obtained from the solution of the electron Boltzmann equation [3, 8, 32, 103]. The model's results based on LMEA but having the Maxwellian EEDF are also presented and discussed.

3.2 Description of Gas Discharge Model

3.2.1 Governing equations

The gas discharge model involves the continuity equations for each of the plasma components,

$$\frac{\partial n_l}{\partial t} + \frac{\partial \Gamma_l}{\partial x} = S_l, \quad (3.2.1)$$

with the "drift-diffusion" approximation for the particle flux densities Γ_l ,

$$\Gamma_l = \text{sgn}(q_l) \mu_l n_l E - D_l \frac{\partial n_l}{\partial x}, \quad (3.2.2)$$

the Poisson equation for the electric field in electrostatic approximation,

$$-\epsilon_0 \frac{\partial^2 \varphi}{\partial x^2} = \sum_l q_l n_l, \quad (3.2.3)$$

and the electron energy equation is also included

$$\begin{aligned} \frac{\partial n_e}{\partial t} + \frac{\partial \Gamma_e}{\partial x} = & - e\Gamma_e E - \frac{3}{2} \frac{m_e}{m_g} \nu_{ea} n_e k_B (T_e - T_g) \\ & - \sum_j \Delta E_j R_j. \end{aligned} \quad (3.2.4)$$

In these equations, n is the particle number density, φ and $E = -\partial\varphi/\partial x$ represent the electric potential and field. μ and D indicate the particle mobility and diffusion coefficients, and the subscript l stands for the sort of species (such as electrons, ions, metastable molecules, etc.). In the electron energy equation, $n_e = n_e \bar{\epsilon}$ is the electron energy density, where $\bar{\epsilon} = (3/2)k_B T_e$ is the mean electron energy, and Γ_e denotes the electron energy flux density,

$$\Gamma_e = -D_e \partial n_e / \partial x - \mu_e E n_e. \quad (3.2.5)$$

Energy transport coefficients for electrons are related to the electron transport coefficients via $D_e = 5/3 D_e$ and $\mu_e = 5/3 \mu_e$ [8]. In the electron energy equation, ν_{ea} is the electron-atomic elastic collision frequency, R_j describes the reaction rate of the j th reaction, ΔE_j is the corresponding energy loss (or gain) due to inelastic collision, m is the particle mass, and kinetic temperature is T .

The source terms S_l located on the right-hand side of the continuity equations (3.2.1), are determined by plasma-chemical reactions occurring in the discharge,

$$S_l = \sum_i R_i - \sum_j R'_j, \quad (3.2.6)$$

where R_i and R'_j are the creation and destruction rates in i th and j th reactions. These rate terms are proportional to the number densities of the reacting components and the rate coefficients of the corresponding reactions.

Within this model, the transport coefficients for electrons (mobility and diffusion coefficients) as well as electron-induced rate coefficients are defined as functions of the mean electron energy utilizing "lookup tables" obtained from the solution of the electron Boltzmann equation [8]. Calculations are performed for a nitrogen gas, and seven plasma species, namely, electrons, N_2^+ ions, excited nitrogen molecules $N_2(a'1)$, $N_2(A3, v0 - 4)$, $N_2(B3)$, $N_2(C3)$, and N_2 molecules in ground state are taken into account. The set of reactions is summarized in table 3.1, where the Phelps

Table 3.1: Set of plasma chemical reactions used in the "extended fluid model".

Index	Reaction	$\Delta E(eV)$	Rate coefficient	Ref.
R1	$e + N_2 \rightarrow e + N_2$	0	Cross section	[235]
R2	$e + N_2 \rightarrow 2e + N_2^+$	15.60	Cross section	[235]
R3	$e + N_2 \rightarrow e + N_2(a'1)$	8.40	Cross section	[235]
R4	$e + N_2 \rightarrow e + N_2(A3, v0 - 4)$	6.17	Cross section	[235]
R5	$e + N_2 \rightarrow e + N_2(B3)$	7.35	Cross section	[235]
R6	$e + N_2 \rightarrow e + N_2(C3)$	11.03	Cross section	[235]
R7	$e + N_2 \rightarrow e + N_2(A3, v5 - 9)$	7.00	Cross section	[235]
R8	$e + N_2 \rightarrow e + N_2(A3, v10 -)$	7.80	Cross section	[235]
R9	$e + N_2 \rightarrow e + N_2(a''1)$	12.25	Cross section	[235]
R10	$e + N_2 \rightarrow e + N_2(a1)$	8.55	Cross section	[235]
R11	$e + N_2 \rightarrow e + N_2(B'3)$	8.16	Cross section	[235]
R12	$e + N_2 \rightarrow e + N_2(E3)$	11.87	Cross section	[235]
R13	$e + N_2 \rightarrow e + N_2(w1)$	8.89	Cross section	[235]
R14	$e + N_2 \rightarrow e + N_2(W3)$	7.36	Cross section	[235]
R15	$e + N_2 \rightarrow e + N_2(rot)$	0.02	Cross section	[235]
R16	$e + N_2 \rightarrow e + N_2(v1res)$	0.29	Cross section	[235]
R17	$e + N_2 \rightarrow e + N_2(v2)$	0.59	Cross section	[235]
R18	$e + N_2 \rightarrow e + N_2(v8)$	2.35	Cross section	[235]
R19	$e + N_2 \rightarrow e + N_2(v3)$	0.88	Cross section	[235]
R20	$e + N_2 \rightarrow e + N_2(v4)$	1.17	Cross section	[235]
R21	$e + N_2 \rightarrow e + N_2(v5)$	1.47	Cross section	[235]
R22	$e + N_2 \rightarrow e + N_2(v6)$	1.76	Cross section	[235]
R23	$e + N_2 \rightarrow e + N_2(v7)$	2.06	Cross section	[235]
R24	$e + N_2 \rightarrow e + N_2(sum)$	13	Cross section	[235]
R25	$N_2(A3, v0 - 4) + N_2(A3, v0 - 4) \rightarrow N_2(B3) + N_2$	0	$7.7 \times 10^{-17} m^3 s^{-1}$	[236]
R26	$N_2(B3) + N_2 \rightarrow N_2(A3, v0 - 4) + N_2$	0	$5.0 \times 10^{-17} m^3 s^{-1}$	[236]
R27	$N_2(A3, v0 - 4) + N_2(A3, v0 - 4) \rightarrow N_2(C3) + N_2$	0	$3.0 \times 10^{-16} m^3 s^{-1}$	[236]
R28	$N_2(C3) + N_2 \rightarrow N_2(a'1) + N_2$	0	$1.0 \times 10^{-17} m^3 s^{-1}$	[236]
R29	$N_2(a'1) + N_2 \rightarrow N_2(B3) + N_2$	0	$2.0 \times 10^{-19} m^3 s^{-1}$	[236]
R30	$N_2(a'1) + N_2 \rightarrow 2N_2$	0	$2.0 \times 10^{-19} m^3 s^{-1}$	[236]
R31	$N_2(A3, v0 - 4) \rightarrow N_2 + h\nu(293nm)$	0	$5.0 \times 10^{-1} s^{-1}$	[237]
R32	$N_2(B3) \rightarrow N_2(A3, v0 - 4) + h\nu(1045nm)$	0	$1.5 \times 10^5 s^{-1}$	[237]
R33	$N_2(C3) \rightarrow N_2(B3) + h\nu(336.5nm)$	0	$2.7 \times 10^7 s^{-1}$	[237]
R34	$N_2(a'1) \rightarrow N_2 + h\nu(177.1nm)$	0	$1.0 \times 10^2 s^{-1}$	[237]

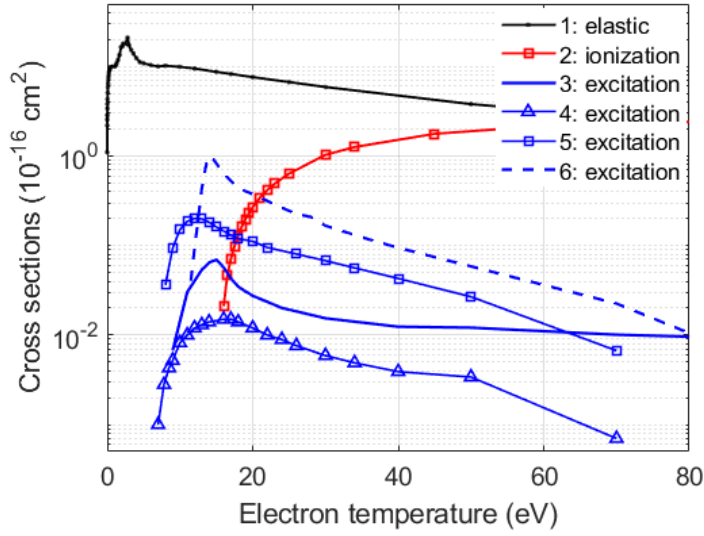


Figure 3.2: Electron cross sections for the first six reactions in table 3.1: (1) elastic, (2) ionization, (3), (4), (5), and (6) excitation collisions in nitrogen used in the model. Numbering corresponds to positions of corresponding processes in table 3.1.

database [235] notation is used. The electron collision cross sections for the first several reactions in table 3.1 are also demonstrated in figure 3.2. The transport coefficients of heavy species are approximated by constants. The mobility coefficient of nitrogen ions is $1140/p \text{ cm}^2 \text{ Torr V}^{-1} \text{ s}^{-1}$, and it is related to diffusion coefficient via the Einstein relation, $D/\mu = k_B T/e$, where p and T indicate the background gas pressure and kinetic temperature. The diffusion coefficient values of the excited nitrogen molecules are defined as $107.19/p \text{ cm}^2 \text{ Torr s}^{-1}$. It should be pointed out that computed current-voltage characteristics, performed in [3] by considering the similar plasma-chemical model of DC-supplied nitrogen discharge, but including fewer reactions, has an almost quantitative agreement with experimental results.

3.2.2 Boundary conditions

The boundary conditions are displayed in table 3.2 [3, 101]. Here, $v_l = \sqrt{8k_B T_l / \pi m_l}$ is the thermal velocity of species l , particle flux density Γ is defined by the equations (3.2.2) and (3.2.5), and γ is the secondary emission coefficient. a is a switching function, which is set to one if the direction of the electric field is toward the surface;

Table 3.2: Boundary conditions used in the "extended fluid model".

	Anode ($x = 0$)	Cathode ($x = d$)
Electrons, n_e	$\Gamma_e = -\frac{1}{4}v_e n_e$	$\Gamma_e = \frac{1}{4}v_e n_e - \gamma \Gamma_i$
Ions, n_i	$\Gamma_i = -\frac{1}{4}v_i n_i + a n_i \mu_i E$	$\Gamma_i = \frac{1}{4}v_i n_i + a n_i \mu_i E$
Excited species, n_m	$\Gamma_m = -\frac{1}{4}v_m n_m$	$\Gamma_m = \frac{1}{4}v_m n_m$
Electron energy density, n_e	$\Gamma_\epsilon = -\frac{1}{3}v_e n_e$	$\Gamma_\epsilon = \frac{1}{3}v_e n_e - 2k_B T_e \gamma \Gamma_i$
Electric potential, φ	U	0

otherwise, it is zero. The electron energy boundary conditions are consistent with the boundary conditions for the electron density via the relation $\Gamma_\epsilon = 2k_B T_e \Gamma_e$.

3.2.3 "Simple fluid model"

We also added the results derived from the "simple fluid model" for gas discharge in nitrogen, which was employed in many numerical models of GDSS [71,223,226,234, 238–241]. Only two species (electrons and positive nitrogen ions) are considered in this modeling approach. Their mobility coefficients are approximated by constants, $\mu_e p = 2 \times 10^5 \text{ cm}^2 \text{ Torr V}^{-1} \text{ s}^{-1}$ and $\mu_i p = 699 \text{ cm}^2 \text{ Torr V}^{-1} \text{ s}^{-1}$, and related diffusion coefficients D_e and D_i are determined by using the respective Einstein relations. Townsend formula is used to approximate the ionization rate as a function of the reduced electric field, $\alpha = A p \exp(-B p / |E|)$, where p is the gas pressure, A and B are the constants specified as $A = 12 \text{ cm}^{-1} \text{ Torr}^{-1}$ and $B = 343 \text{ V cm}^{-1} \text{ Torr}^{-1}$ [224].

In the "simple fluid model", boundary conditions for the electrons and ions at the anode ($x = 0$) are represented by the equations

$$D_e \frac{\partial n_e}{\partial x} = 0 \text{ and } n_i = 0, \quad (3.2.7)$$

and at the cathode ($x = d$)

$$\mu_e n_e = \gamma \mu_i n_i \text{ and } D_i \frac{\partial n_i}{\partial x} = 0, \quad (3.2.8)$$

where γ is the secondary emission coefficient.

3.2.4 Coupling of a semiconductor cathode layer

To establish a one-dimensional GDSS model, an external circuit equation, in the form of the RC circuit, must be added to the system of the gas discharge equations. When a semiconductor layer with a thickness d_s , conductivity σ_s , and dielectric constant ϵ_s is considered, the total current density, $J(t)$, is

$$\epsilon_s \epsilon_0 \frac{\partial E_s(t)}{\partial t} + J_s(t) = J(t), \quad (3.2.9)$$

where $J_s(t) = \sigma_s E_s(t)$. Since the coupled system forms a series-connected circuit, the total current density $J(t)$ does not change while passing from the gas discharge region to the semiconductor layer within the one-dimensional approximation. Then, we have

$$\epsilon_0 \frac{\partial E}{\partial t} + e(\Gamma_i - \Gamma_e) = J(t). \quad (3.2.10)$$

By using the relations $U_s(t) = R_s J_s(t)$ and $U_s(t) = E_s(t) d_s$, the equation (3.2.9) becomes

$$\tau_s \frac{\partial U(t)}{\partial t} = U_t - U(t) - R_s J(t), \quad (3.2.11)$$

where $U(t)$ is the discharge voltage, $U_t = U(t) + U_s(t)$ corresponds to the total stationary voltage, $R_s = d_s / \sigma_s$ is the resistance of the semiconductor, $C_s = \epsilon_s \epsilon_0 / d_s$ denotes the capacitance per area, and $\tau_s = C_s R_s = \epsilon_s \epsilon_0 / \sigma_s$ is the Maxwell time scale of the semiconductor.

3.2.5 Parameter regime

The input parameters were chosen by considering the parameters defined as in [71, 234]. The pressure of the nitrogen gas is $p = 40$ mbar, the discharge gap is $d = 1.4$ mm, GaAs semiconductor layer with a length of $d_s = 1.5$ mm and dielectric constant $\epsilon_s = 13.1$ is used, and the background gas is at the room temperature, $T_g = 297$ K. The semiconductor conductivity value takes values in the interval from $\sigma_s = (6.50 \times 10^6 \Omega \text{ cm})^{-1}$ to $(1.11 \times 10^5 \Omega \text{ cm})^{-1}$. In the computations, the secondary emission coefficient is defined as $\gamma = 0.08$.

The applied total voltage U_t and semiconductor resistance R_s act as control parameters. By aligning these two, we constrain the discharge system to operate in the

regime where it corresponds to the transition from Townsend to glow discharge. Note that the product $pd \approx 4.2$ Torr cm produces a greater value than the critical value of $e \cdot (pd)_{\min}$, where p and d are pressure and discharge gap, and $(pd)_{\min} \approx 1$ Torr cm is the minimum of the Paschen curve. Because of this, our gas discharge system corresponds to the right-hand side of the Paschen curve's minimum, which implies the appearance of the N-type NDR in the system [224].

3.3 Results and discussions

In section 3.1, we have generally introduced the destabilizing mechanisms responsible for exhibiting complex dynamical structures and patterns in planar gas discharges. Before representing and discussing the results, we focus on determining the basic destabilization mechanisms, which play a vital role in the nonlinear nature of the developed models, or can be excluded from our attention. Firstly, all the models used in this chapter are spatially one-dimensional, which means that there is homogeneity in the transverse directions (parallel to the gas discharge and semiconductor layers). Therefore, the spatial variations in the transversal direction and any filamentation are beyond the scope of this chapter. Secondly, the effects of the semiconductor on destabilizing are excluded because the semiconductor layer is assumed to be linear. Thirdly, the gas temperature is defined as a constant value, $T_g = 297$ K, due to ignoring the background gas heating; consequently, the thermal destabilization mechanism stays irrelevant. Finally, a constant and field-independent secondary emission coefficient $\gamma = 0.08$ is used; thereby, its effect on the NDR of the discharge is not taken into account. As a result, the space-charge distortion stemming from the electric field remains an effective destabilization mechanism on the NDR of the discharge system.

Since the sustainability of the near-cathode space-charge sheath is based on the mechanism, named as, *the cathode sheath instability* in [62], the physical principles of this maintenance are needed to be addressed. This particular region of a glow discharge is self-sustained by recurring processes in order: (i) electrons are emitted from the cathode surface by the bombardment of the ions, (ii) these ejected electrons are accelerated by the electric field of the sheath region (iii) the accelerated and high energetic electrons lead to ionizations of the neutral atoms and molecules, and (iv) the released

ions are accelerated by sheath electric field. These four processes in a given order play a vital role in the self-sustainability of a glow discharge. In [62], the glow discharge system having an N-type of CVC is seen as a typical bistable system having a strong positive feedback stemming from the near-cathode space-charge sheath. However, in the literature, this approach, known as a redistribution of the field because of space-charge, serves as an alternative way of interpreting the same destabilizing mechanism based on the increasing dependence between the Townsend ionization coefficient and local reduced electric field.

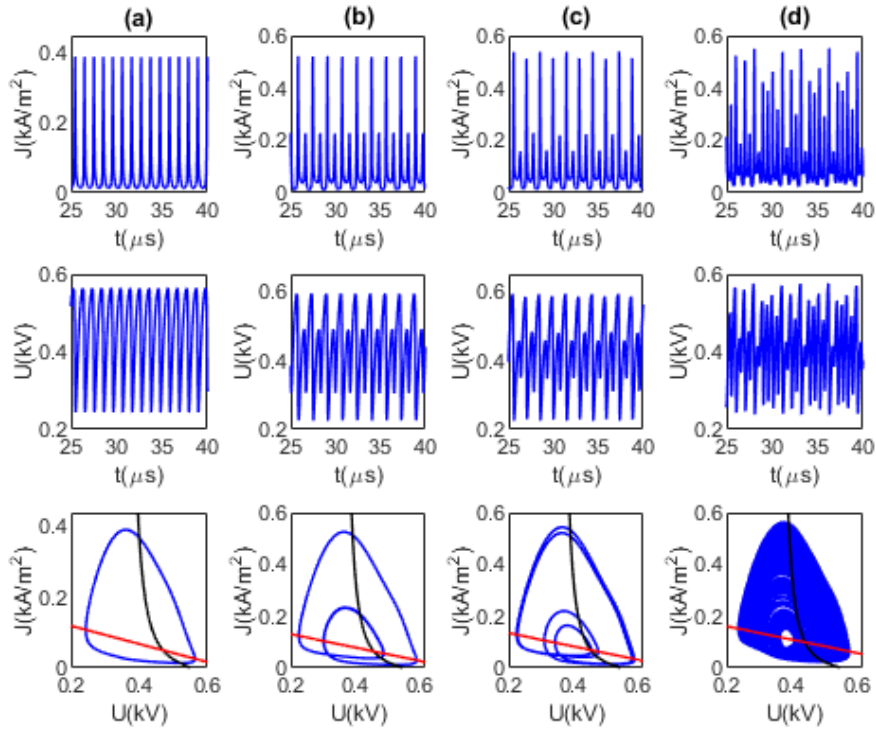


Figure 3.3: The first two rows show the time oscillations of the discharge current J and the voltage U , and the last row represents the corresponding phase portraits of these oscillations. The calculations are carried out at the values of the total applied voltage $U_t = 665$ V (a), 708 V (b), 725 V (c), and 827 V (d), and the semiconductor conductivity $\sigma_s = (2.6 \times 10^5 \Omega \text{ cm})^{-1}$. The two additional lines are the load line $U = U_t - R_s J$ and the current-voltage characteristics (CVC) of the gas discharge $U = U(J)$.

Figure 3.3 illustrates the numerical results of the transition from a periodic oscillatory

regime to a fully chaotic state in the GDSS model via the period-doubling bifurcations. In these calculations, the conductivity of the semiconductor was chosen as $\sigma_s = (2.6 \times 10^5 \Omega \text{ cm})^{-1}$. The applied total voltage U_t is a bifurcation parameter because R_s holds constant. The temporal oscillations of the discharge current J and voltage U at $U_t = 665 \text{ V}$ (a), 708 V (b), 725 V (c), and 827 V (d) are demonstrated in the first and second rows of this figure. The corresponding phase portraits, including two additional lines, namely, the current-voltage characteristics (CVC) $U = U(J)$ and the load line $U = U_t - R_s J$, are shown in the third row. The curves for each modeling approach are also plotted separately in figure 3.4. The intersection point of these two additional lines represents the stationary solution of the GDSS model regarding discharge current and voltage. Indeed, the calculations initiated at this point represent the system's equilibrium state. It should be pointed out that the calculations are started in the close neighborhood of the equilibrium point, and the initial transient regimes are omitted in all plots of the figure 3.3. The regime displayed in panels (d) shows fully chaotic oscillations, and this shows us, as it was stated in section 3.1, a series of period-doubling bifurcations is generally considered a typical route to temporal chaos.

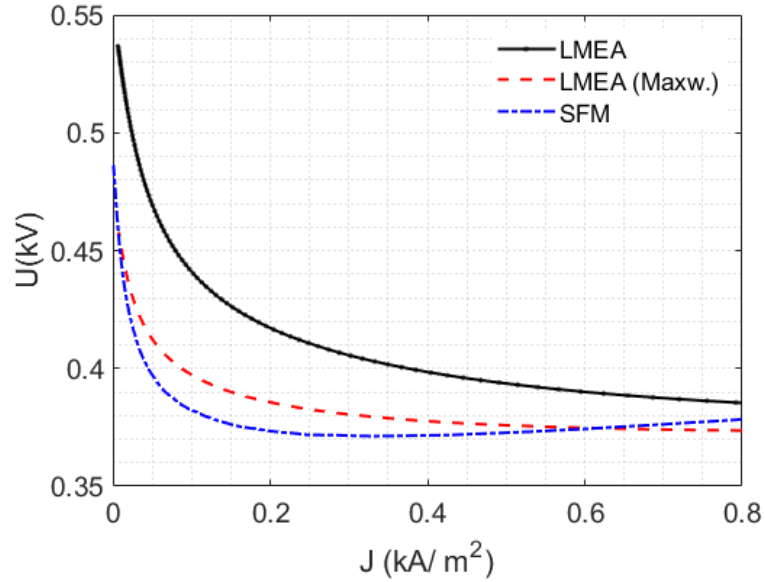


Figure 3.4: Current-voltage characteristics (CVCs) curves derived from the "simple" and "extended" fluid models.

Together with the periodicity in the time evolution of the ion density profile $n_i(x, t)$

at the total applied voltage $U_t = 665 \text{ V}$ and the semiconductor conductivity $\sigma_s = (2.6 \times 10^5 \Omega \text{ cm})^{-1}$, which corresponds to the regime (a) in figure 3.3, the aperiodicity of the respective profile at $U_t = 827 \text{ V}$ is demonstrated in figure 3.5, which corresponds to the regime (d) in figure 3.3.

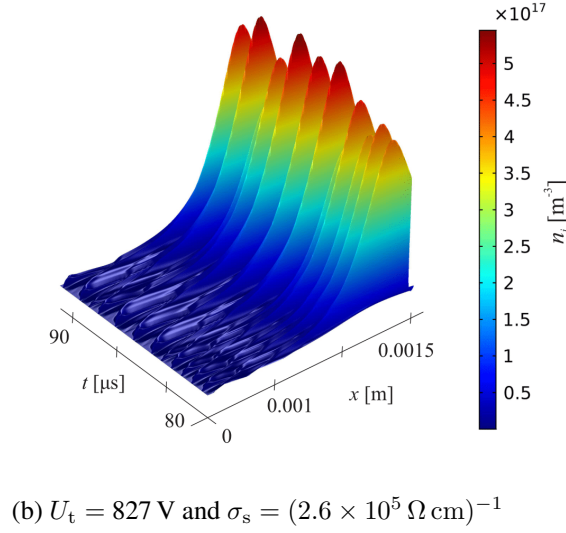
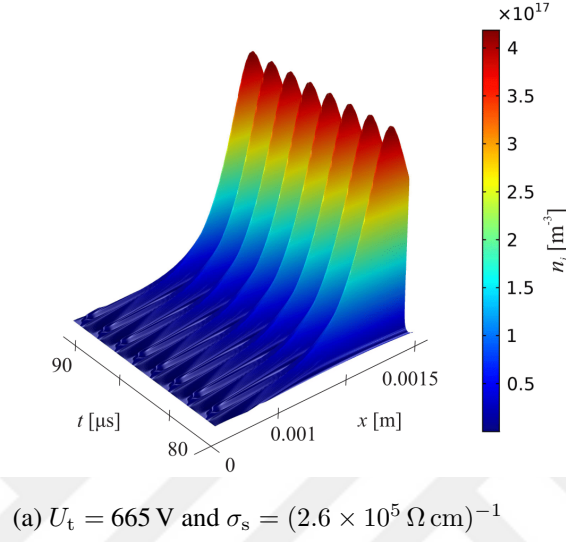


Figure 3.5: Profile of the ion density $n_i(x, t)$ as functions of time and spatial position at the total applied voltages $U_t = 665 \text{ V}$ and 827 V and the semiconductor conductivity $\sigma_s = (2.6 \times 10^5 \Omega \text{ cm})^{-1}$. These results correspond to (a) and (d) regimes in figure 3.3.

In figure 3.6, the onset of period-doubling bifurcation cascade is demonstrated in the space of discharge current density $J(t)$ and the total applied voltage U_t . This bifurcation diagram shows the local maximums of the current density oscillations against the bifurcation parameter, U_t . The driving of a periodic system with increasing the bifurcation parameter U_t leads to the system's transition to chaotic oscillations. It is a process where a new limit cycle emerges from the old one such that the period of the new limit cycle is twice that of the existing one [242] as it is seen from the regimes (a)–(c) depicted in figure 3.3. The numerical procedure includes obtaining the system's equilibrium (stationary) state for each value of the control parameter U_t for a constant semiconductor resistance. The corresponding oscillations, shown in figures 3.3(a)–(c), are derived by a small perturbation from equilibrium states, and the vertical lines in figure 3.6 denote the values of $U_t = 665, 708$, and 725 V corresponding to the regimes (a)–(c) in figure 3.3.

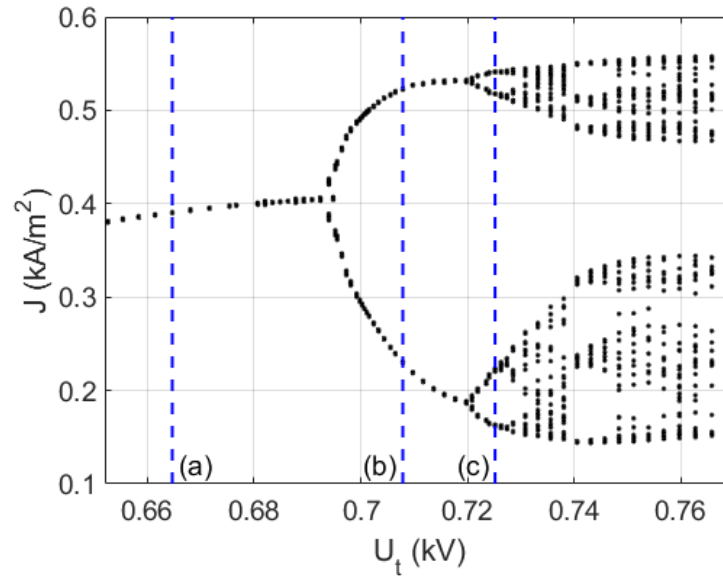


Figure 3.6: The period-doubling bifurcation diagram in the space of the discharge current density J and total applied voltage U_t . The regimes (a), (b), and (c) correspond to the regimes specified with the same labels in figure 3.3.

The Lorenz map, displaying the data points of the $(n+1)$ th local maximum of the discharge current $J(t)$ against its n th maximum, derived for the regimes corresponding to (a)–(d) with applied voltages $U_t = 665, 708, 725$, and 827 V. This map is illustrated in figure 3.7. In accordance with the results demonstrated in figures 3.3 and

3.6, the computed data points at $U_t = 665, 708, 725$, and 827 V in figure 3.7 indicate a "period" of corresponding Lorenz attractor. Indeed, the one point at $U_t = 665$ V, two points at $U_t = 708$ V, four points at $U_t = 725$ V correspond to period-1, period-2, and period-4 attractors, respectively. In the case of $U_t = 827$ V (corresponding to the regime (d) in figure 3.3), when the system has fully chaotic oscillations, the data points do not have the shape of a well-structured one-dimensional curve. This curve resembles the well-known logistic map in agreement with Feigenbaum's universality theory [243]. In figure 3.7, the maps obtained from the corresponding chaotic regimes of the LMEA model with Maxwellian EEDF and "simple fluid model" are also presented.

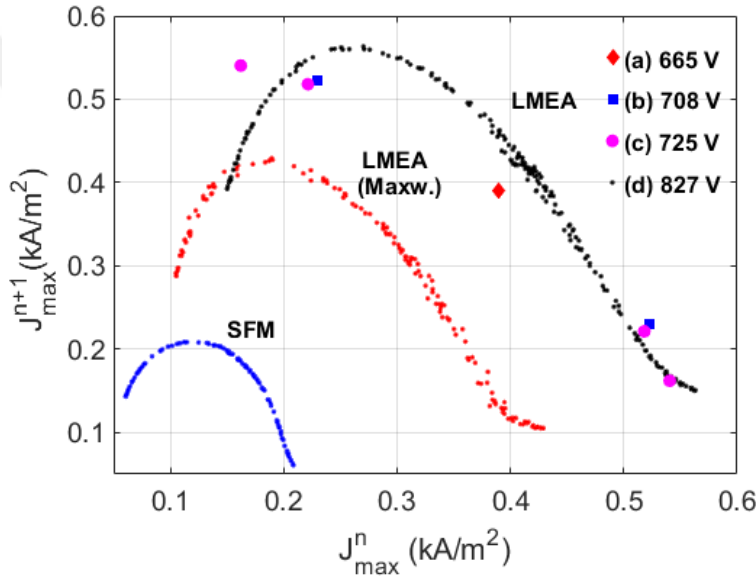


Figure 3.7: Lorenz map, corresponding to the regimes (a)–(d) with total applied voltages $U_t = 665, 708, 725$, and 827 V from figure 3.3, depicts the data points of the $(n+1)$ th maximum of the discharge current $J(t)$ versus its n th maximum. This figure also includes the maps derived from the LMEA model with the Maxwellian EEDF and the "simple fluid model".

Figure 3.8 presents the bifurcation diagram, where the curves separate the stable stationary states of the system from the oscillatory states in the parameter space of U_t and $1/R_s$. To perform the calculations systematically, we consider the total applied potential U_t and semiconductor resistance R_s as two control parameters, while all the other parameters are kept constant as defined in section 3.2.5. The calculations for

each particular value of U_t and R_s include two main steps. (i) the stationary solution of the gas discharge system is determined, and (ii) a new calculation is started from a regime corresponding to a sufficiently small perturbation of this stationary state. In the course of calculations, if the solution relaxes to a limit cycle oscillation about a stationary solution, this stationary state is classified as dynamically unstable. In dynamical terms, this implies a Hopf bifurcation. If the solution is pulled back to this stationary state, this state is classified as stable.

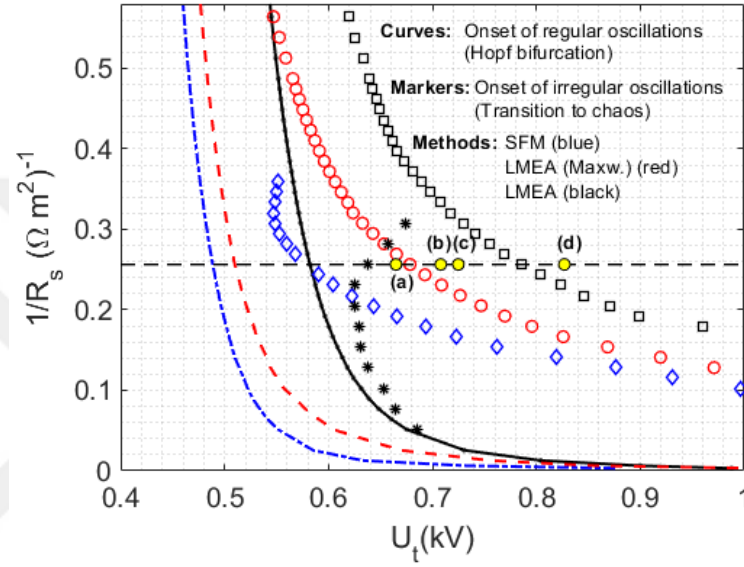


Figure 3.8: The bifurcation diagram is defined in the parameter space of U_t and $1/R_s$. The curves depict the onset of regular (periodic) oscillations via Hopf bifurcation. The markers, representing the transition from periodic to chaotic oscillations, indicate the onset of irregular oscillations. The computations are performed using the "simple" fluid (LFA) and "extended" fluid (LMEA) models. The black asterisk marker * data points indicate the bifurcation curve obtained from the instability development time [7].

In figure 3.8, the onset of irregular oscillations for each considered modeling approach is indicated by the markers (diamonds, circles, and squares), representing the regime where regular oscillations are separated from chaotic ones. The horizontal line corresponds to a constant conductivity value $\sigma_s = (2.6 \times 10^5 \Omega \text{ cm})^{-1}$, which represents the regime, at which the results were given in figure 3.3 (and also in figures 3.5, 3.6, and 3.7), and the points denoted by (a), (b), (c), and (d) along the line represents the

regimes (a)-(d) in the figure 3.3.

The black asterisk markers in figure 3.8 depict the bifurcation points, separating the stable states of the system from the unstable oscillatory states in the parameter space. These data points are determined using the characteristic time of instability development and the criterion in the Refs. [7, 244] is used

$$\tau_{\text{inst}} = \frac{12}{(4\pi en_i)^2 \alpha'' d^3} \tau_i. \quad (3.3.1)$$

In this equation, $\tau_i = d/v_i$ stands for the ion drift time over discharge gap, $v_i = \mu_i E$, where n_i is the ion density, and E is the spatial average of the electric field, the derivative relation $\alpha'' = \alpha''(E_0)$ is calculated with respect to E , and α represents the ionization coefficient described in section 3.2.3, $E_0 = U_{\text{br}}/d$ defines the breakdown field strength, and U_{br} is the discharge initiation (breakdown) voltage. In the presence of an external circuit equation (in the one-dimensional case, it is in the form of RC equation, see section 3.2.4), when the instability time τ_{inst} is shorter than the Maxwell time scale of the semiconductor τ_s , the developed instability may lead to the discharge oscillations [7, 244]

$$\tau_{\text{inst}} < \tau_s = C_s R_s. \quad (3.3.2)$$

It is interesting to note (see figure 3.8) that the approximate bifurcation curve, obtained from the relations 3.3.1 and 3.3.2, is in good agreement with the Hopf bifurcation curve of LMEA (black) at relatively large semiconductor resistance values.

As can be seen from the figures 3.4, 3.7 and 3.8, the results of the "simple fluid model" defined in the section 3.2.3 and abbreviated as SFM in these figures, and those of the "extended fluid model", at which the electron transport and electron-induced rate coefficients (given in table 3.1) are calculated with the assumption of Maxwellian EEDF (it is denoted on these figures with the abbreviation LMEA (Maxw.)), are demonstrated. Note that the results derived from all three methods have quantitatively different. CVCs in figure 3.4 and the Hopf bifurcation curves in figure 3.8, derived from the SFM and the LMEA (Maxw.) methods, are closer to each other when compared to those obtained by the LMEA method with EEDF, which is actually non-Maxwellian, calculated from a separate electron Boltzmann solver [235]. However, the results derived from all considered methods are qualitative agreement in terms of the dynamical behavior of the system as functions of two control parameters, namely the

total applied voltage U_t and resistance of the semiconductor layer R_s .

3.4 Conclusions to this chapter

This chapter investigates the transition from regular (periodic) to irregular chaotic oscillations in a laterally extended DC-driven GDSS in nitrogen. The analysis focuses on the oscillations of the subnormal glow regime, a phase that emerges between the Townsend and normal glow discharge.

The developed numerical models are based on plasma fluid equations with the "drift-diffusion" approximation of particle fluxes. Two modeling approaches are considered, namely the "simple fluid model" and the more advanced "extended fluid model", based on the LMEA technique. In the "simple fluid model", the reaction rate of particle creation was determined by the "Local Field Approximation" using the Townsend formula. Within the "extended fluid model", transport (mobility and diffusion) coefficients for electrons and electron-induced rate coefficients are defined as functions of the mean electron energy obtained from the solution of the electron Boltzmann equation. We have also considered the results of a modified model, which is again based on the LMEA approach; however, the EEDF to compute electron transport and rate coefficients for this model is chosen as the Maxwellian distribution function.

A Lorenz map and the bifurcation diagram, which demonstrates the transition of the system to a fully chaotic state through period-doubling bifurcations, are derived. Even though the results obtained from different modeling approaches lead to a poor quantitative agreement, these results show excellent qualitative similarities in terms of the system's dynamical behavior when the total applied voltage and semiconductor resistance act as control parameters.



CHAPTER 4

THE DELTA UNPREDICTABLE SYNCHRONIZATION ANALYSIS OF CHAOS IN GAS DISCHARGE-SEMICONDUCTOR SYSTEMS

This chapter applies the "Delta Synchronization of Chaos" method to the more detailed "extended fluid model", defined in chapter 3 as the "LMEA" model. Recall that this model is more advanced in terms of plasma chemistry and electron transport treatment than the "simple fluid model" and LMEA with Maxwellian EEDF. Delta synchronization is a newly developed method to detect chaotic synchronization in glow discharge systems. In the literature, the conventional method, known as the generalized synchronization method, is implemented in various discharge systems; however, the appearance of synchronization is not observed with this traditional method. In [4], we have previously introduced the delta synchronization method and implemented it to a chaotic regime of a one-dimensional "simple fluid model" of the GDSS in nitrogen. Fortunately, we have proved that a synchronization exists, which could not be detected by the generalized synchronization method in [89] before. In this chapter, for the first time, we will implement this new method to the chaotic regime of the more detailed "extended fluid model". This analysis allows us to confirm the developed synchronization method on the "extended fluid model" and compare the results of the "simple fluid model" to those of the "extended fluid model". The unpredictable and the delta synchronized motion are verified with simulations, and corresponding algorithms are given. The time sequences of the "unpredictability" and "delta synchronization" are illustrated in tables. The absence of generalized synchronization in the "extended fluid model" is also proved using the auxiliary system approach. The numerical characteristics, denoting the degree of chaos and synchronization, are defined and calculated, which are employed as mathematical tools to compare the detailed model results with the simpler one in [4]. The statement of the problem

is introduced in section 4.1. The mathematical definitions of "unpredictability" and "delta synchronization" are presented in the preliminaries section 4.2. The numerical description of the "unpredictability" and the DSC method with generic forms of the algorithms is given in section 4.3. In section 4.4, the transition to the chaotic regime in the "extended fluid model" has been revised again; however, the results are presented as dimensionless values this time. In section 4.5, the drive and response GDSSs are constructed, and their unpredictability is investigated. The analysis of DSC in the coupled system is made, and the absence of generalized synchronization is demonstrated. The conclusions to the problem are presented in section 4.6.

4.1 The statements of the problem

Synchronization of chaotic behavior in dynamic systems has become an active research area among scientists from different fields involving engineering, mathematics, and physics for the last three decades. Various methods have been developed concerning the scope of the dynamical system to analyze the chaotic synchronization [4, 81, 86, 245–251]. These techniques are mainly discussed in [75, 249] and classified as identical synchronization, phase synchronization, anticipated synchronization, lag synchronization, amplitude envelope synchronization, and generalized synchronization. However, in the literature, there are insufficient studies investigating the chaotic synchronization in glow discharge systems, particularly in a GDSS. Although the implementation and testing of these techniques on a GDSS have not been explicitly acted, the absence of generalized synchronization was recently presented for a GDSS in nitrogen [89]. The absence of generalized synchronization has encouraged us to formulate a novel synchronization technique to detect chaotic synchronization within a comparable system, as defined in [89]. This new method is introduced and named "Delta Synchronization of Chaos" (DSC) in our previous study [4]. This study proves the existence of DSC in GDSSs and shows that the DSC technique is more elaborate than the generalized synchronization method.

The "unpredictability" analysis is usually employed to verify that a dynamical system possesses Poincaré chaos [95, 252–254]. In this technique, the non-closed trajectories, converging to their initial points (described by the Poisson stability method) and

diverging from initial orbital sections at specific times, are considered. This method can be applied to a wide range of dynamic systems, provided the simulation time is allowed to be sufficiently long. A set of time values corresponding to the instants when trajectories converge to the initial points are called a "sequence of convergence", and a set of time values corresponding to the instants when the separation of trajectories starts to increase defines the "sequence of separation". Within this analysis, unpredictable solutions and chaos in a system are decided with respect to the existence of both sequences. Obtaining these two sequences will also form the backbone of the DSC method. The degree of numerical "unpredictability", which we will define in the next sections, allows us to compare the strength of chaos between the systems.

For the first time in Ref. [4], the method of DSC is introduced, and its relation with the "unpredictability" analysis is clearly explained. The implementation of the DSC technique requires two features: (i) the coupled systems (drive and response systems) must have unpredictable solutions, and (ii) the computation time must be sufficiently long. The main idea behind the DSC analysis is to find a common "sequence of finite convergence" and "sequence of separation" between the drive and response systems. Suppose the Poisson stability of the coupled systems, which means that the trajectories for both systems completely return to their initial points at common time values, is satisfied. In that case, the synchronization method is called "complete synchronization" [4]. As a result, the existence of common time sequences for convergence and divergence of the trajectories in the coupled systems confirms the delta synchronization of chaos. Moreover, the degree of numerical synchronization, which we will define in the preliminary section, allows us to describe the degree of the synchronous motion of the systems.

In this chapter, the investigations of DSC in a DC-supplied, planar GDSS in nitrogen are conducted. In the study [4], the one-dimensional "simple fluid model" with a "drift-diffusion" approximation was employed to prove the validity of the DSC method. Here, the more realistic and detailed one-dimensional fluid model, defined as the "extended fluid model", or the LMEA model in chapter 3, is selected to perform similar analyses of the DSC technique. The absence of generalized synchronization in this advanced fluid model is also confirmed, which was done for the "simple fluid model" in the Refs. [4, 89]. Understanding the reasons to perform the DSC

method to the more realistic fluid model, the "simple" and "extended" fluid modeling approaches, which are defined in chapter 3, can be compared in many aspects as follows:

1. Particle types:

- (a) "Simple fluid model" only considers two sorts of particles: electrons and ions.
- (b) Seven species, including electrons, ions, excited molecules, and ground state molecules, are taken into account in the "extended fluid model".

2. Plasma chemistry:

- (a) The secondary electron emission from the cathode wall and the direct ionization reactions are the only mechanisms responsible for the maintenance of a glow discharge.
- (b) In addition to the above two mechanisms, the "extended fluid model" also considers elastic collisions, electron-induced excitations, molecular vibrations and rotations, collisions among molecules, and relaxations of excited molecules.

3. Set of equations:

- (a) "Simple fluid model" solves the continuity equations for electrons and ions within the "drift-diffusion" approximation and Poisson equation for the electric field.
- (b) In addition to continuity equations for each of the seven species and Poisson equation, the electron energy equation is included in the set of equations of the "extended fluid model".

4. Electron transport and rate coefficients:

- (a) The diffusion and mobility coefficients of electrons are approximated by constants, and ionization rate coefficient (Townsend ionization coefficient) is defined as functions of the local electric field [27]. Therefore, the "simple fluid model" is also known as "Local Field Approximation," (LFA) [32].

- (b) The transport coefficients of electrons and rate coefficients of reactions initiated by electrons are defined based on their dependence on the mean electron energy calculated from the electron Boltzmann equation. Therefore, the "extended fluid model" is also known as "Local Mean Energy Approximation," (LMEA) [32].

5. Nonlocal nature of electrons:

Since the temperature of electrons tends to be higher than of ions and background gas in DC-supplied glow discharges, the electrons have a highly nonlocal nature, which means that they are far from equilibrium over the discharge gap [98,255].

- (a) The ionization rate coefficient in the LFA modeling is defined as an increasing function of the local electric field; therefore, it completely ignores the ionizations occurring in the region where the electric field is weak.
- (b) The electron-related transport and rate coefficients are defined as functions of the mean electron energy in the LMEA modeling, the effects of nonlocal electron transport, to some extent, are incorporated in the "extended fluid model".

6. Practical importance:

Controlling and understanding chaotic behaviors and instabilities of the plasma systems are essential because these regimes are practically undesirable cases in plasma devices such as plasma display panels, energy-saving lamps, and excimer lamps [12,143]. Therefore, developing a reliable model considering the computational efficiency and accuracy emphasizes the significance of implementing an appropriate fluid modeling approach. Although there is no substantial computational speed difference between the "simple" and "extended" fluid models, the latter, which is more detailed and realistic in the treatment of electron dynamics, allows us to reach more accurate results in future analysis. Furthermore, in [3], it is explicitly concluded that the one-dimensional LMEA modeling approach for the GDSS with a similar parameter regime agrees better with the experimental data than the LFA modeling.

4.2 Preliminaries

The "unpredictability" analysis is a valuable tool to detect the chaotic behavior of a dynamical system [252, 253]. Let X represent a flow or semi-flow, and $\mathbb{N}$ indicates the set of natural numbers, then the definition of the "unpredictability" is given below,

Definition 1 *The trajectory through a point $p \in X$ and the point itself are unpredictable if there exists a positive number Δ and the sequences of convergence t_n and separation s_n , both of which diverge to infinity, such that $\lim_{n \rightarrow \infty} f(t_n, p) = p$ and $d[f(s_n, f(t_n, p)), f(s_n, p)] \geq \Delta$ for each natural number n .*

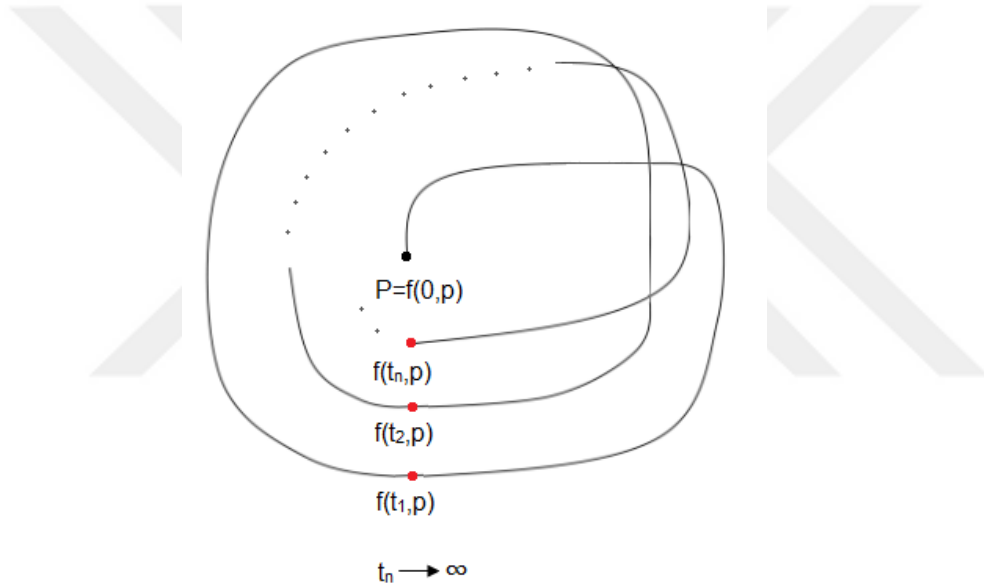


Figure 4.1: The schematic illustration of a Poisson stability.

Figure 4.1 presents a schematic description of Poisson stability. The initial point is denoted by $P = f(0, p)$, in which the first argument of the function f represents the elapsed time, and the second argument is the flow of the concerned point, p . Poisson stability is provided when the system's trajectory completely returns to its initial point as $n \rightarrow \infty$. In figure 4.2, let δ_n indicates the distance between the initial point $P = f(0, p)$ and $f(t_n, p)$ for $n \in \mathbb{Z}^+$, then the Poisson stability is verified by $\delta_n \rightarrow 0$ as $n \rightarrow \infty$. As a result, in the definition 1, the existence of the sequence of convergence $\{t_n\}$ ensures the Poisson stability. Besides Poisson stability, the divergence

of the trajectories at specific moments is a second requirement for the existence of unpredictability. In figure 4.2, the two trajectories with initial points $P = f(0, p)$ and $f(t_n, p)$ move away from each other after s_n time has elapsed such that the distance between them is greater than Δ , which is $\Delta > \delta_n$. The sequence of separation is defined by $\{s_n\}$, and the corresponding two points after s_n time are represented as $f(s_n, p)$ and $f(s_n, f(t_n, p))$.

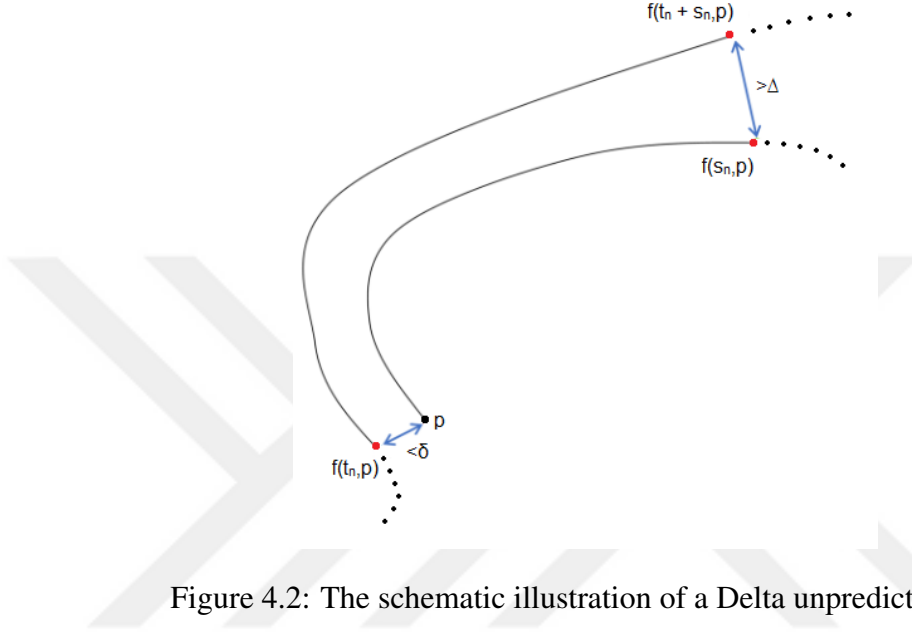


Figure 4.2: The schematic illustration of a Delta unpredictability.

The new chaos synchronization method inspired by the "unpredictability" analysis, the DSC, is clearly explained in [4]. Consider that there exist two unidirectionally coupled systems:

$$\dot{x} = f(x), \quad (4.2.1a)$$

$$\dot{y} = g(y, h(x)), \quad (4.2.1b)$$

where $x(t)$ and $y(t)$ are unpredictable solutions and let the systems 4.2.1a and 4.2.1b be the drive and response systems, then the definition of DSC is made as follows,

Definition 2 *The systems 4.2.1a and 4.2.1b with unpredictable motions $x(t)$ and $y(t)$ admits the delta synchronization of chaos, if there exist positive numbers $\delta, \Delta_1, \Delta_2, \delta < \Delta_{1,2}$, the sequence of finite convergence u_n and sequence of separation v_n such that $\|x(u_n) - x(0)\| + \|y(u_n) - y(0)\| = \delta_n \leq \delta$, $\|x(u_n + v_n) - x(v_n)\| \geq \Delta_1$ and $\|y(u_n + v_n) - y(v_n)\| \geq \Delta_2, n = 1, 2, \dots$*

In the numerical calculations, the existence and strength of the "unpredictability" and the DSC analyzes are determined by the numerical characteristics, namely "degree of numerical unpredictability" and "degree of numerical synchronization".

- Degree of numerical unpredictability,

$$\alpha_k = \frac{\min_{1,2,\dots,k} \delta_n}{\Delta} \quad (4.2.2)$$

where k is a finite number. This expression presents the existence and strength of the unpredictability for a k value. If α_k becomes smaller and goes to zero as k increases, then the "unpredictability" existence is approved. The strength of "unpredictability" is related to the smallness of α_k .

- Degree of numerical synchronization,

$$\alpha_k^{sync} = \frac{\delta_k}{\min(\Delta_1, \Delta_2)} \quad (4.2.3)$$

where δ_k is a non-increasing sequence for $n = 1, 2, \dots, k$. In the sufficiently long simulations, if α_k takes smaller positive values ($\alpha_k^{sync} < 1$) as k increases, the existence of DSC is numerically approved. With $\alpha_k^{sync} \rightarrow 0$ for large values of k , the synchronization is complete since Poisson stability is satisfied. Similarly, the strength of synchronization is related to the smallness of α_k .

Generalized synchronization is the well-known technique to determine the synchronization between the drive and response systems [81, 86]. Consider the coupled systems given in the equations 4.2.1a, 4.2.1b, if there exist initial condition sets of I_x, I_y such that all $x_0 \in I_x$ and $y_{10}, y_{20} \in I_y$ satisfy

$$\lim_{t \rightarrow \infty} \|y(t, x_0, y_{10}) - y(t, x_0, y_{20})\| = 0, \quad (4.2.4)$$

then the generalized synchronization occurs between these two systems.

4.3 The algorithms of the "unpredictability" and DSC methods

Suppose that a dynamical system, represented by a differential equation, has the time-dependent solution $\mathbf{x}$, which can also be multidimensional, in the time domain $T \in$

$[T^i, T^f]$ with a time difference ΔT . The determination of the unpredictability of this system can be provided by algorithm 1. In the algorithm, $\mathbf{x}_0$ specifies the starting point of system motion, and the smallness of δ and the magnitude of Δ are aligned with the coordinate axes covering all possible movement of the system. In the first

Algorithm 1: Algorithm for Unpredictability

```

Input:  $\delta$ 
Input:  $\Delta\delta$ 
Input:  $\Delta$ 
 $n = 1;$ 
 $q = 0;$ 
for  $T = T^i + \Delta T : \Delta T : T^f$  do
    if  $\|\mathbf{x}(T) - \mathbf{x}_0\| < \delta$  then
         $t_n = T$  % Sequence of convergence;
         $n = n + 1;$ 
         $\delta = \delta - \Delta\delta;$ 
    end
end
for  $T = T^i + \Delta T : \Delta T : T^f$  do
    for  $i = 1 : 1 : n - 1$  do
        if  $\|\mathbf{x}(t_i + T) - \mathbf{x}(T)\| > \Delta$  then
            if  $T > q$  then
                 $s_i = T$  % Sequence of separation;
                 $q = T;$ 
                break;
            end
        end
    end
end
end

```

for-loop block, n is incremented by one at each step; thereby, the sequence of finite convergence includes $n - 1$ elements at the last step of the loop. Since the element numbers in the sequence of separation must be less than or equal to those of the

sequence of convergence, the incrementation of i stops at the $(n - 1)^{th}$ element in the second for-loop block.

Algorithm 2: Algorithm for Delta (Complete) Synchronization of Chaos

```

Input:  $\delta$ 
Input:  $\Delta\delta$ 
Input:  $\Delta_1$ 
Input:  $\Delta_2$ 
 $n = 1$ ;
 $q = 0$ ;
for  $T = T^i + \Delta T : \Delta T : T^f$  do
    if  $\|\mathbf{x}(T) - \mathbf{x}_0\| + \|\mathbf{y}(T) - \mathbf{y}_0\| < \delta$  then
         $t_n = T$  % Sequence of finite convergence;
         $n = n + 1$ ;
         $\delta = \delta - \Delta\delta$ ;
    end
end
for  $T = T^i + \Delta T : \Delta T : T^f$  do
    for  $i = 1 : 1 : n - 1$  do
        if  $\|\mathbf{x}(t_i + T) - \mathbf{x}(T)\| > \Delta_1$  &&  $\|\mathbf{y}(t_i + T) - \mathbf{y}(T)\| > \Delta_2$  then
            if  $T > q$  then
                 $s_i = T$  % Sequence of separation;
                 $q = T$ ;
                break;
            end
        end
    end
end
end

```

Suppose that the drive and response systems, given in 4.2.1a and 4.2.1b, are time-dependent systems and have unpredictable solutions $\mathbf{x}$, $\mathbf{y}$, respectively. Note that the solutions $\mathbf{x}$, $\mathbf{y}$ can also be multi-dimensional. The Delta synchronization of chaos, or complete synchronization in the case of Poisson stability, is detected via the algorithm

2. In this algorithm, the corresponding time values T in both systems, 4.2.1a and 4.2.1b, are common because these two systems are run with the same simulation duration (between T^i and T^f) and time difference ΔT . The δ value of the DSC algorithm is the summation of the δ_1 and δ_2 , which are defined by corresponding δ values of the drive and response systems' "unpredictability" analysis. Similarly, Δ_1 and Δ_2 values correspond to the Δ values of the drive and response systems.

4.4 The "extended fluid model"

The present chapter employs the "extended fluid model", defined as the more detailed and realistic model in chapter 3, to develop drive and response systems in future sections. The governing equations of this model are described in the equations 3.2.1, 3.2.2, 3.2.3, 3.2.4, 3.2.5 and 3.2.11. The set of plasma chemical reactions is summarized in table 3.1, the boundary conditions are specified in table 3.2, and the input parameters are given in section 3.2.5.

In chapter 3, the "extended fluid model" is dimensionally developed. However, the results for the "simple fluid model" in [4] are presented in the dimensionless form. To compare this chapter's results with those shown in [4], we must scale our results by following a non-dimensionalization procedure. The procedure of a GDSS model is mainly performed in [70] and [256], and the scaling factors can be written in terms of dimensional parameters as follows

$$\mathbf{L} = \frac{x}{X_0}, \quad \mathbf{n}_{e,i} = \frac{n_{e,i}}{n_0}, \quad \boldsymbol{\tau} = \frac{t}{t_0}, \quad \mathbf{E} = \frac{E}{E_0}, \quad (4.4.1)$$

$$\mathbf{U} = \frac{U}{E_0 X_0}, \quad \mathbf{J} = \frac{J}{en_0 X_0 / t_0}, \quad \mathbf{R}_s = \frac{R_s}{E_0 t_0 / en_0}, \quad (4.4.2)$$

where the intrinsic parameters are

$$\begin{aligned} X_0 &\approx 2.78 \times 10^{-5} \text{ m}, \quad t_0 \approx 4.05 \times 10^{-11} \text{ s}, \\ n_0 &\approx 2.05 \times 10^{18} \text{ m}^{-3}, \quad E_0 \approx 1.03 \times 10^6 \text{ V/m}. \end{aligned} \quad (4.4.3)$$

The dimensionless parameters $\mathbf{L}$, $\mathbf{n}_{e,i}$, $\boldsymbol{\tau}$ and $\mathbf{E}$ are calculated simply by dividing their physical equivalents by intrinsic values. Furthermore, $\mathbf{U}$, $\mathbf{J}$ and $\mathbf{R}_s$ are obtained by dividing U , J and R_s by 28.63 V , $2.25 \times 10^5 \text{ A/m}^2$ and $1.27 \times 10^{-4} \text{ } \Omega \text{ m}^2$, respectively. It should be pointed out that the results in all future sections of this chapter are represented as dimensionless results.

4.4.1 The stable and unstable stationary states

The CVC curve of a gas discharge system is a significant integral characteristic to determine discharge regimes (abnormal, normal, and subnormal regimes) and to decide the adequacy of the developed model relative to the actual discharge system [27]. In figure 4.3, the dimensionless CVC ($U = U(J)$), previously presented with dimensional quantities in the figure 3.4, and load line $U = U_t - R_s J$, which is a time-independent correspondence of the external circuit equation 3.2.11, of the LMEA model are demonstrated in the (U, J) plane.

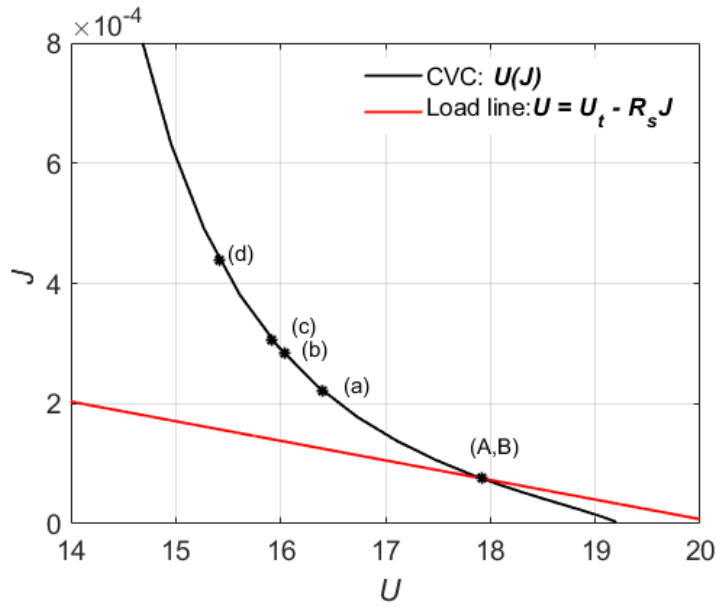


Figure 4.3: The CVC curve: $U = U(J)$ and the load line: $U = U_t - R_s J$ of the "extended fluid model". The intersection point (A, B) shows the equilibrium point at the regime $U_t = 20.23$ and $R_s = 30709$. The points (a), (b), (c), and (d) denote stationary solutions with $U_t = 23.2, 24.7, 25.3$ and 28.9 at $R_s = 30709$. Note that the load lines for (a), (b), (c), and (d) are not depicted for clarity of demonstration.

The stationary solutions, or equilibrium points, corresponding to intersection points between $U = U(J)$ and $U = U_t - R_s J$, are also presented. Here, points (a), (b), (c), and (d) on the CVC curve indicate unstable stationary solutions, which are previously specified in figure 3.3. Note that their correspondence load lines are not displayed for clarity of illustration.

When the calculation is started in the close neighborhood of $(A, B) = (17.92, 7.55 \times 10^{-5})$, which is located at $(A_0, B_0) = (17.81, 8.22 \times 10^{-5})$, this perturbed solution is pulled back to the equilibrium point (A, B) . As a result, the equilibrium point (A, B) at the regime $U_t = 20.23$, and $R_s = 30709$ is dynamically stable. The behavior of this perturbed calculation is illustrated in figure 4.4.

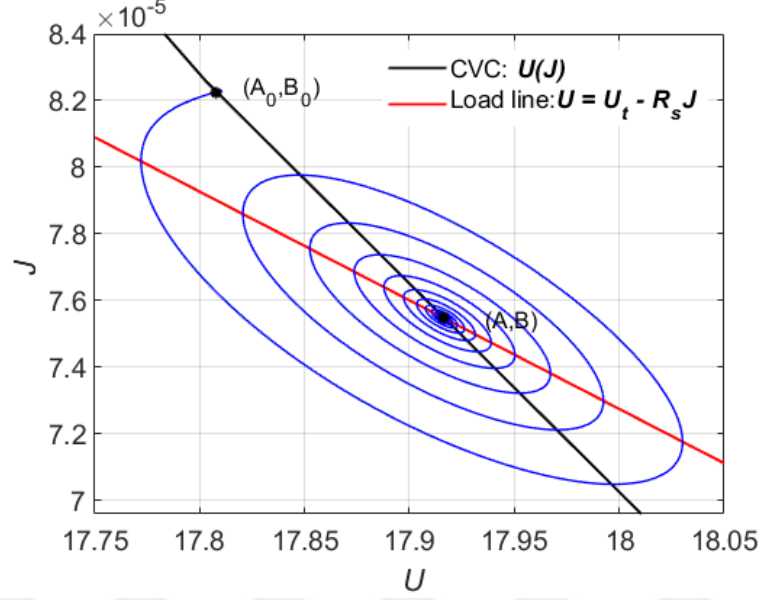


Figure 4.4: Phase trajectories initiated at $(A_0, B_0) = (17.81, 8.22 \times 10^{-5})$ spiral inwards towards the stationary point $(A, B) = (17.92, 7.55 \times 10^{-5})$ in the dimensionless (U, J) plane.

Table 4.1: The values of the control parameters (U_t, R_s) and the intersection points of the load line with the CVC curve (U, J) for (a), (b), (c) and (d) points in figure 4.3.

	U_t	R_s	U	J
(a)	23.2	30709	16.40	2.22×10^{-4}
(b)	24.7	30709	16.04	2.83×10^{-4}
(c)	25.3	30709	15.92	3.07×10^{-4}
(d)	28.9	30709	15.42	4.38×10^{-4}

If the calculations are started from the close neighborhood of the equilibrium points (a), (b), (c), and (d), denoted in figure 4.3, the perturbed solutions are developed into

limit cycle oscillations around these stationary solutions. As a result, these solutions are dynamically unstable with corresponding control parameters U_t , R_s and intersection points of the load line with the CVC curve, namely U , J values, which are summarized in table 4.1. The corresponding phase portraits, which is the dimensionless correspondence of the last row of the figure 3.3 of chapter 3, are demonstrated in figure 4.5. Here, in regimes (a), (b), and (c), the phase trajectories have periodic oscillations repeating themselves in regular cycles, unlike regime (d), which shows the fully chaotic oscillations of the system.

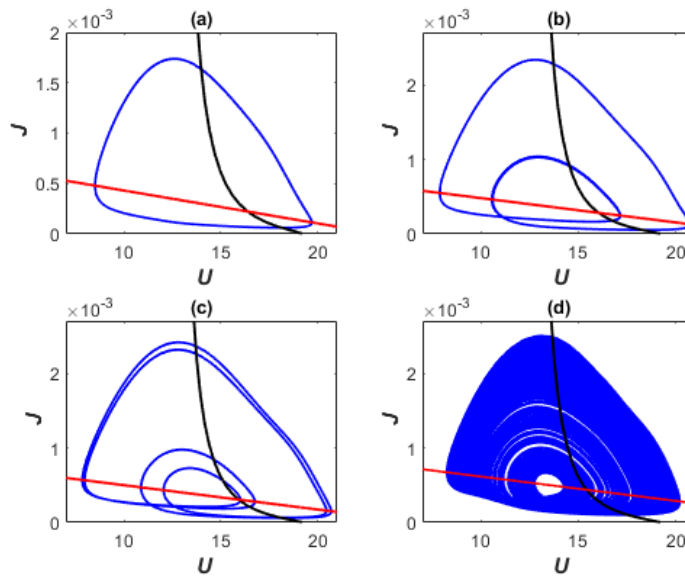


Figure 4.5: The phase portraits include the trajectories of the system oscillations, the CVC curve $U = U(J)$ and the load line $U = U_t - R_s J$ for the corresponding regimes (a), (b), (c) and (d), indicated in figure 4.3.

4.4.2 The dimensionless bifurcation diagram

The dimensionless correspondence of the bifurcation diagram for the "extended fluid model" is demonstrated in figure 4.6, which is previously defined in figure 3.8 with dimensional quantities. In the figures 4.3, 4.5 and table 4.1, the calculations are made by varying the U_t values and holding $R_s = 30709$ constant to observe the transition of the system from a regular to a chaotic state. These calculations correspond to (a), (b), (c), and (d) labels located on a horizontal line in figure 4.6. When U_t and R_s act

as two varying control parameters, the bifurcation curve, separating stable stationary states from the unstable ones, and the onset of chaotic oscillations, indicated by square markers, are obtained in figure 4.6.

In the following sections of this chapter, the system model corresponding to chaotic oscillations, shown as regime (d) in figures 4.3, 4.5, 4.6 and table 4.1, are considered to define the drive and response systems for series of analysis.

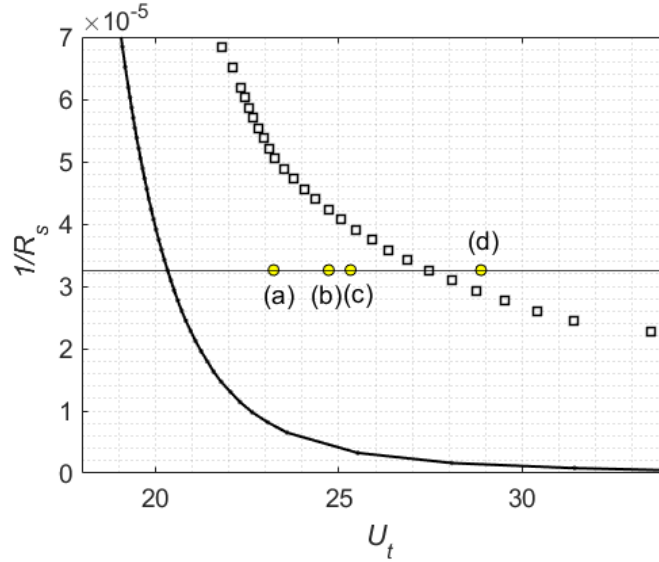


Figure 4.6: Bifurcation diagram of the LMEA model with onsets of periodic and chaotic oscillations in the dimensionless parameter space. Points denoted by (a), (b), (c) and (d) denote the regimes with $U_t = 23.2, 24.7, 25.3$ and 28.9 at $R_s = 30709$.

4.5 Delta synchronization in the "extended fluid model"

In this section, the drive and response GDSS models are constructed using the "extended fluid model" at its chaotic regime (d). The drive system corresponds to the same system defined in chapter 3 and previous sections of this chapter. However, the response system is guided by defining the discharge potential of the drive system as the perturbation term with a small multiplier. In the following subsections, the "unpredictability" analysis is separately examined in both systems, the DSC method is implemented in the coupled system, and the absence of the generalized synchronization in the LMEA model at regime (d) is proved.

4.5.1 The "unpredictability" analysis of drive and response systems

The drive system of the GDSS with the "extended" fluid (LMEA) modeling approach employs the same equations and boundary conditions defined in chapter 3. The set of equations is collectively rewritten as the governing equations of the drive system as follows.

$$\begin{aligned}
\frac{\partial n_k}{\partial t} + \frac{\partial \Gamma_k}{\partial x} &= S_k, \\
\Gamma_k &= \text{sgn}(q_k) \mu_k n_k E - D_k \frac{\partial n_k}{\partial x}, \\
-\epsilon_0 \frac{\partial^2 \varphi}{\partial x^2} &= \sum_k q_k n_k, \\
\frac{\partial n_\epsilon}{\partial t} + \frac{\partial \Gamma_\epsilon}{\partial x} &= -e \Gamma_e E - \frac{3}{2} \frac{m_e}{m_g} \nu_{\text{ea}} n_e k_B (T_e - T_g) - \sum_j \Delta E_j R_j \\
\Gamma_\epsilon &= -D_\epsilon \frac{\partial n_\epsilon}{\partial x} - \mu_\epsilon E n_\epsilon \\
\tau_s \frac{\partial U(t)}{\partial t} &= U_t - U(t) - R_s J(t),
\end{aligned} \tag{4.5.1}$$

with the boundary conditions given in table 4.2.

To define the response system of the GDSS with the "extended" fluid (LMEA) modeling approach, the discharge potential U , which is obtained by simultaneous calculations of the equations 4.5.1, or drive system, is considered as a perturbation term by multiplying it with a small constant δ . The set of equations for the response system is as follows:

Table 4.2: Boundary conditions for the drive GDSS.

	Anode ($x = 0$)	Cathode ($x = d$)
Electrons, n_e	$\Gamma_e = -\frac{1}{4} v_e n_e$	$\Gamma_e = \frac{1}{4} v_e n_e - \gamma \Gamma_i$
Ions, n_i	$\Gamma_i = -\frac{1}{4} v_i n_i + a n_i \mu_i E$	$\Gamma_i = \frac{1}{4} v_i n_i + a n_i \mu_i E$
Excited species, n_m	$\Gamma_m = -\frac{1}{4} v_m n_m$	$\Gamma_m = \frac{1}{4} v_m n_m$
Electron energy density, n_ϵ	$\Gamma_\epsilon = -\frac{1}{3} v_e n_\epsilon$	$\Gamma_\epsilon = \frac{1}{3} v_e n_\epsilon - 2 k_B T_e \gamma \Gamma_i$
Electric potential, φ	U	0

$$\begin{aligned}
\frac{\partial \tilde{n}_k}{\partial t} + \frac{\partial \tilde{\Gamma}_k}{\partial x} &= \tilde{S}_k, \\
\tilde{\Gamma}_k &= \text{sgn}(q_k) \mu_k \tilde{n}_k \tilde{E} - \tilde{D}_k \frac{\partial \tilde{n}_k}{\partial x}, \\
-\epsilon_0 \frac{\partial^2 \tilde{\varphi}}{\partial x^2} &= \sum_k q_k \tilde{n}_k, \\
\frac{\partial \tilde{n}_\epsilon}{\partial t} + \frac{\partial \tilde{\Gamma}_\epsilon}{\partial x} &= -e \tilde{\Gamma}_\epsilon \tilde{E} - \frac{3}{2} \frac{m_e}{m_g} \nu_{\text{ea}} \tilde{n}_\epsilon k_B (T_e - T_g) - \sum_j \Delta E_j \tilde{R}_j \\
\tilde{\Gamma}_\epsilon &= -D_\epsilon \frac{\partial \tilde{n}_\epsilon}{\partial x} - \mu_\epsilon \tilde{E} \tilde{n}_\epsilon \\
\tau_s \frac{\partial V(t)}{\partial t} &= V_t - V(t) - R_s \tilde{J}(t) + \delta U(t),
\end{aligned} \tag{4.5.2}$$

with the boundary conditions given in table 4.3.

Table 4.3: Boundary conditions for the response GDSS.

	Anode ($x = 0$)	Cathode ($x = d$)
Electrons, $\tilde{n}_e$	$\tilde{\Gamma}_e = -\frac{1}{4} v_e \tilde{n}_e$	$\tilde{\Gamma}_e = \frac{1}{4} v_e \tilde{n}_e - \gamma \tilde{\Gamma}_i$
Ions, $\tilde{n}_i$	$\tilde{\Gamma}_i = -\frac{1}{4} v_i \tilde{n}_i + a \tilde{n}_i \mu_i \tilde{E}$	$\tilde{\Gamma}_i = \frac{1}{4} v_i \tilde{n}_i + a \tilde{n}_i \mu_i \tilde{E}$
Excited species, $\tilde{n}_m$	$\tilde{\Gamma}_m = -\frac{1}{4} v_m \tilde{n}_m$	$\tilde{\Gamma}_m = \frac{1}{4} v_m \tilde{n}_m$
Electron energy density, $\tilde{n}_\epsilon$	$\tilde{\Gamma}_\epsilon = -\frac{1}{3} v_e \tilde{n}_\epsilon$	$\tilde{\Gamma}_\epsilon = \frac{1}{3} v_e \tilde{n}_\epsilon - 2k_B T_e \gamma \tilde{\Gamma}_i$
Electric potential, $\tilde{\varphi}$	V	0

Note that the discharge voltage of the drive system is applied as a perturbation to the external circuit equation of the response system, which is the last equation of the system 4.5.2. Moreover, the plasma variables in the response system, which have a tilde symbol above them, emphasize that they are not the same variable as the drive system due to the given perturbation.

The "unpredictability" in both drive and response systems is investigated by separately acting the algorithm 1 to the coupled system. The analysis utilizes the dimensionless $\mathbf{U}(\tau)$ and $\mathbf{J}(\tau)$ solutions for each system, which are derived from the system of equations 4.5.1, 4.5.2. Indeed, these systems are dimensionally solved as in chapter 3, and the results are converted into dimensionless values by following the scaling procedure defined in section 4.4. The numerical idea behind the implementation of algorithm 1 is based on putting a circle with radius δ_n in the $(\mathbf{U}, \mathbf{J})$ phase space,

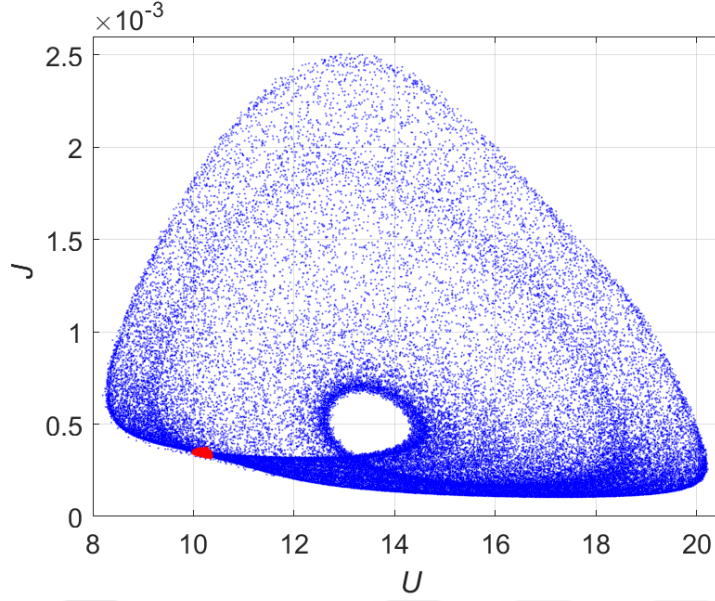


Figure 4.7: Phase portrait of the drive system at $U_t = 28.9$ and $R_s = 30709$ in the dimensionless (U, J) plane. The points denoted by red markers represent the corresponding U and J values at the time values of the sequence of convergence $\{t_n\}$.

which is centered at the initial point. The equation of this circle for each n values can be defined by $\sqrt{(U - U_0)^2 + ((J - J_0) \times 10^4)^2} = \delta_n$. Note that J is multiplied by a factor 10^4 . The reason for this scaling is that U and J have values with different order of magnitudes to define a circle in the phase space (see figures 4.5 at (d), 4.7 and 4.8) such that, approximately, $U \in [8, 21]$ and $J \in [0, 2.5 \times 10^{-3}]$. As a result, one of the purposes of the algorithm 1 is the determination of time values of $\{t_n\}$ sequence, satisfying $\sqrt{(U - U_0)^2 + ((J - J_0) \times 10^4)^2} < \delta_n$ with having a smaller radius δ_n at each iteration. The second purpose of the algorithm 1 is the determination of time values of $\{s_n\}$ sequence, which corresponds to the instant at which the divergence of the phase trajectories has started. Note that these calculations accounting for the separation of trajectories have initiated at the time values of $\{t_n\}$ sequence. Therefore, the number of time values belonging to $\{s_n\}$ sequence must be smaller than those of $\{t_n\}$ sequence.

The set of equations of the drive system, which is defined in the system 4.5.1, are solved at the chaotic regime of two control parameters $U_t = 28.9$ and $R_s = 30709$,

Table 4.4: The elements of $\{t_n\}$, $\{s_n\}$ and $\{\delta_n\}$ sequences of the drive system.

n	t_n	s_n	δ_n
1	12350	9468	0.180
2	61338	20583	0.179
3	88096	32110	0.178
4	150669	148199	0.177
5	164666	148611	0.176
	$\vdots$		
	$\vdots$		
176	47442737	3021209	0.005
177	54925159	3032736	0.004
178	70891584	3071020	0.003
179	87914341	3083782	0.002
180	111008744	-	0.001

as explained in the previous sections. The dimensionless simulation time is $\tau \in [0, 1.73 \times 10^8]$ with a time difference $\Delta\tau = 411$. As a result of the calculations, the sequence of convergence $\{t_n\}$ includes 180-time elements, and the sequence of convergence $\{s_n\}$ includes 179-time elements for $\Delta = 6.4$ and radii takes the values of $\delta_n = 0.18, 0.179, \dots, 0.001$. Note that the δ_n values decrease for each iteration of n . The existence of $\{t_n\}$ and $\{s_n\}$ sequences confirms the unpredictability, which implies that the drive system possesses Poincaré chaos. The solution of the system is demonstrated in figure 4.7 with red marked points, which indicates the corresponding U , J values at the time elements of $\{t_n\}$ sequence. The corresponding dimensionless data for the elements of $\{t_n\}$, $\{s_n\}$ and $\{\delta_n\}$ values are given in table 4.4. The degree of numerical unpredictability, a characteristic showing the strength of the unpredictability, is calculated as $\alpha_{179} = 0.0003125$. Here, 179^{th} element of δ_n , which is 0.002, is used as a minimum radius of convergence to calculate the α_{179} because there is no s_n correspondence of 180^{th} element (see table 4.4).

In the response system, the set of equations is defined in the system 4.5.2 such that the external circuit equation includes a perturbation term, $\delta U(t)$ where $U(t)$ is derived

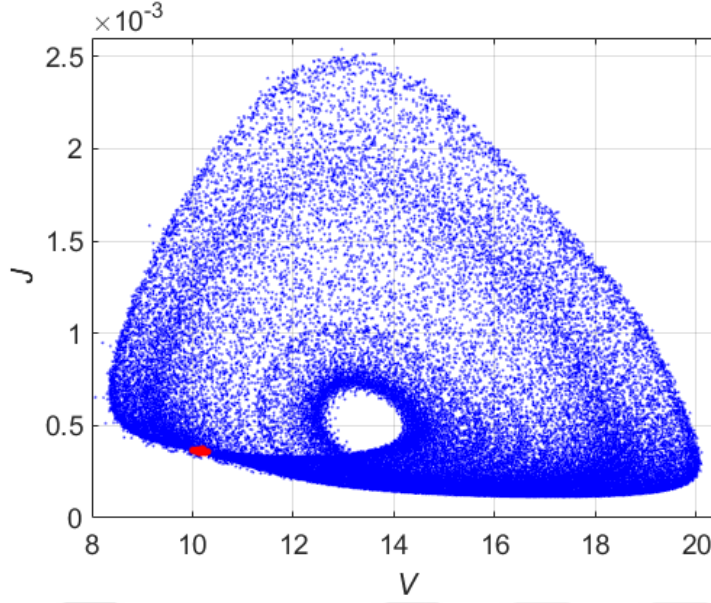


Figure 4.8: Phase portrait of the response system at $V_t = 28.9$ and $R_s = 30709$ in the dimensionless (V, J) plane. The points denoted by red markers represent the corresponding V and J values at the time values of the sequence of convergence $\{t_n\}$.

from the drive system, and δ is a small multiplier, or perturbation constant. The perturbation constant δ is taken as 0.047, which is the same as in the previous study [4] for making a consistent comparison. As in the drive system, the equations are solved at the chaotic regime of two control parameters $V_t = 28.9$ and $R_s = 30709$. The dimensionless simulation time with corresponding time difference and sequence of radius $\{\delta_n\}$ are defined the same as the drive system to establish compatibility. As a result, time elements in the sequence of convergence $\{t_n\}$ and the sequence of separation $\{s_n\}$ are 175 for $\Delta = 5$. The existence of these two sequences confirms that the response system is unpredictable and essentially chaotic. The corresponding solutions in the (V, J) phase space are demonstrated in figure 4.8. The corresponding V and J values at the time values of the sequence of convergence $\{t_n\}$ are indicated by red markers. When the figures 4.7 and 4.8 are compared, one can conclude that corresponding discharge potential and current density values for time elements of the sequence of convergence occupy the same region in the corresponding dimensionless parameter space. This is not true because time values of $\{t_n\}$ and $\{s_n\}$ in table 4.5 are not same those in table 4.4. Moreover, the "degree of numerical unpredictabil-

ity" is $\alpha_{175} = 0.0012$, which implies that the response system is more unpredictable (chaotic) than the drive system in the concerned simulation time.

Table 4.5: The elements of $\{t_n\}$, $\{s_n\}$ and $\{\delta_n\}$ sequences of the response system.

n	t_n	s_n	δ_n
1	22642	7410	0.180
2	35403	7822	0.179
3	47753	8233	0.178
4	60926	8645	0.177
5	86450	18937	0.176
	$\vdots$		
	$\vdots$		
171	52707520	1724464	0.010
172	72399924	1724876	0.009
173	74360273	1733933	0.008
174	88937328	1745048	0.007
175	162001680	1756986	0.006

4.5.2 Delta synchronization between the drive and response systems

The unpredictable analyzes confirm that the drive and response systems have chaotic solutions at the same regime at which U_t and V_t are 28.9 and $R_s = 30709$. The chaos synchronization in the coupled system is investigated using the DSC method based on the unpredictability analysis [95]. The algorithm 2 is implemented in the drive and response systems.

Here, the non-dimensionalized simulation time, the elements in $\{\delta_n\}$ sequence, and the corresponding initial points of both systems are defined the same as the values used in the "unpredictability" analysis due to providing consistency in the algorithm 2. However, in the "unpredictability" analysis, the Δ values for drive and response systems are chosen as $\Delta_1 = 6.4$ and $\Delta_2 = 5$, respectively. The different Δ values are preferred because these Δ values provide the maximum number of elements in both systems' corresponding $\{s_n\}$ sequence. In figure 4.9, the

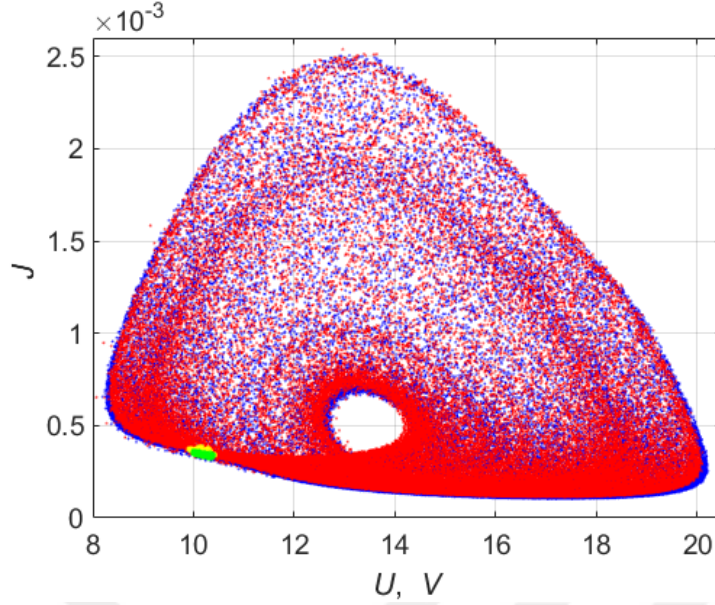


Figure 4.9: Phase portraits of the drive (blue) and response (red) systems at $V_t = 28.9$ and $R_s = 30709$ in the dimensionless $((U, V), J)$ plane. The points denoted by green and yellow markers represent the corresponding U, V and J values at common time values u_n of the sequence of convergence for the drive and response systems, respectively.

phase portraits of the drive and response systems in the dimensionless space are demonstrated with blue and red points, respectively. These two-phase portraits do not overlap due to the perturbation term defined in the response system. The green and yellow markers represent the corresponding U, V and J values at the common time values in the sequence of convergence of the coupled system. There are 73 and 72 common time values in the sequence of finite convergence and sequence of separation, which is illustrated with u_n and v_n in table 4.6. The common time values between the drive and response systems do not mean the same time values found in both tables 4.4 and 4.5. They imply corresponding time values satisfying the relation $\|\mathbf{x}(T) - \mathbf{x}_0\| + \|\mathbf{y}(T) - \mathbf{y}_0\| < \delta$ for convergence sequence $\{u_n\}$ and $\|\mathbf{x}(u_i + T) - \mathbf{x}(T)\| > \Delta_1$ & $\|\mathbf{y}(u_i + T) - \mathbf{y}(T)\| > \Delta_2$ for separation sequence $\{v_n\}$, which are defined in algorithm 2, where $\mathbf{x}(T)$ and $\mathbf{y}(T)$ represent the unpredictable solutions of the drive and response systems in the all-time domain. The degree of numerical synchronization is $\alpha_{72}^{sync} = 0.0218$, which confirms the delta synchronization of chaos.

Table 4.6: Common dimensionless u_n , v_n , and δ_n values for the coupled system.

n	u_n	v_n	δ_n
1	1773453	299280	0.180
2	2123368	831975	0.179
3	2961518	838973	0.178
4	3166115	839385	0.177
5	3726803	844736	0.176
	$\vdots$		
	$\vdots$		
69	145735563	5559123	0.112
70	147405688	5785951	0.111
71	160527096	5788832	0.110
72	167032225	5810651	0.109
73	170450691	-	0.108

4.5.3 The generalized synchronization in the coupled system

In [4, 89], the absence of the generalized synchronization in the coupled system is demonstrated, which is based on a "simple" fluid (LFA) modeling approach with the same parameter regime as in this chapter. Additionally, in [4], the DSC method proves the existence of synchronization, although the generalized synchronization does not reveal any synchronization. Thus, it has been verified that the DSC method is more inclusive than the generalized synchronization method. A similar approach to show the absence of generalized synchronization in the "extended fluid model" is followed such that the auxiliary system approach is utilized for the coupled system. The set of equations for the auxiliary system is as follows.

$$\begin{aligned}
\frac{\partial \bar{n}_k}{\partial t} + \frac{\partial \bar{\Gamma}_k}{\partial x} &= \bar{S}_k, \\
\bar{\Gamma}_k &= \text{sgn}(q_k) \mu_k \bar{n}_k \bar{E} - \bar{D}_k \frac{\partial \bar{n}_k}{\partial x}, \\
-\epsilon_0 \frac{\partial^2 \bar{\varphi}}{\partial x^2} &= \sum_k q_k \bar{n}_k, \\
\frac{\partial \bar{n}_\epsilon}{\partial t} + \frac{\partial \bar{\Gamma}_\epsilon}{\partial x} &= -e \bar{\Gamma}_\epsilon \bar{E} - \frac{3}{2} \frac{m_e}{m_g} \nu_{\text{ea}} \bar{n}_\epsilon k_B (T_e - T_g) - \sum_j \Delta E_j \bar{R}_j \\
\bar{\Gamma}_\epsilon &= -D_\epsilon \frac{\partial \bar{n}_\epsilon}{\partial x} - \mu_\epsilon \bar{E} \bar{n}_\epsilon \\
\tau_s \frac{\partial W(t)}{\partial t} &= W_t - W(t) - R_s \bar{J}(t) + \delta U(t),
\end{aligned} \tag{4.5.3}$$

Table 4.7: Boundary conditions for the auxiliary GDSS.

	Anode ($x = 0$)	Cathode ($x = d$)
Electrons, $\bar{n}_e$	$\bar{\Gamma}_e = -\frac{1}{4} v_e \bar{n}_e$	$\bar{\Gamma}_e = \frac{1}{4} v_e \bar{n}_e - \gamma \bar{\Gamma}_i$
Ions, $\bar{n}_i$	$\bar{\Gamma}_i = -\frac{1}{4} v_i \bar{n}_i + a \bar{n}_i \mu_i \bar{E}$	$\bar{\Gamma}_i = \frac{1}{4} v_i \bar{n}_i + a \bar{n}_i \mu_i \bar{E}$
Excited species, $\bar{n}_m$	$\bar{\Gamma}_m = -\frac{1}{4} v_m \bar{n}_m$	$\bar{\Gamma}_m = \frac{1}{4} v_m \bar{n}_m$
Electron energy density, $\bar{n}_\epsilon$	$\bar{\Gamma}_\epsilon = -\frac{1}{3} v_e \bar{n}_\epsilon$	$\bar{\Gamma}_\epsilon = \frac{1}{3} v_e \bar{n}_\epsilon - 2k_B T_e \gamma \bar{\Gamma}_i$
Electric potential, $\bar{\varphi}$	W	0

The corresponding boundary conditions for the auxiliary system are defined in table 4.7. In the auxiliary system of the equations, the two control parameters are $\mathbf{W}_t = 28.9$, $\mathbf{R}_s = 30709$, and the perturbation constant is $\delta = 0.047$, which are the same values as in the response system. Indeed, the equations of the system 4.5.3 are similar to those of the response system defined in the system 4.5.2, except for the initial conditions. In the preliminaries section of this chapter, the existence of the generalized synchronization between the drive and response systems is equated with satisfying the asymptotic stability condition 4.2.4. This condition is satisfied for $y(0, x_0, y_{10}) = y_{10} = y_0^{\text{response}}$, $y(0, x_0, y_{20}) = y_{20} = y_0^{\text{auxiliary}}$ with the same $x(t)$ values and $x(0) = x_0$ [81].

The behavior of $\mathbf{V}$ and $\mathbf{W}$ is depicted in the dimensionless $\mathbf{V} - \mathbf{W}$ plane in figure 4.10. The asymptotic condition is not satisfied because the corresponding results do not occur on the $\mathbf{V} = \mathbf{W}$ line. As a result, the implementation of the auxiliary

approach reveals that the drive and response systems, based on the "extended" fluid (LMEA) modeling approach, do not possess generalized synchronization.

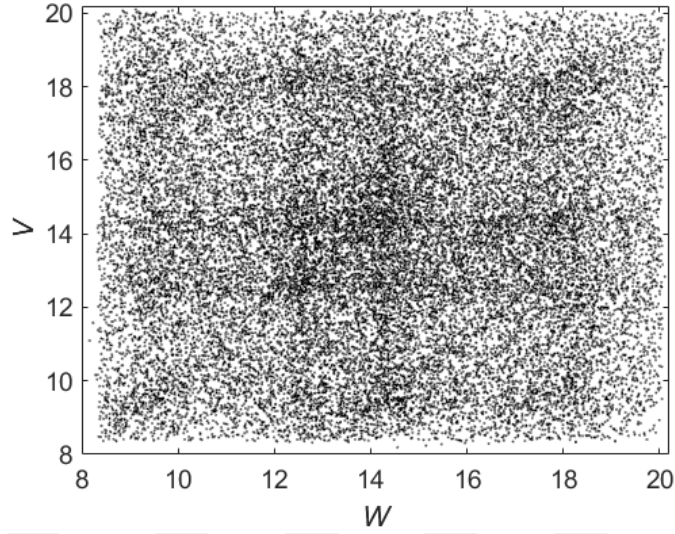


Figure 4.10: Implementation of the auxiliary system approach at $W_t = 28.9$, $R_s = 30709$.

4.6 Conclusions to this chapter

This chapter describes the "unpredictability" and "Delta Synchronization of Chaos" (DSC) techniques with the algorithms and numerical characteristics (degree of numerical unpredictability and synchronization). A GDSS is considered, which is based on a one-dimensional "extended fluid model" approach. The calculations of the concerned model are dimensionally done in chapter 3. This chapter converts the dimensional results into dimensionless quantities by applying defined scale laws. The transition from a periodic to a completely chaotic state is demonstrated via phase portraits and bifurcation diagrams in the dimensionless spaces. The drive and response systems are developed only focusing the chaotic regime of the GDSS model. By implementing the "unpredictability" algorithm, the existence of chaotic solutions is proved by obtaining the sequences of convergence and separation. Although generalized synchronization is absent, the coupled system still exhibits chaos synchronization. This is achieved by deriving shared time values by applying the DSC algorithm, indicating

instances when the coupled system behaves similarly.

The similar analyses are conducted in the prior study [4], which focused on a more straightforward one-dimensional fluid modeling approach within the same parameter regime. In the present study, an advanced one dimensional fluid model is considered, which is detailed in terms of the plasma chemistry and treatment of electron dynamics. Although there is approximately 7% simulation time difference between these two studies, the degree of numerical unpredictability of the drive and response systems in the present study have lower values compared to the previous study. This implies that the detailed fluid model is *more unpredictable* and essentially *more chaotic* than the simpler fluid model. The degree of numerical synchronization is almost two times smaller than the previous study, which implies that the detailed fluid model is *more synchronous*. As a result, when the aforementioned analyzes are applied to the more realistic models, the results would be more unpredictable and synchronous.

CHAPTER 5

ANALYSIS OF BOUNDARY CONDITIONS FOR FLUID MODELS OF GAS DISCHARGES: IMPLEMENTATION OF THE ION BOUNDARY FLUX

This chapter discusses the correct definition of boundary conditions in the fluid model of gas discharges. Several sets of boundary conditions are considered for a radio frequency capacitively coupled plasma (RF CCP) discharge in argon. One-dimensional "extended" fluid modeling (LMEA) approach described in the chapters 2, 3 and 4 is used in the analyses. In the literature, the suggested boundary conditions for ion flux in fluid modeling of the discharges include a non-physical switching factor. Therefore, the numerical investigations mainly focus on the ion boundary flux, which forms the backbone of this study. The statement of the problem is introduced in section 5.1. Section 5.2 describes governing equations, transport and rate coefficients, set of plasma chemical reactions, and boundary conditions of the model. In section 5.3, the results for implementing defined boundary conditions are discussed. The presented boundary conditions' accuracies are also evaluated by comparing obtained results with PIC/MCC simulation results. The conclusions to the problem are presented in section 5.4.

5.1 The statements of the problem

Radio frequency plasmas are common in many practical situations, such as laboratory experiments and manufacturing processes. The etching and deposition of thin films on semiconductors, creation of quantum dots [257], plasma synthesis [258], producing carbon nanotubes [259], PECVD (Plasma Enhanced Chemical Vapor Deposition) [260], and coating of nanoparticles are the processes where RF plasmas

are widely used. They are also used in medical applications such as equipment sterilization [18, 261], healing wounds [198], and treatment of cancer [262]. Understanding the fundamentals of RF plasmas and applied plasma setups, it is essential to develop a fluid modeling approach to perform efficient calculations. In this chapter, a one-dimensional "extended fluid model" (LMEA), based on the solutions of "drift-diffusion" equations of each plasma species, is employed to model the RF CCP discharge in argon.

Although boundary conditions (BCs) are required to complete the fluid model equations, there needs to be a consensus in the existing literature for the formulation of correct BCs in RF plasma systems. Generally, BCs in RF discharge systems were conventionally defined as those of DC systems. Studies have been carried out for the correct description of BCs used in DC-supplied discharge systems [101, 117]; however, different approaches to the formulation of BCs are used for RF plasmas [96, 106]. In [106], it has been shown that the choice of BC can significantly affect the sheath and bulk regions of the RF-supplied discharge. The thermal and diffusion fluxes directed to the boundary walls were also important. A lacking consensus on the exact form of BCs leads to one of the sources of numerical uncertainty around and including the boundaries of the fluid modeling calculations of the discharge systems. Indeed, the applicability of fluid modeling for the k -type plasma component depends on the comparison between the small mean free path λ_k and the characteristic size L of the discharge system [263–267]

$$L > \lambda_k.$$

Therefore, fluid modeling approaches are inapplicable in regions where the distances from the system's boundaries are less than λ_k . Consequently, it is crucial to formulate appropriate boundary conditions to complete equations of fluid modeling to develop an effective numerical model of gas discharges.

Regions in the vicinity of boundaries have both high and small electric fields. In regions with a low electric field, the thermal velocity of particles is comparable to the drift velocity of particles. For this reason, particles in low electric field region around the boundary have an almost isotropic velocity distribution in their direction of motion. In regions with a high electric field, charged particles' drift velocity exceeds the particles' thermal velocity. In this case, the moving particles' velocity distribution

functions are far from being isotropic. As a result, BCs must be set at all boundaries such that these can consider the effects of low and high electric field regions in the vicinity of the walls. This situation requires a formulation of a universal boundary condition, which includes all the information related to field magnitudes and particle types when simulating an RF discharge system.

In some studies in the literature, it is assumed that the surfaces absorb charged particles. In this case, the number and energy densities at the walls are defined as zero [120,121]. In other studies, the boundary fluxes are defined in terms of drift [119,122] and thermal [29,123] terms, or both are included using a nonphysical switch function. For example, in [29,101,106], the generalized forms of boundary conditions include this strange quantity, which is turned on the drift term if the electric field is toward the wall; otherwise, the contribution of this term is not taken into account. Moreover, the significant effects of BCs on the results of discharge modeling are demonstrated in the [106]. Therefore, formulating a universal boundary condition without a non-physical switch function is an urgent problem in the existing literature.

Another point to be clarified is the secondary electron emission (SEE) coefficient, which is responsible for ionization due to ion impacts on the surfaces. Unlike BCs used in DC-powered plasma discharge systems [101,118], RF discharges often ignore this coefficient even though it is known to affect the sheath and bulk regions. The following analyses also include the SEE mechanism's significance on the bulk and sheath regions in the RF-supplied discharge gap.

5.2 The description of gas discharge model

5.2.1 Governing equations

The numerical calculations of the RF CCP discharge system are based on a one-dimensional "extended fluid model", also known as LMEA [32]. This modeling approach is described in detail in chapters 2, 3, and 4. For the completeness of this chapter, it is also appropriate to define the equations here. The gas discharge model

involves the continuity equations

$$\frac{\partial n_k}{\partial t} + \frac{\partial \Gamma_k}{\partial x} = S_k \quad (5.2.1)$$

where the particle flux densities Γ_k are defined with "drift-diffusion" flux densities

$$\Gamma_k = \text{sgn}(q_k) \mu_k n_k E - D_k \frac{\partial n_k}{\partial x}, \quad (5.2.2)$$

the Poisson equation for the electric field

$$\epsilon_0 \frac{\partial E}{\partial x} = \sum_k q_k n_k \quad \text{and} \quad E = -\frac{\partial \varphi}{\partial x}, \quad (5.2.3)$$

and the electron energy equation

$$\begin{aligned} \frac{\partial n_\varepsilon}{\partial t} + \frac{\partial \Gamma_\varepsilon}{\partial x} = & - e \Gamma_e E - \frac{3}{2} \frac{m_e}{m_g} \nu_{ea} n_e k_B (T_e - T_g) \\ & - \sum_j \Delta E_j R_j. \end{aligned} \quad (5.2.4)$$

In these equations, n_k stands for the number density, E and φ denote the electric field and potential, μ_k and D_k are the mobility and diffusion coefficients, S_k indicates the rate of creation and destruction of species, and subscript k is the sort of species such as electrons, ions, and metastable atoms. In the electron energy equation, $n_\varepsilon = n_e \bar{\varepsilon}$ is the electron energy density, where $\bar{\varepsilon} = \frac{3}{2} k_B T_e$ is the mean electron energy, and Γ_ε is the density of the electron energy flux

$$\Gamma_\varepsilon = -D_\varepsilon \frac{\partial n_\varepsilon}{\partial x} - \mu_\varepsilon E n_\varepsilon. \quad (5.2.5)$$

The terms on the right-hand side of equation 5.2.4 describe the Joule heating (or cooling) of electrons in the electric field and the energy losses in the elastic and inelastic collisions, where ν_{ea} is the frequency of the electron-atomic elastic collision, R_j represents the reaction rate of j th reaction, ΔE_j denotes the corresponding energy loss (or gain), m is the particle mass, and T indicates the kinetic temperature.

5.2.2 Reaction rate and transport coefficients

The calculations are performed for the gas discharge in argon. The set of reactions is summarized in table 5.1, which is similar to that in Ref. [103]. Four plasma species

are considered: electrons, Ar^+ ions, excited argon atoms Ar^* , and Ar atoms in the ground state. The mobility and diffusion coefficients are obtained from the relations

$$\mu_e = -\frac{e}{n_e m_e} \int_0^\infty D_r \sqrt{\varepsilon_k} \frac{\partial F_0}{\partial \varepsilon_k} d\varepsilon_k, \quad (5.2.6)$$

$$D_e = \frac{1}{n_e} \int_0^\infty D_r \sqrt{\varepsilon_k} F_0 d\varepsilon_k, \quad (5.2.7)$$

where $D_r = 2\varepsilon_k/3m_e\nu_{ea}$ represents the space diffusion coefficient, $\varepsilon_k = m_e v_e^2/(2e)$ is the electron kinetic energy (in eV units), F_0 denotes the EEDF, calculated from the solution of local Boltzmann equation. This distribution function is normalized to unity according to

$$\int_0^\infty F_0(\varepsilon) \sqrt{\varepsilon} d\varepsilon = 1. \quad (5.2.8)$$

The electron-atomic elastic collision frequency is obtained from

$$\nu_{ea} = \frac{e}{m_e} \frac{1}{\mu_e}. \quad (5.2.9)$$

Table 5.1: Set of plasma chemical reactions.

Index	Reaction	Type	$\Delta E(\text{eV})$	Rate coefficient
R1	$e + Ar \rightarrow e + Ar$	Elastic collision	0	Cross section
R2	$e + Ar \rightarrow 2e + Ar^+$	Direct ionization	15.8	Cross section
R3	$e + Ar \rightarrow e + Ar^*$	Excitation	11.4	Cross section
R4	$e + Ar \rightarrow e + Ar$	Excitation	13.1	Cross section
R5	$e + Ar^* \rightarrow 2e + Ar^+$	Stepwise ionization	4.4	Cross section
R6	$2Ar^* \rightarrow e + Ar^+ + Ar$	Penning ionization	-	$6.2 \times 10^{-10} \text{cm}^3 \text{s}^{-1}$
R7	$Ar^* \rightarrow h\nu + Ar$	Radiation	-	$1.0 \times 10^7 \text{s}^{-1}$

The source terms S_k in the continuity equation 5.2.1 are determined by the chemical reactions in the bulk of the discharge

$$S_k = \sum_i R_i - \sum_j R'_j, \quad (5.2.10)$$

where R_i and R'_j correspond to the creation and destruction rates in i th and j th reactions. Direct, stepwise, and penning ionizations in table 5.1 determine the source

terms for electrons and ions

$$S_e = S_i = R_2 + R_5 + R_6. \quad (5.2.11)$$

These rate terms are proportional to the rate coefficients and number densities of species of respective reactions such that $R_2 = K_2 n_e n_0$, $R_5 = K_5 n_e n_m$, and $R_6 = K_6 n_m n_m$, where n_0 is the concentration of neutral atoms. The reactions of excitation, stepwise ionization, penning ionization, and radiation reactions determine the balance of excited *Ar* atoms

$$S_m = R_3 - R_5 - 2R_6 - R_7 = K_3 n_e n_0 - K_5 n_e n_m - 2K_6 n_m n_m - K_7 n_m. \quad (5.2.12)$$

For electron-induced reactions ($R_1 - R_5$ in table 5.1), the corresponding rate coefficients are calculated as functions of T_e as follows

$$K_R = \int_0^\infty \sqrt{\varepsilon_k} \sigma_R F_0 d\varepsilon_k, \quad (5.2.13)$$

where σ_R indicates the collision cross sections of reaction R . The dependence of σ_R on energy with related reactions in table 5.1 is demonstrated in figure 5.1

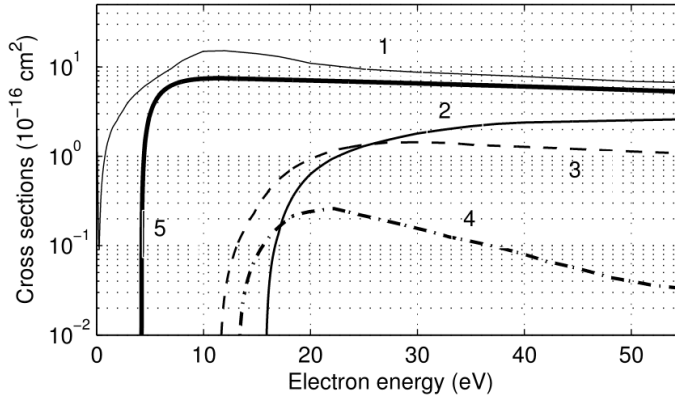


Figure 5.1: Electron cross sections for (1) elastic, (2) direct ionization, (3) excitation, (4) excitation, and (5) stepwise ionization reactions in argon, included in the numerical model. The numbering on the curves corresponds to the indices of reactions in table 5.1.

Table 5.2: The set of boundary conditions.

	BC set 1 (BC-1)	BC set 2 (BC-2)	BC set 3 (BC-3)	BC set 4 (BC-4)
$\hat{\mathbf{n}} \cdot \boldsymbol{\Gamma}_e$	$\frac{1}{4}v_e n_e - \gamma \hat{\mathbf{n}} \cdot \boldsymbol{\Gamma}_i$	$\frac{1}{2}v_e n_e - \gamma \hat{\mathbf{n}} \cdot \boldsymbol{\Gamma}_i$	$\frac{1}{2}v_e n_e - 2\gamma \hat{\mathbf{n}} \cdot \boldsymbol{\Gamma}_i$	$\frac{1}{2}v_e n_e - 2\gamma \hat{\mathbf{n}} \cdot \boldsymbol{\Gamma}_i$
$\hat{\mathbf{n}} \cdot \boldsymbol{\Gamma}_i$	$\frac{1}{4}v_i n_i + a n_i \mu_i (\hat{\mathbf{n}} \cdot \mathbf{E})$	$\frac{1}{2}v_i n_i + a n_i \mu_i (\hat{\mathbf{n}} \cdot \mathbf{E})$	$n_i \mu_i (\hat{\mathbf{n}} \cdot \mathbf{E})$	Univ. condition (5.3.1)
$\hat{\mathbf{n}} \cdot \boldsymbol{\Gamma}_m$	$\frac{1}{4}v_m n_m$	$\frac{1}{2}v_m n_m$	$\frac{1}{2}v_m n_m$	$\frac{1}{2}v_m n_m$
$\hat{\mathbf{n}} \cdot \boldsymbol{\Gamma}_\varepsilon$	$\frac{1}{3}v_e n_\varepsilon - 2k_B T_e \gamma \hat{\mathbf{n}} \cdot \boldsymbol{\Gamma}_i$	$\frac{5}{6}v_e n_\varepsilon - \frac{3}{2}k_B T_e \gamma \hat{\mathbf{n}} \cdot \boldsymbol{\Gamma}_i$	$\frac{2}{3}v_e n_\varepsilon - 3k_B T_e \gamma \hat{\mathbf{n}} \cdot \boldsymbol{\Gamma}_i$	$\frac{2}{3}v_e n_\varepsilon - 3k_B T_e \gamma \hat{\mathbf{n}} \cdot \boldsymbol{\Gamma}_i$

5.2.3 Boundary conditions

The boundary conditions employed in this chapter are listed in table 5.2. Conditions defined in BC-1 are taken from Ref. [103], which is based on the CFD-ACE+ solver [268]. BC-2 considers predefined boundary conditions for discharge simulations in the COMSOL's built-in Plasma Module [269]. BC-3 is based on the conditions formulated in [96]. This boundary condition is critical because, unlike BC-1 and BC-2, it does not consider the non-physical switching function in ion boundary fluxes. BC-4 differs from BC-3 only in the definition of ion boundary flux. BC-4 defines a universal expression including drift and diffusion information without using a switching function. Therefore, the most precise one among the specified boundary conditions is BC-4.

In table 5.2, the thermal velocity of the species j is defined as $v_j = \sqrt{8k_B T_j / \pi m_j}$. Constant γ is the secondary emission coefficient. The flux density $\boldsymbol{\Gamma}$ is defined by the equations 5.2.2 and 5.2.5. Note that a switching function a is employed to regulate the contribution of drift term in the ion boundary flux in the BC-1 and BC-2. It is equal to one if the electric field direction is toward the wall; otherwise, it is set to zero:

$$a = \begin{cases} 1, & (\hat{\mathbf{n}} \cdot \mathbf{E}) > 0, \\ 0, & (\hat{\mathbf{n}} \cdot \mathbf{E}) \leq 0. \end{cases} \quad (5.2.14)$$

where $\hat{\mathbf{n}}$ is a normal unit vector pointing towards the boundary wall.

The right side electrode is feeded by a sinusoidal RF voltage, while the left side electrode is grounded:

$$\varphi = 0 \text{ at } x = 0, \quad \varphi = V_0 \sin(2\pi f t) \text{ at } x = d,$$

where V_0 is the amplitude, f is the frequency of the total applied voltage, and the

distance d is the gap length of the discharge.

5.3 Results and discussion

Numerical calculations investigate the effects of different sets of boundary conditions defined in table 5.2 on the plasma discharge properties. Calculations are carried out for the gas discharge in argon at pressure $p = 3$ torr. The RF voltage drives the system with an amplitude $V_0 = 300$ V. The frequency of the supplied RF field f is taken as 0.85 MHz. The discharge gap width is $d = 1$ cm. The background gas temperature is $T_g = 0.025$ eV, and the thermal mechanism is ignored. Diffusion coefficient for ions and metastable atoms are $D_i = 0.0021$ m²/s and $D_m = 0.0036$ m²/s, respectively. These are related to corresponding mobilities via the Einstein relation, $D/\mu = k_B T/e$, where p and T indicate the background gas pressure and kinetic temperature. The secondary electron emission coefficient $\gamma = 0.07$ is constant and field-independent.

5.3.1 Effects of the different forms for the ion boundary flux

In this section, the effects of using different ion boundary fluxes on the models are tested. To do this, the set of BC-3 conditions, containing only a "drift" term as an ion boundary flux, and BC-4, employing a "universal" ion boundary flux, are considered. Only the ion boundary flux terms of BC-3 and BC-4 are different, and the remaining terms are the same, as seen in table 5.2. In addition, "purely thermal" ion boundary flux is also defined, provided the remaining conditions are kept the same as in the sets BC-3 and BC-4. These three formulations for the ion boundary flux Γ_i are given as follows:

"universal" BC

$$\Gamma_i = n_i v_{d,i} \left[\frac{1}{U_{d,i}} \frac{1}{2\sqrt{\pi}} \exp(-U_{d,i}^2) + \frac{1}{2} \operatorname{erfc}(-U_{d,i}) \right], \quad (5.3.1)$$

"purely drift" BC (it is identical to that in table 5.2)

$$\Gamma_i = n_i v_{d,i}, \quad (5.3.2)$$

"purely thermal" BC

$$\Gamma_i = \frac{1}{4} n_i v_i, \quad (5.3.3)$$

where $v_{d,i} = \mu_i E$, $U_{d,i} = v_{d,i} \sqrt{m_i / (2k_B T_i)}$, $\text{erfc}(x) = 1 - \text{erf}(x)$ that $\text{erf}(x)$ is the error function.

In figure 5.2, spatial distributions of the time-averaged ion density, n_i , derived from these three formulations ("universal", "purely drift", and "purely thermal") are demonstrated. Although the "purely drift" and "universal" BCs almost overlap along the computation domain, there is a slight difference in the close neighborhood of the boundary (see the inset of the figure 5.2). This minor difference shows that the diffusion information included in the "universal" condition is ignorable when compared to the drift term. However, the results of purely thermal BC show a dramatic peak at the edges of the domain, which seems more detailed at the inset of figure 5.2.

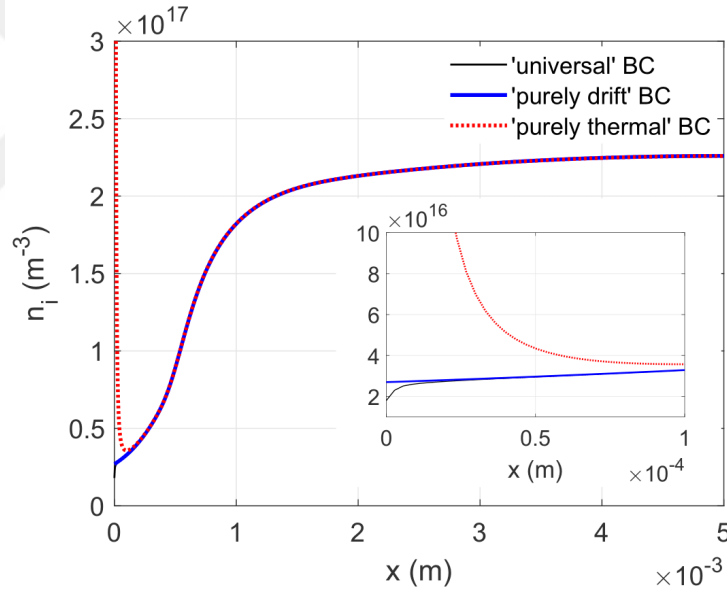


Figure 5.2: Spatial profiles of the time-averaged ion density, n_i , obtained from three formulations for the boundary ion flux Γ_i ("universal", "purely drift", "purely thermal"). Inset displays the same profiles in the immediate vicinity of the left boundary of the domain. Note that only half of the whole domain is plotted because the other side shows the same behavior due to symmetry. Discharge conditions are pressure $p = 3$ torr, the total applied voltage amplitude and frequency are $V_0 = 300$ V and $f = 0.85$ MHz, and the discharge gap is $d = 1$ cm.

5.3.2 Effect of approximation for T_i

In order to understand the dramatic increase of the ion density, n_i , at the edges of the domain when "purely thermal" BC is defined for the ion flux (see figure 5.2), we firstly intended to analyse the effect of the ion temperature, T_i approximations. The results in figure 5.2 are obtained as a result of the computations made by assuming the condition $T_i = T_g$. Alongside with this condition, two forms of the Wannier's [33] formula are taken into account as follows

$$\text{i: } T_i = T_g, \quad (5.3.4)$$

$$\text{ii: } T_i = T_g + \frac{m_i + m_g}{5m_i + 3m_g} \frac{m_g (\mu_i E)^2}{k_B}, \quad (5.3.5)$$

$$\text{iii: } T_i = T_{imax} \tanh \left[\frac{1}{T_{imax}} \left(T_g + \frac{(m_i + m_g) (\mu_i E)^2}{3k_B} \right) \right]. \quad (5.3.6)$$

In the above definitions, m_i and m_g are the masses of the ion and gas particles. These are assumed to be almost equal, $m_i \approx m_g$. At relatively small electric field values, the ion temperatures defined in cases (ii) and (iii) are very close to each other.

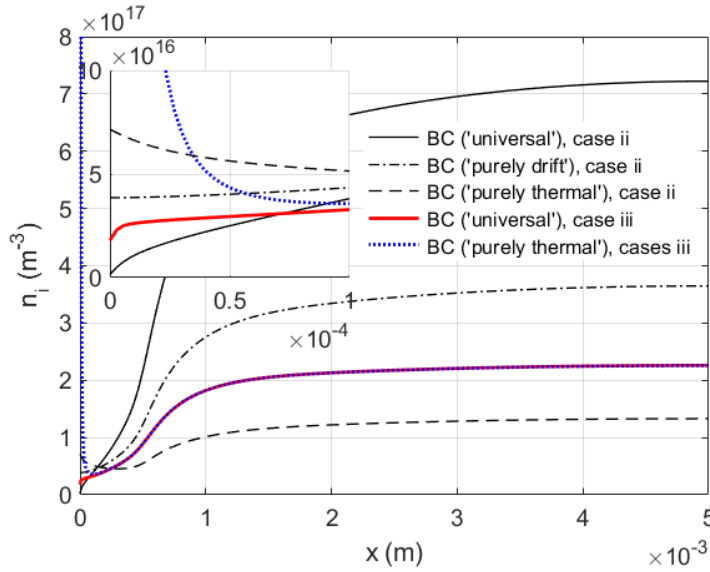


Figure 5.3: Spatial profiles of the time-averaged ion density, n_i , obtained from the models based on three formulations of ion boundary flux Γ_i ("universal", "purely drift", "purely thermal") with defined T_i approximations (indicated as case (i), (ii), and (iii)). Discharge conditions are the same as in figure 5.2.

However, in case (iii), an uncontrolled increase in ion temperature with an electric

field has bounded with an upper limit value. The calculations define it as $T_{imax} = 0.52$ eV. Figure 5.3 shows spatial distributions of time-averaged ion density n_i for each ion temperature conditions (i), (ii), and (iii) of the models describing the ion boundary flux Γ_i as in the equations 5.3.1, 5.3.2, and 5.3.3 ("universal", "purely drift", "purely thermal"). The inset of figure 5.3 represents the results enlarged at a distance of 1×10^{-4} m from the left boundary. As can be observed from this figure, ion temperature defined by the case (ii) for the models including "universal", "purely drift", and "purely thermal" conditions as ion boundary fluxes lead to significant ion density deviations in the bulk region of discharge and vicinity of the left boundary. Because the ion temperature defined by the case (ii) reaches 10^2 eV orders, which is even higher than the temperature of the electrons demonstrated in figure 5.4. As a result, the definition of ion temperature defined in case (ii) causes non-physical consequences regardless of which ion boundary flux is utilized.

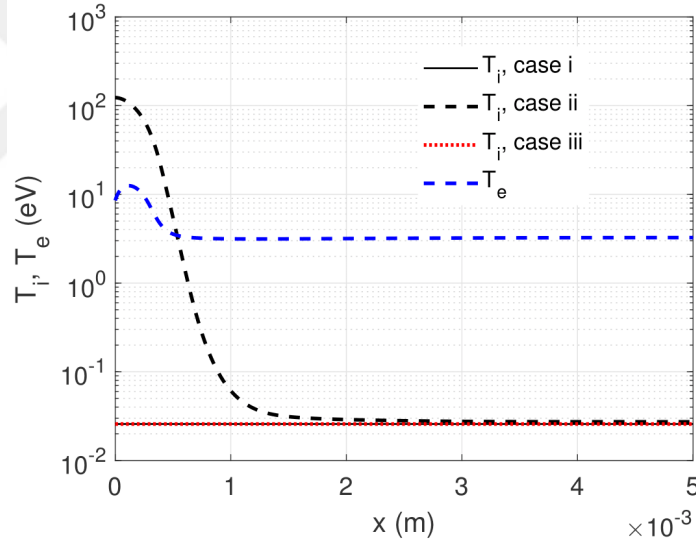


Figure 5.4: Spatial profiles of the time-averaged electron and ion temperatures T_e and T_i , derived from different ion temperature approximations (case (i), (ii), and (iii)). The discharge conditions are the same as in figure 5.2.

Note that cases (i) and (iii) practically yield similar results for each corresponding modeling approach in the given computation domain. The peaks in the model with "purely thermal" ion boundary flux do not disappear for cases (i) and (iii). It means that ion temperature approximations are not related to the edge behavior of the mod-

els. Referring to figure 5.4, the spatial profiles of time-averaged ion temperatures for cases (i) and (iii) overlap and are smaller than the electron temperatures, as expected. As a result, this analysis reveals that

- The ion and electron density profiles of the models in which BC-3 ("purely drift") and BC-4 ("universal") ion boundary fluxes are defined give similar results regardless of which ion temperature approach (case (i) or (iii)) is used. It means that these two forms of boundary conditions are practically equivalent.
- The ion temperature approximation defined by $T_i = T_g$ (case (i)) gives similar results to those obtained with modified form of Wannier's formula (case (iii)). Either case (i) or (iii) is sufficient to define the ion temperature in the RF CCP discharge system in a given parameter regime.
- For the "purely thermal" ion flux model, the ion density peaks at the edges of the boundaries is not related to which ion temperature approximations are employed.

5.3.3 Effect of low and high electric field regions

In light of these results, we are now confident that different ion temperature approaches do not lead to dramatic increases around the edges. In that case, the attention is directed to the effects of variations in the electric field's magnitude on the charged particles around the boundaries. In contrast to low electric field regions with an isotropic distribution function [8, 27], the velocity distribution function of charged particles will not be isotropic in regions with a high electric field [27]. Thus, the drift velocity of the particles significantly exceeds the thermal velocity, and the model with a "purely drift" boundary condition gives a result closer to the universal one.

To test the above claim, ion boundary conditions are investigated at small and high electric field limits. Firstly, the expressions in the equation 5.3.1 ("universal"), 5.3.2 ("purely drift"), and 5.3.3 ("purely thermal") are multiplied by a factor $\frac{1}{n_i} \sqrt{\frac{m_i}{2k_B T_i}}$ to make them dimensionless. The dimensionless forms of ion boundary fluxes are as follows.

"universal" BC

$$\tilde{\Gamma}_i = \frac{1}{2\sqrt{\pi}} \exp(-U_{d,i}^2) + \frac{1}{2} \operatorname{erfc}(-U_{d,i}), \quad (5.3.7)$$

"purely drift" BC

$$\tilde{\Gamma}_i = U_{d,i}, \quad (5.3.8)$$

"purely thermal" BC

$$\tilde{\Gamma}_i = \frac{1}{2\sqrt{\pi}}. \quad (5.3.9)$$

Secondly, the dependence of dimensionless ion boundary fluxes, $\tilde{\Gamma}_i$, on the dimensionless drift velocity, $U_{d,i} = v_{d,i} \sqrt{m_i / (2k_B T_i)}$ where $v_{d,i} = \mu_i E$, is demonstrated in figure 5.5. It is a classical illustration of a flux of particles to the wall [100]. Unlike low $U_{d,i}$ values, corresponding to the isotropic case of charged particles, the ion boundary flux of "purely drift" overlaps with the "universal" one at high $U_{d,i}$ values, that is, at high E-field values.

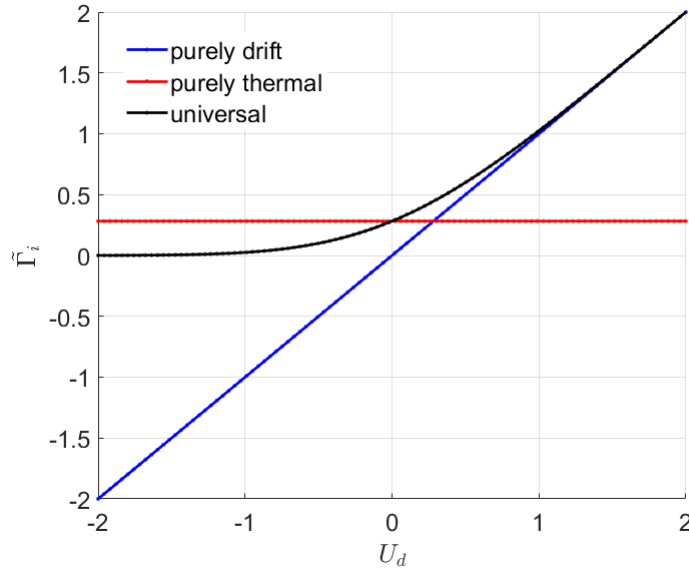


Figure 5.5: The dependence of three dimensionless formulations of ion boundary $\tilde{\Gamma}_i$ ("universal", "purely drift", and "purely thermal") on the dimensionless drift velocity $U_{d,i}$

As a result, in contrast to the highly overlapping bulk region (see Fig 5.2), the fact that the velocity distribution of the charged particles is not isotropic is found to be the

reason why the model with purely thermal BC give inconsistent results in the sheath regions (high electric field regions).

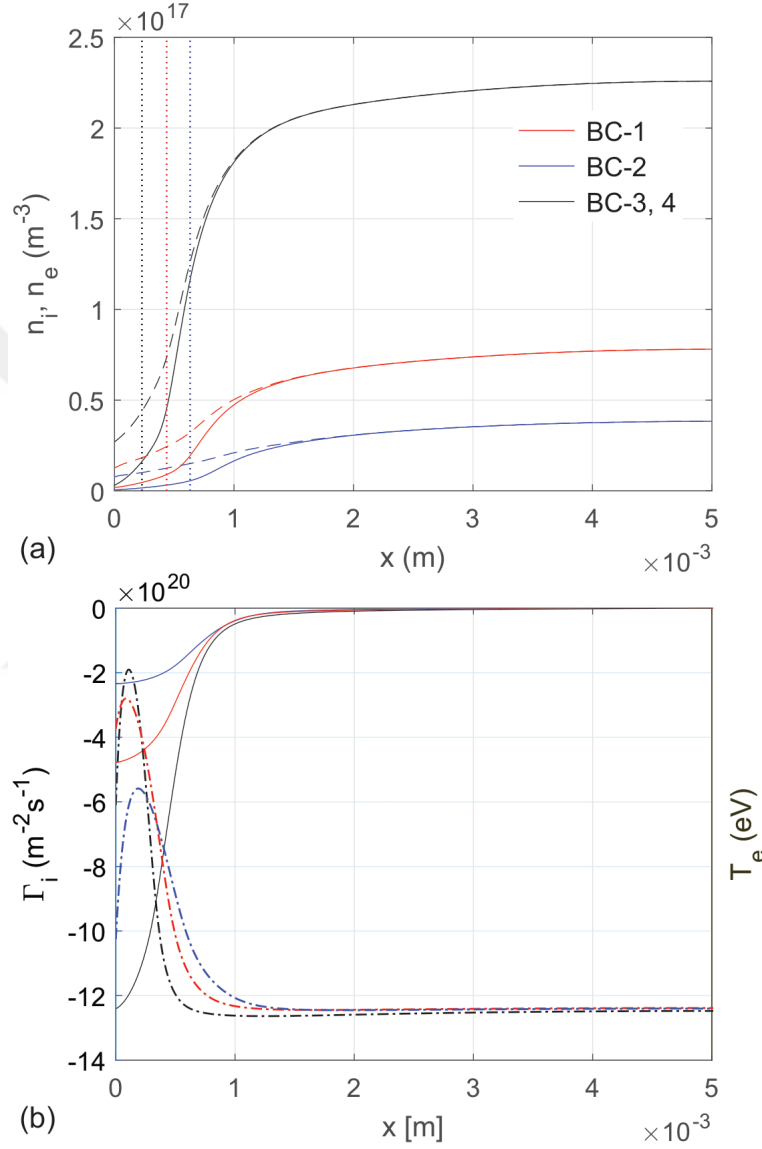


Figure 5.6: Spatial profiles of the time-averaged (a) electron n_e (solid lines) and ion n_i (dashed lines) number densities, and (b) of the ion flux density Γ_i (solid lines) and electron temperature T_e (dash-dot lines), obtained from the models implementing BCs from table 5.2. The vertical lines in panel (a) indicate the sheath layers. The conditions are the same as in figure 5.2.

5.3.4 Effect of different BCs

After understanding how the drift and pure components of ion boundary fluxes affect the modeling results in the previous section, the results of the models based on the BCs given in table 5.2 can be compared. Spatial distributions of typical plasma variables obtained by implementing boundary conditions given in table 5.2 are compared in figure 5.6. The spatial profiles of time-averaged ion n_i and electron n_e densities are shown in figure 5.6(a). Vertical lines indicate the corresponding sheath layers of each model with different BCs. Calculations reveal no difference in plasma parameters derived from BC-3 and BC-4. However, the results of BC-1 and BC-2 differ significantly. Figure 5.6(b) also confirms this in terms of the spatial distribution of ion flux density Γ_i and electron temperature T_e .

5.3.5 Effect of secondary electron emission coefficient

In previous sections, BC-3 and BC-4 (the most accurate one in table 5.2) give identical results. Therefore, the model using BC-3 as the ion boundary flux can be utilized to observe the effects of secondary electron emission by implementing different coefficient values $\gamma = 0.04, 0.07$, and 0.1 . As the γ value takes larger values, the ionization rate in the sheath region increases with narrower sheath widths due to the fact that emitted electrons from the surface become more probable. Both ion and electron number densities, n_e and n_i , experience an increase (refer to figure 5.7(a)), leading to a significant increase in the electron temperature, T_e , particularly in the vicinity of the sheath regions. It is depicted in figure 5.7(b).

In table 5.3, the maximum point of plasma density in the bulk region, where the ion number density equals the electron number density, is given for the BCs specified in table 5.2. The percentage differences of BC-3 for various secondary emission coefficients are compared to BC-3 ($\gamma = 0.07$). There is a %75 difference between the maximum plasma densities for $\gamma = 0.04$ and $\gamma = 0.07$, and a %172 difference for $\gamma = 0.07$ and $\gamma = 0.1$. The increase in maximum plasma densities with γ is related to the increase in electron temperature and ionization rate in the sheath region. These lead to generating more secondary electrons, which are accelerated toward the bulk

region under the influence of the electric field in the sheath region. Therefore, the plasma medium with higher ion and electron number densities is formed in the bulk region of the domain.

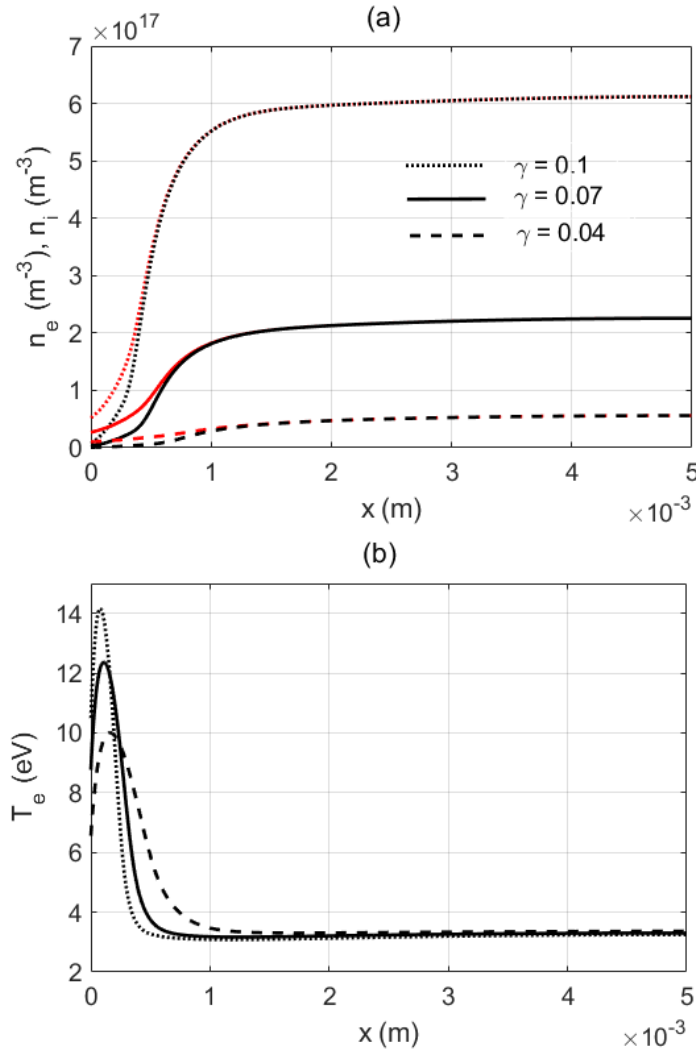


Figure 5.7: Spatial profiles of the time-averaged (a) ion n_i (red colored lines) and electron n_e (black colored lines) number densities, and (b) electron temperature T_e , obtained from the model of BC-3 with different values of γ . The conditions are the same as in figure 5.2.

In figure 5.8(a) and 5.8(b), the spatial distributions of the electron and ion number densities n_e and n_i are normalized by the corresponding maximum plasma densities. Note that the maximum of bulk plasma density in BC-2 case is the smallest one ($3.83 \times 10^{16} \text{ m}^{-3}$) in table 5.3, which corresponds to the largest sheath region. In

Table 5.3: Effect of boundary conditions on the maximum bulk plasma density. The pressure is $p = 3$ torr, the applied voltage amplitude and frequency are $V_0 = 300$ V and $f = 0.85$ MHz, the discharge gap is $d = 1$ cm.

	Maximum of bulk plasma density $\times 10^{16}$ (m $^{-3}$)	% Difference (compared to BC-3)
BC-1 ($\gamma = 0.07$)	7.81	%65
BC-2 ($\gamma = 0.07$)	3.83	%83
BC-3 ($\gamma = 0.07$)	22.5	<i>N/A</i>
BC-4 ($\gamma = 0.07$)	22.6	%0.4
BC-3 ($\gamma = 0.04$)	5.61	%75
BC-3 ($\gamma = 0.10$)	61.2	%172

other words, the BC-2 case becomes normalized to unity in a more distant part of the domain when compared to the other normalized cases in figures 5.8(a) and 5.8(b). In contrast, BC-3 with $\gamma = 0.1$ leads to the smallest sheath width. Thus, the increases or decreases of the γ value cause a significant impact on sheath width. Notice from table 5.3 and figure 5.8 that the maximum bulk plasma density difference between BC-3 and BC-4 is negligible.

5.3.6 Comparison to PIC/MCC simulation results

To test the accuracy of the BCs used in this study, the calculated results are compared with particle models obtained by PIC/MCC (Particle In Cell / Monte Carlo Collision) methods. Here, the background gas pressure of argon is $p = 0.1$ torr. The gas remains near room temperature, with $T_g = 0.03$ eV (348 K), and the discharge gap width is $d = 2$ cm. The amplitude and frequency of the supplied RF field are $V_0 = 270$ V and $f = 13.56$ MHz. Constant secondary emission coefficients are zero in the PIC/MCC models used in the references [270,271]. The γ values are set to $\gamma = 0.16$ in calculations implementing BC-1, BC-2, and BC-3. Furthermore, the γ is adjusted as 0.1745 to obtain a better fit of BC-3 with the PIC/MCC simulation results. Figure 5.9 (a) shows the spatial profiles of time-averaged ion density n_i for various modeling approaches. The PIC/MCC simulations described in [270,271] give results closer to the model implementing BC-3 ($\gamma = 0.16$) compared to BC-1 and BC-2. The constant γ for BC-3 is taken as 0.1745 to fit the fluid modeling n_i data with those

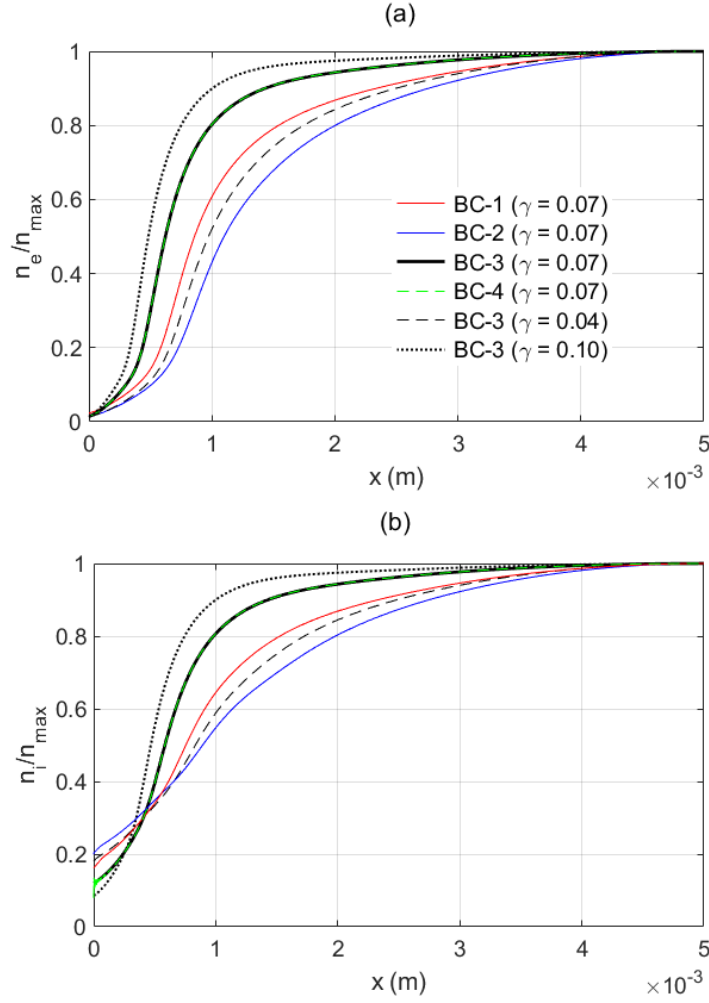


Figure 5.8: Spatial profiles of the time-averaged, normalized (a) electron and (b) ion number densities n_e and n_i . The conditions are the same as in figure 5.2.

of the particle simulations. As the maximum n_i of the BC-3 model approaches the peaks of the particle simulations in figure 5.9(a), it starts to diverge in the sheath region (see figure 5.9(b)). Note that the fluid modeling results in the parameter, as mentioned earlier, are sensitive to the secondary emission coefficient. When the γ value is increased from 0.16 to 0.17, the maximum ion density increases by %32.5. If the γ value is increased from 0.17 to 0.1745, the maximum ion density increases by %30. Compared to BC-1 and BC-2, the model with BC-3 ($\gamma = 0.16$) is in good agreement with those of PIC/MCC simulations in the sheath region of the discharge. Figure 5.9 (c) shows the spatial profile of the discharge potential. Although all the considered models demonstrate a similar trend with the discharge potentials derived

from PIC/MCC methods, the models with BC-3 are closer to the particle simulations than the BC-1 and BC-2.

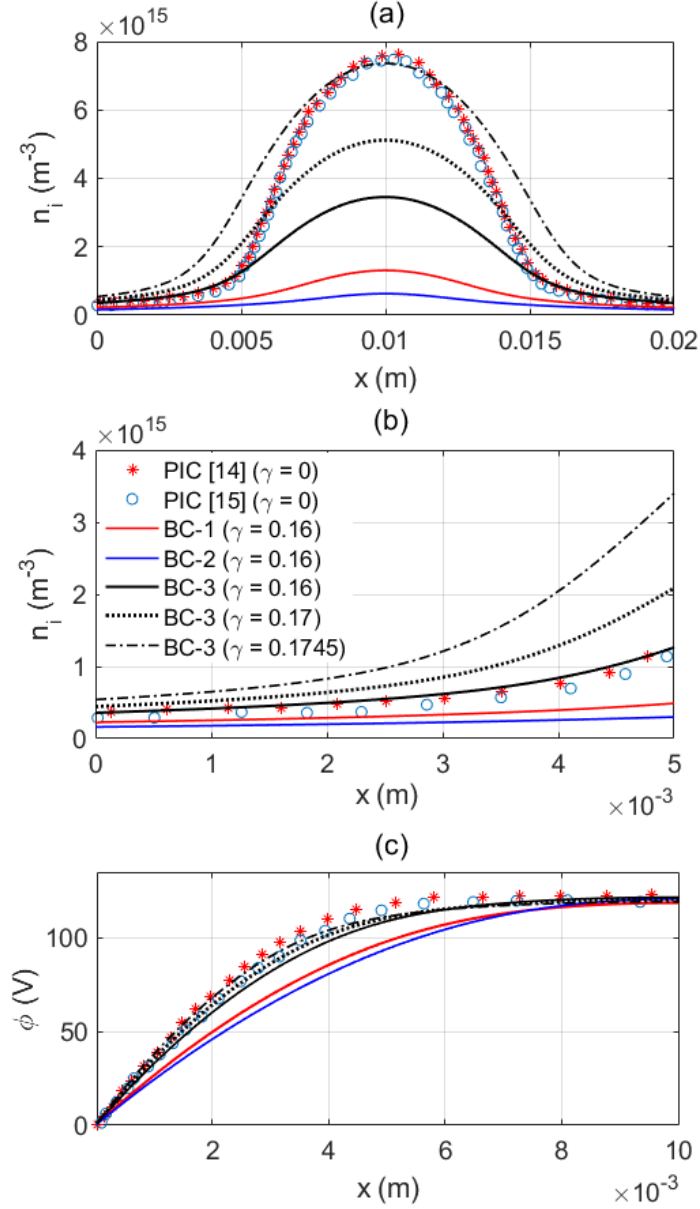


Figure 5.9: (a) Spatial profiles of the time-averaged ion density n_i , (b) enlarged form of the panel (a), and (c) the time-averaged profiles of the potential for different modeling approaches and boundary conditions. Discharge conditions are pressure of the argon gas $p = 0.1$ torr, the total applied voltage amplitude and frequency $V_0 = 270$ V and $f = 13.56$ MHz, the discharge gap $d = 2$ cm, and the background gas temperature $T_g = 0.03$ eV

Developing exact compatibility between the PIC/MCC methods and fluid models is not possible because the fluid modeling approaches inherently lead to results with less accuracy than fully kinetic particle simulations. However, particle simulations are not computationally efficient; therefore, the fluid modeling approaches have become a practical alternative for numerical calculations. Moreover, a more realistic and detailed fluid model can be derived by considering more plasma chemical reactions. Thus, the different plasma chemical reaction sets may increase the compatibility between the fluid model data and PIC/MCC results.

5.4 Conclusions to this chapter

Several sets of boundary conditions are considered for numerical fluid models of an argon RF CCP discharge. More precisely, the four sets of boundary conditions have been defined, and two of them are widely used in the gas discharge simulations in the literature. In particular, these are based on the default boundary conditions of the computational packages of the COMSOL's built-in Plasma Module [269] and CFD-ACE+ solver [268]. These four BCs are implemented on a one-dimensional "extended fluid model" to test the validation and verification of the considered boundary conditions.

The analyses are focused on investigating the effects of various ion boundary fluxes. Calculations show that spatial profiles of typical discharge variables obtained from the claimed boundary conditions in this chapter significantly deviate from the results of conventional ones. One necessary consequence of this study is that conventional boundary conditions include such a non-physical switching function in ion boundary flux terms that it has yet to be fully understood in the literature. However, this switching function is removed in our defined ion boundary fluxes.

The "universal" ion boundary flux is defined to determine the contributions of the thermal and drift flux components in the given parameter regime. Similar models are developed by separately considering the "purely thermal" and "purely drift" terms as ion boundary fluxes. It is seen that the "purely drift" model shows very similar results to the "universal" one. Moreover, ion temperature approximations are analyzed to

intend to understand the unusual edge behavior of the "purely thermal" model. Under considered discharge conditions, it is concluded that the effect of Wannier's approximation is negligible. Later, it is revealed that the reason why purely thermal BC does not give compatible results with universal ones near the edges is that the drift velocity dominates thermal velocity attributable to the high electric field in the sheath region.

The critical influence of the secondary electron emission coefficient on the discharge characteristics of the models with several sets of boundary conditions has been revealed.

It is noteworthy that the suggested universal approximation for the ion boundary flux almost gives identical results with the model, including only a drift component as the ion boundary flux.

Compared to particle models (PIC/MCC methods), the claimed boundary conditions lead to more consistent results than BCs, including a non-physical switching factor. These consequences suggest a revision of the boundary conditions in the computational packages.



CHAPTER 6

ANALYSIS OF INPUT PARAMETERS FOR A DIELECTRIC BARRIER DISCHARGE IN CO_2 FLOW AT ATMOSPHERIC PRESSURE

In fluid models of gas discharges, the transport and rate coefficients must be defined as input parameters such that how these coefficients are derived in a self-consistent manner significantly affects modeling results. Since electron dynamics is decisive in gas discharge systems, transport and rate coefficients of electrons are more critical than those of heavy species. In order to achieve maximum consistency between the kinetic and fluid description of electron dynamics, these coefficients for electrons are calculated from collision cross-section data by solving the Boltzmann equation with a two-term approximation. This chapter develops an electron Boltzmann solver by modifying the COMSOL Multiphysics, "The Boltzmann Equation, Two-Term Approximation Interface" [269] and discusses employed physical approximations and numerical techniques. The transport and rate coefficients of electrons from this solver and the results from an online solver (BOLSIG+) are compared. Moreover, spatially one-dimensional coaxial DBD fluid models in CO_2 flow at atmospheric pressure are constructed by inputting derived transport and rate coefficients as lookup tables. The numerical models are based on plasma fluid equations with the "drift-diffusion" theory of gas discharges. This chapter is organized as follows. The statement of the problem is introduced in section 6.1. Section 6.2 describes model definitions of the Boltzmann equation solver, calculations of electron transport and rate coefficients, and comparing these coefficients with those derived from different solvers and approximations commonly used in the literature. The gas discharge model of the CO_2 DBD system, including governing equations, parameter regime, plasma-chemical model, and boundary conditions, are represented in section 6.3. The DBD model's integral characteristics with temporal evolution of discharge variables are investigated in sec-

tion 6.4. Finally, conclusions to the problem are presented in section 6.5.

6.1 The statements of the problem

Fluid equations for gas discharges are formulated by considering the first few moments of the Boltzmann equation. These contain the continuity, the momentum (frequently employed with the "drift-diffusion" approach), and the electron energy equations. These equations define each species' transport and rate coefficients as input data [28, 205, 207]. Therefore, the error in the calculated coefficients can limit the accuracy of the fluid model. Although user-friendly online Boltzmann solvers exist for the coefficients of fluid modeling, there are also different definitions using approximations in the literature to define transport and rate coefficients. Electron coefficients are primarily determined by the characteristics of the EEDF. It defines how electrons acquire energy from the field and the energy gained (or lost) in inelastic collisions with the background gas. This chapter considers Maxwellian, non-Maxwellian EEDFs, and the EEDF derived from the collision cross-section data by solving the developed Boltzmann equation. The calculated data for transport and rate coefficients are organized in tabular form, varying with the mean electron energy. These tabulated values are intended for utilization in CO_2 DBD fluid models.

The fundamental method for generating plasma at low temperatures and atmospheric pressure involves the development of a DBD system [27]. Barrier discharges are used in many industrial applications. These are methane conversion, vapor deposition, pollution control, ozone generation, excitation of CO_2 lasers, surface modification, excimer lamps, and plasma medicine [11, 13, 14, 27]. A distinguishing feature of barrier discharges is their incorporation of a dielectric layer, typically composed of materials such as glass, quartz, and ceramics, that interfaces with the gas discharge between the two electrodes. This configuration fed with an AC power source prevents the discharge system from going through the regime of a sharp increase in current density and gas temperature. The general features and modes of DBD systems are studied in the Refs. [27, 142, 143, 199]. Typical operational conditions for DBD systems contain a gas pressure range extending from several tens of torr to atmospheric pressure, discharge gaps ranging from fractions of millimeters to several millimeters, voltage

amplitudes varying from several hundred to several kilovolts, and field frequencies ranging from tens to several hundreds of kilohertz. Depending on the discharge parameters and gas type, DBD systems at high pressure can show either filamentary or diffuse character [130, 131, 133, 143]. DBD regimes are generally grouped under two headings. The first is the streamer type, which shows itself as a filamentary discharge, and the second is a glow discharge, which also occurs as a homogeneous discharge. The mechanism of secondary electron emission by the dielectric wall leads to an electron avalanche, which maintains the plasma medium in the discharge cell.

CO_2 gas poses a problem for the environment as it plays a vital role in global warming [272]. For this reason, studies on the use and conversion of CO_2 gas have gained significant importance in recent years. Plasma technology is one of the famous gas conversion techniques with great potential. DBD system implementations are commonly used for CO_2 conversion because there is no need for extreme conditions to operate the system such that plasma chemical reactions can occur under normal conditions (such as at room temperature). The literature does not clearly explain the effects of essential discharge parameters, including applied voltage, driving frequency, and reactor configuration on CO_2 DBD systems [167, 194, 197, 204, 273–276]. The main reason for the lack of this issue is that, due to Paschen's law, the discharge gap must be shrunk to a millimeter or even a micrometer size at atmospheric pressure in order to see the discharge ignition and maintenance such that experimental studies on plasma properties are limited. Therefore, numerical studies on modeling CO_2 DBD system are of great importance regarding accuracy, adequacy, and reliability.

In this chapter, an electron Boltzmann solver with a two-term approximation is developed by using COMSOL Multiphysics [269]. The electron and electron energy transport coefficients and some electron-induced reactions' rate coefficients are calculated from this solver. The results are compared with those derived from an online Boltzmann solver (BOLSIG+) [8] with different EEDFs, namely the Maxwellian and Nonmaxwellian EEDFs. In addition, the data calculated from a frequently used approximation between electron transport and electron energy transport coefficients ($\mu_e = 5/3\mu_e$, $D_e = 5/3D_e$) [8] are taken into account in the coefficient comparisons. Then, lookup data tables of electron coefficients versus mean electron energy from each solver are created to be used in the one-dimensional CO_2 DBD fluid models.

These data are separately inputted into the models with the interpolation method and labeled as Model-1 and Model-2 for comparison. The models' integral characteristics are investigated by calculating the RMS values of the discharge current and applied potential and comparing them with the experimental results. The temporal evolution of discharge variables is also compared with experimental data. Similar investigations to test the validity and applicability of the models are conducted in different discharge gaps to understand the effects of electron-induced reactions and electron transport coefficients on the fluid modeling of the CO_2 DBD system. This detailed study aims to conclude how different definitions of electron dynamics affect one-dimensional CO_2 DBD modeling results.

6.2 The Boltzmann equation solver

6.2.1 Model definition

This section defines physical assumptions and numerical techniques in the Boltzmann equation solver developed in this chapter. The Boltzmann equation for the ensemble of electrons in a closed ionized gas system is

$$\frac{\partial f}{\partial t} + v \cdot \nabla f - \frac{e}{m} \mathbf{E} \cdot \nabla_v f = C[f], \quad (6.2.1)$$

where f describes the evolution of the electron energy distribution function in six-dimensional phase space, v is the coordinates of the velocity, e is the electron charge, m is the mass of the electrons, $\mathbf{E}$ denotes the electric field, ∇_v is a gradient operator for particle velocity, and C is the rate of change in f due to collisions.

To solve equation 6.2.1 for electrons and obtain corresponding EEDF, it is necessary to perform drastic simplifications [8]. The first is to expand the distribution function in velocity space in terms of spherical harmonic functions, and six or more terms are needed to obtain a high precision [216]. Due to the significant mass difference between electrons and heavier species, transfer of energy between them are very small. Therefore, electrons scatter randomly due to elastic collisions. Thus, EEDF becomes spherically symmetric (isotropic), and only a two-term expansion of f is sufficient for fluid modeling of the gas discharge system in usual discharge conditions [8]. An-

other simplification is separating the resulting distribution function of the two-term approximation into the isotropic and the anisotropic perturbation terms when inelastic collisions are considered. These terms' temporal and spatial dependence is further simplified by the assumptions described in [8]. The studies [8,277] have studied these simplifications within the two-term approach in detail.

As a result, the stationary two-term Boltzmann equation in energy space [8] is

$$\frac{\partial}{\partial \varepsilon} \left(W f - D \frac{\partial f}{\partial \varepsilon} \right) = S, \quad (6.2.2)$$

where

$$W = -\gamma \varepsilon^2 \sigma_\varepsilon - 3a \left(\frac{n_e}{N} \right) A_1, \quad (6.2.3)$$

$$D = \frac{\gamma}{3} \left(\frac{E}{N} \right)^2 \left(\frac{\varepsilon}{\sigma_m} \right) + \frac{\gamma k_B T_g}{q} \varepsilon^2 \sigma_\varepsilon + 2a \left(\frac{n_e}{N} \right) (A_2 + \varepsilon^{3/2} A_3), \quad (6.2.4)$$

$$a = \frac{q^2 \gamma}{24 \pi \varepsilon_0^2} \ln \Lambda, \quad \Lambda = \frac{12 \pi (2 \varepsilon_0 q \bar{\varepsilon} / 3)^{3/2}}{q^3 n_e^{1/2}}, \quad (6.2.5)$$

$$\gamma = \sqrt{\left(\frac{2q}{m_e} \right)}, \quad \sigma_m = \sum_{k=\text{elastic}} x_k \sigma_k, \quad \sigma_\varepsilon = \sum_{k=\text{elastic}} 2 \frac{m_e}{M} x_k \sigma_k, \quad (6.2.6)$$

$$A_1 = \int_0^\varepsilon u^{1/2} f(u) du, \quad A_2 = \int_0^\varepsilon u^{3/2} f(u) du, \quad A_3 = \int_\varepsilon^\infty f(u) du. \quad (6.2.7)$$

$$S = \sum_{k=\text{inelastic}} = C_k - \gamma \lambda \varepsilon^{1/2} f. \quad (6.2.8)$$

Here, ε is the electron energy, σ_ε is the cross-section of elastic collisions, σ_m is the total collision cross sections, q is the electron charge, m_e is the electron mass, M is the target species' mass, ε_0 is the permittivity of free space, T_g is the background gas temperature, k_B is the Boltzmann constant, n_e is the density of electrons, the background gas density is N , and Λ is the Coulomb logarithm. The source term, S , denotes the energy loss due to inelastic collisions. C_k is the collision term, including different collision processes k , such that it can represent excitation, de-excitation, ionization,

attachment, and electron-electron collisions. The term λ is a renormalization factor such that EEDF is normalized with the following property

$$\int_0^\infty f(\varepsilon) \sqrt{\varepsilon} d\varepsilon = 1. \quad (6.2.9)$$

As a result, since the Boltzmann equation 6.2.1 is challenging to solve, it is reduced to equation 6.2.2 with a two-term approximation. Note that equation 6.2.2 in energy space is very similar to the convection-diffusion continuity equation. This resemblance provides us to numerically solve this equation by taking various grids in the energy domain of the electron. In this way, the EEDF is obtained to calculate electrons' transport and rate coefficients for fluid modeling of discharges.

6.2.2 Fluid model coefficients

In this section, the electron's transport and rate coefficients for standard fluid equations are associated with the EEDF derived from the two-term Boltzmann equation. In fluid equations, transport coefficients (mobility and diffusion) are input parameters in a "drift-diffusion" approximation of electron flux. The well-known equation for electron flux density is

$$\Gamma_e = -\mu_e E n_e - \frac{\partial D_e n_e}{\partial x}, \quad (6.2.10)$$

where the reduced mobility and diffusion coefficients are calculated using the following integrals [3, 8]

$$\mu_e N = -\frac{\gamma}{3} \int_0^\infty \frac{\varepsilon}{\tilde{\sigma}_m} \frac{\partial f}{\partial \varepsilon} d\varepsilon, \quad (6.2.11)$$

$$D_e N = \frac{\gamma}{3} \int_0^\infty \frac{\varepsilon}{\tilde{\sigma}_m} f d\varepsilon, \quad (6.2.12)$$

where $\tilde{\sigma}_m$ is the effective momentum-transfer cross-section, $\varepsilon = \frac{m_e v_e^2}{2e}$ is the electron kinetic energy in eV.

The "drift-diffusion" equation for electron energy flux density is

$$\Gamma_\varepsilon = -\mu_\varepsilon E n_\varepsilon - \frac{\partial D_\varepsilon n_\varepsilon}{\partial x}, \quad (6.2.13)$$

where electron energy mobility and diffusion coefficients are computed

$$\mu_\varepsilon N = -\frac{\gamma}{3\bar{\varepsilon}} \int_0^\infty \frac{\varepsilon^2}{\bar{\sigma}_m} \frac{\partial f}{\partial \varepsilon} d\varepsilon, \quad (6.2.14)$$

$$D_\varepsilon N = \frac{\gamma}{3\bar{\varepsilon}} \int_0^\infty \frac{\varepsilon^2}{\bar{\sigma}_m} f d\varepsilon, \quad (6.2.15)$$

where $\bar{\varepsilon} = n_\varepsilon/n_e$ is the mean electron energy [8].

Rate coefficients are calculated from the EEDF by solving the following integral

$$k_R = \gamma \int_0^\infty \varepsilon \sigma_R f d\varepsilon, \quad (6.2.16)$$

where σ_R corresponds to cross-section of reaction R .

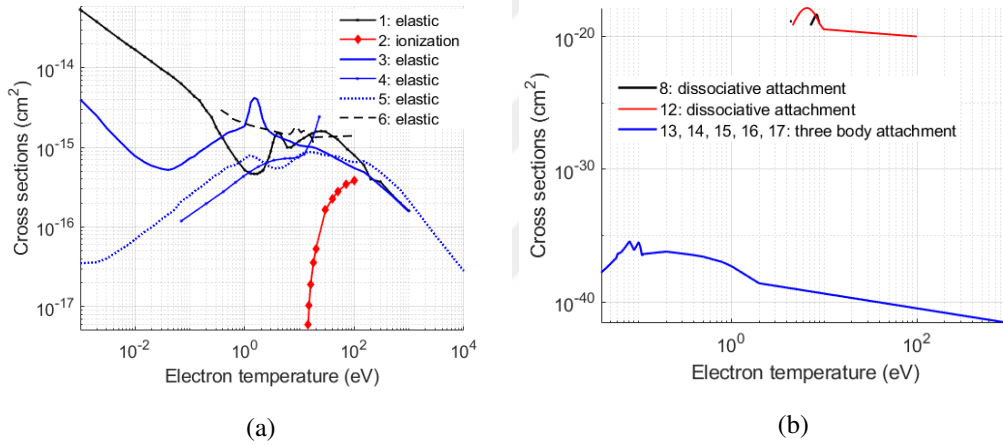


Figure 6.1: Electron cross sections for the reactions in table 6.1: (1) elastic, (2) ionization, (3), (4), (5), and (6) elastic collisions in figure 6.1a; (8), (12) dissociative attachments, (13), (14), (15), (16), and (17) three body attachments in figure 6.1b, used in the hand-made Boltzmann solver model. Numbering corresponds to positions of corresponding processes in table 6.1

The integrals in equations 6.2.11, 6.2.12, 6.2.14, 6.2.15, and 6.2.16 are solved with the EEDF derived from the solution of our hand-made Boltzmann equation solver. The mobility, diffusion and rate coefficients are defined as functions of the mean electron energy in the lookup tables. Moreover, the online Boltzmann solver (BOLSIG+) is used for Maxwellian and Nonmaxwellian EEDFs to calculate these coefficients and

Table 6.1: Set of plasma chemical reactions used in DBD model for CO_2 gas at atmospheric pressure.

Index	Reaction	Rate coefficient	Reaction type
R1	$e + CO_2 \rightarrow e + CO_2$	Cross section	Elastic scattering
R2	$e + CO_2 \rightarrow 2e + CO_2^+$	Cross section	Direct Ionization
R3	$e + CO \rightarrow e + CO$	Cross section	Elastic scattering
R4	$e + O \rightarrow e + O$	Cross section	Elastic scattering
R5	$e + O_2 \rightarrow e + O_2$	Cross section	Elastic scattering
R6	$e + O_3 \rightarrow e + O_3$	Cross section	Elastic scattering
R7	$e + CO_2 \rightarrow e + CO + O$	$5.8 \times 10^{-11} cm^3 s^{-1}$	Impact dissociation
R8	$e + CO_2 \rightarrow CO + O^-$	Cross section	Dissociative attachment
R9	$e + CO_2^+ \rightarrow CO + O$	$6.5 \times 10^{-7} cm^3 s^{-1}$	Recombination
R10	$e + O_3 \rightarrow e + O + O_2$	$2.0 \times 10^{-9} cm^3 s^{-1}$	Impact dissociation
R11	$e + O_2 \rightarrow e + O + O$	$2.0 \times 10^{-9} cm^3 s^{-1}$	Impact dissociation
R12	$e + O_2 \rightarrow O + O^-$	Cross section	Dissociative attachment
R13	$e + O_2 + CO_2 \rightarrow O_2^- + CO_2$	Cross section	Three body attachment
R14	$e + O_2 + CO \rightarrow O_2^- + CO$	Cross section	Three body attachment
R15	$e + O_2 + O_2 \rightarrow O_2^- + O_2$	Cross section	Three body attachment
R16	$e + O_2 + O \rightarrow O_2^- + O$	Cross section	Three body attachment
R17	$e + O_2 + O_3 \rightarrow O_2^- + O_3$	Cross section	Three body attachment
R18	$O^- + CO \rightarrow e + CO_2$	$5.5 \times 10^{-10} cm^3 s^{-1}$	Detachment
R19	$O^- + O_2 \rightarrow e + O_3$	$1.0 \times 10^{-12} cm^3 s^{-1}$	Detachment
R20	$O^- + O_3 \rightarrow e + O_2 + O_2$	$3.0 \times 10^{-10} cm^3 s^{-1}$	Detachment
R21	$O_2^- + CO_2^+ \rightarrow O + O_2 + CO$	$6.0 \times 10^{-7} cm^3 s^{-1}$	Recombination
R22	$O + O + CO_2 \rightarrow O_2 + CO_2$	$5.2 \times 10^{-35} exp(900/T_0) cm^6 s^{-1}$	Neutral reactions
R23	$O + O_2 + CO_2 \rightarrow O_3 + CO_2$	$4.5 \times 10^{-34} (T_0/298)^{-270} cm^6 s^{-1}$	Neutral reactions
R24	$O + O_3 \rightarrow O_2 + O_2$	$8.2 \times 10^{-12} exp(-17.3/T_0) cm^3 s^{-1}$	Neutral reactions
R25	$O + CO + CO_2 \rightarrow CO_2 + CO_2$	$1.7 \times 10^{-33} exp(-1510/T_0) cm^6 s^{-1}$	Neutral reactions
R26	$O_3 + CO_2 \rightarrow O_2 + O + CO_2$	$4.1 \times 10^{-10} exp(-11430/T_0) cm^3 s^{-1}$	Neutral reactions
R27	$O + O + CO \rightarrow O_2 + CO$	$5.2 \times 10^{-35} exp(900/T_0) cm^6 s^{-1}$	Neutral reactions
R28	$O + O + O_2 \rightarrow O_2 + O_2$	$5.2 \times 10^{-35} exp(900/T_0) cm^6 s^{-1}$	Neutral reactions
R29	$O + O + O \rightarrow O_2 + O$	$5.2 \times 10^{-35} exp(900/T_0) cm^6 s^{-1}$	Neutral reactions
R30	$O + O + O_3 \rightarrow O_2 + O_3$	$5.2 \times 10^{-35} exp(900/T_0) cm^6 s^{-1}$	Neutral reactions
R31	$O + O_2 + CO \rightarrow O_3 + CO$	$4.5 \times 10^{-34} (T_0/298)^{-270} cm^6 s^{-1}$	Neutral reactions
R32	$O + O_2 + O_2 \rightarrow O_3 + O_2$	$4.5 \times 10^{-34} (T_0/298)^{-270} cm^6 s^{-1}$	Neutral reactions
R33	$O + O + O_2 \rightarrow O_3 + O$	$4.5 \times 10^{-34} (T_0/298)^{-270} cm^6 s^{-1}$	Neutral reactions
R34	$O + O_2 + O_3 \rightarrow O_3 + O_3$	$4.5 \times 10^{-34} (T_0/298)^{-270} cm^6 s^{-1}$	Neutral reactions
R35	$O + CO + CO \rightarrow CO_2 + CO$	$1.7 \times 10^{-33} exp(-1510/T_0) cm^6 s^{-1}$	Neutral reactions
R36	$O + CO + O_2 \rightarrow CO_2 + O_2$	$1.7 \times 10^{-33} exp(-1510/T_0) cm^6 s^{-1}$	Neutral reactions
R37	$O + CO + O \rightarrow CO_2 + O$	$1.7 \times 10^{-33} exp(-1510/T_0) cm^6 s^{-1}$	Neutral reactions
R38	$O + CO + O_3 \rightarrow CO_2 + O_3$	$1.7 \times 10^{-33} exp(-1510/T_0) cm^6 s^{-1}$	Neutral reactions
R39	$O_3 + CO \rightarrow O_2 + O + CO$	$4.1 \times 10^{-10} exp(-11430/T_0) cm^3 s^{-1}$	Neutral reactions
R40	$O_3 + O_2 \rightarrow O_2 + O + O_2$	$4.1 \times 10^{-10} exp(-11430/T_0) cm^3 s^{-1}$	Neutral reactions
R41	$O_3 + O \rightarrow O + O + O_2$	$4.1 \times 10^{-10} exp(-11430/T_0) cm^3 s^{-1}$	Neutral reactions
R42	$O_3 + O_3 \rightarrow O_3 + O + O_2$	$4.1 \times 10^{-10} exp(-11430/T_0) cm^3 s^{-1}$	Neutral reactions

compare them with those from the hand-made Boltzmann equation solver. It shows how and when these solvers significantly affect fluid model coefficients.

To solve the Boltzmann equation in COMSOL Multiphysics' two-term interface [269], cross-section data must be entered as input with interpolation. Plasma chemical reactions to be used in the CO_2 DBD model are given in table 6.1. Figure 6.1 shows the corresponding cross-section data as a function of mean electron energy, which is implemented in our Boltzmann equation solver. The numbers in the figure correspond to the reaction numbers indicated in table 6.1.

6.2.3 Comparison of the coefficients

This section compares electrons' transport and rate coefficients derived from various Boltzmann solvers while considering the approaches commonly employed in the existing literature. Figure 6.2 compares mobility and diffusion coefficients for electrons and electron energies. Here, the hand-made solver corresponds to the results of the solver developed in this chapter. BOLSIG+ is an online Boltzmann solver, using similar approximations as our solver but assuming different EEDFs.

For electron energy transport coefficients, many fluid models in existing literature use the following relations

$$\mu_\varepsilon = \frac{5}{3}\mu_e, \quad D_\varepsilon = \frac{5}{3}D_e. \quad (6.2.17)$$

It is also included to obtain an idea of the accuracy of the approximations. As noted in the references [8, 9], the relations expressed in equation 6.2.17 are obtained under the assumptions of Maxwellian EEDF, constant momentum-transfer frequency, and constant kinetic pressure. The mobility and diffusion electron energy coefficients from the BOLSIG+ with Maxwellian EEDF are implemented to the above relations by multiplying the electron energy coefficients with 3/5, directly giving electron mobility and diffusion coefficients. The difference between the results of BOLSIG+ with Maxwellian EEDF and those of the defined approximation is that the online solver's kinetic pressure and momentum-transfer frequency are not constant. Indeed, these values vary depending on the electron's energy.

Einstein relation is also widely used to define the transport coefficients of plasma

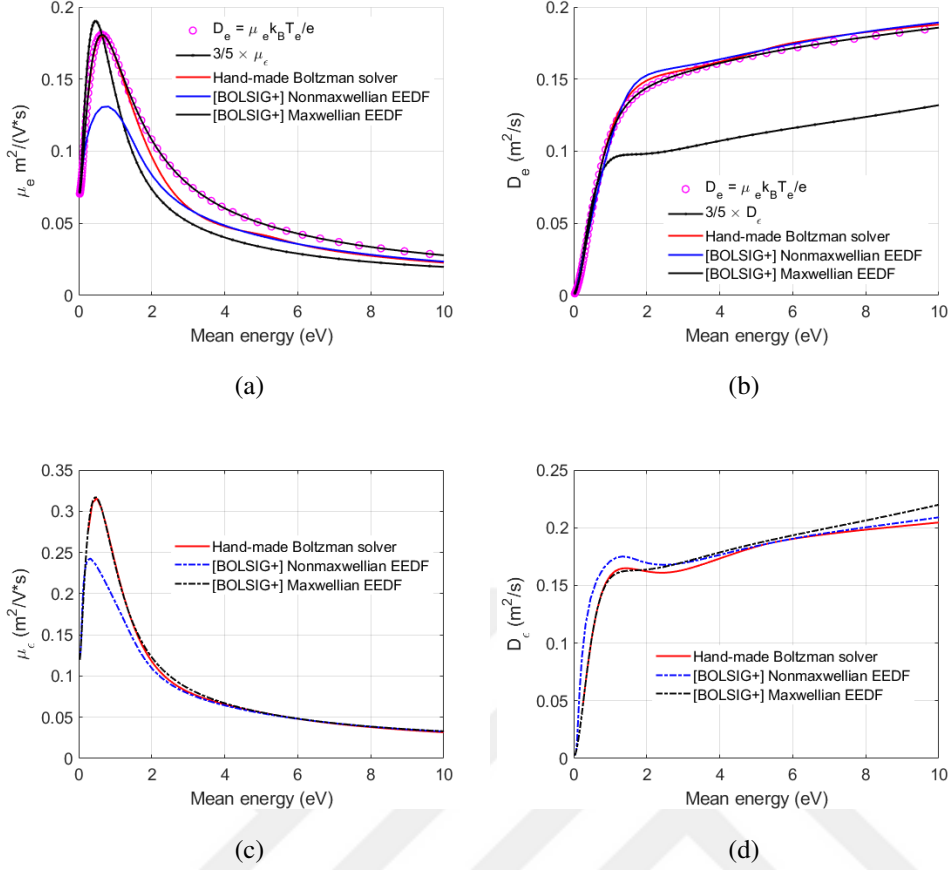


Figure 6.2: Mobility and diffusion coefficients for electron and electron energy transport as functions of mean electron energy. In 6.2a and 6.2b, the Einstein relation and the relation defined in [8,9] are also considered.

species in fluid modeling of gas discharge systems. Since electrons reach very high energies, their transport is generally nonlocal and far from equilibrium with the gas medium. Therefore, the applicability of the Einstein relation's approximation to real discharge situations highly depends on the system's operating conditions. The commonly used Einstein relation for an electron is [9]

$$D_e = \mu_e k_B T_e / e, \quad (6.2.18)$$

where T_e is the electron temperature defined in the Kelvin unit. The transport coefficients from equation 6.2.18 are exact with the results derived from a Maxwellian EEDF [8], confirmed in figure 6.2. Unlike electron and electron energy diffusivities, the mobilities decrease when the mean electron energy increases. Note that, at small

electron mean energy values, the results of BOLSIG+ with Maxwellian EEDF are more in agreement with the results of the hand-made solver. However, as the mean electron energy increases, BOLSIG+ with Nonmaxwellian EEDF has more consistency with the handmade (realistic) solver.

Figure 6.3 compares the rate coefficients derived from different solvers. The number indices of the rate coefficients correspond to the reaction numbers in table 6.1. As seen more clearly in k_2 and k_8 , the rate coefficients of BOLSIG+ with Nonmaxwellian EEDF give more consistent behavior with the results of the handmade solver when compared to BOLSIG+ with Maxwellian. The calculations show that rate features depend more on the EEDF than transport coefficients.

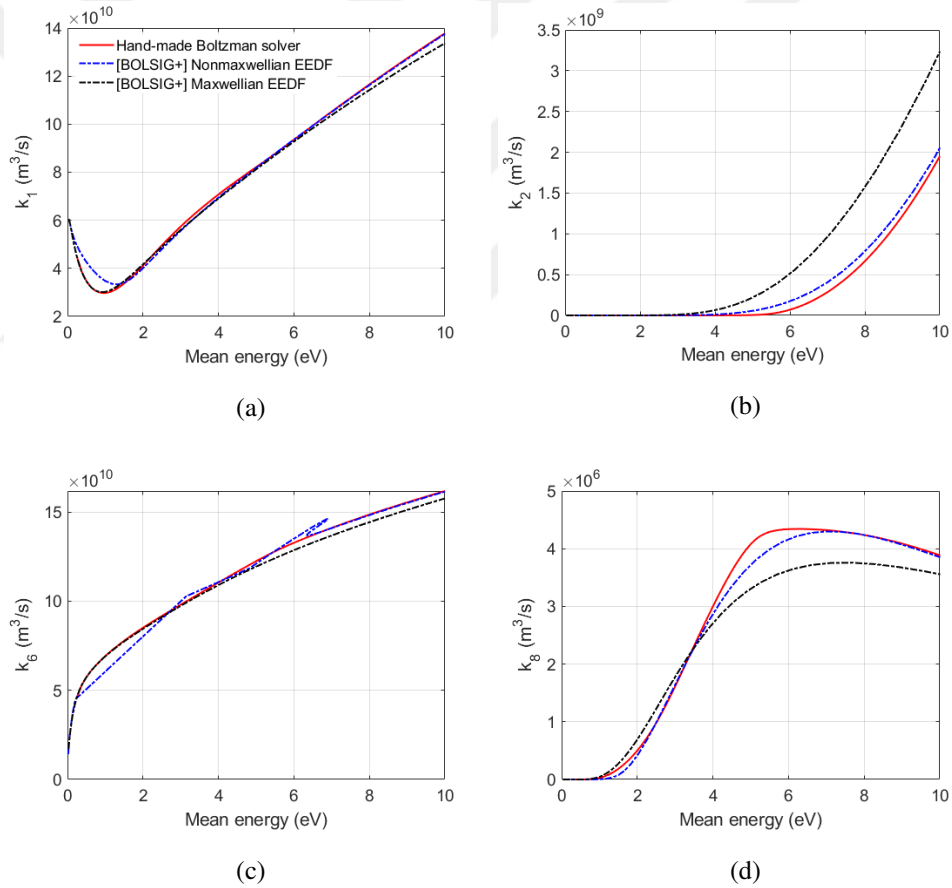


Figure 6.3: The rate coefficients vs. mean electron energy for different EEDFs derived from different Boltzmann solvers. The number indices of the rate coefficients correspond to the reaction numbers in table 6.1.

Although there is a slight difference among the coefficients derived from different solvers in figures 6.2 and 6.3, this may show a significant difference in the results of fluid modeling. For this reason, the effects of input parameters (transport and rate coefficients) on fluid modeling of DBD are investigated in the following sections.

6.3 Gas discharge model

6.3.1 Governing equations

The numerical DBD fluid model includes the particle balance equations for each plasma species,

$$\frac{\partial n_k}{\partial t} + \nabla_r \cdot \Gamma_k = S_k, \quad (6.3.1)$$

where the "drift-diffusion" approximation for the particle flux densities Γ_k ,

$$\Gamma_k = \text{sgn}(q_k) \mu_k n_k \mathbf{E} - D_k \nabla_r n_k, \quad (6.3.2)$$

the Poisson equation for the electrostatic field

$$-\epsilon_0 \nabla^2 \varphi = \sum_k q_k n_k, \quad (6.3.3)$$

and the electron energy equation is also included

$$\begin{aligned} \frac{\partial n_e}{\partial t} + \nabla_r \cdot \Gamma_e = & - e \Gamma_e \cdot E - \frac{3}{2} \frac{m_e}{m_g} \nu_{ea} n_e k_B (T_e - T_g) \\ & - \sum_j \Delta E_j R_j. \end{aligned} \quad (6.3.4)$$

In these equations, ∇_r denotes the divergence operator in a radial direction of cylindrical coordinates, n is particle number density, φ and $\mathbf{E} = -\nabla \varphi$ stand for the electric potential and field. μ and D are the particle mobility and diffusion coefficients. The subscript k indicates the sort of species (such as electrons, ions, metastable molecules, etc.). In the electron energy equation, ν_{ea} is the electron-atomic elastic collision frequency, R_j describes the reaction rate of the j th reaction, ΔE_j is the corresponding energy loss (or gain) due to inelastic collision, m is the particle mass, and T is the kinetic temperature. $n_e = n_e \bar{\epsilon}$ is the electron energy density, where $\bar{\epsilon} = (3/2)k_B T_e$ is the mean electron energy, and Γ_e denotes the electron energy flux density,

$$\Gamma_e = -D_e \nabla n_e - \mu_e E n_e, \quad (6.3.5)$$

where D_e is the electron energy diffusion coefficient, and μ_e is the electron energy mobility coefficient.

6.3.2 Parameter regime

The input parameters of CO_2 DBD system are defined as follows. The pressure of the CO_2 gas is specified as atmospheric, $p = 760$ torr. The gas temperature is $T_g = 300$ K (room temperature). The schematic of the coaxial DBD tube is displayed in figure 6.4. Since the three different configurations are considered, the geometrical features of the coaxial discharge cell configurations are given in table 6.2.

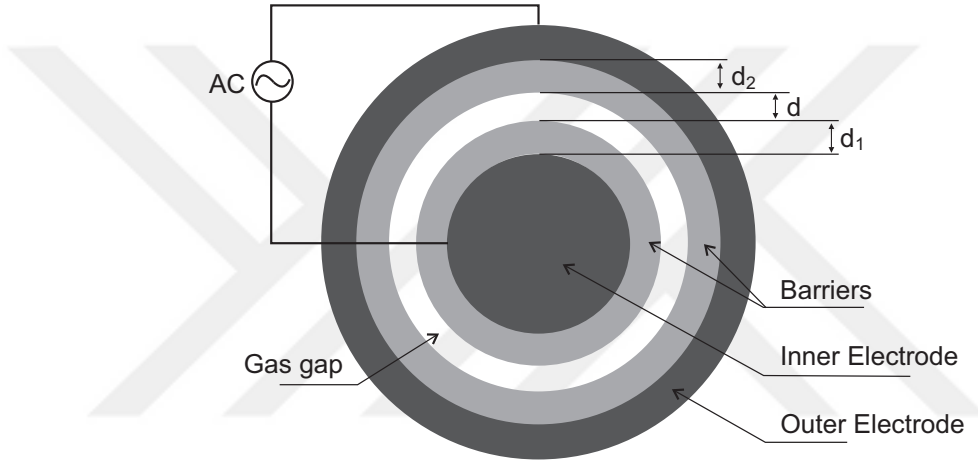


Figure 6.4: The schematic illustration of a coaxial DBD tube. Geometrical features for three different coaxial discharge tube configurations are given in table 6.2.

The results are derived by three different discharge gaps: $d = 1.9$ mm, $d = 3.5$ mm, and $d = 5.5$ mm. The external electrode length is 3.7 cm, and the inner electrode length is 30 mm for all three discharge configurations. Both outer and inner dielectric widths are $d_1 = d_2 = 1$ mm for $d = 1.9$ mm and $d = 3.5$ mm gaps, and $d_1 = d_2 = 2$ mm for $d = 5.5$ mm gap. The radius of inner electrodes are $r_0 = 4$ mm for $d = 1.9$ mm and $d = 3.5$ mm gaps, and $r_0 = 15.25$ mm for $d = 5.5$ mm. The voltage amplitudes and field frequencies of the applied voltage are $f = 106$ kHz with $V_0 = 7$ kHz, $f = 111$ kHz with $V_0 = 8.5$ kHz, and $f = 104$ kHz with $V_0 = 9$ kHz, respectively. The dielectric constant of two quartz barriers is set as $\epsilon_s = 4.05$.

In figure 6.4, the position of electrodes is defined at the boundaries $r = r_0$ and $r =$

Table 6.2: The geometric properties of different coaxial DBD cells. The inner electrode diameter (I.E.D.), inner dielectric diameter (I.D.D.), outer dielectric diameter (O.D.D.), and outer electrode diameter (O.E.D.) values are defined for three different configurations.

d (mm)	I.E.D (mm)	I.D.D (mm)	O.D.D (mm)	O.E.D (mm)
1.9	8	8-10	13.80-15.80	15.80
3.5	8	8-10	17-19	19
5.5	30.5	30.5-34.5	44.5-48.5	48.5

r_1 , and the discharge gap is defined between positions $r = r_0 + d_1$ and $r = r_1 - d_2$. Since the numerical fluid models are developed as spatially one-dimensional, the calculations are only carried out in the radial direction of the discharge gap.

6.3.3 Plasma-chemical model

Calculations are carried out for the plasma-chemical model of a DBD in CO_2 gas. This model follows a reduced set of plasma chemical reactions from Ref. [194, 278], summarized in table 6.1. Nine species, namely, electrons, the positive ions CO_2^+ , the negative ions O^- and O_2^- , the neutral atoms and molecules CO , O , O_3 , O_2 and CO_2 are taken into account. Vibrationally and electronically excited species are neglected within this model because their importance have a minor importance in DBD plasmas [183, 195, 198, 278].

As seen in table 6.1, the reaction R_2 represents direct ionization such that CO_2^+ ions are produced from the CO_2 molecule by electron-impact ionization. It is the only considered electron-impact ionization reaction, and this process is more significant than the dissociative ionization of CO_2 [194, 195]. Recombination reactions of CO_2^+ and O_2^- ions are indicated by R_9 and R_{21} . Indeed, R_{21} is the neutralization of the O_2^- ions with CO_2^+ . The electron impact dissociation of CO_2 , O_3 , and O_2 molecules corresponds to the reaction R_7 , R_{10} , and R_{11} , respectively. Reactions R_8 , $R_{12}-R_{17}$ are account for the process of electron attachment. The detachment process producing

electrons is described by $R_{18}-R_{20}$. Reactions from $R_{22}-R_{42}$ are responsible for the processes between the neutral species.

The source terms in the continuity equation 6.3.1 are determined by the reactions occurring in the bulk of the discharge,

$$S_k = \sum_i R_i - \sum_j R'_j, \quad (6.3.6)$$

where R_i and R'_j are the creation and destruction rates in i th and j th reactions. These rates are proportional to the number densities of species involved in these reactions and the rate coefficients of the corresponding reactions. The model defines electron-induced reactions' rate coefficients and the mobility and diffusion coefficients of electrons as look-up tables. The electron coefficient versus mean electron energy data in the look-up table is derived by the Boltzmann solvers, described in section 6.2.

The transport coefficients of heavy species are approximated by constants. The mobility coefficients of the ions CO_2^+ , O^- and O_2^- are 5.7×10^{-4} , 1.0×10^{-3} and $6.7 \times 10^{-4} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively. The diffusion coefficients of heavy species are defined according to the "mixture-averaged" diffusion model [269].

6.3.4 Boundary conditions

The boundary conditions are displayed in table 6.3 [101,269,278]. In these equations, $v_k = \sqrt{\frac{8k_B T_k}{\pi m_k}}$ is thermal velocity of k^{th} species. Γ is the flux density, defined by equations 6.3.2 and 6.3.5. a is a switching factor: it is set equal to 1 if the direction of the electric field is toward the boundary; otherwise, it is equal to zero. γ_i is the secondary electron emission coefficient, set to 0.1 for both dielectric surfaces. In the electron energy boundary flux, ϵ_i defines the mean energy of the secondary electrons.

In the ion boundary flux, $\zeta_i = \frac{\gamma_f}{1-0.5\gamma_f}$, where γ_f is the forward sticking coefficient, set to 0.1 for all ionic components. In the boundary conditions for the surface charge σ , the relation $\mathbf{J}_g = \sum_k q_k \Gamma_k$ determines the current density in the discharge gap.

Electric potential is continuous across the interfaces between the dielectric barriers

Table 6.3: Boundary conditions at the walls of the discharge gap.

Electron flux	$\hat{\mathbf{n}} \cdot \mathbf{\Gamma}_e = \frac{1}{2}v_e n_e - \sum_i \gamma_i (\hat{\mathbf{n}} \cdot \mathbf{\Gamma}_i)$
Ion flux for CO_2^+ , O^- , O_2^-	$\hat{\mathbf{n}} \cdot \mathbf{\Gamma}_i = \frac{1}{4}\zeta_i v_i n_i + a_i n_i \mu_i (\hat{\mathbf{n}} \cdot \mathbf{E})$
Neutral species flux for CO , O , O_3 , O_2	$\hat{\mathbf{n}} \cdot \mathbf{\Gamma}_m = \frac{1}{2}v_m n_m$
Electron energy flux	$\hat{\mathbf{n}} \cdot \mathbf{\Gamma}_\epsilon = \frac{5}{6}v_e n_e - \sum_i \gamma_i \epsilon_i (\hat{\mathbf{n}} \cdot \mathbf{\Gamma}_i)$
Surface charge density	$\sigma = (\epsilon_r \epsilon_0 \mathbf{E}_d - \epsilon_0 \mathbf{E}_g) \cdot \hat{\mathbf{n}}$ and $\frac{\partial \sigma}{\partial t} = \mathbf{J}_g \cdot \hat{\mathbf{n}}$

and gas discharge gap; on the electrode surfaces, it is defined as

$$\varphi = 0 \quad (6.3.7)$$

on the ground electrode and

$$\varphi = V_0 \cos(2\pi f t) \quad (6.3.8)$$

on the powered electrode.

6.4 Validation and verification of the models

6.4.1 Integral characteristics of CO_2 DBD

The integral characteristics of DBD show whether the developed numerical models are realistic enough to represent the experimental discharge system. Since the events of the barrier discharges include many random breakdowns, it is more convenient comparing the computed and experimental root mean squares (RMS) values of the discharge current and applied voltage instead of amplitudes of them [14]. The RMS current and voltage are defined as

$$I_{rms} = \left[\frac{1}{T} \int_0^T I^2(t) dt \right]^{0.5}, \quad U_{rms} = \left[\frac{1}{T} \int_0^T U^2(t) dt \right]^{0.5}, \quad (6.4.1)$$

where $I(t)$ and $U(t)$ indicate the instantaneous values of the discharge current and total applied potential.

Figure 6.5 demonstrates the RMS values of discharge current plotted against applied potential, comparing experimental data and numerical model predictions. In addition

to Model-2, based on Maxwellian EEDF (BOLSIG+), Model-1 also represents a one-dimensional CO_2 DBD fluid model on the basis of a more realistic EEDF calculated from our Boltzmann solver, defined in section 6.2. Other approaches given in section 6.2 are not displayed because the results of the two models almost overlap in three different configurations. Therefore, the results of other approaches are not presented in this section because they do not reveal anything new. In other words, their results are also similar to those of Model-1 and Model-2.

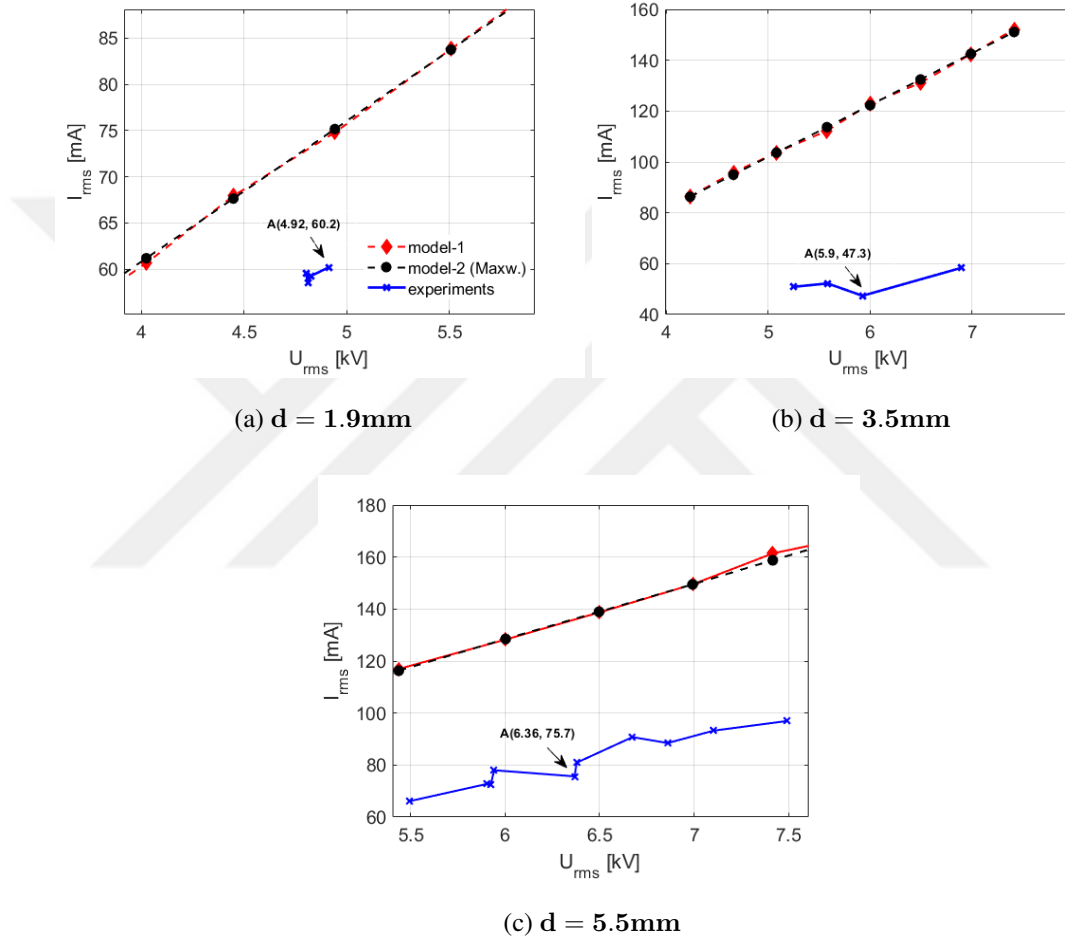


Figure 6.5: Computed and measured RMS of the discharge current vs. RMS of the applied potential. The numerical and experimental results of three different discharge gaps, namely figure 6.5a, 6.5b, and 6.5c are 1.9 mm, 3.5 mm, and 5.5 mm, respectively. The configuration parameters are based on table 6.2.

The numerical and experimental results of three configurations based on the param-

eters given in table 6.2 are compared. As can be observed, both of Model-1 and Model-2 in figures 6.5b, 6.5c stay apart from the profile derived from the experiment more than those in figure 6.5a. Indeed, the results of the models become more consistent with the experimental data as the discharge gap decreases. Possible reasons for the inconsistencies in figure 6.5 are explained in the following subsection.

6.4.2 Temporal evolution of discharge variables

In Figure 6.6, the calculations are carried out in the regimes indicated by point *A* in each of the figure 6.5. The time progression of discharge current and applied potential derived from the models is compared with corresponding experimental data. The current data from the models in $d = 1.9$ mm gap show more agreement with the experimental data than the other discharge gaps $d = 3.5$ mm and $d = 5.5$ mm, which supports the results in figure 6.5. Indeed, the larger the discharge gap is, the greater discrepancy with the experimental results. In addition, the models' sinusoidal voltage is applied according to the voltage of the corresponding experiments, shown in all three figures in figure 6.6.

Possible reasons for the inconsistencies between the models and corresponding experiments can be given as follows: The test calculations indicate that the modeling results could fit into the experimental data by adjusting the secondary electron emission and reflection coefficients in defining the boundary conditions. The fact that only the discharge gap is 1.9 mm gives a more consistent result with the experiment is related to the taken value of these coefficients. It is not a correct inference to say that the one-dimensional fluid model gives correct results for the small gaps because the consistency in small gaps is related to the adjustments of fitting parameters. Gas heating is ignored in the developed models. The reference [278] considers gas heating for a similar model to ours and investigates its effect on discharge properties in detail. As such, not including the gas heating equation in the modeling equations can be seen as one of the reasons for the discrepancy with experimental results. It has been seen in the experiments that after the ignition of the DBD system, the discharge mode is strongly filamentary. Indeed, many filaments stochastically occupy a particular part of the discharge cell instead of the entire cell. Therefore, a three-dimensional and

sophisticated numerical treatment is necessary instead of a spatially one-dimensional model that only considers the radial direction changes and limits the model's computation accuracy. Moreover, the slight deviations from the sinusoidal waveform of the experimental supply voltage, seen in figure 6.6, lead to the discrepancy between observed and calculated discharge parameters.

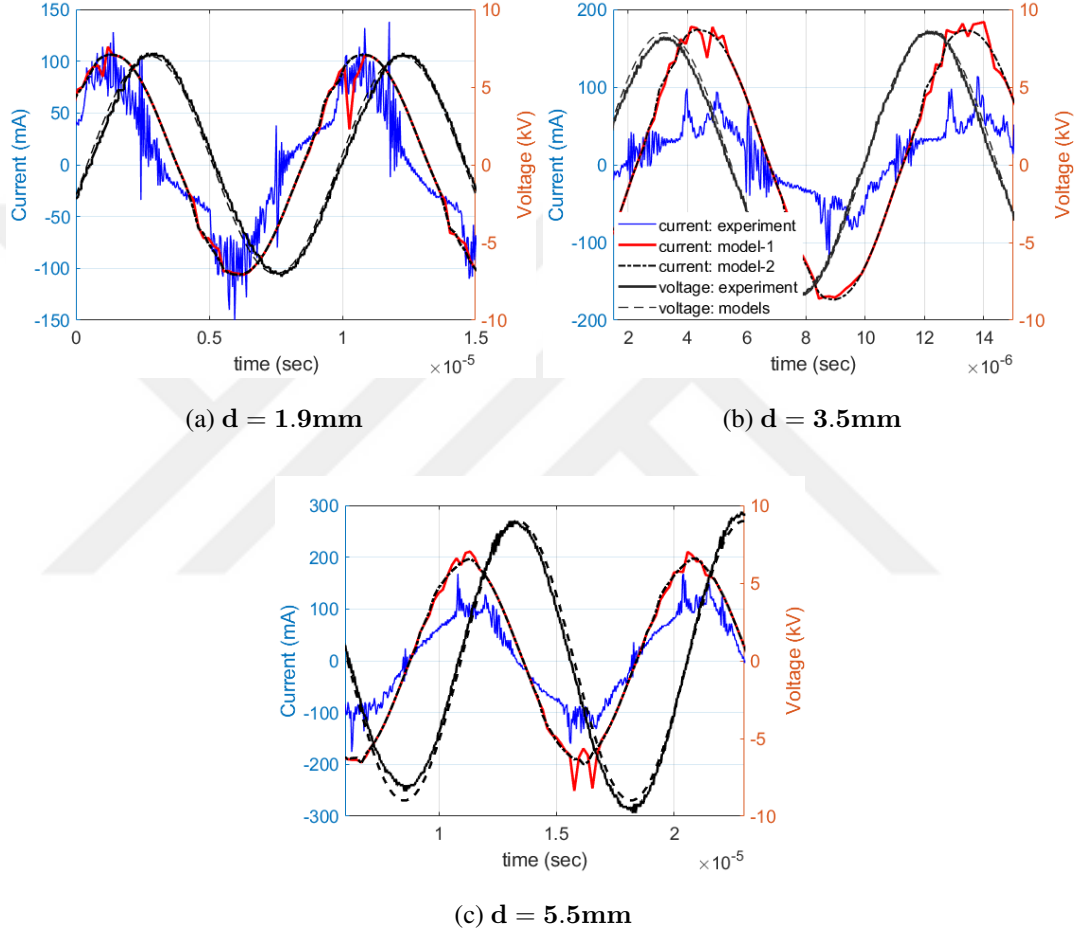


Figure 6.6: Temporal evolution of discharge current and applied voltage in the regimes indicated by point *A* in the figures 6.5a, 6.5b, and 6.5c. The excitation frequency and applied voltage amplitudes are (a) $f = 106$ kHz and $V_0 = 7$ kV, (b) $f = 111$ kHz and $V_0 = 8.5$ kV, (c) $f = 104$ kHz and $V_0 = 9$ kV.

6.5 Conclusions to this chapter

This chapter develops an electron Boltzmann solver to compute the electron transport and rate coefficients utilized as input data in the fluid models. The COMSOL Multiphysics, "Boltzmann Equation, Two-term Approximation Interface" [269] module is modified to construct this solver. The solver's physical approaches and numerical techniques are founded on the classical two-term expansion, as outlined in reference [8]. To calculate the transport (including mobility and diffusion) and rate coefficients of electrons, the essential integrals involving the EEDF derived from the Boltzmann solver are defined. The electron coefficients obtained from this realistic solver are compared with those from an online solver (BOLSIG+) [8]. Indeed, BOLSIG+ considers similar approximations with our solver but with different EEDFs: Maxwellian and Non-Maxwellian. The validness and exactness of the commonly used relation defined in [8, 9] and the Einstein relation are also tested. The results obtained from the Einstein relation overlap precisely with the BOLSIG+ with Maxwellian EEDF, as expected. Although the difference between the electron coefficients obtained from the realistic solver and those from other solvers and approximations seems very small, these minor differences may lead to significant differences in the results of gas discharge fluid models.

Due to limited experimental studies in CO_2 DBD (Dielectric Barrier Discharge) systems, the effects of electron coefficients on numerical modeling of CO_2 DBD are investigated because the error in the calculated coefficients can limit the accuracy, adequacy, and reliability of the fluid model. Spatially one-dimensional coaxial DBD fluid models in CO_2 flow at atmospheric pressure are developed by inputting derived transport and rate coefficients from different solvers as lookup tables. The models are based on plasma fluid equations with the "drift-diffusion" theory of gas discharges. To test these models' validity and applicability, the DBD model's integral characteristics – the calculation of Root Mean Square (RMS) values for discharge current and applied voltage – are compared with experimental data for three different discharge configurations. Initially, when the discharge gap diminishes, the developed models will probably yield more consistent outcomes with experimental results. However, it is understood that this situation relates to the specific choices made for the constants,

such as secondary electron emission and reflection coefficients, used when defining the boundary conditions. Possible reasons for the discrepancies between the models and experiments are mainly based on omitting gas heating equations in the governing equations of the developed models and that the discharge mode is stochastically filamentary; thus, a more advanced three-dimensional treatment is needed to model the system.





CHAPTER 7

CONCLUSION

This thesis begins with analyzing fluid modeling techniques applied to glow discharge phenomena. The foundation of this investigation lies in deriving fluid equations from the moments of the kinetic Boltzmann equation. A comprehensive understanding of plasma behavior through a "two-fluid" modeling approach is achieved by carefully considering the continuity, momentum, and energy equations. A significant contribution of this work is developing a self-consistent model for the electric field, which is constructed by integrating the Poisson equation with "two-fluid" equations. Considering the "drift-diffusion" approximation to characterize particle flux densities, a key outcome emerges from combining the momentum and continuity equations, which yields a particular equation that serves as the basis for defining both the "simple" and "extended" fluid models. The comparative examination is then conducted to explain the strengths and limitations inherent in these two distinct fluid modeling approaches. This critical evaluation revolves around their respective abilities to address the fundamental requirements of glow discharge phenomena. Through this comprehensive investigation, the thesis sets the stage for a deeper understanding of complex plasma behavior and its modeling in the context of glow discharge.

Chapter 3 presents a detailed exploration of the transition from periodic to chaotic oscillations in a laterally extended DC-supplied gas discharge–semiconductor system (GDSS) operating in a nitrogen environment. The study focuses on understanding oscillations within the subnormal glow regime, a significant phase bridging the Townsend and normal glow discharge regimes. Numerical models based on plasma fluid equations, utilizing the "drift-diffusion" approximation of particle fluxes, are developed to delve into this phenomenon. Two distinct modeling approaches are em-

ployed: the "simple fluid model" (LFA) and the more sophisticated "extended fluid model" (LMEA). These models provide a qualitative understanding of the system's dynamic behavior during the evolution from regular to chaotic oscillations. Essential outcomes include the creation of a Lorenz map and a bifurcation diagram, which vividly illustrate the system's journey towards a fully chaotic state through the occurrence of period-doubling bifurcations. The interplay between the total applied voltage and semiconductor resistance emerges as crucial control parameters influencing this transition. These findings significantly contribute to solving the complex dynamics underlying the GDSS system's evolution into chaotic oscillations. By employing diverse modeling techniques and adjusting control parameters, this comprehensive analysis clarifies the intricate mechanisms driving this complex behavior, paving the way for future investigations in GDSSs and nonlinear dynamics.

Chapter 4 investigates "unpredictability" and "Delta Synchronization of Chaos" (DSC) techniques. Centered around a GDSS and utilizing a one-dimensional "extended fluid model", the calculations from Chapter 3 are transformed into dimensionless quantities using predefined scale laws. This chapter focuses on the GDSS model's chaotic regime, investigating the "unpredictability" algorithm's role in validating chaotic solutions. Moreover, the DSC algorithm proved synchronization within the coupled system (drive and response) by identifying common time values with similar behavior. A comparison with a previous study utilizing a simpler one-dimensional fluid model highlights the increased chaos and synchronicity demonstrated by the detailed fluid model. These insights provide a basis for future research, particularly in complex systems characterized by chaotic behavior, offering valuable perspectives on unpredictability and synchronization phenomena.

Chapter 5 comprehensively focuses on various boundary conditions applied to numerical fluid models of an argon RF CCP discharge. The investigation has four sets of boundary conditions, with a particular focus on two commonly utilized conditions in commercial computational packages. The analysis uses a one-dimensional "extended fluid model" for validation, investigating the influence of ion boundary fluxes on the discharge's behavior. The findings emphasize substantial differences in the spatial profiles of discharge variables between the proposed and conventional boundary conditions. Notably, the elimination of a non-physical switching function in ion boundary

flux terms reveals crucial insights. Introducing a "universal" ion boundary flux sheds light on the interplay between thermal and drift flux components. The "purely drift" model closely mirrors the "universal" model, underlining their functional equivalence. This chapter emphasizes the importance of reevaluating boundary conditions within computational packages, enhancing the accuracy and insight of plasma simulations. It contributes to advancing suitable boundary conditions for numerical fluid models, with implications for plasma physics and computational simulations, recommending further research for refined conditions and heightened reliability.

Chapter 6 presents the detailed development of an electron Boltzmann solver, a cornerstone for calculating electron transport and rate coefficients required for fluid models. The solver is constructed by modifying the "Boltzmann Equation, Two-term Approximation Interface" module in COMSOL Multiphysics, combining physical principles with advanced numerical techniques. Calculating electron transport and rate coefficients involves complicated integrals incorporating the EEDF derived from the Boltzmann solver. A comparison with an online solver, BOLSIG+, verifies the accuracy of electron coefficients. At the same time, validation analyses test the precision of the approximations commonly used in the literature, such as the Einstein relation. Although the calculations show minor differences among the results derived from the solvers and approximations, they seriously impact the outcomes of gas-discharge fluid models. Fluid models for one-dimensional coaxial DBD in CO_2 , utilizing input parameters obtained from the developed solvers, are created and verified through comparison with experimental data. The study concludes with the need for the necessity of advanced three-dimensional modeling to capture the stochastic nature of the discharge model and the difficulties of accurate numerical modeling of CO_2 DBD systems. Furthermore, it is essential to consider the gas heating equations within the governing equations of the developed models. This chapter provides a basis for future investigations and refining our understanding of complex plasma phenomena.

This thesis offers a comprehensive and detailed investigation into various aspects of gas discharges, including chaotic oscillations, chaos synchronization, boundary conditions, electron transport, and interactions within diverse gas discharge systems. Through rigorous investigations, this work provides valuable insights into these systems' complex behaviors and dynamic characteristics. As a result, this thesis con-

tributes significantly to our understanding of gas discharge phenomena and their complex nature.



REFERENCES

- [1] V. Kolobov and R. Arslanbekov, “Deterministic boltzmann solver for electron kinetics in plasma reactors for microelectronics applications,” *Microelectronic engineering*, vol. 69, no. 2-4, pp. 606–615, 2003.
- [2] F. P. Universität Münster, “Institute of applied physics.” <https://www.uni-muenster.de/Physik.AP/Purwins/DC/index-en.html>.
- [3] I. Rafatov and C. Yesil, “Transition from homogeneous stationary to oscillating state in planar gas discharge–semiconductor system in nitrogen: Effect of fluid modelling approach,” *Physics of Plasmas*, vol. 25, no. 8, p. 082107, 2018.
- [4] M. Akhmet, K. Başkan, and C. Yeşil, “Delta synchronization of poincaré chaos in gas discharge-semiconductor systems,” *Chaos: An Interdisciplinary Journal of Nonlinear Science*, vol. 32, no. 8, p. 083137, 2022.
- [5] Y.-X. Liu, Q.-Z. Zhang, K. Zhao, Y.-R. Zhang, F. Gao, Y.-H. Song, and Y.-N. Wang, “Fundamental study towards a better understanding of low pressure radio-frequency plasmas for industrial applications,” *Chinese Physics B*, vol. 31, no. 8, p. 085202, 2022.
- [6] R. Brandenburg, “Dielectric barrier discharges: progress on plasma sources and on the understanding of regimes and single filaments,” *Plasma Sources Science and Technology*, vol. 26, no. 5, p. 053001, 2017.
- [7] A. Kudryavtsev and L. Tsendin, “Townsend discharge instability on the right-hand branch of the paschen curve,” *Technical Physics Letters*, vol. 28, no. 12, pp. 1036–1039, 2002.
- [8] G. Hagelaar and L. C. Pitchford, “Solving the boltzmann equation to obtain electron transport coefficients and rate coefficients for fluid models,” *Plasma Sources Science and Technology*, vol. 14, no. 4, p. 722, 2005.

- [9] J. A. Bittencourt, *Fundamentals of plasma physics*. Springer Science & Business Media, 2004.
- [10] D. C. Schram, “Is plasma unique? the presence of electrons and the importance of charge,” *Plasma Sources Science and Technology*, vol. 18, no. 1, p. 014003, 2008.
- [11] A. A. Fridman and G. G. Friedman, *Plasma medicine*. John Wiley & Sons Chichester, UK:, 2013.
- [12] U. Kogelschatz, “Dielectric-barrier discharges: their history, discharge physics, and industrial applications,” *Plasma chemistry and plasma processing*, vol. 23, no. 1, pp. 1–46, 2003.
- [13] G. Park, S. Park, M. Choi, I. Koo, J. Byun, J. Hong, J. Sim, G. Collins, and J. Lee, “Atmospheric-pressure plasma sources for biomedical applications,” *Plasma Sources Science and Technology*, vol. 21, no. 4, p. 043001, 2012.
- [14] D. Li, V. Rohani, F. Fabry, A. P. Ramaswamy, M. Sennour, and L. Fulcheri, “Direct conversion of CO_2 and CH_4 into liquid chemicals by plasma-catalysis,” *Applied Catalysis B: Environmental*, vol. 261, p. 118228, 2020.
- [15] A. Von Keudell and V. Schulz-Von Der Gathen, “Foundations of low-temperature plasma physics—an introduction,” *Plasma Sources Science and Technology*, vol. 26, no. 11, p. 113001, 2017.
- [16] U. S. Inan and M. Gołkowski, *Principles of plasma physics for engineers and scientists*. Cambridge University Press, 2010.
- [17] G. Schmidt, *Physics of high temperature plasmas*. Elsevier, 2012.
- [18] M. Laroussi, “Low temperature plasma-based sterilization: overview and state-of-the-art,” *Plasma processes and polymers*, vol. 2, no. 5, pp. 391–400, 2005.
- [19] G. S. Oehrlein and S. Hamaguchi, “Foundations of low-temperature plasma enhanced materials synthesis and etching,” *Plasma Sources Science and Technology*, vol. 27, no. 2, p. 023001, 2018.

- [20] M. G. Kong, G. Kroesen, G. Morfill, T. Nosenko, T. Shimizu, J. Van Dijk, and J. Zimmermann, "Plasma medicine: an introductory review," *new Journal of Physics*, vol. 11, no. 11, p. 115012, 2009.
- [21] X. Rao, S. Hammack, T. Lee, C. Carter, and I. B. Matveev, "Combustion dynamics of plasma-enhanced premixed and nonpremixed flames," *IEEE Transactions on Plasma Science*, vol. 38, no. 12, pp. 3265–3271, 2010.
- [22] V. I. Pârvulescu, M. Magureanu, and P. Lukes, *Plasma chemistry and catalysis in gases and liquids*. John Wiley & Sons, 2012.
- [23] H. Conrads and M. Schmidt, "Plasma generation and plasma sources," *Plasma Sources Science and Technology*, vol. 9, no. 4, p. 441, 2000.
- [24] Y. P. Raizer, M. N. Shneider, and N. A. Yatsenko, *Radio-frequency capacitive discharges*. CRC press, 2017.
- [25] I. Rafatov and A. Kudryavtsev, *Introduction to Simulation Methods for Gas Discharge Plasmas: Accuracy, Reliability and Limitations*. IOP Publishing, 2020.
- [26] V. A. Rozhansky and L. D. Tsendin, *Transport phenomena in partially ionized plasma*. CRC Press, 2001.
- [27] Y. P. Raizer and J. E. Allen, *Gas discharge physics*, vol. 2. Springer Berlin, 1997.
- [28] J.-P. Boeuf, "Numerical model of rf glow discharges," *Physical review A*, vol. 36, no. 6, p. 2782, 1987.
- [29] J. Boeuf and L. Pitchford, "Two-dimensional model of a capacitively coupled rf discharge and comparisons with experiments in the gaseous electronics conference reference reactor," *Physical Review E*, vol. 51, no. 2, p. 1376, 1995.
- [30] A. Derzsi, P. Hartmann, I. Korolov, J. Karacsony, G. Bánó, and Z. Donkó, "On the accuracy and limitations of fluid models of the cathode region of dc glow discharges," *Journal of Physics D: Applied Physics*, vol. 42, no. 22, p. 225204, 2009.

- [31] R. R. Arslanbekov and V. I. Kolobov, “Two-dimensional simulations of the transition from townsend to glow discharge and subnormal oscillations,” *Journal of Physics D: Applied Physics*, vol. 36, no. 23, p. 2986, 2003.
- [32] G. Grubert, M. Becker, and D. Loffhagen, “Why the local-mean-energy approximation should be used in hydrodynamic plasma descriptions instead of the local-field approximation,” *Physical Review E*, vol. 80, no. 3, p. 036405, 2009.
- [33] Y. Sakiyama, D. Graves, and E. Stoffels, “Influence of electrical properties of treated surface on rf-excited plasma needle at atmospheric pressure,” *Journal of Physics D: Applied Physics*, vol. 41, no. 9, p. 095204, 2008.
- [34] E. Bogdanov, S. Adams, V. Demidov, A. Kudryavtsev, and J. Williamson, “Influence of the transverse dimension on the structure and properties of dc glow discharges,” *Physics of Plasmas*, vol. 17, no. 10, 2010.
- [35] E. Bogdanov, K. Kapustin, A. Kudryavtsev, and A. Chirtsov, “Different approaches to fluid simulation of the longitudinal structure of the atmospheric-pressure microdischarge in helium,” *Technical Physics*, vol. 55, pp. 1430–1442, 2010.
- [36] M. M. Becker, D. Loffhagen, and W. Schmidt, “A stabilized finite element method for modeling of gas discharges,” *Computer Physics Communications*, vol. 180, no. 8, pp. 1230–1241, 2009.
- [37] R. Winkler, S. Arndt, D. Loffhagen, F. Sigeneger, and D. Uhrlandt, “Progress of the electron kinetics in spatial and spatiotemporal plasma structures,” *Contributions to Plasma Physics*, vol. 44, no. 5-6, pp. 437–449, 2004.
- [38] V. I. Kolobov and R. R. Arslanbekov, “Simulation of electron kinetics in gas discharges,” *IEEE transactions on plasma science*, vol. 34, no. 3, pp. 895–909, 2006.
- [39] R. Robson, R. D. White, and Z. L. Petrović, “Colloquium: Physically based fluid modeling of collisionally dominated low-temperature plasmas,” *Reviews of modern physics*, vol. 77, no. 4, p. 1303, 2005.

- [40] R. D. White, R. Robson, S. Dujko, P. Nicoletopoulos, and B. Li, “Recent advances in the application of boltzmann equation and fluid equation methods to charged particle transport in non-equilibrium plasmas,” *Journal of Physics D: Applied Physics*, vol. 42, no. 19, p. 194001, 2009.
- [41] C. Yuan, E. Bogdanov, S. Eliseev, and A. Kudryavtsev, “1d kinetic simulations of a short glow discharge in helium,” *Physics of Plasmas*, vol. 24, no. 7, 2017.
- [42] Z. Donkó, P. Hartmann, and K. Kutasi, “On the reliability of low-pressure dc glow discharge modelling,” *Plasma Sources Science and Technology*, vol. 15, no. 2, p. 178, 2006.
- [43] Z. Donkó, “Particle simulation methods for studies of low-pressure plasma sources,” *Plasma Sources Science and Technology*, vol. 20, no. 2, p. 024001, 2011.
- [44] A. Fiala, L. Pitchford, and J. Boeuf, “Two-dimensional, hybrid model of low-pressure glow discharges,” *Physical Review E*, vol. 49, no. 6, p. 5607, 1994.
- [45] J.-P. Boeuf and E. Marode, “A monte carlo analysis of an electron swarm in a nonuniform field: the cathode region of a glow discharge in helium,” *Journal of Physics D: Applied Physics*, vol. 15, no. 11, p. 2169, 1982.
- [46] M. Surendra, D. Graves, and G. Jellum, “Self-consistent model of a direct-current glow discharge: Treatment of fast electrons,” *Physical Review A*, vol. 41, no. 2, p. 1112, 1990.
- [47] M. J. Kushner, “Hybrid modelling of low temperature plasmas for fundamental investigations and equipment design,” *Journal of Physics D: Applied Physics*, vol. 42, no. 19, p. 194013, 2009.
- [48] J. Van Dijk, G. Kroesen, and A. Bogaerts, “Plasma modelling and numerical simulation,” *Journal of Physics D: Applied Physics*, vol. 42, no. 19, p. 190301, 2009.
- [49] A. Bogaerts, R. Gijbels, and W. Goedheer, “Hybrid monte carlo-fluid model of a direct current glow discharge,” *Journal of applied physics*, vol. 78, no. 4, pp. 2233–2241, 1995.

- [50] L. Tsendin, “Electron kinetics in non-uniform glow discharge plasmas,” *Plasma Sources Science and Technology*, vol. 4, no. 2, p. 200, 1995.
- [51] J. P. Trelles, “Pattern formation and self-organization in plasmas interacting with surfaces,” *Journal of Physics D: Applied Physics*, vol. 49, no. 39, p. 393002, 2016.
- [52] M. C. Cross and P. C. Hohenberg, “Pattern formation outside of equilibrium,” *Reviews of modern physics*, vol. 65, no. 3, p. 851, 1993.
- [53] E. Gurevich, Y. A. Astrov, and H. Purwins, “Pattern formation in planar dc-driven semiconductor–gas discharge devices: two mechanisms,” *Journal of Physics D: Applied Physics*, vol. 38, no. 3, p. 468, 2005.
- [54] H. Purwins and L. Stollenwerk, “Synergetic aspects of gas-discharge: lateral patterns in dc systems with a high ohmic barrier,” *Plasma Physics and Controlled Fusion*, vol. 56, no. 12, p. 123001, 2014.
- [55] H. Willebrand, T. Hünteler, F.-J. Niedernostheide, R. Dohmen, and H.-G. Purwins, “Periodic and turbulent behavior of solitary structures in distributed active media,” *Physical Review A*, vol. 45, no. 12, p. 8766, 1992.
- [56] C. Strümpel, Y. A. Astrov, and H.-G. Purwins, “Multioscillatory patterns in a hybrid semiconductor gas-discharge system,” *Physical Review E*, vol. 65, no. 6, p. 066210, 2002.
- [57] C. Strümpel, Y. A. Astrov, and H.-G. Purwins, “Nonlinear interaction of homogeneously oscillating domains in a planar gas discharge system,” *Physical Review E*, vol. 62, no. 4, p. 4889, 2000.
- [58] C. Yuan, C. Yesil, J. Yao, Z. Zhou, and I. Rafatov, “Transition from periodic to chaotic oscillations in a planar gas discharge-semiconductor system,” *Plasma Sources Science and Technology*, vol. 29, no. 6, p. 065009, 2020.
- [59] P. Manneville, “Dissipative structures and weak turbulence,” in *Chaos—The Interplay Between Stochastic and Deterministic Behaviour: Proceedings of the XXXIst Winter School of Theoretical Physics Held in Karpacz, Poland 13–24 February 1995*, pp. 257–272, Springer, 2005.

- [60] T. Klinger, C. Schröder, D. Block, F. Greiner, A. Piel, G. Bonhomme, and V. Naulin, “Chaos control and taming of turbulence in plasma devices,” *Physics of Plasmas*, vol. 8, no. 5, pp. 1961–1968, 2001.
- [61] C. Radehaus, R. Dohmen, H. Willebrand, and F.-J. Niedernostheide, “Model for current patterns in physical systems with two charge carriers,” *Physical Review A*, vol. 42, no. 12, p. 7426, 1990.
- [62] M. Benilov, “Multiple solutions in the theory of dc glow discharges and cathodic part of arc discharges. application of these solutions to the modeling of cathode spots and patterns: a review,” *Plasma Sources Science and Technology*, vol. 23, no. 5, p. 054019, 2014.
- [63] A. Phelps, Z. L. Petrović, and B. Jelenković, “Oscillations of low-current electrical discharges between parallel-plane electrodes. iii. models,” *Physical Review E*, vol. 47, no. 4, p. 2825, 1993.
- [64] B. Jelenković, K. Rózsa, and A. Phelps, “Oscillations of low-current electrical discharges between parallel-plane electrodes. ii. pulsed discharges in h_2 ,” *Physical Review E*, vol. 47, no. 4, p. 2816, 1993.
- [65] V. Kolobov and A. Fiala, “Transition from a townsend discharge to a normal discharge via two-dimensional modeling,” *Physical Review E*, vol. 50, no. 4, p. 3018, 1994.
- [66] Y. P. Raizer and M. Mokrov, “Physical mechanisms of self-organization and formation of current patterns in gas discharges of the townsend and glow types,” *Physics of Plasmas*, vol. 20, no. 10, p. 101604, 2013.
- [67] E. Ott, *Chaos in dynamical systems*. Cambridge university press, 2002.
- [68] H.-G. Purwins, H. Bödeker, and S. Amiranashvili, “Dissipative solitons,” *Advances in Physics*, vol. 59, no. 5, pp. 485–701, 2010.
- [69] D. Mansuroglu, I. Uzun-Kaymak, and I. Rafatov, “An evidence of period doubling bifurcation in a dc driven semiconductor-gas discharge plasma,” *Physics of Plasmas*, vol. 24, no. 5, p. 053503, 2017.

- [70] D. D. Šijačić, U. Ebert, and I. Rafatov, “Period doubling cascade in glow discharges: Local versus global differential conductivity,” *Physical Review E*, vol. 70, no. 5, p. 056220, 2004.
- [71] I. Rafatov, “Three-dimensional numerical modelling of temporal and spatial pattern formation in a dc-driven gas discharge-semiconductor system,” *Plasma Sources Science and Technology*, vol. 25, no. 6, p. 065014, 2016.
- [72] R. M. May, “Simple mathematical models with very complicated dynamics,” *Nature*, vol. 261, no. 5560, pp. 459–467, 1976.
- [73] A. S. Hornby and A. P. Cowie, “Oxford advanced learner’s dictionary of current english,” 1977.
- [74] C. Huguenii, H. Oscilatorium, and A. F. Muquet, “Parisiis, 1673, english translation: The pendulum clock,” 1986.
- [75] A. Pikovsky, M. Rosenblum, and J. Kurths, *Synchronization, a Universal Concept in Nonlinear Sciences*. Cambridge, UK: Cambridge University Press, 2001.
- [76] H. Fujisaka and T. Yamada, “Stability theory of synchronized motion in coupled-oscillator systems,” *Progress of theoretical physics*, vol. 69, no. 1, pp. 32–47, 1983.
- [77] L. M. Pecora and T. L. Carroll, “Synchronization in chaotic systems,” *Physical review letters*, vol. 64, no. 8, p. 821, 1990.
- [78] M. G. Rosenblum, A. S. Pikovsky, and J. Kurths, “Phase synchronization of chaotic oscillators,” *Physical review letters*, vol. 76, no. 11, p. 1804, 1996.
- [79] E. Rosa Jr, E. Ott, and M. H. Hess, “Transition to phase synchronization of chaos,” *Physical review letters*, vol. 80, no. 8, p. 1642, 1998.
- [80] M. G. Rosenblum, A. S. Pikovsky, and J. Kurths, “From phase to lag synchronization in coupled chaotic oscillators,” *Physical Review Letters*, vol. 78, no. 22, p. 4193, 1997.

- [81] L. Kocarev and U. Parlitz, “Generalized synchronization, predictability, and equivalence of unidirectionally coupled dynamical systems,” *Physical review letters*, vol. 76, no. 11, p. 1816, 1996.
- [82] N. F. Rulkov, M. M. Sushchik, L. S. Tsimring, and H. D. Abarbanel, “Generalized synchronization of chaos in directionally coupled chaotic systems,” *Physical Review E*, vol. 51, no. 2, p. 980, 1995.
- [83] S. Boccaletti and D. Valladares, “Characterization of intermittent lag synchronization,” *Physical Review E*, vol. 62, no. 5, p. 7497, 2000.
- [84] M. A. Zaks, E.-H. Park, M. G. Rosenblum, and J. Kurths, “Alternating locking ratios in imperfect phase synchronization,” *Physical review letters*, vol. 82, no. 21, p. 4228, 1999.
- [85] R. Femat and G. Solis-Perales, “On the chaos synchronization phenomena,” *Physics Letters A*, vol. 262, no. 1, pp. 50–60, 1999.
- [86] H. D. Abarbanel, N. F. Rulkov, and M. M. Sushchik, “Generalized synchronization of chaos: The auxiliary system approach,” *Physical review E*, vol. 53, no. 5, p. 4528, 1996.
- [87] S. J. Schiff, P. So, T. Chang, R. E. Burke, and T. Sauer, “Detecting dynamical interdependence and generalized synchrony through mutual prediction in a neural ensemble,” *Physical Review E*, vol. 54, no. 6, p. 6708, 1996.
- [88] M. Breakspear and J. Terry, “Detection and description of non-linear interdependence in normal multichannel human eeg data,” *Clinical neurophysiology*, vol. 113, no. 5, pp. 735–753, 2002.
- [89] M. Akhmet, I. Rafatov, and M. O. Fen, “Extension of spatiotemporal chaos in glow discharge-semiconductor systems,” *Chaos: An Interdisciplinary Journal of Nonlinear Science*, vol. 24, no. 4, p. 043127, 2014.
- [90] M. Akhmet, C. Yeşil, and K. Başkan, “Synchronization of chaos in semiconductor gas discharge model with local mean energy approximation,” *Chaos, Solitons & Fractals*, vol. 167, p. 113035, 2023.

- [91] M. Akhmet, K. Başkan, and C. Yeşil, “Revealing chaos synchronization below the threshold in coupled mackey–glass systems,” *Mathematics*, vol. 11, no. 14, p. 3197, 2023.
- [92] M. Akhmet and M. O. Fen, “Unpredictable points and chaos,” *Communications in Nonlinear Science and Numerical Simulation*, vol. 40, pp. 1–5, 2016.
- [93] M. Akhmet, M. O. Fen, and A. Tola, “The sequential test for chaos,” *arXiv preprint arXiv:1904.09127*, 2019.
- [94] M. Akhmet, M. O. Fen, and A. Tola, “Strange non-chaotic attractors with unpredictable trajectories,” *arXiv preprint arXiv:2110.15805*, 2021.
- [95] M. Akhmet, M. O. Fen, and E. M. Alejaily, *Dynamics with Chaos and Fractals*. Cham, Switzerland: Springer, 2020.
- [96] V. Gorin, A. Kudryavtsev, J. Yao, C. Yuan, and Z. Zhou, “Boundary conditions for drift-diffusion equations in gas-discharge plasmas,” *Physics of Plasmas*, vol. 27, no. 1, p. 013505, 2020.
- [97] I. Shkarofsky, T. Johnston, M. Bachynski, and J. Hirshfield, “The particle kinetics of plasmas,” *American Journal of Physics*, vol. 35, no. 6, pp. 551–552, 1967.
- [98] A. Kudryavtsev, A. Morin, and L. Tsendin, “Role of nonlocal ionization in formation of the short glow discharge,” *Technical Physics*, vol. 53, no. 8, pp. 1029–1040, 2008.
- [99] U. Kortshagen, C. Busch, and L. Tsendin, “On simplifying approaches to the solution of the boltzmann equation in spatially inhomogeneous plasmas,” *Plasma Sources Science and Technology*, vol. 5, no. 1, p. 1, 1996.
- [100] E. W. McDaniel, “Collision phenomena in ionized gases,” *Wiley Series in Plasmas Physics*, 1964.
- [101] G. Hagelaar, F. De Hoog, and G. Kroesen, “Boundary conditions in fluid models of gas discharges,” *Physical Review E*, vol. 62, no. 1, p. 1452, 2000.

- [102] S. I. Eliseev, A. A. Kudryavtsev, H. Liu, Z. Ning, D. Yu, and A. S. Chirtsov, "Transition from glow microdischarge to arc discharge with thermionic cathode in argon at atmospheric pressure," *IEEE Transactions on Plasma Science*, vol. 44, no. 11, pp. 2536–2544, 2016.
- [103] I. Rafatov, E. Bogdanov, and A. Kudryavtsev, "On the accuracy and reliability of different fluid models of the direct current glow discharge," *Physics of Plasmas*, vol. 19, no. 3, 2012.
- [104] A. Phelps, "The diffusion of charged particles in collisional plasmas: free and ambipolar diffusion at low and moderate pressures," *Journal of research of the National Institute of Standards and Technology*, vol. 95, no. 4, p. 407, 1990.
- [105] P. Chantry, A. Phelps, and G. Schulz, "Theory of electron collision experiments at intermediate and high gas densities," *Physical Review*, vol. 152, no. 1, p. 81, 1966.
- [106] A. Wilson and B. Shotorban, "Investigation of surface boundary conditions for continuum modeling of rf plasmas," *Physics of Plasmas*, vol. 25, no. 5, p. 053509, 2018.
- [107] A. J. Lichtenberg, *Principles of plasma discharges and materials processing*. Wiley-Interscience, 2005.
- [108] G. Hays, C. Tracy, and H. Oskam, "Surface catalytic efficiency of a sputtered molybdenum layer on quartz and pyrex for the recombination of nitrogen atoms," *The Journal of Chemical Physics*, vol. 60, no. 5, pp. 2027–2034, 1974.
- [109] E. Kawamura, V. Vahedi, M. Lieberman, , and C. Birdsall, "Ion energy distributions in rf sheaths; review, analysis and simulation," *Plasma Sources Science and Technology*, vol. 8, no. 3, p. R45, 1999.
- [110] J. Schulze, A. Derzsi, K. Dittmann, T. Hemke, J. Meichsner, and Z. Donkó, "Ionization by drift and ambipolar electric fields in electronegative capacitive radio frequency plasmas," *Physical review letters*, vol. 107, no. 27, p. 275001, 2011.

- [111] Y. Mitsui and T. Makabe, “Review and current status: E, h mode transition in low-temperature icp and related electron dynamics,” *Plasma Sources Science and Technology*, vol. 30, no. 2, p. 023001, 2021.
- [112] H.-C. Lee, “Review of inductively coupled plasmas: Nano-applications and bistable hysteresis physics,” *Applied Physics Reviews*, vol. 5, no. 1, 2018.
- [113] S.-K. Park and D. J. Economou, “Analysis of low pressure rf glow discharges using a continuum model,” *Journal of applied physics*, vol. 68, no. 8, pp. 3904–3915, 1990.
- [114] W. Goedheer, M. Akdim, and Y. I. Chutov, “Hydrodynamic and kinetic modelling of dust free and dusty radio-frequency discharges,” *Contributions to Plasma Physics*, vol. 44, no. 5-6, pp. 395–404, 2004.
- [115] B. Parent, S. O. Macheret, and M. N. Shneider, “Electron and ion transport equations in computational weakly-ionized plasmadynamics,” *Journal of Computational Physics*, vol. 259, pp. 51–69, 2014.
- [116] M. Zakari, H. Caquineau, P. Hotmar, and P. Ségur, “An axisymmetric unstructured finite volume method applied to the numerical modeling of an atmospheric pressure gas discharge,” *Journal of Computational Physics*, vol. 281, pp. 473–492, 2015.
- [117] S. T. Surzhikov and J. S. Shang, “Two-component plasma model for two-dimensional glow discharge in magnetic field,” *Journal of Computational Physics*, vol. 199, no. 2, pp. 437–464, 2004.
- [118] F. Bouanaka and S. Rebiai, “Effect of secondary electron emission on argon glow discharge characteristics,” *Int. Rev. Phys*, vol. 7, no. 1, pp. 81–86, 2013.
- [119] M. Davoudabadi and F. Mashayek, “Numerical modeling of dust particle configurations in a cylindrical radio-frequency plasma reactor,” *Physical Review E*, vol. 76, no. 5, p. 056405, 2007.
- [120] J. Passchier and W. Goedheer, “A two-dimensional fluid model for an argon rf discharge,” *Journal of applied physics*, vol. 74, no. 6, pp. 3744–3751, 1993.

- [121] M. Davoudabadi, J. S. Shrimpton, and F. Mashayek, “On accuracy and performance of high-order finite volume methods in local mean energy model of non-thermal plasmas,” *Journal of Computational Physics*, vol. 228, no. 7, pp. 2468–2479, 2009.
- [122] C. Horn, M. Davoudabadi, and B. Shotorban, “Effects of radiofrequency on dust particle dynamics in a plasma reactor,” *Journal of Applied Physics*, vol. 110, no. 11, p. 113305, 2011.
- [123] T. Paunskaa, A. Shivarova, and K. Tarnev, “Comments on the boundary conditions for metal and dielectric walls in the fluid-plasma modelling,” *Proceedings of the 30th ICPIG*, 2011.
- [124] G. Gozadinos, A. Ivlev, and J. Boeuf, “A fluid model for colloidal plasmas under microgravity conditions,” *New Journal of Physics*, vol. 5, no. 1, p. 32, 2003.
- [125] W. Siemens, “Ueber die elektrostatische induction und die verzögerung des stroms in flaschendrahten,” *Annalen der Physik*, vol. 178, no. 9, pp. 66–122, 1857.
- [126] U. Kogelschatz, “Collective phenomena in volume and surface barrier discharges,” in *Journal of Physics: Conference Series*, vol. 257, p. 012015, IOP Publishing, 2010.
- [127] X. Li, N. Zhao, T. Fang, Z. Liu, L. Li, and L. Dong, “Characteristics of an atmospheric pressure argon glow discharge in a coaxial electrode geometry,” *Plasma Sources Science and Technology*, vol. 17, no. 1, p. 015017, 2008.
- [128] S. Gadkari and S. Gu, “Numerical investigation of co-axial dbd: Influence of relative permittivity of the dielectric barrier, applied voltage amplitude, and frequency,” *Physics of Plasmas*, vol. 24, no. 5, 2017.
- [129] D. Petrović, T. Martens, J. Van Dijk, W. Brok, and A. Bogaerts, “Fluid modelling of an atmospheric pressure dielectric barrier discharge in cylindrical geometry,” *Journal of Physics D: Applied Physics*, vol. 42, no. 20, p. 205206, 2009.

- [130] A. Sublet, C. Ding, J. Dorier, C. Hollenstein, P. Fayet, and F. Coursimault, “Atmospheric and sub-atmospheric dielectric barrier discharges in helium and nitrogen,” *Plasma Sources Science and Technology*, vol. 15, no. 4, p. 627, 2006.
- [131] R. Brandenburg, Z. Navrátil, J. Jánský, P. St’Ahel, D. Trunec, and H.-E. Wagner, “The transition between different modes of barrier discharges at atmospheric pressure,” *Journal of Physics D: Applied Physics*, vol. 42, no. 8, p. 085208, 2009.
- [132] P. Kloc, H.-E. Wagner, D. Trunec, Z. Navrátil, and G. Fedoseev, “An investigation of dielectric barrier discharge in ar and ar/nh₃ mixture using cross-correlation spectroscopy,” *Journal of Physics D: Applied Physics*, vol. 43, no. 34, p. 345205, 2010.
- [133] F. Massines, A. Rabehi, P. Decomps, R. B. Gadri, P. Ségur, and C. Mayoux, “Experimental and theoretical study of a glow discharge at atmospheric pressure controlled by dielectric barrier,” *Journal of Applied Physics*, vol. 83, no. 6, pp. 2950–2957, 1998.
- [134] Y. B. Golubovskii, V. Maiorov, J. Behnke, and J. Behnke, “Modelling of the homogeneous barrier discharge in helium at atmospheric pressure,” *Journal of Physics D: Applied Physics*, vol. 36, no. 1, p. 39, 2002.
- [135] Z. Navrátil, R. Brandenburg, D. Trunec, A. Brablec, P. St’ahel, H.-E. Wagner, and Z. Kopecký, “Comparative study of diffuse barrier discharges in neon and helium,” *Plasma Sources Science and Technology*, vol. 15, no. 1, p. 8, 2005.
- [136] R. Timm and A. Piel, “Hysteresis and transition to chaos in a thermionic plasma discharge,” *Contributions to Plasma Physics*, vol. 32, no. 6, pp. 599–611, 1992.
- [137] P. Cheung and A. Wong, “Chaotic behavior and period doubling in plasmas,” *Physical review letters*, vol. 59, no. 5, p. 551, 1987.
- [138] M. Mikikian, M. Cavarroc, L. Couédel, Y. Tessier, and L. Boufendi, “Mixed-mode oscillations in complex-plasma instabilities,” *Physical Review Letters*, vol. 100, no. 22, p. 225005, 2008.

- [139] Y. T. Zhang, D. Z. Wang, and M. G. Kong, “Complex dynamic behaviors of nonequilibrium atmospheric dielectric-barrier discharges,” *Journal of applied physics*, vol. 100, no. 6, 2006.
- [140] H. Shi, Y. Wang, and D. Wang, “Nonlinear behavior in the time domain in argon atmospheric dielectric-barrier discharges,” *Physics of Plasmas*, vol. 15, no. 12, 2008.
- [141] D. Zhang, Y. Wang, J. Sun, and D. Wang, “Two-dimensional numerical study of a period-two dielectric-barrier discharge in atmospheric argon,” *Physics of Plasmas*, vol. 19, no. 4, 2012.
- [142] F. Massines, N. Ghérardi, N. Naudé, and P. Ségur, “Recent advances in the understanding of homogeneous dielectric barrier discharges,” *The European Physical Journal-Applied Physics*, vol. 47, no. 2, p. 22805, 2009.
- [143] U. Kogelschatz, “Filamentary, patterned, and diffuse barrier discharges,” *IEEE Transactions on plasma science*, vol. 30, no. 4, pp. 1400–1408, 2002.
- [144] K. R. Gurney, D. L. Mendoza, Y. Zhou, M. L. Fischer, C. C. Miller, S. Geethakumar, and S. de la Rue du Can, “High resolution fossil fuel combustion co₂ emission fluxes for the united states,” *Environmental science & technology*, vol. 43, no. 14, pp. 5535–5541, 2009.
- [145] D. P. Van Vuuren, J. Edmonds, M. Kainuma, K. Riahi, A. Thomson, K. Hibbard, G. C. Hurtt, T. Kram, V. Krey, J.-F. Lamarque, *et al.*, “The representative concentration pathways: an overview,” *Climatic change*, vol. 109, pp. 5–31, 2011.
- [146] A. Zecchina, “Energy sources and carbon dioxide waste,” *Rendiconti Lincei*, vol. 25, pp. 113–117, 2014.
- [147] M. Aresta, *Carbon dioxide as chemical feedstock*. John Wiley & Sons, 2010.
- [148] J. Christophe, T. Doneux, and C. Buess-Herman, “Electroreduction of carbon dioxide on copper-based electrodes: activity of copper single crystals and copper–gold alloys,” *Electrocatalysis*, vol. 3, pp. 139–146, 2012.

- [149] T. Sakakura, J.-C. Choi, and H. Yasuda, "Transformation of carbon dioxide," *Chemical reviews*, vol. 107, no. 6, pp. 2365–2387, 2007.
- [150] J. H. Jones, "The cativa™ process for the manufacture of acetic acid," *Platinum Metals Review*, vol. 44, no. 3, pp. 94–105, 2000.
- [151] G. J. Sunley and D. J. Watson, "High productivity methanol carbonylation catalysis using iridium: The cativa™ process for the manufacture of acetic acid," *Catalysis Today*, vol. 58, no. 4, pp. 293–307, 2000.
- [152] M. E. Dry, "The fischer–tropsch process: 1950–2000," *Catalysis today*, vol. 71, no. 3-4, pp. 227–241, 2002.
- [153] L. F. Spencer and A. D. Gallimore, "Efficiency of co₂ dissociation in a radio-frequency discharge," *Plasma Chemistry and Plasma Processing*, vol. 31, pp. 79–89, 2011.
- [154] M.-S. Fan, A. Z. Abdullah, and S. Bhatia, "Catalytic technology for carbon dioxide reforming of methane to synthesis gas," *ChemCatChem*, vol. 1, no. 2, pp. 192–208, 2009.
- [155] M. Scapinello, L. M. Martini, and P. Tosi, "Co₂ hydrogenation by ch₄ in a dielectric barrier discharge: Catalytic effects of nickel and copper," *Plasma Processes and Polymers*, vol. 11, no. 7, pp. 624–628, 2014.
- [156] X. Tu, H. J. Gallon, M. V. Twigg, P. A. Gorry, and J. C. Whitehead, "Dry reforming of methane over a ni/al₂o₃ catalyst in a coaxial dielectric barrier discharge reactor," *Journal of Physics D: Applied Physics*, vol. 44, no. 27, p. 274007, 2011.
- [157] X. Tu and J. Whitehead, "Plasma-catalytic dry reforming of methane in an atmospheric dielectric barrier discharge: Understanding the synergistic effect at low temperature," *Applied Catalysis B: Environmental*, vol. 125, pp. 439–448, 2012.
- [158] A. Ghorbanzadeh, R. Lotfalipour, and S. Rezaei, "Carbon dioxide reforming of methane at near room temperature in low energy pulsed plasma," *international journal of hydrogen energy*, vol. 34, no. 1, pp. 293–298, 2009.

- [159] D. Li, X. Li, M. Bai, X. Tao, S. Shang, X. Dai, and Y. Yin, "Co₂ reforming of ch₄ by atmospheric pressure glow discharge plasma: a high conversion ability," *International Journal of Hydrogen Energy*, vol. 34, no. 1, pp. 308–313, 2009.
- [160] Z. Bo, J. Yan, X. Li, Y. Chi, and K. Cen, "Plasma assisted dry methane reforming using gliding arc gas discharge: effect of feed gases proportion," *International Journal of Hydrogen Energy*, vol. 33, no. 20, pp. 5545–5553, 2008.
- [161] A. Indarto, D. R. Yang, J.-W. Choi, H. Lee, and H. K. Song, "Gliding arc plasma processing of co₂ conversion," *Journal of hazardous materials*, vol. 146, no. 1-2, pp. 309–315, 2007.
- [162] T. Nunnally, K. Gutsol, A. Rabinovich, A. Fridman, A. Gutsol, and A. Kermoun, "Dissociation of co₂ in a low current gliding arc plasmatron," *Journal of Physics D: Applied Physics*, vol. 44, no. 27, p. 274009, 2011.
- [163] K. Pornmai, A. Jindanin, H. Sekiguchi, and S. Chavadej, "Synthesis gas production from co₂-containing natural gas by combined steam reforming and partial oxidation in an ac gliding arc discharge," *Plasma Chemistry and Plasma Processing*, vol. 32, pp. 723–742, 2012.
- [164] X. Tu and J. C. Whitehead, "Plasma dry reforming of methane in an atmospheric pressure ac gliding arc discharge: Co-generation of syngas and carbon nanomaterials," *International journal of hydrogen energy*, vol. 39, no. 18, pp. 9658–9669, 2014.
- [165] L. Spencer and A. Gallimore, "Co₂ dissociation in an atmospheric pressure plasma/catalyst system: a study of efficiency," *Plasma Sources Science and Technology*, vol. 22, no. 1, p. 015019, 2012.
- [166] A. Vesel, M. Mozetic, A. Drenik, and M. Balat-Pichelin, "Dissociation of co₂ molecules in microwave plasma," *Chemical Physics*, vol. 382, no. 1-3, pp. 127–131, 2011.
- [167] F. Brehmer, S. Welzel, M. Van De Sanden, and R. Engeln, "Co and byproduct formation during co₂ reduction in dielectric barrier discharges," *Journal of Applied Physics*, vol. 116, no. 12, p. 123303, 2014.

- [168] X. Duan, Y. Li, W. Ge, and B. Wang, "Degradation of CO_2 through dielectric barrier discharge microplasma," *Greenhouse Gases: Science and Technology*, vol. 5, no. 2, pp. 131–140, 2015.
- [169] D. Mei, X. Zhu, Y.-L. He, J. D. Yan, and X. Tu, "Plasma-assisted conversion of CO_2 in a dielectric barrier discharge reactor: understanding the effect of packing materials," *Plasma Sources Science and Technology*, vol. 24, no. 1, p. 015011, 2014.
- [170] R. Snoeckx, Y. Zeng, X. Tu, and A. Bogaerts, "Plasma-based dry reforming: improving the conversion and energy efficiency in a dielectric barrier discharge," *RSC advances*, vol. 5, no. 38, pp. 29799–29808, 2015.
- [171] Q. Yu, M. Kong, T. Liu, J. Fei, and X. Zheng, "Characteristics of the decomposition of CO_2 in a dielectric packed-bed plasma reactor," *Plasma Chemistry and Plasma Processing*, vol. 32, pp. 153–163, 2012.
- [172] B. Eliasson, M. Hirth, and U. Kogelschatz, "Ozone synthesis from oxygen in dielectric barrier discharges," *Journal of Physics D: Applied Physics*, vol. 20, no. 11, p. 1421, 1987.
- [173] U. Kogelschatz, B. Eliasson, and W. Egli, "Plasma chem. plasma process," *Plasma Chem. Plasma Process*, vol. 23, no. 1, pp. 1–46, 2003.
- [174] U. Kogelschatz, B. Eliasson, and W. Egli, "From ozone generators to flat television screens: history and future potential of dielectric-barrier discharges," *Pure and Applied Chemistry*, vol. 71, no. 10, pp. 1819–1828, 1999.
- [175] A. Ozkan, T. Dufour, G. Arnoult, P. De Keyser, A. Bogaerts, and F. Reniers, " CO_2 - CH_4 conversion and syngas formation at atmospheric pressure using a multi-electrode dielectric barrier discharge," *Journal of CO_2 utilization*, vol. 9, pp. 74–81, 2015.
- [176] X. Tu, H. J. Gallon, and J. C. Whitehead, "Transition behavior of packed-bed dielectric barrier discharge in argon," *IEEE Transactions on Plasma Science*, vol. 39, no. 11, pp. 2172–2173, 2011.

- [177] K. Van Laer and A. Bogaerts, “Improving the conversion and energy efficiency of carbon dioxide splitting in a zirconia-packed dielectric barrier discharge reactor,” *Energy Technology*, vol. 3, no. 10, pp. 1038–1044, 2015.
- [178] Y. Zhang, H.-y. Wang, W. Jiang, and A. Bogaerts, “Two-dimensional particle-in cell/monte carlo simulations of a packed-bed dielectric barrier discharge in air at atmospheric pressure,” *New Journal of Physics*, vol. 17, no. 8, p. 083056, 2015.
- [179] V. D. Rusanov, A. Fridman, and G. V. Sholin, “The physics of a chemically active plasma with nonequilibrium vibrational excitation of molecules,” *Soviet Physics Uspekhi*, vol. 24, no. 6, p. 447, 1981.
- [180] D. Loffhagen and F. Sigeneger, “Advances in boltzmann equation based modelling of discharge plasmas,” *Plasma Sources Science and Technology*, vol. 18, no. 3, p. 034006, 2009.
- [181] L. C. Brown and A. T. Bell, “Kinetics of the oxidation of carbon monoxide and the decomposition of carbon dioxide in a radiofrequency electric discharge. ii. theoretical interpretation,” *Industrial & Engineering Chemistry Fundamentals*, vol. 13, no. 3, pp. 210–218, 1974.
- [182] A. Cenian, A. Chernukho, and V. Borodin, “Modeling of plasma-chemical reactions in gas mixture of co₂ lasers. ii. theoretical model and its verification,” *Contributions to Plasma Physics*, vol. 35, no. 3, pp. 273–296, 1995.
- [183] T. Kozák and A. Bogaerts, “Splitting of co₂ by vibrational excitation in non-equilibrium plasmas: a reaction kinetics model,” *Plasma Sources Science and Technology*, vol. 23, no. 4, p. 045004, 2014.
- [184] T. Kozák and A. Bogaerts, “Evaluation of the energy efficiency of co₂ conversion in microwave discharges using a reaction kinetics model,” *Plasma Sources Science and Technology*, vol. 24, no. 1, p. 015024, 2014.
- [185] W. Wiegand and W. L. Nighan, “Plasma chemistry of co₂–n₂–he discharges,” *Applied Physics Letters*, vol. 22, no. 11, pp. 583–586, 1973.
- [186] H. Hokazono and H. Fujimoto, “Theoretical analysis of the co₂ molecule decomposition and contaminants yield in transversely excited atmospheric co₂

- laser discharge,” *Journal of applied physics*, vol. 62, no. 5, pp. 1585–1594, 1987.
- [187] H. Hokazono and M. Obara, “Reduction of the co₂ decomposition in the transversely excited atmospheric co₂ laser discharge plasma by a very small amount of the water vapor,” *Applied physics letters*, vol. 57, no. 1, pp. 4–6, 1990.
- [188] H. Hokazono, N. Kobayashi, and M. Obara, “Catalytic control requirements for the stable operation of the closed-cycle, transversely excited atmospheric co₂ laser,” *Journal of applied physics*, vol. 70, no. 4, pp. 1962–1965, 1991.
- [189] H. Hokazono, M. Obara, K. Midorikawa, and H. Tashiro, “Theoretical operational life study of the closed-cycle transversely excited atmospheric co₂ laser,” *Journal of applied physics*, vol. 69, no. 10, pp. 6850–6868, 1991.
- [190] S. R. Byron and H. Apter, “Model of gas composition and plasma properties in sealed cw co₂ lasers,” *Journal of applied physics*, vol. 71, no. 4, pp. 1976–1991, 1992.
- [191] A. Cenian, A. Chernukho, P. Kukiello, R. Zaremba, V. Borodin, and G. Sliwinski, “Improvement of self-regeneration of gas mixtures in a convection-cooled 1.2 kw laser,” *Journal of Physics D: Applied Physics*, vol. 30, no. 7, p. 1103, 1997.
- [192] A. Yamamoto, S. Mori, and M. Suzuki, “Scale-up or numbering-up of a micro plasma reactor for the carbon dioxide decomposition,” *Thin solid films*, vol. 515, no. 9, pp. 4296–4300, 2007.
- [193] B. Eliasson, W. Egli, and U. Kogelschatz, “Modelling of dielectric barrier discharge chemistry,” *Pure and applied chemistry*, vol. 66, no. 6, pp. 1275–1286, 1994.
- [194] R. Aerts, W. Somers, and A. Bogaerts, “Carbon dioxide splitting in a dielectric barrier discharge plasma: a combined experimental and computational study,” *ChemSusChem*, vol. 8, no. 4, pp. 702–716, 2015.
- [195] R. Aerts, T. Martens, and A. Bogaerts, “Influence of vibrational states on co₂ splitting by dielectric barrier discharges,” *The Journal of physical chemistry C*, vol. 116, no. 44, pp. 23257–23273, 2012.

- [196] R. Aerts, T. Martens, and A. Bogaerts, “Correction to “influence of vibrational states on co₂ splitting by dielectric barrier discharges”,” *The Journal of Physical Chemistry C*, vol. 118, no. 48, pp. 28330–28330, 2014.
- [197] S. Ponduri, M. Becker, S. Welzel, M. Van De Sanden, D. Loffhagen, and R. Engeln, “Fluid modelling of co₂ dissociation in a dielectric barrier discharge,” *Journal of Applied Physics*, vol. 119, no. 9, p. 093301, 2016.
- [198] G. Fridman, G. Friedman, A. Gutsol, A. B. Shekhter, V. N. Vasilets, and A. Fridman, “Applied plasma medicine,” *Plasma processes and polymers*, vol. 5, no. 6, pp. 503–533, 2008.
- [199] M. Becker, T. Hoder, R. Brandenburg, and D. Loffhagen, “Analysis of microdischarges in asymmetric dielectric barrier discharges in argon,” *Journal of Physics D: Applied Physics*, vol. 46, no. 35, p. 355203, 2013.
- [200] H. Höft, M. Kettlitz, M. Becker, T. Hoder, D. Loffhagen, R. Brandenburg, and K. Weltmann, “Breakdown characteristics in pulsed-driven dielectric barrier discharges: influence of the pre-breakdown phase due to volume memory effects,” *Journal of Physics D: Applied Physics*, vol. 47, no. 46, p. 465206, 2014.
- [201] M. Ramakers, I. Michielsen, R. Aerts, V. Meynen, and A. Bogaerts, “Effect of argon or helium on the co₂ conversion in a dielectric barrier discharge,” *Plasma Processes and Polymers*, vol. 12, no. 8, pp. 755–763, 2015.
- [202] R. Snoeckx, S. Heijckers, K. Van Wesenbeeck, S. Lenaerts, and A. Bogaerts, “Co₂ conversion in a dielectric barrier discharge plasma: N₂ in the mix as a helping hand or problematic impurity?,” *Energy & Environmental Science*, vol. 9, no. 3, pp. 999–1011, 2016.
- [203] R. Snoeckx, M. Setareh, R. Aerts, P. Simon, A. Maghari, and A. Bogaerts, “Influence of n₂ concentration in a ch₄/n₂ dielectric barrier discharge used for ch₄ conversion into h₂,” *international journal of hydrogen energy*, vol. 38, no. 36, pp. 16098–16120, 2013.
- [204] A. Ozkan, T. Dufour, A. Bogaerts, and F. Reniers, “How do the barrier thickness and dielectric material influence the filamentary mode and co₂ conversion

- in a flowing dbd?," *Plasma Sources Science and Technology*, vol. 25, no. 4, p. 045016, 2016.
- [205] J. H. Ingold, "Moment method applied to gaseous electronics," *Physical Review A*, vol. 40, no. 7, p. 3855, 1989.
- [206] D. B. Graves and K. F. Jensen, "A continuum model of dc and rf discharges," *IEEE Transactions on plasma science*, vol. 14, no. 2, pp. 78–91, 1986.
- [207] A. Salabas, G. Gousset, and L. Alves, "Two-dimensional fluid modelling of charged particle transport in radio-frequency capacitively coupled discharges," *Plasma Sources Science and Technology*, vol. 11, no. 4, p. 448, 2002.
- [208] L. Frost and A. Phelps, "Rotational excitation and momentum transfer cross sections for electrons in H_2 and N_2 from transport coefficients," *Physical Review*, vol. 127, no. 5, p. 1621, 1962.
- [209] J. Wilhelm and R. Winkler, "Convergence questions in calculating velocity distribution function of electrons of lorentz plasma in a homogenous, stationary electrical field," *ANNALEN DER PHYSIK*, vol. 23, no. 1-2, p. 28, 1969.
- [210] S. D. Rockwood, "Elastic and inelastic cross sections for electron-hg scattering from hg transport data," *Physical Review A*, vol. 8, no. 5, p. 2348, 1973.
- [211] P. Luft, "Jila information center report no. 14 university of colorado," 1975.
- [212] H. Tagashira, Y. Sakai, and S. Sakamoto, "The development of electron avalanches in argon at high e/n values. ii. boltzmann equation analysis," *Journal of Physics D: Applied Physics*, vol. 10, no. 7, p. 1051, 1977.
- [213] W. Morgan, "Jila information center report no. 19 university of colorado," 1979.
- [214] W. Morgan and B. Penetrante, "Elendif: A time-dependent boltzmann solver for partially ionized plasmas," *Computer Physics Communications*, vol. 58, no. 1-2, pp. 127–152, 1990.
- [215] S. Lin, R. Robson, and E. Mason, "Moment theory of electron drift and diffusion in neutral gases in an electrostatic field," *The Journal of Chemical Physics*, vol. 71, no. 8, pp. 3483–3498, 1979.

- [216] L. Pitchford, S. O'Neil, and J. Rumble Jr, "Extended boltzmann analysis of electron swarm experiments," *Physical Review A*, vol. 23, no. 1, p. 294, 1981.
- [217] P. Segur, M.-C. Bordage, J.-P. Balaguer, and M. Yousfi, "The application of a modified form of thesn method to the calculation of swarm parameters of electrons in a weakly ionised equilibrium medium," *Journal of Computational Physics*, vol. 50, no. 1, pp. 116–137, 1983.
- [218] "Bolsig+." <https://us.lxcat.net/solvers/BOLSIG+/index.php?step=0>. Retrieved 30 2023.
- [219] S. Surzhikov and J. Shang, "Normal glow discharge in axial magnetic field," *Plasma Sources Science and Technology*, vol. 23, no. 5.
- [220] P. Manneville, *Dissipative Structures and Weak Turbulence*. San Diego: Academic Press, 1990.
- [221] T. Klinger, C. Schröder, D. Block, F. Greiner, A. Piel, G. Bonhomme, and V. Naulin, "Chaos control and taming of turbulence in plasma devices," *Physics of Plasmas*, vol. 8, no. 5, pp. 1961–1968, 2001.
- [222] A. Figueroa, R. Salgado-García, J. Rodríguez, F. Y. Bashir, M. A. Rivera, and F. Vázquez, "Stochastic and nonlinear dynamics in low-temperature plasmas," in *Plasma Science and Technology-Progress in Physical States and Chemical Reactions*, IntechOpen, 2016.
- [223] Y. P. Raizer and M. Mokrov, "Physical mechanisms of self-organization and formation of current patterns in gas discharges of the townsend and glow types," *Physics of Plasmas*, vol. 20, no. 10, p. 101604, 2013.
- [224] Y. P. Raizer, *Gas Discharge Physics*. Berlin: Springer-Verlag, 1991.
- [225] M. Mokrov and Y. P. Raizer, "3d simulation of hexagonal current pattern formation in a dc-driven gas discharge gap with a semiconductor cathode," *Plasma Sources Science and Technology*, vol. 27, no. 6, p. 065008, 2018.
- [226] I. Rafatov, "Multiple stationary filamentary states in a planar dc-driven gas discharge-semiconductor system," *Physics of Plasmas*, vol. 23, no. 12, p. 123506, 2016.

- [227] R. Wilson IV and N. Podder, “Observation of period multiplication and instability in a dc glow discharge,” *Physical Review E*, vol. 76, no. 4, p. 046405, 2007.
- [228] E. A. Jackson, *Perspectives of Nonlinear Dynamics*, vol. 1. Cambridge: Cambridge University Press, 1989.
- [229] T. Braun, J. A. Lisboa, R. E. Francke, and J. A. Gallas, “Observation of deterministic chaos in electrical discharges in gases,” *Physical review letters*, vol. 59, no. 6, p. 613, 1987.
- [230] E. Pugliese, R. Meucci, S. Euzzor, J. G. Freire, and J. A. Gallas, “Complex dynamics of a dc glow discharge tube: Experimental modeling and stability diagrams,” *Scientific reports*, vol. 5, p. 8447, 2015.
- [231] J. Qin, L. Wang, D. Yuan, P. Gao, and B. Zhang, “Chaos and bifurcations in periodic windows observed in plasmas,” *Physical review letters*, vol. 63, no. 2, p. 163, 1989.
- [232] T. Klinger, F. Greiner, A. Latten, A. Piel, T. Pierre, G. Bonhomme, C. A. Capeau, G. Bachet, and F. Doveil, “Nonlinear dynamics and chaos in gas discharge systems,” *Le Journal de Physique IV*, vol. 5, no. C6, pp. C6–131, 1995.
- [233] Y. Wang, Y. Zhang, D. Z. Wang, and M. G. Kong, “Period multiplication and chaotic phenomena in atmospheric dielectric-barrier glow discharges,” *Applied physics letters*, vol. 90, no. 7, p. 071501, 2007.
- [234] D. D. Šijačić, U. Ebert, and I. Rafatov, “Period doubling cascade in glow discharges: Local versus global differential conductivity,” *Physical Review E*, vol. 70, no. 5, p. 056220, 2004.
- [235] “Phelps database.” <https://www.lxcat.net/Phelps>. Retrieved 02 2022.
- [236] Y. Choi, J. Kim, and Y. Hwang, “One-dimensional discharge simulation of nitrogen dbd atmospheric pressure plasma,” *Thin solid films*, vol. 506, pp. 389–395, 2006.

- [237] M. Capitelli, C. Ferreira, B. Gordiets, and A. Osipov, *Plasma Kinetics in Atmospheric Gases*. Berlin: Springer, 2000.
- [238] D. D. Šijačić, U. Ebert, and I. Rafatov, “Oscillations in dc driven barrier discharges: Numerical solutions, stability analysis, and phase diagram,” *Physical Review E*, vol. 71, no. 6, p. 066402, 2005.
- [239] I. R. Rafatov, D. D. Šijačić, and U. Ebert, “Spatiotemporal patterns in a dc semiconductor-gas-discharge system: Stability analysis and full numerical solutions,” *Physical Review E*, vol. 76, no. 3, p. 036206, 2007.
- [240] Y. P. Raizer and M. Mokrov, “A simple physical model of hexagonal patterns in a townsend discharge with a semiconductor cathode,” *Journal of Physics D: Applied Physics*, vol. 43, no. 25, p. 255204, 2010.
- [241] M. Mokrov and Y. P. Raizer, “Simulation of current filamentation in a dc-driven planar gas discharge–semiconductor system,” *Journal of Physics D: Applied Physics*, vol. 44, no. 42, p. 425202, 2011.
- [242] M. J. Feigenbaum, “The onset spectrum of turbulence,” *Physics Letters A*, vol. 74, no. 6, pp. 375–378, 1979.
- [243] M. J. Feigenbaum, “Quantitative universality for a class of nonlinear transformations,” *Journal of statistical physics*, vol. 19, no. 1, pp. 25–52, 1978.
- [244] V. Melekhin and N. Y. Naumov, “Critical current measurement and dynamic theory of townsend discharge stability,” *Sov. Phys. Tech. Phys.*, vol. 29, no. 8, pp. 888–892, 1984.
- [245] L. M. Pecora and T. L. Carroll, “Driving systems with chaotic signals,” *Physical review A*, vol. 44, no. 4, p. 2374, 1991.
- [246] M. J. Ogorzalek, “Taming chaos. i. synchronization,” *IEEE Transactions on Circuits and Systems I: Fundamental Theory and Applications*, vol. 40, no. 10, pp. 693–699, 1993.
- [247] N. F. Rulkov, M. M. Sushchik, L. S. Tsimring, and H. D. Abarbanel, “Generalized synchronization of chaos in directionally coupled chaotic systems,” *Physical Review E*, vol. 51, no. 2, p. 980, 1995.

- [248] K. Pyragas, “Weak and strong synchronization of chaos,” *Physical Review E*, vol. 54, no. 5, p. R4508, 1996.
- [249] J. M. González-Miranda, *Synchronization and Control of Chaos*. London, UK: Imperial College Press, 2004.
- [250] F. Min and A. C. Luo, “Complex dynamics of projective synchronization of chua circuits with different scrolls,” *International Journal of Bifurcation and Chaos*, vol. 25, no. 05, p. 1530016, 2015.
- [251] F. Min and H. Ma, “The mechanism of switching combination synchronization for three distinct nonautonomous systems under sinusoidal constraints,” *Nonlinear Dynamics*, vol. 100, no. 1, pp. 475–492, 2020.
- [252] M. Akhmet and M. O. Fen, “Unpredictable points and chaos,” *Communications in Nonlinear Science and Numerical Simulation*, vol. 40, pp. 1–5, 2016.
- [253] M. Akhmet, M. O. Fen, and A. Tola, “The sequential test for chaos,” *arXiv preprint arXiv:1904.09127*, 2019.
- [254] M. Akhmet, *Domain Structured Dynamics: Unpredictability, Chaos, Randomness, Fractals, Differential Equations and Neural Networks*. Bristol, UK: IOP publishing, 2021.
- [255] V. Kolobov and L. Tsendin, “Analytic model of the cathode region of a short glow discharge in light gases,” *Physical Review A*, vol. 46, no. 12, p. 7837, 1992.
- [256] D. D. Šijačić, U. Ebert, and I. Rafatov, “Oscillations in dc driven barrier discharges: Numerical solutions, stability analysis, and phase diagram,” *Physical Review E*, vol. 71, no. 6, p. 066402, 2005.
- [257] X. Michalet, F. F. Pinaud, L. A. Bentolila, J. M. Tsay, S. Doose, J. J. Li, G. Sundaresan, A. Wu, S. Gambhir, and S. Weiss, “Quantum dots for live cells, in vivo imaging, and diagnostics,” *science*, vol. 307, no. 5709, pp. 538–544, 2005.
- [258] X. Pi, R. Liptak, J. D. Nowak, N. Wells, C. Carter, S. Campbell, and U. Kortshagen, “Air-stable full-visible-spectrum emission from silicon nanocrystals

- synthesized by an all-gas-phase plasma approach,” *Nanotechnology*, vol. 19, no. 24, p. 245603, 2008.
- [259] M. Meyyappan, L. Delzeit, A. Cassell, and D. Hash, “Carbon nanotube growth by pecvd: a review,” *Plasma sources science and technology*, vol. 12, no. 2, p. 205, 2003.
- [260] A. Stoffel, A. Kovacs, W. Kronast, and B. Müller, “Lpcvd against pecvd for micromechanical applications,” *Journal of Micromechanics and Microengineering*, vol. 6, no. 1, p. 1, 1996.
- [261] M. Moisan, J. Barbeau, S. Moreau, J. Pelletier, M. Tabrizian, and Y. L’H, “Low-temperature sterilization using gas plasmas: a review of the experiments and an analysis of the inactivation mechanisms,” *International journal of Pharmaceutics*, vol. 226, no. 1-2, pp. 1–21, 2001.
- [262] J. Schlegel, J. Köritzer, and V. Boxhammer, “Plasma in cancer treatment,” *Clinical Plasma Medicine*, vol. 1, no. 2, pp. 2–7, 2013.
- [263] V. Ivanov, O. Proshina, T. Rakhimova, A. Rakhimov, D. Herrebout, and A. Bogaerts, “Comparison of a one-dimensional particle-in-cell–monte carlo model and a one-dimensional fluid model for a $4\pi/\hbar^2$ capacitively coupled radio frequency discharge,” *Journal of applied physics*, vol. 91, no. 10, pp. 6296–6302, 2002.
- [264] T. Nitschke and D. Graves, “A comparison of particle in cell and fluid model simulations of low-pressure radio frequency discharges,” *Journal of applied physics*, vol. 76, no. 10, pp. 5646–5660, 1994.
- [265] N. Balcon, G. Hagelaar, and J. Boeuf, “Numerical model of an argon atmospheric pressure rf discharge,” *IEEE transactions on plasma science*, vol. 36, no. 5, pp. 2782–2787, 2008.
- [266] S. Rauf and M. J. Kushner, “Argon metastable densities in radio frequency ar, ar/o₂ and ar/cf₄ electrical discharges,” *Journal of applied physics*, vol. 82, no. 6, pp. 2805–2813, 1997.

- [267] Y. Zhang, W. Jiang, Q. Zhang, and A. Bogaerts, “Computational study of plasma sustainability in radio frequency micro-discharges,” *Journal of Applied Physics*, vol. 115, no. 19, p. 193301, 2014.
- [268] E. Group *et al.*, “Cfd-ace+ module manual.” <https://www.esi.com.au/software/cfd-ace/>, 2006.
- [269] C. Multiphysics and C. Module, “Comsol multiphysics user’s guide,” *Version: COMSOL Multiphysics*, vol. 3, 2014.
- [270] E. Kawamura, C. K. Birdsall, and V. Vahedi, “Physical and numerical methods of speeding up particle codes and paralleling as applied to rf discharges,” *Plasma Sources Science and Technology*, vol. 9, no. 3, p. 413, 2000.
- [271] T. Lafleur and R. Boswell, “Particle-in-cell simulations of hollow cathode enhanced capacitively coupled radio frequency discharges,” *Physics of Plasmas*, vol. 19, no. 2, p. 023508, 2012.
- [272] M. AK-BHD, “Wmo greenhouse gas bulletin,” *World Meteorological Organization: Geneva, Switzerland*, 2021.
- [273] A. Ozkan, T. Dufour, T. Silva, N. Britun, R. Snyders, A. Bogaerts, and F. Reniers, “The influence of power and frequency on the filamentary behavior of a flowing dbd—application to the splitting of co₂,” *Plasma Sources Science and Technology*, vol. 25, no. 2, p. 025013, 2016.
- [274] A. Ozkan, T. Dufour, T. Silva, N. Britun, R. Snyders, F. Reniers, and A. Bogaerts, “Dbd in burst mode: solution for more efficient co₂ conversion?,” *Plasma Sources Science and Technology*, vol. 25, no. 5, p. 055005, 2016.
- [275] S. Paulussen, B. Verheyde, X. Tu, C. De Bie, T. Martens, D. Petrovic, A. Bogaerts, and B. Sels, “Conversion of carbon dioxide to value-added chemicals in atmospheric pressure dielectric barrier discharges,” *Plasma Sources Science and Technology*, vol. 19, no. 3, p. 034015, 2010.
- [276] A. Ozkan, A. Bogaerts, and F. Reniers, “Routes to increase the conversion and the energy efficiency in the splitting of co₂ by a dielectric barrier discharge,” *Journal of Physics D: Applied Physics*, vol. 50, no. 8, p. 084004, 2017.

- [277] T. Holstein, "Energy distribution of electrons in high frequency gas discharges," *Physical Review*, vol. 70, no. 5-6, p. 367, 1946.
- [278] C. Zhou, C. Yuan, A. Kudryavtsev, T. Y. Katircioglu, I. Rafatov, and J. Yao, "Self-consistent treatment of gas heating in modeling of a coaxial dbd in atmospheric pressure co₂," *Plasma Sources Science and Technology*, 2023.





CURRICULUM VITAE

EDUCATION

■ Ph.D. in Physics **September 2018 - September 2023**
Middle East Technical University

- **Thesis Title:** Comprehensive Investigation of Plasma Behavior in Diverse Gas Discharge Phenomena: Fluid Modeling, Chaotic Dynamics, and Synchronization
- **Advisor:** Prof. Dr. İsmail Rafatov

■ M.Sc. in Physics **September 2015 - July 2018**
Middle East Technical University

- **Thesis Title:** Investigation of Nonlinear Oscillations in the Gas Discharge-Semiconductor System
- **Advisor:** Prof. Dr. İsmail Rafatov

■ B.Sc. in Physics **September 2009 - June 2015**
Middle East Technical University

- **Graduation Project:** Quantum Hall Effect
- **Advisor:** Prof. Dr. Seçkin Kürkçüoğlu

WORK EXPERIENCE

■ Teaching Assistant **December 2015 - November 2022**
Middle East Technical University

Physics I & II (First-year courses for physics students)

General Physics Laboratory I & II (First-year laboratory courses for physics students)

Introductory Electronics (Second year laboratory course for physics students)

Principles of Measurement and Instrument I (Fourth year laboratory course for physics students)

Mathematical Methods for Physicists (Graduate level course for physics students)

Statistical Mechanics (Graduate level course for physics students)

■ Engineer ASELSAN A.Ş. November 2022 - Ongoing

Navigation Systems Design Engineer

RESEARCH INTERESTS

- Numerical Modelling of Gas Discharge Plasma, Numerical Modelling of Heat Transfer Systems, Bifurcation and Chaos in Differential Equations, Chaos Synchronization in Nonlinear Systems, Inertial Measurement Unit (IMU) Sensors

AWARDS

- Performance award for the student with the highest GPA (May 2017)

PUBLICATIONS

- Rafatov, I., Yesil, C. (2018). Transition from homogeneous stationary to oscillating state in planar gas discharge–semiconductor system in nitrogen: Effect of fluid modelling approach. *Physics of Plasmas*, 25(8), 082107.
- Şahin Kürekçi, Gilda Afshari, Mehmet Tabak, Cihan Yesil and Emre Yüce. “Focusing light through scattering medium”. Poster presented at: National Workshop on Optics, Electro-optics and Photonics, September 2018, Bilkent University in Ankara (2018)

- Cınar, K., **Yesil, C.**, Bek, A. (2019). Revealing laser crystallization mechanism of silicon thin films via pulsed IR lasers. *The Journal of Physical Chemistry C*, 124(1), 976-985.
- **Yesil, C.**, Rafatov, İ. (2019, September). Transition to Chaos in Planar Gas Discharge-Semiconductor System in Nitrogen: Effect of Fluid Modelling Approach. In *BOOK OF ABSTRACTS* (p. 61).
- Yuan, C., **Yesil, C.**, Yao, J., Zhou, Z., Rafatov, I. (2020). Transition from periodic to chaotic oscillations in a planar gas discharge-semiconductor system. *Plasma Sources Science and Technology*, 29(6), 065009.
- Akhmet, M., Başkan, K., **Yesil, C.** (2022). Delta synchronization of Poincaré chaos in gas discharge-semiconductor systems. *Chaos: An Interdisciplinary Journal of Nonlinear Science*, 32(8), 083137.
- Akhmet, M., **Yesil, C.**, Başkan, K. (2023). Synchronization of chaos in semiconductor gas discharge model with local mean energy approximation. *Chaos, Solitons and Fractals*, 167, 113035.
- Akhmet, M., Başkan, K., **Yesil, C.** (2023). Revealing Chaos Synchronization Below the Threshold in Coupled Mackey–Glass Systems. *Mathematics*, 11(14), 3197.
- Rafatov, I., Islamov, G., Eylenceoglu, E., **Yesil, C.**, Bogdanov, E. (2023). Analysis of different modeling approaches for simulation of glow discharge in helium at atmospheric pressure. *Physics of Plasmas*, 30(9).

SCHOOLS, WORKSHOPS and CONFERENCES

- International School on Low Temperature Plasma Physics: Basics and Applications, Program of the PhD Class, October 2017, Physikzentrum Bad Honnef
- International School on Low Temperature Plasma Physics: Basics and Applications, Program of the Master Class, October 2017, Physikzentrum Bad Honnef
- National Workshop on Optics, Electro-optics and Photonics, September 2018, Bilkent University in Ankara

- BEYOND 2019, Computational Science and Engineering Conference, September 2019, Middle East Technical University in Ankara

TECHNICAL COMPUTATION, SIMULATION SKILLS

COMSOL MULTIPHYSICS, MATLAB, C++, Python, Octave, LTspice, L^AT_EX

