The Dynamics of the Plasma Boundary Sheath

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Abstract

The bombardment of substrates by energetic ions is a central element of plasma based surface modification such as plasma etching, deposition, and sputtering. For the technology computer-aided design of such processes, the calculation of the flux, energy and angular distributions of the surface incident particles is of great importance, as they determine simultaneously the rate, the anisotropy, and the selectivity of the plasma processes. In that respect, stochastic solutions of the kinetic equations, based on the Particle-in-Cell approach (PIC) complemented with Monte Carlo collisions calculation (MCC), provide a realistic and accurate calculations. However, they require large computational resources and are only feasible for simple discharge geometries. If the latter requirement is an issue, one possibility is the hybrid schemes which combine fluid and kinetic arguments in a clever way. They are computationally efficient, however, not fully self-consistent on the kinetic level.

Therefore, in this thesis, we present a mathematical model which enables the efficient, kinetically self-consistent simulation of DC and RF modulated plasma boundary sheaths in all technically relevant discharge regimes, namely, the algorithm “Ensemble-in-Spacetime” (EST). The model consists of a set of kinetic equations for ions, Boltzmann’s relation for electrons, and Poisson’s equation for the electrical field. Boundary conditions specify the ion flux at a point deep in the plasma bulk and a periodically modulated sheath voltage $V_{sh}(t)$ or sheath charge $Q(t)$. These boundary conditions could be imported from fluid models (such as, HPEM, nonPDPsim, Comsol, and CFD-ACE+), global simulations, or experiments. The equations are solved based on iterative scheme. Unlike PIC, EST does not follow the transient evolution until it reaches a ”converged” (= periodic) state, but rather seeks within the space of all periodic sheath states for a solution of the equations of motion. The iteration is started with the potential values
of a self-consistent fluid model and terminates when the updates become sufficiently small, i.e. when self-consistency is achieved. A subsequent post-processing determines important quantities, in particular the phase-resolved and phase-averaged values of the ion energy and angular distributions and the total energy flux at the electrode. A drastic reduction of the computational effort compared with PIC calculations is achieved.

When compared against experiments, PIC/MCC models, and analytical models, the EST model shows a good agreement. The model is applicable for virtually all important parameter regimes and has no practically relevant restrictions regarding the amplitude, waveform, and frequency of the applied RF, the number of the ion species, and the pressure and composition of the neutral background. The model is viable to be used as a post-processing tool.
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Introduction

1.1 Motivation and aim of study

A plasma is a quasineutral gas containing freely and randomly moving neutral and charged particles. Applying a negative potential to a substrate in contact with a plasma creates a depleted layer of electrons—termed "sheath"—above the substrate. The ions diffusing or drifting out of the plasma bulk are accelerated through the sheath toward the substrate. When the sheath potential is large compared to the thermal energy of ions in the plasma bulk and there are only few collisions within the sheath, most of the ions are directed normally to the substrate and the ions’ energies can be utilized to achieve specific material modifications. Plasma processing technology is crucial in many manufacturing industries, for example the surface treatment of physical components for the automotive, aerospace, steel, biomedical treatments, solar cells, and microelectronics industries. It enables unique ways for the material surface treatment. For instance, integrated circuits (ICs) consist of many layers of thin films of semiconductors, dielectrics, and conductors. Here, thin films are deposited and etched in order to form patterns of the order of a few tens nanometers, i.e., a hundred times smaller than a human hair.

The energy and angular distribution of ions striking an electrode and/or a substrate—which are often difficult to be measured in many applications—are of paramount importance for the development and optimization of plasma processes. Conventional capacitively coupled plasmas, driven by a single RF power source (1f-CCPs), offer little opportunity in that respect: They have only one adjustable ”knob”, namely the value of the RF power; the setting of
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**Figure 1.1:** A graph shows etch rates as a function of the ion energy for different species in a reactive ion beam etching system (using CF$_4$) (13).

**Figure 1.2:** Deposition rates of the prepared ZrO$_2$ films as a function of ion bombardment energy (14).
1.1 Motivation and aim of study

Figure 1.3: The hardness of the prepared ZrO$_2$ films as a function of ion bombardment energy (14).

1f-CCPs influences the plasma density and the ion energies alike. Dual and triple frequency sources (2f-CCPs, 3f-CCPs) and inductive-capacitive combinations offer more possibilities such as independent control of ion energy and ion flux (1, 2). However, recent investigations have shown that this separate control is limited due to coupling of frequencies (3, 4, 5, 6) and the effect of the secondary electrons (7, 8).

Fully arbitrary waveforms of the applied RF, finally, may even enable ”tailoring” of the ion energy distribution function (IEDF) (9, 10, 11, 12). The IEDF can be controlled via manipulation of the sheath voltage waveform. Instead of the traditional sinusoidal waveform, a bias waveform with specially tailored shape could be used. The tailored IEDF exhibits a narrower distribution than the sinusoidal case which is a broad and saddle-shaped distribution. Therefore ”tailoring” of the ion energy distribution function can be used to design specific IEDF shapes for particular purposes, such as,

- Etching selectivity: When ions have energies between the etching thresholds of two materials, infinite etching selectivity is possible. Figure 1.1 demonstrates the concept of etching selectivity. Bombarding the substrate with ions have energy between 125 – 150 eV achieves infinite etching rate of SiO$_2$ with respect to the etching rate of Si (13). In addition, it is important to report that controlling the ion energy is needed to facilitate proper etching and prevent damage of the substrate.

- Adjusting the deposition rates and the hardness of thin films (14): As shown in Figure 1.2 and Figure1.3, the maximum deposition rate and
the hardness of the SiO$_2$ thin films correspond to ion energy around 250 eV.

To fully exploit the flexibility of the new sources it is necessary to clearly understand how the amplitude and waveform of the applied RF affect the resulting ion energy distribution. Luckily, owing to a wealth of experimental and theoretical research conducted in the past, this is possible. Experimentalists employed retarding field analyzers (RFAs), energy resolving mass spectrometers (plasma monitors), and other devices to measure the ion energy distributions first at sinusoidally and later at arbitrarily biased substrates (11, 12, 15, 16, 17, 18, 19, 20, 21, 22). Theoreticians have studied the situation with analytical models (23, 24), hybrid models (25, 26, 27, 28), and completely self-consistent particle-in-cell (PIC) models (29, 30, 31). (For an extended review, see references (32) and (33).) All approaches have concurred that the energy distribution function of ions behind an RF modulated sheath is the result of both the acceleration by the time-varying field as well as of the collisions with the background neutrals. The field, in turn, is determined by the distribution of the charges. Mathematical models taking this self-consistency problem properly into account yield IEDFs that align with experimental results.

In spite of the vast amount of research that has been done in the field of plasma processing, researchers who actually endeavor to "tailor" ion energies will soon find that there is no tool available to calculate the IEDF for a given sheath voltage $V_{sh}(t)$ or sheath charge $Q(t)$ in a quick and accurate fashion. (That is, quick and accurate at the same time. There are of course models that evaluate efficiently; for instance, analytical models (23, 24) or hybrid codes (25, 26, 27, 28), but they are not self-consistent on the kinetic level and involve various restrictions. PIC codes which are fully self-consistent require large computational resources and are slow nonetheless (7, 29, 30, 33).) So it is highly required in the context of plasma technology to find a model that is computationally efficient and simultaneously provides reliable results, i.e., keeping the kinetic information. Moreover, the model should be applicable for virtually all important parameter regimes; it should has no practically relevant restrictions regarding the amplitude, waveform, the frequency of the applied RF, the number of the ion species, and the pressure and composition of the neutral background.

The mathematical foundation, the numerical implementation and coding, and the verification of such a model are the scope of this thesis. Furthermore, we explore the use of this model to study the ion transit effects in the intermediate radio-frequency regime and utilizes this model as a post-processing
tool to calculate the sheath dynamics, the ion energy distribution, and the ion angular distribution.

1.2 Outline of the dissertation

This dissertation is organized into the following sections: A literature review in chapter 2 will be presented to place briefly various fundamentals of plasma discharges. This summary will include background literature on plasma description, sheath theory, ion energy distribution, and ion angular distribution. Chapter 3 is the backbone of this thesis. It explains the basis and the mathematical foundations of our new model, termed, “Ensemble-In-Spacetime” (EST). The chapter ends with a comparison of the EST results and two analytical models to prove the validity of EST model. Next, in Chapter 4, a discussion of the effects of the ions’ inertia on the sheath dynamics will be given. The influence of the ion inertia on the temporal sheath dynamics will be discussed employing the EST model and a semi-analytical model. In Chapter 5 the results of the EST model are compared with PIC/MCC results and with experiments. In chapter 6, the effect of a magnetic field on the characteristics of capacitively coupled radio frequency discharges is demonstrated. It was found that geometrically and/or electrically symmetric discharges can be asymmetrized by applying a spatially inhomogeneous magnetic field. It is of interest to use EST model to calculate the IED in situations in which the measurements become difficult. One of these situations is the deposition of Alumina on thin films. The Alumina is insulator, so during the deposition process the Alumina will deposit on the collector of the Retarding Field Analyzer and consequently the collector current will drop to zero. Therefore, the EST is presented in chapter 7 as a part of a deposition experiment procedure –the experiment is carried out by M. Prenzel and A. von Keudell– to clarify the correlation between arbitrary RF-waveforms and the formation of crystalline Alumina phases. Finally, chapter 8 concludes the dissertation and presents some ideas for the future work.
1. INTRODUCTION
2

Fundamentals

2.1 Introduction

Plasma-based material processing technology aims to modify the chemical and physical properties of a surface. To achieve this goal, different plasma reactors have been developed. The ideal plasma source should enable uniform plasma over the substrate, stability during the whole processing period, and independent control of plasma processing parameters. Nonetheless, in reality some compromises are always necessary. The classical radio frequency reactors used in industry are capacitively coupled plasmas (CCPs), they simply consist of a vacuum chamber containing two planar electrodes separated by a gap of few centimeters and driven by an RF power in the frequency range of 1MHz to 200MHz. In geometrically asymmetric CCP discharges, a DC self bias is built up in front of the electrode with the smaller area. Therefore, ions bombarding the smaller area electrodes will have higher energies. For arbitrary electrodes, if $a$ and $b$ having the areas $A_a$ and $A_b$ and voltage drops $V_a$ and $V_b$, most experimental observations have typically found $V_a/V_b = (A_b/A_a)^c$ (34), where $c$ is determined by the plasma parameters. Unfortunately, the dependence of $c$ on plasma conditions is a great obstacle in achieving an independent control of the ion flux and the ion energy over the substrate. A reasonable level of the independent control of the ion flux and the ion energy has been carried out using a dual RF-frequency CCPs (1). The idea behind using two frequencies is that the lower frequency offers the ability to control the ion energy while the high frequency controls the ion flux. Furthermore, if the RF waveform applied to the plasma contains an even harmonic of the fundamental, then the
two sheaths will not be electrically symmetric. This asymmetry gives rise to a DC self bias. The DC self bias magnitude is maximized by using the fundamental frequency and the second harmonic and can be adjusted by the phase angle between the two frequencies (2). However, the separation between the ion flux and the ion energy is not always ideal (3, 4, 5, 6, 7, 8). The performance of conventional CCPs has been also improved by applying a magnetic field to increase the efficiency of power transfer from the RF source to the plasma by means of enhancing the plasma electron confinement (35). Thus, the discharge could be sustained even if the plasma pressure is under 1 Pa. The main drawback of magnetically enhanced RF CCPs is the inhomogeneity of the plasma over the substrate. However, the plasma inhomogeneity effects can be minimized by rotating the magnetic field in the plane of the substrate.

The limitations of capacitive RF discharges (i.e., capacitive RF discharges often operate at high sheath voltages with low ion flux and high ion-bombarding energy at a given power level) have led to the development of various low-pressure and high-density plasma discharges, for instance, electron cyclotron resonance (ECR), Helicon, inductive, and inductive/capacitive sources. The common feature of these high density sources is that the RF or microwave power is coupled to the plasma across a dielectric window or wall, rather than by direct connection to an electrode in the plasma as it would be for a capacitive discharge. To control the ion energy, the electrode on which the substrate is placed can be independently driven by a capacitively coupled RF source. Hence independent control of the ion/radical fluxes (through the source power) and the ion-bombarding energy (through the substrate electrode power) is possible.

2.2 Plasma modeling

2.2.1 Kinetic description

The most accurate description of a plasma is the kinetic description. (The word kinetic means “of or relating to motion”.) Thus this description of the plasma would be developed by adding up the behavior and effects of all the individual particles in a plasma. However, this is impracticable due to the huge number of particles in typical RF plasma reactors (of the order of $10^{10} - 10^{13} \text{ cm}^{-3}$) and the equation of motion of each particle must be solved self-consistently with the fields within the plasma. Furthermore, particles experience collisions, modify their velocities and energies on very short time scales. Therefore, an appropriate average must be considered. For a given
species and in a statistical sense, a distribution function \( f(r, v, t) \) in the six-dimensional phase space \((r,v)\) of particle positions and velocities could be introduced \((36)\). Where the six coordinates \((r, v)\) are independent variables and \( f(r, v, t) \, d^3r \, d^3v = \text{number of particles inside a six-dimensional phase space volume } d^3r d^3v \text{ at time } t \).

Distribution functions must obey a continuity equation that takes into account the probability of particles entering and leaving the plasma volume. The distribution functions should also include information about particles that can be produced by ionization or destroyed by recombination within this volume. The equation governing the evolution of the distribution is called the Boltzmann equation (or Vlasov equation when the collision term is vanished) and is given by

\[
\frac{\partial f}{\partial t} + \vec{v} \cdot \nabla_r f + \frac{\vec{F}}{m} \cdot \nabla_v f = \frac{\partial f}{\partial t} \bigg|_c, \tag{2.1}
\]

where \( \vec{F} = q[\vec{E} + \vec{v} \times \vec{B}] \) stands for the Lorenz force acting on the charged particles. \( q, \vec{E}, \) and \( \vec{B} \) are the particle charge, the electric field, and the magnetic field, respectively. To understand the full meaning of Boltzmann equation, let us assume an elementary volume in the phase space and write the total derivative of \( f \) with respect to time:

\[
\frac{df}{dt} = \frac{\partial f}{\partial t} + \frac{\partial f}{\partial r} \frac{\partial r}{\partial t} + \frac{\partial f}{\partial v} \frac{\partial v}{\partial t}, \tag{2.2}
\]

where \( \partial f/\partial t \) - the explicit dependence on time - gives the change of the distribution function at a fixed point in the elementary volume and termed the Eulerian derivative. However, this quantity will not be a measure of how the plasma distribution increases or decreases in the elementary volume. The reason is that the plasma is swept by a prevailing flow field, i.e., the mean velocity of all the individual particles. The total derivative \( df/dt \) can be interpreted as the rate of change as seen in the frame of moving particle, i.e., \( df/dt \) is the convective derivative (or the material derivative) in phase space and equal to zero in collisionless plasmas. The right hand side of 2.1 is a symbolic representation of collision processes and is known as the Boltzmann integral. It is a big challenge to set up a model for the collision term. For instance, the collision term of an elastic collision between an incident and target particles can be modeled based on the Stosszahlansatz as

\[
\left. \frac{\partial f_1}{\partial t} \right|_c = \int d^3v_2 \int_0^{2\pi} d\varphi_1 \int_0^\pi (f'_1 f'_2 - f_1 f_2) |v_1 - v_2| I \sin \vartheta_1 d\vartheta_1, \tag{2.3}
\]
in a laboratory system \((34)\). Here the incident and target particles have \( f_1 \) and \( f_2 \) distributions and velocities \( v_1 \) and \( v_2 \), respectively. And they are
scattered to velocities \( v_1' \) and \( v_2' \) in distributions \( f_1' \) and \( f_2' \), respectively. \( I(|v_1 - v_2|, \vartheta_1) \) is the differential cross section for scattering through angle \( \vartheta_1 \). \( \varphi \) is the azimuthal angle from 0 to \( 2\pi \). However, it is worth mentioning that it is not easy to evaluate the Boltzmann integral under the action of arbitrary forces. The Boltzmann equation is a non-linear and integro-differential equation. It is too difficult to solve it in a general way.

The complexity of getting a kinetic description to a plasma is decreased into a reasonable level using PIC simulation (37). Recently, the PIC approach becomes more and more attractive due to the availability of faster and cheaper workstations and GPUs. Nonetheless, PIC simulation is time consuming, particularly in the simulation of 2D and 3D discharges. The basic idea behind the PIC method is indeed to represent a large number of charged particles as a super-particle; each super-particle represents, for instance, \( 10^6 - 10^9 \) real particles. The super-particle is always multiply-charged and has the same charge-to-mass ratio as that of the actual particles. The large number of charges in a plasma are thus replaced by a much smaller number of these super-particles. Therefore, the simulation can be carried by solving the equation of motion \( \vec{F} = q(\vec{E} + \vec{v} \times \vec{B}) \) and Maxwell equation for the \( (\vec{E}, \vec{B}) \) fields generated the super-particles densities and currents \( (\rho, \vec{J}) \). However, the simulation must run with a sufficient number of super-particles in order to minimize the statistical noise, i.e. the number of super-particles/number of grid cells must be much greater than one.

The iteration cycle of a 1d3v PIC complemented with Monte Carlo Collisions is shown in Figure 2.1 (38). The cycle of conventional PIC is mainly consists of the following procedures:

---

**Figure 2.1:** Schematic of the computational PIC/MCC cycle, one time step.
1. Moving particles: Integration of the equations of motion to push all particles simultaneously using the same time-step.

2. Weighting: Interpolation of charge and current source terms to the field mesh.

3. Field: Computation of the fields on mesh points.

4. Weighting: Interpolation of the fields from the mesh to the particle locations.

Moreover, the loss or the gain of super-particles at the boundaries as well as the binary collisions are included. The binary collisions are random, thus a Monte Carlo (MC) method is used to include probable collisions. Let the probability that an particle collides with another particle in time $\Delta t$ is

$$P_c = \nu \Delta t,$$

where $\nu$ is the collision rate given by

$$\nu = N_g v \sigma_T.$$  \hspace{1cm} (2.5)

Here, $N_g$ is the number density of the plasma, $v$ is the speed of the particle, and $\sigma_T$ is the total collision cross section, which is primarily a function of the particle energy. Note that $\nu$ depends on time due to the dependence of particle speed on time. Let consider a collision in time interval $(0, t + \Delta t)$ and $P_{\text{null}}(t + \Delta t)$ represents the probability of no collision in this time interval. Then

$$P_{\text{null}}(t + \Delta t) = P_{\text{null}}(t) [1 - \nu(t) \Delta t],$$

where $[1 - \nu(t) \Delta t]$ is the probability of no collision in $(t, t + \Delta t)$. In the limit of $\Delta t \to 0$, the equation takes the solution (39):

$$P_{\text{null}}(t) = \exp[-\int_0^t \nu(t) dt].$$

Let $T_c$ be the time of collision and $f_c$ be its probability density function, then

$$\int_0^{t_c} f_c(t) dt = 1 - P_{\text{null}}(t_c) = R \text{ for } T_c < t_c,$$

where $R$ is a random number between 0 and 1.0. Once $R$ is known, $t_c$ can be determined utilizing 2.7. However, finding $t_c$ is time consuming where $\nu$ is time dependent. This difficulty can be overcome by introducing a constant
trial collision frequency \( \nu' \) which is greater than the actual collision frequency \( \nu(t) \) for all the velocities considered and at any \( t \) (40). If we replace \( \nu \) by \( \nu' \) in 2.7 we have

\[
t'_c = -\ln R/\nu'.
\] (2.9)

When a collision is considered at \( t = t'_c \), the collision probability for the rates \( \nu \) and \( \nu' \), respectively, read as

\[
P_c = 1 - \exp\left[ - \int_0^{t'_c} \nu(t) dt \right],
\] (2.10)

\[
(P_c)_{\text{max}} = 1 - \exp(-\nu't'_c).
\] (2.11)

The collision at \( t = t'_c \) is regarded as a real collision with probability of \( P_c/(P_c)_{\text{max}} \) and as a null collision with a probability of \( 1 - P_c/(P_c)_{\text{max}} \). This method is computationally efficient because of no need of finding \( t_c \) from 2.7. When the particle undergoes many events with a maximum number of \( K \), then the total cross section reads as

\[
\sigma_T = \sum_{k=0}^{K} \sigma_k.
\] (2.12)

A collisional event at time \( t'_c \) is chosen as follows: The probability of occurrence of a \( k^{th} \) event in the interval \( (0, t'_c) \) is

\[
P_k = 1 - \exp\left[ - \int_0^{t'_c} \nu_k(t) dt \right],
\] (2.13)

with total probability

\[
P_T = \sum_{k=1}^{K} P_k.
\] (2.14)

The event \( k \) should be sampled with a probability of \( P_k/P_T \) (39).

The computational saving can be quite significant if the sampling technique avoids the calculation of the collision probabilities 2.13 and 2.14. A schematic of an efficient sampling technique is shown in Figure 2.2 (38). Each particle is checked for the type of collision by a random number \( R \) between zero and one as follows:
2.2 Plasma modeling

Figure 2.2: The addition of the null collision process results in a constant collision frequency over all energies.

\[ R \leq \frac{\nu_1}{\nu'} \text{ the collision is type 1,} \quad (2.15) \]

\[ \frac{\nu_1}{\nu'} < R \leq \left(\frac{\nu_1 + \nu_2}{\nu'}\right) \text{ the collision is type 2,} \quad (2.16) \]

\[ \vdots \]

\[ \sum_{j=1}^{N} \frac{\nu_j}{\nu'} < R \text{ null collision.} \quad (2.17) \]

Once a collision occurs the scattering angle and energy of the particle after collision are determined based on the model assumed for that type of collision.

Finally, the time step and the grid spacing -in explicit PIC schemes- must be used precisely to minimize numerical instabilities (i.e., accuracy criterion) and ensure that most particles will not travel more than one cell per time-step and will sample accurate electric fields, such as the Courant condition (37). The time-step also should be chosen in such a way that the probability of having a particle collide more than once per time step is low.
2.2.2 Fluid description

In various applications, the macroscopic description (i.e., the collective description) of the plasma is required instead of the microscopic description which is too detailed and computationally complicated. Because the distribution function of plasma particles contains detailed information about the system, one can obtain the macroscopic parameters of the plasma by averaging the distribution over particle velocities, as for example:

1. Particle density: \( n(\vec{r}, t) = \int \int \int f(\vec{r}, \vec{v}, t) d^3v. \)

2. Flow velocity: \( \vec{u}(r, t) = (1/n(\vec{r}, t)) \int \int \int \vec{v} f(\vec{r}, \vec{v}, t) d^3v. \)

3. Pressure tensor component: \( \Pi_{ij}(\vec{r}, t) = m \int \int \int \vec{v}_i \vec{v}_j f(\vec{r}, \vec{v}, t) d^3v, \) where the subscripts \( i, j \) give the component direction. \( m \) is the particle mass.

4. Average kinetic energy \( \varepsilon(\vec{r}, t) = (1/n(\vec{r}, t)) \int \int \int \frac{m |\vec{v}|^2}{2} f(\vec{r}, \vec{v}, t) d^3v. \)

In the event the particle distribution is unknown and one does not want to pay the computational cost of the microscopic description, the macroscopic quantities could be calculated using the magnetohydrodynamic (i.e., fluid) equations. The fluid equations are simply moments of the Boltzmann equation 2.1 (34, 41, 42). To get the moment \( l \), simply multiply Boltzmann equation 2.1 with \( mv^l \) and then integrate over all particle velocities. The lowest moment (zero moment) is the continuity equation:

\[
\frac{\partial n}{\partial t} + \nabla . (n \vec{u}) = G - L, \quad (2.18)
\]

where \( G \) and \( L \) are the gain and loss term, respectively. These terms present the variation of the number of particles of a given species per unit volume and per unit time due to production or consumption of particles, respectively. The next moment is the equation of motion or the equation for the conservation of momentum:

\[
m n \left( \frac{\partial \vec{u}}{\partial t} + (\vec{u} \cdot \nabla) \vec{u} \right) = q n (E + \vec{u} \times B) - \nabla \cdot \Pi + \vec{S}_c. \quad (2.19)
\]

Of the two terms on the left, the first term \( \frac{\partial \vec{u}}{\partial t} \) is the acceleration term due to an explicitly time-varying \( \vec{u} \). The second term -inertial term- \( (\vec{u} \cdot \nabla) \vec{u} \) represents an acceleration due to a spatially varying fluid flow \( \vec{u} \). On the write hand side, \( \Pi \) stands for the pressure tensor. The divergence of the pressure tensor is simplified if the plasma is isotropic to a pressure gradient with \( \nabla \cdot \Pi = \nabla p \). The third term \( \vec{S}_c \) on the right hand side represents the
time rate of momentum transfer per unit volume due to collisions with other species. For a non-magnetized plasma, if we assume the collision rate of \( \nu \):

\[
m_n \left( \frac{\partial \vec{u}}{\partial t} + (\vec{u} \cdot \vec{V})\vec{u} \right) = q_n \vec{E} - \vec{V}p + mn\nu \vec{u}.
\] (2.20)

Equations 2.18 and 2.20 together do not form a closed set even with Maxwell equations, since the scaler pressure \( p \) is not determined. One can write an energy equation in the same way as momentum by multiplying Boltzmann equation by \( \frac{1}{2} m|\vec{v}|^2 \) and integrating over velocity. However, doing so would then contain an unknown term for the heat flux which is related to the particle random velocity. In general, the \( l^{th} \) moment equation contains a term which is a \( (l+1)^{th} \) moment. In order to get a sensible result this hierarchy must be truncated. The usual procedure is to use the equation of state which relates \( p \) to \( n \):

\[
p n^{-\gamma} = \text{const} \quad \text{or} \quad \frac{\Delta p}{p} = \gamma \frac{\Delta n}{n}.
\] (2.21)

\( \gamma \) is the ratio of the specific heat at constant pressure to that at constant volume. The value of \( \gamma \) to be taken depends on the heat flux assumption and on the isotropy of the energy distribution. If the plasma is isotropic (unmagnetized), then the isothermal approximation is \( \gamma = 1 \). For adiabatic situations when the fluid does not exchange energy with its surrounding, \( \gamma \) is given in terms of degrees of freedom \( N \);

\[
\gamma = \frac{2 + N}{N}.
\] (2.22)

Where for a monatomic gas with three degrees of freedom \( \gamma = 5/3 \), while a diatomic gas with five degree of freedom \( \gamma = 7/5 \).

It is worth reporting that the diffusion and transport of particles could be more simplified based on the plasma conditions. At sufficient high pressure and under the quasineutrality condition, the momentum equation would be read as

\[
n\vec{u} = \pm \mu n \vec{E} - D \vec{V} n,
\] (2.23)

where \( \mu = |q|/mv \) and \( D = T/mv \) are the particle mobility and the particle diffusion constants, respectively. If the particles are assumed to be inertialess (i.e, \( m \to 0 \)), then \( \mu \to \infty \) and \( D \to \infty \). This leads to Boltzmann relation.
2.2.3 Global description

The global models – which are space averaged fluid equations – are computationally efficient. The main advantage of utilizing the global models is that they can describe multi-species plasma efficiently. They usually solve the particle balance and species energy equation for the plasma chemistry, i.e., the temporal variation of species population and species energy \((43, 44, 45)\).

Lumped model circuits, a kind of global model, provide another technique to calculate the temporal plasma dynamics via elements as capacitors, inductors, and resistances \((46)\). An example of 2RF geometrically asymmetric CCP discharge in the high frequency regime (i.e., \(\omega_{pe} \gg \omega_{RF} \gg \omega_{pi}\)) is shown in Figure 2.3, \((47)\). Where \(\omega_{pe}\), \(\omega_{RF}\), and \(\omega_{pi}\) are the electron plasma frequency, the RF frequency, and the ion plasma frequency, respectively. The plasma sheath and the plasma bulk are subject to the same current and their voltages obey Kirchhoff’s voltage law. The RF current in the plasma sheath consists of three components in parallel: A displacement current is represented as a nonlinear capacitor, a constant ion current, and a time dependent electron current (represented as a diode). In the bulk, quasi-neutrality holds and consequently the RF current is mainly carried by electron conduction. The current-voltage characteristic of electrons in the bulk is determined by the momentum balance equation of electrons; electron dynamics could be represented as a resistance for ohmic and stochastic heating and inductance for electron inertia. Combining the sheath and the bulk voltages yields a nonlinear second order differential equation in the sheath charge \(Q(t)\). Different phenomena could be demonstrated in terms of the model outcome, for instance, electron plasma series resonance, the stochastic heating, and the energy dissipation over the RF period \((47, 48, 49, 50, 51, 52, 53, 54)\).

2.2.4 Hybrid description

Although the kinetic model describes the physics accurately, it is more complex (and in the case of numerical simulations, more computationally intensive) than the fluid model and global model. The hybrid model is a combination of kinetic models and fluid models or global models, treating some components of the system as a fluid or global, and others kinetically. The hybrid descriptions are supposed to be computationally efficient and to restore, somehow, part of the kinetic information of the system.
2.3 Plasma sheaths

The research in the plasma boundaries started roughly around the beginning of the last century. A wide variety of theories have been developed over the last ten decades. Nevertheless, the need of a plasma boundary sheath model – which is not restricted to the real plasma discharge conditions – is an obligatory demand for plasma based industry. In this section we will highlight in brief some fundamentals and cornerstones of the sheath theory. The sheath models distributed in the literature are manifold, crowded, and differ in entities. Of course, it is not possible to cover every detail concerning sheath theory in depth in this thesis. However, more physical and mathematical insights can be found in several reviews of the physics of the plasma boundary sheath (55, 56, 57, 58). The textbook of Lieberman and Lichtenberg (34) and the textbook of Chabert and Braithwait (41) are also instructive.

2.3.1 Direct current sheaths

Quasi-neutral plasmas are joined to absorbing walls across thin positively charged layers (i.e., sheaths). Due to the ion and electron fluxes directed outside the plasma, the walls will be biased negatively with respect to the plasma. The sheath potential will increase until the ion current balances the electron current. If we assume a constant ion flux and a Maxwellian electron...
where $T_e$, $m_i$, $m_e$ stand for the electron temperature, ion mass, and electron mass, respectively. The wall potential (i.e., floating potential) $\Phi_f$ is negative and depends linearly on the electron temperature with a factor depends on the ion mass. One could educe that, the plasma potential is always higher than the instantaneous wall potential ($59, 60$). This of course due to the high mobility of electrons, they will leave the plasma bulk to neutralize the electrode if the electrode is positively charged with respect to the plasma bulk. In order to maintain the continuity of ion flux a transition layer (namely, presheath) must be exist between the plasma bulk and the sheath giving rise to an ion velocity at the plasma-sheath edge as shown in Figure 2.4. The quasi-neutrality is breakdown when the ion speed becomes close to the Bohm speed, i.e., $u_i \rightarrow \sqrt{T_e/m_i}$ ($34, 41$). For the limiting case of a collisionless sheath, the presheath (having $u_i < \sqrt{T_e/m_i}$) matches the collisionless sheath (having $u_i > \sqrt{T_e/m_i}$) exactly at $u_i = \sqrt{T_e/m_i}$. This result is known as the Bohm sheath criterion where the ion speed must exceed the ion acoustic speed in order to ensure a stable sheath solution.

For collisional plasmas a unique edge position is not exactly defined. There is no agreed upon a specific definition of Bohm criterion when collisions are included. Riemann ($55$) and Franklin ($61$) argued that the definition of a collisionally defined Bohm criterion is not possible. On the contrary, Godyak ($62$), Valentini ($63$), and Chen ($64$) proposed different definitions of the collisionally modified Bohm criterion. In order to solve this conflict—the existence

\[ \Phi_f = \frac{-T_e}{e} \ln \left( \frac{m_i}{2\pi m_e} \right), \]
or non-existence of collisionally modified Bohm criterion—Brinkmann (65) investigated the plasma sheath transition for arbitrary levels of collisionality. He came to the conclusion that there is a collisionally modified Bohm criterion which is exactly the opposite of what Riemann and Franklin said. However, Brinkmann stated that there is no physical contradiction between the two views. Because in general, Bohm criterion is only present in asymptotic regimes and absent in the unabridged model. Moreover, the conflict is only about words and is a result of using different assumptions and entities, for more details see (65).

The dynamics of high voltage sheaths could be analytically described employing the matrix sheath model. In such cases, the electrons will be strongly repelled and Poisson’s equation can easily be integrated twice assuming a uniform ion density:

\[ \Phi(x) = -\frac{e n_i}{\epsilon_0} \left( \frac{x^2}{2} + c_1 x + c_2 \right), \]  \hspace{1cm} (2.25)

the constants \( c_1 \) and \( c_2 \) are determined by the sheath boundary conditions. The matrix sheath is the simplest space charge sheath model. It is not self-consistent since it does not account for the inhomogeneity of the ion density and neglect the electron space charge and the ion-neutral collisions in the sheath. Including the ion flow via the consideration of continuity and momentum equations of ions yields Child-Langmuir law (66, 67). In the collisionless regime Child-Langmuir law reads as

\[ \Phi(x) = -\left( \frac{3}{2} \right)^{4/3} \left( \frac{J_i}{\epsilon_0} \right)^{4/6} \left( \frac{2e}{m_i} \right)^{-1/3} (s - x)^{4/3}, \]  \hspace{1cm} (2.26)

while in the collisional regime (with ion mobility inversely proportional to the fluid speed, \( \mu_i \propto \lambda_i/|u| \)) it becomes

\[ \Phi(x) = -\frac{3}{5} \left( \frac{3}{2\epsilon_0} \right)^{2/3} \frac{J_i^{2/3}}{(2e\lambda_i/\pi m_i)^{1/3}} (s - x)^{5/3}. \]  \hspace{1cm} (2.27)

Where we have set \( \Phi(s) = \Phi'(s) = 0 \). Note that \( J_i \) is the ion current and \( \lambda_i \) is the mean free path. It worth noting that, in the full collisional regime where the ion mobility could be considered constant, Child-Langmuir law reads as

\[ \Phi(x) \propto -J_i^{1/2} (s - x)^{3/2}. \]  \hspace{1cm} (2.28)

Comparing 2.26, 2.27, and 2.28 one can conclude that they represent the same features but with different scalings. The different forms of Child-Langmuir
law covers collisionless and collisional regime. However, what remains unsatisfactory: The electrons are excluded, the assumption of zero ion speed at the sheath edge which causes a singularity at the sheath edge, and the neglection of the ambipolar electric field in the bulk.

2.3.2 Radio-frequency sheaths

The dynamics of RF sheaths in general is nonlinear dynamics. The relations constitute a system of nonlinear integro-differential equations for which no analytical solution exist. Consequently, some assumptions must be imposed to make the problem easy to be solved, i.e., simplifications which bypass certain mathematical difficulties. One of the very popular assumptions the so called “step model” which was introduced by Godyak (68). The step model has found in many subsequent studies (19, 69, 70, 71). The step model ignores thermal effects and assumes abrupt transition from electron depletion region to quasi-neutrality region. On the other hand Brinkmann described a sophisticated 1D-RF sheath model (65, 72, 73) where thermal effects are included and gradient of the sheath edge is not necessary sharp. To explore the differences between Brinkmann model and step models, later we will compare the Brinkmann model results with the step model proposed by Lieberman (69, 70).

In Brinkmann model, the electrons assumed to have Boltzmann relation, i.e., their diffusion pressure \( p_e = n_e T_e \) balances the electric force \( -e n_e E \). So the electric field inside the bulk can be written as

\[
E = \frac{T_e}{e n_e} \frac{\partial n_e}{\partial x}. \tag{2.29}
\]

In the high frequency limit (i.e., \( \omega_{RF} \gg \omega_i \), the RF frequency is greater than the ion plasma frequency) the ions inertia effects may be neglected and the ions assumed to respond to the time averaged electric field

\[
\bar{E}(x) = \frac{1}{T_{RF}} \int_0^{T_{RF}} E(x, t) dt. \tag{2.30}
\]

Furthermore, the ions are assumed to only experience elastic scattering and charge exchange. In this case, there is no ionization, recombination, excitation, or de-excitation in the sheath. The ion flux conservation can be written as

\[
n_i v_i = -\Psi_i, \tag{2.31}
\]

where \( \Psi_i \) is the ion flux in the sheath and the negative sign reflects that ions move opposite to \( x \) direction. Equation 2.31 tells that the flux of ions in the
sheath is constant. The momentum conservation equation for ions is simplified by assuming that the ions only respond to the time averaged electric field. The simplified form of the ion momentum conservation equation becomes:

\[ v_i \frac{\partial v_i}{\partial x} = \frac{e}{m_i} \vec{E}(x) - \frac{T_i}{m_i} \frac{1}{n_i} \frac{\partial n_i}{\partial x} - \nu_i(v_i)v_i, \quad (2.32) \]

where \( \nu_i(v_i)v_i \) represents the momentum transfer due to collisions. The instantaneous electric field is related to the charge density via Poisson’s equation,

\[ \epsilon_0 \frac{\partial E}{\partial x} = e[n_i - n_e]. \quad (2.33) \]

The RF modulation is introduced by setting the total RF current and the position of the electrode is defined by the equality of the ion flux and the time averaged electron flux.

Under the same conditions (a 47 m Torr Argon discharge with \( \omega_{RF} = 2\pi \times 13.56 \text{ MHz} \), ion Flux of 0.5mA/cm\(^2\), RF current of 10.8mA/cm\(^2\), electron temperature of 3 eV), the sheath dynamics is solved using Brinkmann model and Lieberman step model \((73)\). Figure 2.5 shows a comparison between the ion and the electron density calculated by the two models. Within the sheath, the models provide a good agreement. However, Lieberman model provides a much steeper transition from the electron depletion to quasi-neutrality than

![Figure 2.5: A comparison between the calculated particle density within an RF sheath using Brinkmann and Lieberman models. The black line presents Lieberman step model results, while the gray presents Brinkmann model results (73).](image)
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Figure 2.6: A comparison between the calculated field within an RF sheath using Brinkmann and Lieberman models. The black line presents Lieberman step model results, while the gray presents Brinkmann model results (73).

...the gradient of the ion density. Moreover, the assumption of an electric field equals to zero in the plasma bulk causes a singularity at the sheath edge, i.e., the electron and ion density diverge to infinity. One possible fix to avoid this singularity at the sheath edge was proposed by Godyak and Sternberg (71) by introducing an ad-hoc boundary condition for the electrical field at the sheath edge and within the bulk (|E| = T_e/e\lambda_D). This assumption removes the divergence of the plasma density at the sheath edge, but the assumed electric field value in the bulk is spatially constant (this is not realistic) and is too large compared to the ambipolar field given by 2.29. Figure 2.6 shows a comparison between the calculated electric field by Lieberman model and Brinkmann model. The models agree on the spatial distribution of the electric field within the sheath. The discrepancy again is a result of neglecting the thermal effects in the bulk. Therefore, as a simple conclusion, the sheath cannot be self-consistently matched to the quasi-neutral region using the step model approximation. On the contrary, Brinkmann model treats the sheath and the presheath in a unified way.

The self-consistency of the solution of 2.29-2.33 is an important and difficult issue. It implies that the densities and the fields are calculated from each others as shown in Figure 2.7. Starting from a known average electric field and assuming that the ions are affected only by the average electric field, then the ion density as a function of position can be calculated. Next, using Poisson’s equation, Boltzmann’s relation, and the applied voltage, one could calculate the time dependent electric field. Then the average electric field could be calculated. The advanced algebraic approximation model (AAA) is a sheath...
2.3 Plasma sheaths

Figure 2.7: Structure of the Brinkmann RF sheath model (73).

The model breaks this loop –Figure 2.7– by making several transformations and approximating the solution with a perturbation analysis. The AAA model enables an accurate (the output of the AAA model has an excellent agreement with the exact solution of the set 2.29-2.33) and provides a computationally fast simulation of DC and RF plasma sheaths. Several investigations have verified the ideas behind the AAA model and have been used for practical work (26, 74, 75, 76, 77, 78, 79). A mathematically rigorous description of the model has been presented by Brinkmann (65, 72, 73). However, an outline of the AAA model is presented at a level necessary for understanding the basis of the model and how it functions. Although the author did not participate in the developing of the AAA model, he wrote C++ code to solve it for an initial guess solution for EST model. If the electrons are in mechanical equilibrium with the electric field, Boltzmann-Poisson equation can be written as

\[-\varepsilon_0 \frac{\partial^2 \Phi}{\partial x^2} = e [n_i(x) - \hat{n} \exp(\frac{e\Phi}{T_e})], \tag{2.34}\]

where \(\hat{n}\) is a reference density. The ion density \(n_i\) is assumed to be a given function, defined for all values of \(x\), differentiable, positive, and terminated at the positive \(x_E\) of the sheath-inducing electrode or surface, and reaches a maximum somewhere in the bulk. Certain auxiliary conditions are also imposed: In the limit \(x \to \infty\), the density is assumed to grow slower than exponentially, so that

\[\lim_{x \to \infty} \frac{1}{n_i} \frac{\partial n_i}{\partial x} = 0. \tag{2.35}\]

For negative \(x\), it is assumed to fall off slowly so that the integral diverges

\[\lim_{x \to -\infty} n_i(x')dx' = \infty. \tag{2.36}\]
The electron and ion density related to the sheath solution of Boltzmann-Poisson equation is depicted schematically in Figure 2.8. They are monotonically increasing with $x$, and show three distinctive zones, namely electron depletion in the unipolar sheath (zone I), quasi-neutrality in the ambipolar region (zone III), and a rapid transition between the two regimes in the transition zone II (which has an extension of only a few Debye lengths). Mathematically, this behavior is captured by

\[
\lim_{x \to -\infty} \hat{n} \exp\left[\frac{\epsilon \Phi}{T_e}\right] = 0, \quad (2.37)
\]

\[
\lim_{x \to \infty} \hat{n} \exp\left[\frac{\epsilon \Phi}{T_e}\right] - n_i(x) = 0. \quad (2.38)
\]

The equations (2.37) and (2.38) work as a boundary conditions. But to specify certain solution another degree of freedom is required to locate the formal electron edge at a given point. The “formal electron edge” is defined as the point $s$ where the total electron charge in the interval $(-\infty, s)$ equals the positive charge excess in $(s, \infty)$,

\[
\int_{-\infty}^{s} \hat{n} \exp\left(\frac{\epsilon \Phi}{T_e}\right) dx = \int_{s}^{\infty} [n_i(x) - \hat{n} \exp\left(\frac{\epsilon \Phi}{T_e}\right)] dx. \quad (2.39)
\]

This definition for electron edge is different from the "sheath edge" defined in terms of the Bohm criterion, also this definition is valid even when the transition from depletion to quasi-neutrality is not sharp. From the mathematical nature of Boltzmann-Poisson equations, it is a class of elliptical differential equations for which extensive analysis can be found in the literature. It is well known that the solution of the Boltzmann-Poisson equation exist and are unique, but can generally not be expressed in analytical form. A series of approximations to Poisson’s equation could be derived employing a matched asymptotic expansion around the formal sheath edge $s(t)$ (72). The electrical potential related to the approximate solution reads

\[
\Phi(x, s) = \begin{cases} 
\frac{e}{\epsilon_0} \int_{x}^{s} (x - x') n_i(x') dx' \\
+ T_e \left( \ln \frac{n_i(s)}{\bar{n}} - 1 + d_1 \frac{\lambda_D(s)}{n_i(s)} \frac{\partial n_i(s)}{\partial s} \right) \\
+ \Delta \Phi, \\
\frac{T_e}{e} \ln \frac{n_i(x)}{\bar{n}} \\
\end{cases} \quad : x < s(t) \\
\hspace{1cm} + \Delta \Phi, \\
\hspace{1cm} : x > s(t) \\
\end{cases} \quad (2.40)
\]
Figure 2.8: Sketch of the density of the plasma sheath.

where $\Delta \Phi$ is given by

$$\Delta \Phi = \frac{T_e}{e} \Delta \Psi_0 \left( \frac{x - s}{\lambda_D(s)} \right) + \frac{T_e \lambda_D(s)}{e n_i(s)} \frac{\partial n_i(s)}{\partial s} \Delta \Psi_1 \left( \frac{x - s}{\lambda_D(s)} \right). \tag{2.41}$$

While the corresponding field is

$$E(x, s) = \begin{cases} -\frac{e}{\epsilon_0} \int_x^s n_i(x') \, dx & : x < s(t) \\ -\frac{T_e}{e n_i(s)} \frac{\partial n_i}{\partial x} & : x > s(t) \end{cases} + \Delta E, \tag{2.42}$$

where $\Delta E$ is given by

$$\Delta E = -\frac{T_e}{e \lambda_D(s)} \Delta \Psi_0' \left( \frac{x - s}{\lambda_D(s)} \right) - \frac{T_e}{e n_i(s)} \frac{\partial n_i(s)}{\partial s} \Delta \Psi_1' \left( \frac{x - s}{\lambda_D(s)} \right). \tag{2.43}$$

Here $\lambda_D(s)$ is the Debye length and is defined as $\lambda_D(s) = \sqrt{\epsilon_0 T_e/e^2 n_i(s)}$. The functions $\Delta \Psi_0$, $\Delta \Psi_1$, $\Delta \Psi_0'$ and $\Delta \Psi_1'$ are derived during the asymptotic expansion and perform transition in equations 2.40 and 2.42. $\Delta \Psi_0'$ and $\Delta \Psi_1'$ are the piecewise derivatives of $\Delta \Psi_0$ and $\Delta \Psi_1$, with the discontinuities at $\xi = 0$ not considered, and can be defined as:

$$\Delta \Psi_0' \left( \xi \right) = \begin{cases} \Psi_0'(\xi) + \xi & : \xi \ll 0, \\ \Psi_0'(\xi) & : \xi \gg 0, \end{cases} \tag{2.44}$$

$$\Delta \Psi_1 \left( \xi \right) = \begin{cases} \Psi_1'(\xi) + \frac{1}{2} \xi^2 & : \xi \ll 0, \\ \Psi_1'(\xi) - 1 & : \xi \gg 0, \end{cases} \tag{2.45}$$

where $\xi = \frac{x - s}{\lambda_D(s)}$. 
The characteristic functions $\Psi_0$ and $\Psi_1$ are the unique solutions of the following differential equations and asymptotic boundary conditions, where $d_0 = -1$ and $d_1 \approx 0.388$ are nonlinear eigenvalues which ensure the prescribed polynomial behavior for large values of $|\xi|:

\[ -\frac{\partial^2 \Psi_0}{\partial \xi^2} + \exp(\Psi_0) = 1, \] \quad (2.46)

with

\[ \Psi_0 \rightarrow \begin{cases} d_0 - \frac{1}{2} \xi^2 & : \xi \ll 0, \\ 0 & : \xi \gg 0, \end{cases} \] \quad (2.47)

and

\[ -\frac{\partial^2 \Psi_1}{\partial \xi^2} + \exp(\Psi_0)\Psi_1 = \xi, \] \quad (2.48)

with

\[ \Psi_1 \rightarrow \begin{cases} d_1 - \frac{1}{6} \xi^3 & : \xi \ll 0, \\ \xi & : \xi \gg 0. \end{cases} \] \quad (2.49)

The functions $\Psi_0$ and $\Psi_1$ are shown in Figure 2.9 and $\Psi'_0$ and $\Psi'_1$ are displayed in Figure 2.10.

The representation of the electric field using the spatial functions in 2.42 is not suitable, because the relation is not local. The electric field at a point
2.3 Plasma sheaths

Figure 2.10: The spatial functions $\Psi_0$ and $\Psi_1$ as a function of $\xi$.

Figure 2.11: The spatial functions $\Delta \Psi_0$ and $\Delta \Psi_1$ as a function of $\xi$. 
$x$ is not only a function of the value of $x$ itself, but it also depends on the position of the sheath edge $s$ and on the yet unknown ion density $n_i(x)$ in the interval in between. So a transformation from the spatial space $x$ and $s$ to the charge coordinates $q$ and $Q$ should be done. Here

$$q(x) = \int_0^x n_i(x) dx, \quad (2.50)$$

and

$$Q(t) = \int_0^{s(t)} n_i(x) dx. \quad (2.51)$$

Both the depletion field and the ambipolar field can be transformed without any problems. The term $\frac{\partial n_i}{\partial x}$ can be transformed as

$$\frac{\partial n_i}{\partial x} = \frac{\partial n_i}{\partial q} \frac{\partial q}{\partial x} \Rightarrow \frac{1}{n_i(x)} \frac{\partial n_i(x)}{\partial x} = \frac{\partial n_i(q)}{\partial q}, \quad (2.52)$$

similarly

$$\frac{\partial n_i(s)}{\partial s} = \frac{\partial n_i(s)}{\partial Q} \frac{\partial Q}{\partial s} \Rightarrow \frac{1}{n_i(s)} \frac{\partial n_i(s)}{\partial s} = \frac{\partial n_i(Q)}{\partial Q}. \quad (2.53)$$

The next step is to transform the argument of the special functions $\Delta \Psi'_0$ and $\Delta \Psi'_1$ into $q$ coordinates. The $\Delta \Psi'(\xi)$ vanishes quickly for large $|\xi|$, so that all functions of $s = s(Q)$ may be represented by a Taylor expansion around the point $Q = q$. The argument of the field function 2.42 becomes

$$\frac{x - s}{\lambda_D(s)} = \sqrt{\frac{e^2}{\varepsilon_0 T_e} \frac{q - Q}{\sqrt{n_i(q)}}} + O((Q - q)^3), \quad (2.54)$$

where the quadratic term in the expansion is not neglected but vanishes. The Taylor expansion of the pre-factors reads

$$\frac{1}{\lambda_D(s)} = \sqrt{\frac{e^2}{\varepsilon_0 T_e}} \left( \frac{1}{\sqrt{n_i(q)}} - \frac{1}{2 \sqrt{n_i(q)}} \frac{\partial n_i}{\partial q} (q - Q) + O((Q - q)^2) \right). \quad (2.55)$$

Collecting the results, the approximate expression of the field is

$$E(q, Q) = \begin{cases} \frac{e}{\varepsilon_0} (q - Q) & : q < Q, \\ + \Delta E q & \\ - \frac{T_e}{e} \frac{\partial n_i(q)}{\partial q} & : q > Q, \end{cases} \quad (2.56)$$
where $\Delta Eq$ is given by

$$
\Delta Eq = \frac{T_e}{e} \sqrt{\frac{\varepsilon^2}{\varepsilon_0 T_e}} \left( \sqrt{n_i(q)} - \frac{(q - Q) \partial n_i(q)}{2 \sqrt{n_i(q)}} \right) \Delta \Psi'_0 \left( \sqrt{\frac{\varepsilon^2}{\varepsilon_0 T_e}} \frac{q - Q}{\sqrt{n_i(q)}} \right)
\quad - \frac{T_e}{e} \frac{\partial n_i(q)}{\partial q} \Delta \Psi'_1 \left( \sqrt{\frac{\varepsilon^2}{\varepsilon_0 T_e}} \frac{q - Q}{\sqrt{n_i(q)}} \right). \tag{2.57}
$$

The time averaged electric field is calculated by the time averaging of 2.56. Therefore, this equation is re-written in a simplified and more compact form. For this transformation, the original definition $\Delta \Psi'_0$ and $\Delta \Psi'_1$, 2.44 and 2.45, in terms of $\Psi'_0$ and $\Psi'_1$ are first substituted back into 2.56. Note that the cases $q < Q$ and $q > Q$ lead to the same expression. The electrical field $E$ can be expressed as an algebraic function of its arguments:

$$
E(q, Q) = -\frac{T_e}{e \lambda_D(q)} \Xi_0 \left( \sqrt{\frac{\varepsilon^2}{\varepsilon_0 T_e}} \frac{q - Q}{\sqrt{n_i(q)}} \right)
\quad - \frac{T_e}{e} \frac{\partial n_i(q)}{\partial q} \Xi_1 \left( \sqrt{\frac{\varepsilon^2}{\varepsilon_0 T_e}} \frac{q - Q}{\sqrt{n_i(q)}} \right), \tag{2.58}
$$

where $\lambda_D(q) = \sqrt{\varepsilon_0 T_e / e^2 n_i(q)}$, $\Xi_0$ and $\Xi_1$ can be defined as

$$
\Xi_0(\xi) = \Psi'_0(\xi), \tag{2.59}
$$

$$
\Xi_1(\xi) = \Psi'_1(\xi) - \frac{\xi}{2} \Psi'_0(\xi). \tag{2.60}
$$

The given expression of the electric field by 2.58 has a number of interesting features. It is local, i.e. the field at the point $q$ depends only on the ion density and its derivative at that point, the sheath charge state $Q$ appears as a parameter. It is quasi-linear, i.e. the dependence on the derivative is linear, with a prefactor that is a function of the density. The explicit dependence on the charge coordinate $q$ and the sheath charge $Q$ appears as the difference $q - Q$; this ensures translation invariance with respect to the reference point $s$. The functions $\Xi_0$ and $\Xi_1$ are shown in Figure 2.12. They are functions that switch the calculation of the electric field between two different regions. The first being the unipolar region, where there are no electrons,
The special functions \( \Xi_0 \) and \( \Xi_1 \).

\( \Xi_0(\xi) \rightarrow -\xi \), and \( \Xi_1(\xi) \rightarrow 0 \). The second is the ambipolar region, where the field is due to ambipolar diffusion, \( \Xi_0(\xi) \rightarrow 0 \) and \( \Xi(\xi) \rightarrow 1 \). The transition in between, the field is reasonable but not exact. The model error, the absolute deviation between the approximate and the exact field, scales with \( n_i'^2/\sqrt{n_i} \) and \( \sqrt{n_i}n''_i \). In the depletion zone, the model error vanishes exponentially, in the quasineutral zone it behaves like \( n_i n'''_i \) which is the error of the ambipolar field expression itself. The time averaged electric field can be calculated with the following integral:

\[
\bar{E}(q, n_i, n'_i, \{Q\}) = \frac{1}{T_{RF}} \int_0^{T_{RF}} E(q, Q(t)) \, dt. \tag{2.61}
\]

Hence,

\[
\bar{E}(q, n_i, n'_i, \{Q\}) = -\frac{T_e}{e\lambda_D(q)} \frac{1}{T_{RF}} \int_0^{T_{RF}} \Xi_0 \left( \sqrt{\frac{e^2}{\epsilon_0 T_e} \frac{q - Q}{\sqrt{n_i(q)}}} \right) \, dt

- \frac{T_e}{e} \frac{\partial n_i(q)}{\partial q} \frac{1}{T_{RF}} \int_0^{T_{RF}} \Xi_1 \left( \sqrt{\frac{e^2}{\epsilon_0 T_e} \frac{q - Q}{\sqrt{n_i(q)}}} \right) \, dt. \tag{2.62}
\]

Unfortunately, it is not possible to reconstruct an algebraic representation with similar properties for the electrical potential. The potential in the depletion region behaves like \( \int_s^x (x' - x)n_i(x') \, dx' \) which in the charge coordinates will be \( \int_Q^q \int_q^{q'} \frac{1}{n_i(q'')} dq'' \, dq' \). So the potential is not local but depends explicitly on the ion density between \( q \) and \( Q \). But this trick can be avoided by calculating the potential after the calculation of the ion density.
In $q$ coordinates, the first moment equation assuming high frequency regime and no sources (i.e., ionization) or sinks (i.e., recombination) could be written as

$$ n_i(q)v_i(q) = -\Psi_i. \quad (2.63) $$

Using the ion velocity $v_i(q) = \frac{-\Psi_i}{n_i(q)}$, its first derivative $\frac{\partial v_i(q)}{\partial q} = \frac{\Psi_i}{n_i^2(q)} \frac{\partial n_i(q)}{\partial q}$, and the transformation $\frac{\partial}{\partial x} = n_i(q) \frac{\partial}{\partial q}$, the second moment equation can be written as

$$ -\frac{\Psi_i^2}{n_i^2(q)} \frac{\partial n_i(q)}{\partial q} = \frac{Z_i e \bar{E}(q, Q)}{m_i} + \frac{\Psi_i^2}{\lambda n_i^2(q)}, \quad (2.64) $$

with a mean free path $\lambda$.

$$ L(q, n_i) \frac{\partial n_i}{\partial q} = R(q, n_i), \quad (2.65) $$

where

$$ L(q, n_i, Q) = \frac{-\Psi_i^2}{n_i^2(q)} + \frac{Z_i T_e}{m_i} \frac{1}{T_{RF}} \int_0^{T_{RF}} \Xi_1 \left( \sqrt{\frac{e^2}{\varepsilon_0 T_e} \frac{q - Q}{\sqrt{n_i(q)}}} \right) dt, \quad (2.66) $$

and

$$ R(q, n_i, Q) = \frac{\Psi_i^2}{\lambda n_i^2} - \frac{Z_i T_e}{m_i \lambda_D} \frac{1}{T_{RF}} \int_0^{T_{RF}} \Xi_0 \left( \sqrt{\frac{e^2}{\varepsilon_0 T_e} \frac{q - Q}{\sqrt{n_i(q)}}} \right) dt. \quad (2.67) $$
Figure 2.14: A comparison between the calculated particle density within an RF sheath using the exact solution of Brinkmann model and the AAA model. The gray lines show the corresponding densities of the exact numerical solution (73).

To solve a first order differential equation, a starting point needs to be given. Luckily, the equation 2.65 has a critical point (removable singularity at \((q^*, n_i^*)\)) when

\[ L(q^*, n_i^*, Q) = 0, \quad R(q^*, n_i^*, Q) = 0. \] (2.68)

The critical point serves the same function in determining the stable solution to the ion density in the sheath as the Bohm criterion does. A topology for an RF sheath is shown in Figure 2.13. The critical point is presented as a blue point at the intersection of \(L = 0\) (orange, dotted) and \(R = 0\) (orange, dashed). Based on the boundary conditions at the critical point there are two solutions, both are monotonic. One of them increases in the positive direction of the charge coordinate. This solution is physically accepted and is shown as a solid blue line in Figure 2.13. The second, is unphysical solution, is shown as a dotted blue line in Figure 2.13. After the calculation of the ion density as a function of charge coordinate, the potential could be also calculated. Finally a transformation back into \(x\) coordinate is done.

To demonstrate the benefits of the AAA model, let evaluated the AAA model for the set of parameters used in Figure 2.5 and Figure 2.6. The result of the calculations is shown in Figure 2.14. The agreement of the exact solution of Brinkmann model and the AAA model is excellent. The absolute deviations are smaller than the thickness of the curves; the relative deviations (shown in the inset) are localized and never larger than a few per cent. In addition, the AAA model is computationally efficient. Therefore, the AAA model can
be used as a basis of a computationally efficient self-consistent model of the plasma boundary sheath which treats the sheath and the presheath in a unified way.

Despite of these advantages of Brinkmann model— the exact and the AAA solutions—one should not forget that the model is restricted to the high RF frequency regime. Thus one of the aims behind this thesis is to extend Brinkmann model to include the intermediate and the low RF frequency regimes. More details to come later.

### 2.4 Ion energy distribution and ion angular distribution

The calculations of the ion energy distribution rely frequently on numerical methods due to the complexity of RF sheath dynamics as numerical integration of the equation of motion (18, 19, 46, 80, 81, 82), Monte Carlo simulations (83, 84, 85, 86), and particle-in-cell (PIC) methods (29, 87, 88). However, approximate analytical models are present but mostly restricted to collisionless plasma (23, 89, 90). The critical parameter that controls the ion dynamics in the collisionless RF sheaths is the ratio \( \tau_{\text{ion}}/\tau_{\text{RF}} \), where \( \tau_{\text{ion}} \) is the ion transit time and \( \tau_{\text{RF}} \) is the periodic RF time.

In the high frequency regime \( \tau_{\text{ion}}/\tau_{\text{rf}} \gg 1 \), the ions experience many field oscillations across the sheath. The ions will then respond to the time-averaged potential and the phase at which they enter the sheath becomes unimportant. Based on the assumption of a collisionless Child-Langmuir space charge sheath (i.e., \( V_{\text{sh}}(x) \propto x^{4/3} \)) and neglecting the ion inertia, the ion transit time has been estimated as

\[
\tau_{\text{ion}} = 3 \bar{s} \sqrt{\frac{m_i}{2eV_{\text{sh}}}}, \tag{2.69}
\]

where \( \bar{s} \) is the sheath thickness averaged over the RF period, \( m_i \) is the ion mass, and \( V_{\text{sh}} \) is the averaged sheath potential. Consequently,

\[
\frac{\tau_{\text{ion}}}{\tau_{\text{RF}}} = \frac{3 \bar{s} \omega_{\text{RF}}}{2\pi} \sqrt{\frac{m_i}{2eV_{\text{sh}}}}. \tag{2.70}
\]

It is interesting to study the relation between the ion transit frequency \( \omega_{\text{ion}} = 2\pi/\tau_{\text{ion}} \) and the ion plasma frequency \( \omega_{\text{pi}} = \sqrt{e^2n_i/\epsilon_0m_i} \). From the definition of the ion transit frequency and the ion plasma frequency one can say that

\[
\frac{\omega_{\text{pi}}}{\omega_{\text{ion}}} = \frac{3 \bar{s}}{2\pi} \left( \frac{n_0e}{2\epsilon_0V_{\text{sh}}} \right)^{1/2}. \tag{2.71}
\]
The ratio $\frac{\omega_{pi}}{\omega_{\text{ion}}}$ as a function of the sheath potential is shown in Figure 2.15. Where $s = 1$ mm and $n_i = 10^{11}$ cm$^{-3}$. It is clearly seen that the ion plasma frequency is comparable to the ion transit frequency. Thus it is not surprising that $\omega_{pi}$ and $\omega_{\text{ion}}$ are used interchangeably in the literature (32).

The bimodal structure is due to ions which have crossed the sheath without collisions. However, with increasing pressures, the bimodal shape successively disappears and the average ion energy decreases significantly.

Benoit-Cattin and Bernard (23) came up with an analytical expression for a bimodal IED with two peaks symmetric about $e\bar{V}_{sh}$:

$$g(\varepsilon) = \frac{2n_t}{\omega_{RF}\Delta\varepsilon} \left( 1 - \frac{4}{\Delta\varepsilon^2}(\varepsilon - e\bar{V}_{sh})^2 \right)^{-1/2}, \quad (2.72)$$

where $n_t$ is the number of ions entering the sheath per unit time. The width of bimodal shape of the ion energy distribution $\Delta\varepsilon$ is given by

$$\Delta\varepsilon = \frac{4e\bar{V}_{sh}}{\pi} \frac{\tau_{RF}}{\tau_{\text{ion}}}. \quad (2.73)$$

The calculations of 2.72 and 2.73 are carried out assuming a collisionless high frequency regime, constant sheath width, zero initial ion velocity at the plasma sheath boundary, and sinusoidal sheath bias $V_{sh}(t) = \bar{V}_{sh} + \bar{V}_{sh} \sin \omega_{RF}t$. Okamoto and Tamagawa (89) and Kawamura et al (32) did a similar calculation to Benoit-Cattin and Bernard and obtained the same $\Delta\varepsilon$ as in 2.73. The IED function exhibits a saddle shape with a width depends on the value of $(\tau_{RF}/\tau_{\text{ion}})$. The width of the IED distribution is reduced by increasing
Figure 2.16: The ion energy distribution at different pressures as calculated and measured by Wild and Koidl (19).
the ion mass and/or by increasing the RF frequency. The two peaks of the bimodal IED shape are singular—but integrable—due to the assumption of mono-energetic initial velocity distribution at the plasma sheath boundary.

In the low frequency regime \((\tau_{\text{ion}}/\tau_{\text{RF}}) \ll 1\), the ions traverse the sheath in a short time compared with the field oscillations and respond instantaneously to the field. The sheath can be described as a series of infinite DC sheaths at the different moments in time during the RF cycle \((91)\). The energy \(\varepsilon\) of an ion hitting the target depends on the phase at which the ion enters the sheath. Thus, \(g(\varepsilon) \, d\varepsilon = g(\omega_{\text{RF}} t) \, d(\omega_{\text{RF}} t)\); where \(g(\varepsilon) \, d\varepsilon\) is the number of ions hitting the target with energies between \(\varepsilon\) and \(\varepsilon + d\varepsilon\) and \(g(\omega_{\text{RF}} t) \, d(\omega_{\text{RF}} t)\) is the number of ions hitting the target with energies between phases \(\omega_{\text{RF}} t\) and \(\omega_{\text{RF}} t + d(\omega_{\text{RF}} t)\). Because the sheath motion is slow compared to Bohm velocity, the flux of ions entering the sheath from the presheath is constant. This implies that

\[
g(\varepsilon) = \frac{1}{2\pi} \left| \frac{d\varepsilon}{d(\omega_{\text{RF}} t)} \right|^{-1}. \tag{2.74}
\]

Furthermore, for the low frequency regime, we have

\[
\varepsilon(\omega_{\text{RF}} t) = eV_{\text{sh}}(\omega_{\text{RF}} t). \tag{2.75}
\]

Therefore, for sinusoidal driven voltage \(V(t) = V_0 \sin(\omega_{\text{RF}} t)\) and symmetric RF reactor, the IED is

\[
g(\varepsilon) \rightarrow \begin{cases} 
\frac{1}{\pi} \left( (eV_0)^2 - \varepsilon^2 \right)^{-1/2} & : 0 < \omega_{\text{RF}} t < \pi, \\
\frac{1}{2} \delta(\varepsilon) & : \pi < \omega_{\text{RF}} t < 2\pi.
\end{cases} \tag{2.76}
\]
The IED is broad and independent of the ion mass (32). The distribution is singular – however, integrable – at the peaks due to the assumption of monoenergetic initial velocity distribution.

So far, we have seen that the sheath dynamics in the collisionless high frequency regime and the collisionless low frequency regime could be simplified and calculated analytically. In spite of that, in the intermediate regime, where \( \tau_{\text{ion}} \approx \tau_{\text{RF}} \), the analytical representation of the sheath becomes difficult. The partial response of ions to the instantaneous electric field leads to local and memory effects and makes the spatial integration of Poisson equation difficult. Therefore, a numerical tool must be utilized to calculate the sheath dynamics and consequently the ion energy distribution.

It is worth noting that the complexity of the sheath dynamics increases by involving collisions. The effect of collisions of ions with the background gas within the sheath was first studied experimentally as well as theoretically by Wild and Koidl (19). The collisions within the sheath restrain the particle motion to energies lower than the energies of the free fall under the sole influence of sheath field. The ion energy distributions were found to exhibit detailed and pronounced features. These features result from the creation of thermal ions in the sheath, e.g., by charge-exchange processes, and from the RF modulation of the sheath potential. At low pressures the IED shows mainly the well-known bimodal structure and a number of less significant peaks for lower energies due to the charge exchange collisions as displayed in Figure 2.16. At higher pressures, the bimodal distribution of the primary ions (i.e., ions cross the sheath without collisions) is much diminished.

The ion collisions adjust the ion angular distribution (IAD) (26, 74, 92). It is well known that the IAD determines the anisotropy of plasma processes as plasma etching. Ion collisions cause a widespread ion angular distribution as displayed in Figure 2.17: Ions near normal incidence angle are either created by charge exchange or have experienced no collision in the sheath at all. Ions that hit the surface with a larger impact angle have been scattered at least one elastic collision.
The EST Model

3.1 Introduction

In this chapter we intend to present a mathematical model which enables the efficient, kinetically self-consistent simulation of RF modulated plasma boundary sheaths in all technically relevant discharge regimes. It is defined on a one-dimensional geometry where a Cartesian x-axis points from the electrode or wall at $x_E = 0$ towards the plasma bulk. An arbitrary endpoint $x_B$ is chosen deep in the bulk. The model consists of a set of kinetic equations for the ions, Boltzmann’s relation for the electrons and Poisson’s equation for the electrical field. Boundary conditions specify the ion flux at $x_B$ and a periodically –though not necessarily harmonically– modulated sheath voltage $V_{sh}(t)$ or sheath charge $Q(t)$. The equations are solved in a statistical sense. However, it is not the well-known particle-in-cell (PIC) scheme that is employed, but an alternative iterative algorithm termed ensemble-in-spacetime (EST). The basis of the scheme is a discretization of the spacetime, the product of the domain $[x_E, x_B]$ and the RF period $[0, T]$. Three modules are called in a sequence. A Monte Carlo module calculates the trajectories of a large set of ions from their start at $x_B$ until they reach the electrode at $x_E$, utilizing the potential values on the nodes of the spatio-temporal grid. A harmonic analysis module reconstructs the Fourier modes $n_{im(x)}$ of the ion density $n_i(x, t)$ from the calculated trajectories. A field module finally solves the Boltzmann-Poisson equation with the calculated ion densities to generate an updated set of potential values for the spatio-temporal grid. The iteration is started with the potential values of a self-consistent fluid model and terminates when
the updates become sufficiently small, i.e. when self-consistency is achieved. A subsequent post-processing determines important quantities, in particular the phase-resolved and phase-averaged values of the ion energy and angular distributions and the total energy flux at the electrode. A drastic reduction of the computational effort compared with PIC calculations is achieved. As a first application of the new model, the influence of ion inertia on the dynamics of a collisionless sheath is studied and a comparison of the simulated ion energy distribution with published analytical solutions is performed. This chapter is primarily based on the published material in (93).

3.2 Physical model

Our model studies the dynamics of an RF modulated boundary sheath in planar geometry. Cartesian coordinates are erected on the electrode or wall of interest, with the $x$-axis pointing towards the plasma. The solution domain is therefore the interval $[x_E, x_B]$, with $x_E(=0)$ representing the electrode or wall, and $x_B$ denoting an arbitrary point ”deep in the plasma”. For each ion species $s$ a kinetic equation is formulated; $f_s = f_s(x, v_x, v_y, v_z, t) \equiv f_s(x, \vec{v}, t)$ is the distribution function and $q_s$ and $m_s$ are the charge and mass, respectively. The term $\langle f_s \rangle_c$ denotes the collisional interaction with the background neutrals. The considered processes are elastic scattering and charge transfer; Coulomb collisions and ”chemistry” are ignored. $E_x = -\frac{\partial \Phi}{\partial x}$ denotes the electrical field, a magnetic field is neglected:

$$\frac{\partial f_s}{\partial t} + v_x \frac{\partial f_s}{\partial x} + \frac{q_s}{m_s} E_x \frac{\partial f_s}{\partial v_x} = \langle f_s \rangle_c. \quad (3.1)$$

The electrons are not treated kinetically but assumed to be in Boltzmann equilibrium at a given electron temperature $T_e$; $\hat{n}$ is an arbitrary but constant reference density:

$$n_e(x, t) = \hat{n} \exp \left( \frac{e \Phi(x, t)}{T_e} \right). \quad (3.2)$$

The electrical field is governed by Poisson’s equation

$$\epsilon_0 \frac{\partial E_x}{\partial x} = -\epsilon_0 \frac{\partial^2 \Phi}{\partial x^2} = \sum_s q_s \int f_s d^3v - e n_e. \quad (3.3)$$

Suitable boundary conditions complete the model. At the bulk side $x_B$, the influx $\Psi_s$ of the ions for each species $s$ is prescribed. The $\Psi_s$ are positive
quantities, the sign merely reflects the orientation of the coordinate system:

$$\int v_x f_s \, d^3v \bigg|_{xB} = -\Psi_s. \tag{3.4}$$

The corresponding velocity distributions are chosen reasonably: When collisions are present, the exact form of the distributions has no influence as long as $x_B$ is located sufficiently deep in the bulk. For fully collisionless sheaths, beams or shifted Maxwellians are assumed. (Ions which leave the domain at $x_B$ are simply restarted to keep the ion flux $\Psi_s$ constant.) At $x_E$, a sticking factor of unity is assumed; all ions leave the domain.

Another set of boundary conditions applies to the field. At $x_B$, quasineutrality is assumed, this leads to a relation between the ion density and the bulk potential $\Phi_B$:

$$\Phi_B = \frac{T_e}{e} \ln \left( \sum_s \frac{q_s}{e} \int f_s \, d^3v / \hat{n} \right) \bigg|_{xB}. \tag{3.5}$$

For the second field condition, there are two obvious choices. One is to specify the potential difference between $x_B$ and $x_E$ as the sheath voltage $V_{sh}(t)$,

$$\Phi_B - \Phi_E = V_{sh}(t). \tag{3.6}$$

Alternatively, one can prescribe the total charge $Q$ in the sheath, or equivalently the electrical field $E_{x_E}$ at the electrode. When $x_B$ is properly chosen, both quantities are nearly identical, as the electrical field at $x_B$ is very small compared to the value at $x_E$:

$$Q(t) = \int_{x_E}^{x_B} e(n_i - n_e) \, dx = \epsilon_0 \left( E_{xB} - E_{xE} \right). \tag{3.7}$$

In both instances, the prescribed functions can be any periodic function; $V_{sh}(t) = V_{sh}(t + T)$ or $Q(t) = Q(t + T)$. The period length is referred to as $T$, the RF frequency is $\omega_{RF} = 2\pi / T$. Harmonics of any order below the electron plasma frequency $\omega_{pe}$ are allowed.

It is advantageous to use dimensionless variables. The normalization is based on the reference density $\hat{n}$, the electron charge $e$, a typical ion mass $\hat{m}$, and the electron temperature $T_e$; the reference values of the Debye length and the ion plasma frequency are $\lambda_D = \sqrt{\epsilon_0 T_e / e^2 \hat{n}}$ and $\omega_{pi} = \sqrt{e^2 \hat{n} / \epsilon_0 \hat{m}}$, respectively. We employ $x \rightarrow \hat{x} x$, $m_s \rightarrow \hat{m} m_s$, $q_s \rightarrow Z_s e$, $n_s \rightarrow \hat{n} n_s$, $v \rightarrow \sqrt{T_e / \hat{m}} \, v$, $f_s \rightarrow \hat{n} (T_e / \hat{m})^{-3/2} f_s$, $\Psi_s \rightarrow \hat{n} \sqrt{T_e / \hat{m}} \, \Psi_s$, $\Phi \rightarrow (T_e / e) \Phi$. 

$E \rightarrow (T_c/e \dot{\lambda}_D)E$. Time is measured by the inverse of the RF frequency, $t \rightarrow \omega_{RF}^{-1} t$; the period is $T = 2\pi/\omega_{RF}$. The kinetic equation reads then

$$\frac{\omega_{RF}}{\dot{\omega}_p} \frac{\partial f_s}{\partial t} + v_x \frac{\partial f_s}{\partial x} + \frac{Z_s E_x}{m_s} \frac{\partial f_s}{\partial v_x} = \langle f_s \rangle_c. \quad (3.8)$$

The boundary condition for the input flux at the bulk side is as follows, with the form of the distribution function chosen "reasonably":

$$\int v_x f_s \, d^3v \bigg|_{x_B} = -\Psi_s. \quad (3.9)$$

The Boltzmann-Poisson equation assumes the form

$$\frac{\partial E_x}{\partial x} = -\frac{\partial^2 \Phi}{\partial x^2} = \sum_s Z_s \int f_s \, d^3v - \exp(\Phi). \quad (3.10)$$

The field condition at the bulk side $x_B$ is

$$\Phi \bigg|_{x_B} = \ln \left( \sum_s Z_s \int f_s \, d^3v \right) \bigg|_{x_B}. \quad (3.11)$$

The two alternatives for the field condition at the electrode $x_B$ read as follows, where the prescribed total sheath voltage $V_{sh}(t)$ or sheath charge $Q(t)$ are $2\pi$-periodic functions, i.e., $V_{sh}(t) = V_{sh}(t + 2\pi)$ or $Q(t) = Q(t + 2\pi)$:

$$\Phi \bigg|_{x_E} = \Phi \bigg|_{x_B} - V_{sh}(t), \quad (3.12)$$

$$E_x \bigg|_{x_E} = E_x \bigg|_{x_B} - Q(t). \quad (3.13)$$

In both the dimensional and the dimensionless form, the given equations describe the temporal evolution of the sheath. Particularly interesting is, of course, the time-asymptotic solution obtained in the limit $t \rightarrow \infty$. It is clearly reasonable to assume that this solution is also periodic in the period $T \equiv 2\pi$. Of course, there is no general proof of that assumption; periodically driven dynamical systems may very well exhibit non-periodic behavior like drift, period doubling, or hysteresis. Such phenomena are also observed in RF driven discharges, but they seem to be related to the dynamics of the reactor as a whole, not to the dynamics of the sheath as such (94, 95, 96).
Figure 3.1: Flow diagram of the EST algorithm.
3.3 The Ensemble-in-Spacetime algorithm

The described boundary sheath model is clearly too complicated to be solved analytically. It is therefore treated numerically, employing a novel iterative algorithm termed ensemble in spacetime (EST). Similar to the well-known particle in cell (PIC) (37), EST combines a Lagrangian (particle based) stochastic calculation of the ion distribution function with an Eulerian (grid based) determination of the field. Yet, the two schemes are rather different: When RF driven discharges are simulated, PIC follows the physical evolution of the system from an arbitrary − but otherwise physical − initial state until ”convergence” is reached, i.e., until the solution is periodic in time. EST, in contrast, is an iteration scheme which does not correspond to a time evolution. Instead it considers the space of periodic distribution functions and field configurations over a full RF period and constructs within this space a sequence towards self-consistency. In short: PIC searches for the one self-consistent solution that is periodic, EST searches for the one periodic solution that is self-consistent.

A flowchart of the algorithm is given in Figure 3.1. This section describes the scheme in detail: First the simulation domain (”spacetime”), its discretization and the representation of the physical quantities, second the construction of the initial solution with the help of a simplified fluid model, third the kinetic iteration scheme, and finally the termination criterion and the post processing, particularly the calculation of the ion energy distribution.

3.3.1 Simulation domain and representation of physical quantities

For a numerical implementation of the model, the one-dimensional spatial domain $[x_E, x_B]$ is discretized into $K$ intervals $[x_{k-1}, x_k]$ of not necessarily equal size $\Delta_k$; with $k = 1 \ldots K$. The grid points are $x_k$, $k = 0 \ldots K$. (Of course, $x_0 = x_E$, $x_K = x_B$, $\Delta_k = x_k - x_{k-1}$.) The RF period $[0, 2\pi]$ is divided in $L$ equal intervals $[t_{l-1}, t_l]$, with $t_l = 2\pi l/L$, $l = 0 \ldots L$. The core simulation domain is thus the direct product $[x_E, x_B] \times [0, 2\pi]$; it is referred to as the spacetime and schematically depicted in Figure 3.2. It can be easily extended to the full domain $[x_E, x_B] \times \mathbb{R}$ by the assumption of $2\pi$-periodicity in time.

The product grid can be viewed as an auxiliary structure to represent the physical quantities in the spacetime; its size and refinement must be adjusted to the particular problem. (In most cases, $K = 500$ and $L = 64$ proved sufficient.) Each physical quantity is either allocated on the grid points or on the grid intervals. For example, the potential is defined on the grid points;
3.3 The Ensemble-in-Spacetime algorithm

\[ x(t) = \frac{1}{2} \left( \frac{\Phi_{kl-1} - \Phi_{k-1l-1}}{x_k - x_{k-1}} + \frac{\Phi_{kl} - \Phi_{k-1l}}{x_k - x_{k-1}} \right). \]  

(3.14)

3.3.2 Initial solution and iteration start

To provide our iteration scheme with a good initial guess, we first solve the sheath problem within a self-consistent fluid approach. Specifically, we map the system of kinetic equations onto an effective ion fluid model where \( n_i \) and \( v_i \) are the density and the velocity, respectively. The effective flux is \( \Psi_i = \sum_s Z_s \Psi_s \), the effective ion mass is \( m_i = \left( \sum_s \Psi_s \sqrt{Z_s m_s} \right)^2 / \Psi_i^2 \). Pressure terms are neglected and the collisional interaction with the neutral background is modeled by a possibly velocity dependent mean free path \( \lambda_i = \frac{\Psi_i^2 m_i}{\left( \sum_s \Psi_s \sqrt{m_s/Z_s \lambda_s} \right)^2} \). (The \( \lambda_s \) are the mean free paths of the individual ion species \( s \) as calculated from the elastic ion neutral collision term.)
Furthermore, the plasma frequency of the ions is assumed small compared to the RF frequency, $\omega_{pi} \ll \omega_{RF}$. The equivalent fluid model then reads as follows, where $\bar{E}_x$ denotes the phase average of the electrical field:

\begin{align}
    n_i v_i &= -\Psi_i, \quad (3.15) \\
    v_i \frac{\partial v_i}{\partial x} &= \frac{Z_i}{m_i} \bar{E}_x - \frac{|v_i|}{\lambda_i} v_i. \quad (3.16)
\end{align}

To find the phase-averaged field, we employ the advanced algebraic approximation (65, 72, 73). The AAA gives the electrical field in terms of the transformed coordinate $q = \int_{x_0}^x n_i(x') dx'$, the ion density $n_i(q)$, its derivative $\partial n_i/\partial q$, and the instantaneous sheath charge $Q(t)$. Its central element are the special functions $\Xi_0$ and $\Xi_1$ which physically act as ”switches” between the depletion region $q \ll Q(t)$ and the quasi-neutral region $q \gg Q(t)$:

\begin{equation}
    E_x(q, n_i, \frac{\partial n_i}{\partial q}, t) = -\Xi_0 \left( \frac{q - Q(t)}{\sqrt{n_i}} \right) \sqrt{n_i} - \Xi_1 \left( \frac{q - Q(t)}{\sqrt{n_i}} \right) \frac{\partial n_i}{\partial q}. \quad (3.17)
\end{equation}

For a known sheath charge function $Q(t)$, the phase-averaged electrical field $\bar{E}_x$ can be calculated as a local function of $q$, $n_i(q)$, and $\partial n_i/\partial q$:

\begin{equation}
    \bar{E}_x(q, n_i, \frac{\partial n_i}{\partial q}, \{Q\}) = -\frac{1}{2\pi} \int_0^{2\pi} \Xi_0 \left( \frac{q - Q(t)}{\sqrt{n_i}} \right) dt \sqrt{n_i} \\
    - \frac{1}{2\pi} \int_0^{2\pi} \Xi_1 \left( \frac{q - Q(t)}{\sqrt{n_i}} \right) dt \frac{\partial n_i}{\partial q}. \quad (3.18)
\end{equation}

Solving the flux relation for the ion velocity, and transforming into the new coordinates, we can express the ion equation of motion as

\begin{equation}
    -\frac{\Psi_i^2}{n_i^2} \frac{\partial n_i}{\partial q} = \frac{Z_i}{m_i} \bar{E}_x + \frac{\Psi_i^2}{\lambda_i n_i^2}. \quad (3.19)
\end{equation}

We substitute the field and define the abbreviations

\begin{align}
    L(q, n_i, \{Q\}) &= \frac{Z_i}{2\pi m_i} \int_0^{2\pi} \frac{\Xi_1}{\sqrt{n_i}} \left( \frac{q - Q(t)}{\sqrt{n_i}} \right) dt - \frac{\Psi_i^2}{n_i^2}, \quad (3.20) \\
    R(q, n_i, \{Q\}) &= \frac{\Psi_i^2}{\lambda_i n_i^2} - \frac{Z_i}{2\pi m_i} \int_0^{2\pi} \Xi_0 \left( \frac{q - Q(t)}{\sqrt{n_i}} \right) dt \sqrt{n_i}, \quad (3.21)
\end{align}

to finally obtain a first-order quasilinear differential equation for $n_i(q)$:

\begin{equation}
    L(q, n_i, \{Q\}) \frac{\partial n_i}{\partial q} = R(q, n_i, \{Q\}). \quad (3.22)
\end{equation}
The derived equation is singular wherever the function \( L(q, n_i, \{Q\}) \) vanishes. Only when the function \( R(q, n_i, \{Q\}) \) vanishes as well, the singularity becomes removable. This condition reduces the degree of freedom of the equation: There is, in fact only one physical solution \( n_i(q) \), characterized by the fact that it passes through the intersection \((q^*, n_i^*)\) of the curves \( L(q, n_i, \{Q\}) = 0 \) and \( R(q, n_i, \{Q\}) = 0 \) and that it is monotonically growing. (It is interesting to note that \((q^*, n_i^*)\) can be interpreted as a generalized Bohm point (65).) Once the physical solution is calculated, the phase-resolved and the phased averaged electric fields follow from 3.17 and 3.18, respectively. The sheath voltage can be calculated via

\[
V_{sh}(t) = \int_{qE}^{qB} \Xi_0 \left( \frac{q - Q(t)}{\sqrt{n_i}} \right) \frac{1}{\sqrt{n_i}} + \Xi_1 \left( \frac{q - Q(t)}{\sqrt{n_i}} \right) \frac{1}{n_i} \frac{\partial n_i}{\partial q} \, dq. \tag{3.23}
\]

For the second of the alternative boundary conditions, where the electrode field is specified by the sheath charge \( Q(t) \), the constructed approximate sheath solution can be used directly. For the other alternative, where the sheath voltage is given, one has to invert equation 3.23. This task, however, can be carried out quite easily with the help of an iterative algorithm. As a last step, one may transform the solution back into the original spatial coordinates \( x \) by solving the integral

\[
x(q) = x_E + \int_{qE}^{q} \frac{1}{n_i(q')} \, dq'. \tag{3.24}
\]

### 3.3.3 Ion trajectory simulation

The response of the ions to the field distribution is calculated with help of the ensemble. Each species \( s \) is represented by a collection of test particles, numbered by the elements of an index set \( S_s \subset \mathbb{N} \). The ions are started at \( x_B \), uniformly distributed over the phase \([0, 2\pi]\) and drawn from the ”reasonable” distribution specified above (beam or shifted Maxwellian). Solving the normalized equations of motion, each ion is followed by numerical integration until it finally arrives at the electrode \( x_E \):

\[
\frac{dx}{dt} = \frac{\hat{\omega}_{pi}}{\omega_{RF}} v_x, \tag{3.25}
\]

\[
\frac{dv_x}{dt} = \frac{Z_s}{m_s} \frac{\hat{\omega}_{pi}}{\omega_{RF}} E_x, \tag{3.26}
\]

\[
\frac{dv_y}{dt} = \frac{dv_z}{dt} = 0. \tag{3.27}
\]
In each grid cell \( [x_{k-1}, x_k] \times [t_{l-1}, t_l] \), the electrical field is represented by a constant \( E_{kl} \), calculated from the interpolation formula given above. Hence, the equation of motion can be integrated explicitly in form of quadratic parabola elements:

\[
x(t) = \frac{1}{2} Z_s \frac{\hat{\omega}_p^2}{m_s \omega_{RF}^2} E_{kl}(t - t_0)^2 + \frac{\hat{\omega}_p}{\omega_{RF}} v_0 x(t - t_0) + x_0, \tag{3.28}
\]

\[
v_x(t) = \frac{Z_s}{m_s} E_{kl} \frac{\hat{\omega}_p}{\omega_{RF}} (t - t_0) + v_{0x}, \tag{3.29}
\]

\[
v_y(t) = v_{0y}, \tag{3.30}
\]

\[
v_z(t) = v_{0z}. \tag{3.31}
\]

The parabola elements are valid until the ion either reaches the time \( t_l \), leaves the interval at \( x_{k-1} \) or \( x_k \), or undergoes a collision with a neutral of the background. (Whatever is next.) The details of the collisions are described by a separate model; we assume the presence of elastic and charge transfer interactions. It is useful to employ the null collision method \((40)\) where a positive null collision frequency \( \nu_{\text{null}} \) is added to the energy dependent physical collision frequency \( \nu \) so that the sum \( \nu' = \nu + \nu_{\text{null}} \) is constant. The free time of flight is then chosen as follows, where \( R_{\text{flight}} \) is a random drawn from the interval \([0, 1]\):

\[
t_{\text{flight}} = -\frac{\ln(R_{\text{flight}})}{\nu'}. \tag{3.32}
\]

### 3.3.4 Reconstruction of the distribution function, density, and flux of the ions

The set of all trajectories of the ions of species \( s \) – time functions \((x_n(t), \vec{v}_n(t))\), \( n \in S_s \) – represents the response of the ions to the field. We expand the set by including an infinite number of "images" shifted by a multiple of \( 2\pi \) in phase. The resulting ensemble defines a Klimontovich-type distribution function \( f(x, \vec{v}, t) \) which solves the kinetic equation \((97, 98, 99)\). (For brevity, we supress the species index \( s \).) Note that \( f \) is by construction periodic in \([0, 2\pi]\) and that all the sums are finite, as each particle spends only a finite time in the domain. The constant in front of the sum represents the ratio of the physical flux \( \Psi \) to the flux \(|S|/2\pi\) of the discrete test particles:

\[
f(x, \vec{v}, t) = \Psi \frac{2\pi}{|S|} \sum_{j=-\infty}^{\infty} \sum_{n \in S} \delta(x - x_n(t + j2\pi)) \delta^{(3)}(\vec{v} - \vec{v}_n(t + j2\pi)). \tag{3.33}
\]
Derived from $f$, the density $n(x, t)$ and the flux $\Psi(x, t)$ in $x$-direction are

$$n(x, t) = \Psi \frac{2\pi}{|S|} \sum_{j=\infty}^{\infty} \sum_{n \in S} \delta(x - x_n(t + j2\pi)), \quad (3.34)$$

$$\Psi(x, t) = \Psi \frac{2\pi}{|S|} \sum_{j=\infty}^{\infty} \sum_{n \in S} v_{xn}(t + j2\pi) \delta(x - x_n(t + j2\pi)); \quad (3.35)$$

they of course fulfill the equation of continuity:

$$\frac{\partial n}{\partial t} + \frac{\partial \Psi}{\partial x} = 0. \quad (3.36)$$

As periodic functions, $n(x, t)$ and $\Psi(x, t)$ can be represented as Fourier series:

$$n(x, t) = \sum_{m=-\infty}^{\infty} n_m(x) e^{imt}, \quad (3.37)$$

$$\Psi(x, t) = \sum_{m=-\infty}^{\infty} \Psi_m(x) e^{imt}, \quad (3.38)$$

where the Fourier coefficients are

$$n_m(x) = \frac{1}{2\pi} \int_{0}^{2\pi} n(x, t) e^{-imt} dt$$
$$= \frac{\Psi}{|S|} \sum_{n \in S} \sum_{x_n(t) = x} \frac{1}{|v_{xn}(t)|} e^{-imt}, \quad (3.39)$$

$$\Psi_m(x) = \frac{1}{2\pi} \int_{0}^{2\pi} \Psi(x, t) e^{-imt} dt$$
$$= \frac{\Psi}{|S|} \sum_{n \in S} \sum_{x_n(t) = x} \text{sign}(v_{xn}(t)) e^{-imt}. \quad (3.40)$$

The equation of continuity holds for each Fourier mode separately:

$$imn_m(x) + \frac{\partial \Psi_m}{\partial x} = 0. \quad (3.41)$$

These coefficient functions contain the full microscopic information on the $x$-projection of the ion trajectories. To obtain the macroscopic quantities, suitable averages must be taken. First, the Fourier series are restricted to modes $|m| \leq M$, with $M \ll L$ determined later. Second, the density
coefficients are averaged over the spatial intervals; in the third line we use
\( \theta_{[x_{k-1}, x_k]}(x) \) to denote the characteristic function of the interval \([x_{k-1}, x_k]\)
which is unity for \( x \in [x_{k-1}, x_k] \) and zero otherwise:

\[
\begin{align*}
    n_{mk} &= \frac{1}{x_k - x_{k-1}} \int_{x_{k-1}}^{x_k} n_m(x) dx \\
    &= \frac{1}{x_k - x_{k-1}} \int_{x_{k-1}}^{x_k} \int_0^{2\pi} \sum_{j=-\infty}^{\infty} \frac{\Psi}{|S|} \sum_{n \in S} \delta(x - x_n(t + j2\pi)) e^{-imt} dt dx \\
    &= \frac{1}{x_k - x_{k-1}} \frac{\Psi}{|S|} \sum_{n \in S} \int_{-\infty}^{\infty} \theta_{[x_{k-1}, x_k]}(x_n(t)) e^{-imt} dt.
\end{align*}
\]

The flux coefficients are evaluated at the grid points,

\[
\Psi_{mk} = \frac{\Psi}{|S|} \sum_{n \in S} \sum_{x_n(t) = x_k} \text{sign}(v_{xn}(t)) e^{-imt}.
\]

The law of continuity now appears as a sequence of exact algebraic relations:

\[
im(x_k - x_{k-1})n_{mk} + \Psi_{mk} - \Psi_{mk-1} = 0.
\]

The derived expressions require access to the full set of ion trajectories
and are not yet suited for practical implementation. To remove that problem,
recall that the trajectories are calculated as a sequence of parabolic path
elements, with each element lying completely in a spacetime cell \([x_{k-1}, x_k] \times [t_{l-1}, t_l]\). We assume that there are \( N_{kl} \) such elements per cell, enumerated
by the index \( \mu \); the ion shall enter at \( t_{kl}^- \) and leave at \( t_{kl}^+ \). Thus we get the
following expression for the coefficients of the ion density where it is not (yet)
advantageous to evaluate the integral explicitely:

\[
\begin{align*}
    n_{mk} &= \frac{1}{x_k - x_{k-1}} \frac{\Psi}{|S|} \sum_{l=1}^{L} \sum_{\mu=1}^{N_{kl}} \int_{t_{kl}^-}^{t_{kl}^+} e^{-imt} dt \\
    &= \frac{1}{x_k - x_{k-1}} \frac{\Psi}{|S|} \sum_{l=1}^{L} \sum_{\mu=1}^{N_{kl}} e^{-imt_{kl}^*} dt.
\end{align*}
\]

Similarly, call \( N_{kl}^* \) the number of instances \( \mu \) when a trajectory crosses the grid
point \( x_k \) with a speed \( v_{kl}^* \) at a time \( t_{kl}^* \) within the phase interval \([t_{l-1}, t_l]\).
In terms of these quantites, the Fourier coefficients of the flux can be written as

\[
\Psi_{mk} = \frac{\Psi}{|S|} \sum_{l=1}^{L} \sum_{\mu=1}^{N_{kl}^*} \text{sign}(v_{kl}^*) e^{-imt_{kl}^*}.
\]

The reason that it is not suitable to employ these formulas directly is
that they involve the calculation of many different exponentials. However,
assuming that the maximum Fourier mode number $M$ is much smaller than the temporal interval number $L$, the exponentials in each interval can be linearly approximated:

$$e^{-imt} \approx e^{-imt_{l-1}} + \frac{e^{-imt_l} - e^{-imt_{l-1}}}{t_l - t_{l-1}}(t - t_{l-1}).$$  \hspace{1cm} (3.47)

Using this expression, the integral in 3.45 can now be carried out. The resulting formula is correct up to errors quadratic in $M/L$, the ratio of the maximum Fourier mode number $M$ to the number $L$ of the phase intervals. (First evaluating the integral and then using the approximation would have resulted in an error linear in $M/L$.)

$$n_{mk} = \frac{1}{x_k - x_{k-1}} \left\{ \frac{\Psi}{|S|} \sum_{l=1}^{L} \left( \frac{e^{-imt_{l-1}} + e^{-imt_l}}{2} T_{kl} + \frac{e^{-imt_l} - e^{-imt_{l-1}}}{t_l - t_{l-1}} S_{kl} \right) \right\}. \hspace{1cm} (3.48)$$

Here, $T_{kl}$ represents the total time that the ions spend in a cell, and $S_{kl}$ is a sum of typically small correction terms which reflect the possible asymmetry of the trajectory elements with respect to the time intervals:

$$T_{kl} = \sum_{\mu=1}^{N_{kl}} \left( t_{kl\mu}^+ - t_{kl\mu}^- \right), \hspace{1cm} (3.49)$$

$$S_{kl} = \sum_{\mu=1}^{N_{kl}} \left( t_{kl\mu}^+ - t_{kl\mu}^- \right) \left( \frac{t_{kl\mu}^+ + t_{kl\mu}^-}{2} - \frac{t_l + t_{l-1}}{2} \right). \hspace{1cm} (3.50)$$

Also the coefficients of the flux can be found more efficiently. Linearizing the respective exponentials with the help of (3.47) gives

$$\Psi_{mk} = \left\{ \frac{\Psi}{|S|} \sum_{l=1}^{L} \left( W_{kl} + \frac{e^{-imt_{l-1}}}{t_l - t_{l-1}} U_{kl} + \frac{e^{-imt_l}}{t_l - t_{l-1}} V_{kl} \right) \right\}, \hspace{1cm} (3.51)$$

with the coefficients

$$W_{kl} = \sum_{\mu=1}^{N_{kl}^*} \text{sign}(v_{kl\mu}^*) e^{-imt_{l-1}}, \hspace{1cm} (3.52)$$

$$U_{kl} = \sum_{\mu=1}^{N_{kl}^*} \text{sign}(v_{kl\mu}^*)(t_l - t_{kl\mu}^*), \hspace{1cm} (3.53)$$

$$V_{kl} = \sum_{\mu=1}^{N_{kl}} \text{sign}(v_{kl\mu}^*)(t_{kl\mu} - t_{l-1}). \hspace{1cm} (3.54)$$
Note, however, that this formula is one order less accurate than the density expression (3.48); the algebraic continuity relation (3.44) therefore now only holds up to a discretization error. In practice, this does not constitute any problems, as it is the particle density which enters the potential calculation, not the particle flux.

3.3.5 Calculation of the electrical potential

At each time point $t_l$, the Boltzmann-Poisson equation must be discretized and solved for the potential values $\Phi_{kl}$ at the grid points $k$, $0 \leq k \leq K$. The employed equations read as follows, where the $n_{ikl}$ are the averages of the ion density over the grid intervals $[x_{k-1}, x_k]$ evaluated at the temporal nodes $t_l$. Note that the scheme is stable in the limit of large densities or large interval sizes, i.e., when the $\Delta_k$ become larger than $1/\sqrt{n_i}$ (= the Debye length). Under bulk conditions, the equations then reduce to the condition of quasineutrality:

$$\frac{2}{\Delta_k + \Delta_{k+1}} \left( \frac{\Phi_{k+1l} - \Phi_{kl}}{\Delta_{k+1}} - \frac{\Phi_{kl} - \Phi_{k-1l}}{\Delta_k} \right) + \frac{\Delta_k}{\Delta_k + \Delta_{k+1}} n_{ik+1l} + \frac{\Delta_{k+1}}{\Delta_k + \Delta_{k+1}} n_{ikl} - \exp(\Phi_{kl}) = 0. \tag{3.55}$$

In addition, the boundary conditions must be implemented. The electrode point $x_E$ can of course be analyzed under the assumption of (partial) electron depletion, while the point $x_B$ is located ”deep in the bulk” where quasineutrality holds. It is useful to introduce the ion density and its spatial derivative at $x_B$; this information may be obtained from an asymptotic analysis of the kinetic equation or, simpler, by linear extrapolation:

$$n_{il}|_{x_B} = n_{iKl} + \frac{\Delta_K}{\Delta_{K-1} + \Delta_K} (n_{iKl} - n_{iK-1l}), \tag{3.56}$$

$$\frac{\partial n_{il}}{\partial x} \bigg|_{x_B} = \frac{2 (n_{iKl} - n_{iK-1l})}{\Delta_{K-1} + \Delta_K}. \tag{3.57}$$

At the bulk side, the boundary condition is then

$$\Phi_{Kl} = \Phi \bigg|_{x_B} = \ln \left( n_{il}|_{x_B} \right). \tag{3.58}$$

When the sheath voltage $V_{sh}(t)$ is prescribed, the boundary condition at the electrode is

$$\Phi \bigg|_{x_E} = \Phi \bigg|_{x_B} - V_{sh}(t). \tag{3.59}$$
When the sheath charge is prescribed, the boundary condition is

\[
\frac{\Phi_{1t} - \Phi_{0t}}{\Delta_1} = Q(t) + \frac{1}{n_{3t}|x_B|} \frac{\partial n_{3t}}{\partial x}|_{x_B} \tag{3.60}
\]

The numerical solution of the system can be carried out with a damped Newton algorithm; for each linear step only a tri-diagonal matrix equation must be solved. Within the EST loop, the result of each completed iteration usually provides an adequate initial solution for the field calculation of the next iteration step.

### 3.3.6 The termination criterion and the post processing

The iteration loop is terminated when the updates of the potential become sufficiently small in comparison to, e.g., the thermal voltage \(T_e/e \equiv 1\). (Owing to its stochastic nature, the algorithm does not exhibit convergence in the strict sense. In practice, however, it is easy to distinguish the progressively smaller updates of the "convergence phase" from the small stochastic fluctuations around the limit state.) The field and trajectory data of the last iteration are subjected to post-processing. Important quantities like the distribution of the field and the densities and fluxes of the species can be determined. Of particular interest are the phase-resolved and phase-averaged values of the ion energy and angular distributions and the total energy flux at the electrode. For the example, the phase-averaged energy distribution of ion species \(s\) is defined as

\[
\bar{g}_{E_s}(\varepsilon) = \frac{1}{2\pi} \int_0^{2\pi} \int_{v_x} \int_{v_y} \int_{v_z} \delta\left(\frac{1}{2}m_s v^2 - \varepsilon\right) v_x f_s(x_E, \vec{v}) \, d^3v \, dt. \tag{3.61}
\]

It is normalized so that the phase averaged particle flux and the energy flux are

\[
\bar{\Psi}_{E_s} = \frac{1}{2\pi} \int_0^{2\pi} \int_{v_x} \int_{v_y} \int_{v_z} v_x f_s(x_E, \vec{v}) \, d^3v \, dt = \int_0^\infty \bar{g}_{E_s}(\varepsilon) \, d\varepsilon, \tag{3.62}
\]

\[
\bar{Q}_{E_s} = \frac{1}{2\pi} \int_0^{2\pi} \int_{v_x} \int_{v_y} \int_{v_z} \frac{1}{2}m_s v^2 v_x f_s(x_E, \vec{v}) \, d^3v \, dt = \int_0^\infty \varepsilon \, \bar{g}_{E_s}(\varepsilon) \, d\varepsilon. \tag{3.63}
\]

Numerically, \(\bar{g}_{E_s}\) is calculated as follows: The domain \([0, \varepsilon_{\text{max}}]\), where \(\varepsilon_{\text{max}}\) is the maximum incident ion energy, is divided into \(R\) regular intervals \([\varepsilon_{r-1}, \varepsilon_r]\)
with constant energy step $\Delta \varepsilon$. Then a discrete IED function $g^{(D)}_{Es}$ is defined as a step function, with $\theta_{[\varepsilon_{r-1}, \varepsilon_r]}(\varepsilon)$ as the characteristic function of the interval $[\varepsilon_{r-1}, \varepsilon_r]$:

$$g^{(D)}_{Es}(\varepsilon) = \sum_{r=1}^{R} g_{Esr} \theta_{[\varepsilon_{r-1}, \varepsilon_r]}(\varepsilon). \quad (3.64)$$

The step coefficients $g_{Esr}$ are defined as the averages of the continuous function $g_{Es}(\varepsilon)$ over the interval $[\varepsilon_{r-1}, \varepsilon_r]$ (first identity below). By substituting the various definitions above, they turn out to equal the total number of ions of the species $s$ which strike the electrode with an energy in that interval, divided by its width $\varepsilon_r - \varepsilon_{r-1}$ (second identity). In the last expression, $\varepsilon_n(t_E) = \frac{1}{2} m_s \dot{v}(t_E)^2$ denotes the kinetic energy of the particle in the moment $t_E$ when is impinges the electrode located at $x_E$:

$$g_{Esr} = \frac{1}{\varepsilon_r - \varepsilon_{r-1}} \int_{\varepsilon_{r-1}}^{\varepsilon_r} g_{Es}(\varepsilon) \, d\varepsilon = \frac{1}{\varepsilon_r - \varepsilon_{r-1}} \frac{\Psi}{|S|} \sum_{n \in S} \theta_{[\varepsilon_{r-1}, \varepsilon_r]}(\varepsilon_n(t_E)). \quad (3.65)$$

### 3.4 Collisionless plasma

Finally, we present an example, namely a harmonically driven, collisionless sheath with only one positive ion species. The chosen parameters correspond to a capacitively driven low pressure argon discharge; the ion mass is $m_i = 40$ a.m.u., the charge is $e = 1.602 \times 10^{-19}$ As. The reference density is $n_i = 10^{10}$ cm$^{-3}$, and the electron temperature is $T_e = 3$ eV. The reference values of the electron Debye length and the ion plasma frequency are $\lambda_D = \sqrt{e \varepsilon_0 T_e / e^2 n} = 0.13$ mm and $\omega_{pi} = \sqrt{e^2 n / e_0 m} = 2.1 \times 10^7$ s$^{-1} = 2\pi \times 3.3$ MHz, respectively. We assume a sheath voltage of the form $V_{sh}(t) = \tilde{V}_{sh} - \tilde{V}_{sh} \cos(\omega_{RF} t)$, with $\tilde{V}_{sh} = 300$V, $\tilde{V}_{sh}$ and $\omega_{RF}$ to be varied.

In a first set of simulation runs, we keep the modulation amplitude constant, $\tilde{V}_{sh} = 150$V, i.e., $\tilde{V}_{sh} / \tilde{V}_{sh} = 0.5$, and change the modulation frequency $f_{RF}$ from about 400 kHz to 40 MHz. As control parameter we use the frequency ratio $\omega_{pi} / \omega_{RF}$ which corresponds, up to a factor of order unity, to the ratio of the RF period $\tau_{rf} = 2\pi / \omega_{RF}$ to the characteristic ion time $\tau_{ion}$. (The ratios are exactly identical for the choice $\tau_{ion} = 2\pi / \omega_{pi}$; they are nearly identical when $\tau_{ion}$ is defined as the ion transit time as analytically calculated from a simplified sheath model. See (32) for a discussion of this point.)

Figure 3.3 shows the ion energy distributions at the electrode for various values of $\omega_{pi} / \omega_{RF}$. The results are as expected: We obtain the bimodally shaped IEDF which is characteristic for harmonically modulated, collisionless
Figure 3.3: Ion energy distribution at the electrode, for different values of the parameter $\hat{\omega}_{pi}/\omega_{RF}$. The normalized values of the applied voltage are $V_{sh} = 100$ and $V_{sh} = 50$, respectively.

Figure 3.4: The normalized IED width at the electrode as a function of the parameter $\hat{\omega}_{pi}/\omega_{RF}$. Points correspond to EST results, the lines are the low frequency and high frequency asymptotics. The normalized values of the applied voltage are $\hat{V}_{sh} = 100$ and $\hat{V}_{sh} = 50$, respectively.
Figure 3.5: The ion density in the sheath at different moments during the RF cycle, for $\omega_{\text{pi}}/\omega_{\text{RF}} = 0.55$. The dimensionless values of the applied voltage are $\tilde{V}_{\text{sh}} = 100$ and $\tilde{V}_{\text{sh}} = 50$, respectively.

Figure 3.6: The ion density in the sheath at different moments during the RF cycle, for $\omega_{\text{pi}}/\omega_{\text{RF}} = 1.66$. The dimension values of the applied voltage are $\bar{V}_{\text{sh}} = 100$ and $\bar{V}_{\text{sh}} = 50$, respectively.
Figure 3.7: The ion energy distribution at the electrode as a function of the RF potential amplitude. The frequency ratio is $\dot{\omega}_{pi}/\omega_{RF} = 1.66$, the dimensionless applied DC voltage is $\tilde{V}_{sh} = 100$.

Sheaths. The energy spread $\Delta \epsilon$, displayed in Figure 3.4, is a strong function of the radio frequency. In the high frequency regime, the ions experience many field oscillations across the sheath. Their directed motion responds mostly to the phase averaged electric field and the energy spread is small. Our simulation results (points) are in good agreement with the analysis of Benoit-Cattin and Bernard (23) who found a linear dependence on the parameter $\tau_{RF}/\tau_{ion}$ (solid line),

$$\Delta \epsilon = \frac{\tau_{RF}}{\tau_{ion}} \frac{4e\tilde{V}_{sh}}{\pi}.$$  (3.66)

In the low frequency regime $\dot{\omega}_{pi}/\omega_{RF} > 10$ ions respond to the instantaneous electric field. The dynamics can be understood as sequence of DC sheaths, parametrically modulated by the sheath voltage $V_{sh}(t)$. The energy spread corresponds to the extrema of the potential, it is therefore given as follows (see dashed line in Figure 3.4):

$$\Delta \epsilon = 2e\tilde{V}_{sh}.$$  (3.67)

Clearly more interesting is the intermediate regime. When the applied frequency and the ion plasma frequency are comparable, the motion of the ions becomes truly modulated. Figure 3.5 shows the transient ion density $n_{i}(x,t)$ for $\dot{\omega}_{pi}/\omega_{RF} = 0.55$, Figure 3.6 displays the same curves for $\dot{\omega}_{pi}/\omega_{RF} = 1.66$. The ion densities oscillate around their equilibrium distribution in the form of a standing wave, with a wavelength comparable to the sheath thickness $s$. This is not surprising in view of the relation $\omega_{RF}\tau_{ion} \approx \omega_{RF}s/v_{i} \approx 1$. Of
course, the amplitude of the standing wave – and thus the spread of the ion energies – depends on the amplitude of the potential modulation. In first approximation, this relation is linear (see Figure 3.7).

3.5 Summary and conclusion

We have presented an efficient, kinetically self-consistent approach to the simulation of RF modulated plasma boundary sheaths. Our model consists of a set of kinetic equations for the ions, Boltzmann’s relation for the electrons, and Poisson’s equation for the electrical field. Boundary conditions specify the ion flux at a point in the bulk and a periodically modulated sheath voltage $V_{sh}(t)$ or sheath charge $Q(t)$. The equations are solved in a statistical sense, using the novel algorithm ensemble-in-spacetime which operates on a spatio-temporal grid. EST calls three modules in sequence: A Monte-Carlo module calculates the ion trajectories for a set of potential values on the grid, a harmonic analysis module reconstructs the distribution of the ion density, and a Boltzmann-Poisson module calculates the new potential values. The iteration is started with the outcome of a corresponding fluid model and terminated when the updates become sufficiently small. In our experience, this only requires a small number of iterations, usually less than five.

Compared to a PIC code, our novel algorithm EST is numerically much more efficient. Typically, a simulation of $2 \times 10^5$ ions at $p \leq 30$ Pa takes few minutes on a 2 GHz CPU. Several details contribute: The calculation of the ion trajectories on the spatio-temporal grid is done analytically and has a natural time-step control. The Fourier reconstruction of the ion density efficiently separates the true temporal modulation from spurious statistical noise. Most importantly, the scheme has a more efficient convergence strategy: PIC looks for the one self-consistent solution that is periodic; it follows the evolution and must – if explicit – obey the Courant criterion. EST, in contrast, searches for the one periodic solution that is self-consistent and is not constrained by the Courant criterion.

In principle, of course, the fundamental assumption of periodicity constitutes a weakness of the approach: Solutions which are not periodic in $T$ will be missed by the EST algorithm. In practice, however, there is a simple test to check at least for possible period doubling: Take the period $T$ to be $2 \times 2\pi/\omega_{RF}$, add a spurious potential perturbation periodic in $T$, and check whether the odd Fourier modes of the solution vanish indeed uniformly as the amplitude of that perturbation goes to zero. In our numerical experiments, the phenomenon period doubling was never observed.
Aside from that caveat, the EST algorithm has no practically relevant restrictions regarding the amplitude, waveform, and frequency of the applied RF, the number of the ion species, and the pressure and composition of the neutral gas. These features make our code viable as a tool for the technology oriented computer aided design (TCAD) of plasma processes, such as the tailoring of distribution functions for a particular need, or the calculation of the sheath charge-voltage characteristics as required for the analysis of the nonlinear dynamics of capacitively coupled plasmas (50, 51).
4

Ion Dynamic Effects

4.1 Introduction

In this chapter, we adjust our scope to the effect of the ion inertia on temporal sheath dynamics. The problem concerns many applications in science and engineering as the ion energy distribution, the stochastic heating, and the plasma series resonance. The ion inertia was found to cause a phase difference between the expansion phase and the contraction phase of the plasma sheath, when the ion plasma frequency is comparable to the RF frequency, i.e., $\omega_{\text{pi}} \approx \omega_{\text{RF}}$. This phase difference enhances a temporal asymmetry and develops a hysteresis in the temporal sheath dynamics. Also, the ion dynamics in the intermediate regime affects the height of the IED peaks, i.e., the temporal distribution of the ion flux over the electrode. The content of this chapter depends on the published material (100, 101, 102).

4.2 EST simulation and results

A simulation has been carried out for a collisionless Argon plasma using a reference density of $10^{10}\text{cm}^{-3}$, an electron temperature of 3 eV, and radio-frequency of $2\pi \times 3\text{ MHz}$. This yields a sheath in the intermediate regime with $\omega_{\text{pi}}/\omega_{\text{RF}} \approx 1$. The sheath potential is temporally symmetric and assumed to be $V_{\text{sh}}(t) = 50 - 45\cos(\omega_{\text{RF}}t)$. Ions cross the RF sheath partially respond to the instantaneous electric field. Before explaining the effect of the ion inertia on sheath dynamics, let first study the trajectory of ions crossing the
RF sheath in the intermediate regime. Figure 4.1 shows the trajectory of a set of ions as a function of the normalized time. The ions start the motion from the bulk with a uniform temporal distribution, i.e., constant ion flux. Once an ion is in the range of the moving sheath, the ion is pulled toward the electrode and accelerated as a result of the sheath field. Because the speed of the oscillating plasma boundary is comparable with the ion Bohm speed, the ion would cross the sheath in a fraction of the RF period or overtaken by the plasma boundary during the phase when the sheath width decreases. Of course this is mainly determined by the phase at which the ion enters the sheath. The ions eventually hit the electrode with a non-uniform temporal distribution, i.e., with a time dependent ion flux. As can be concluded from Figure 4.1 that more ions cross the sheath in the second half of RF cycle than in the first half. Figure 4.2 shows the ion bombardment energy distribution (IED) as a function of the ion energy. It exhibits a bimodal shape. Figure 4.3 shows the ion bombardment energy as a function of normalized time. Each point in Figure 4.3 presents an ion. The set of ions shown in the figure has a uniform temporal distribution in the bulk. The projection of points on the time axis \( \{t_1, t_2, t_3, \ldots\} \) reflects the temporal distribution of ions on the electrode. In other words, the number of points per unite time scales the ion flux hitting the electrode. It is clearly seen that the maximum ion flux and the maximum ion bombardment energy are synchronized at time \( t \approx 0.65 \tau_{\text{RF}} \). This explains why the IED in Figure 4.2 exhibits an asymmetric bimodal shape with a high concentration of energetic ions. Another effect of the ion inertia in the intermediate regime is the phase shift \( \Delta \phi \) between the maximum ion energy and the maximum sheath potential; in our simulation a phase shift
Figure 4.2: The IED in the intermediate regime, the plasma is driven with a single RF frequency and a sinusoidal waveform.

of $\Delta \phi = 0.12 \tau_{RF}$ is present. This lag has been observed experimentally by measuring the phase resolved ion energy distribution (113). The ion inertia has an influence also on the ion modulation. The ion modulation during the expansion phase is not like the ion modulation in the contraction phase of the plasma sheath. The ion density is plotted in Figure 4.4 as a function of normalized time at different positions within the sheath ($x = 0.7$, $x = 13.33$, $x = 20$). Because the electric field is weakly dependent on time within the bulk, the ion modulation is insignificant as shown in Figure 4.4 at $x = 26.7$. Furthermore, the ion inertia in the intermediate regime has been found to produce a two different spatial distribution of the electric field within the RF sheath, as displayed in Figure 4.5. This spatiotemporal asymmetry of the RF sheath field has been reported (114). The ion dynamics gives rise to a hysteresis in the sheath charge-voltage relation, as in Figure 4.6. It is worth noting that the ion inertia effects disappear in the high frequency regime (100): There is no ion modulation, the distribution of the electric field in the expansion phase coincides with the field distribution in the contraction phase, and finally the area of the hysteresis loop of the sheath charge-voltage relation becomes zero.
Figure 4.3: Ion energy (red) and time averaged energy (blue) as a function of normalized time.

Figure 4.4: Ion modulation as a function of normalized time at different positions.
4.2 EST simulation and results

Figure 4.5: The temporal RF sheath field in the intermediate regime, solid lines corresponding to moments in the expansion phase, while dashed lines correspond to moments in the contraction phase.

Figure 4.6: Sheath potential versus sheath charge in the intermediate regime.
4.3 Semi-analytical approach

4.3.1 The physical model

The model assumes the Boltzmann distribution for electrons, the continuity and momentum equation for ions, as well as Poisson’s equation. Because it is advantageous to use dimensionless variables, we use the following normalizations: 

\[ x \rightarrow x \lambda_D, \quad t \rightarrow t/\omega_{RF}, \quad u_i \rightarrow u_i \sqrt{T_e/m_i}, \quad n_i \rightarrow \hat{n}_n n_i, \quad E \rightarrow E T_e/e \lambda_D, \quad \Phi \rightarrow \Phi T_e/e. \]

\[ \lambda_D = \sqrt{\varepsilon_0 T_e/n_e^2} \] stands for the Debye length, \( n_i \) and \( n_e \) are the ion and electron densities, \( u_i \) is the ion speed, \( E \) is the electric field, and \( \Phi \) is the electric potential. Then, the model reads as

\[
\begin{align*}
\frac{\omega_{RF}}{\omega_{pi}} \frac{\partial n_i}{\partial t} + \frac{\partial n_i u_i}{\partial x} &= 0, \quad (4.1) \\
\frac{\omega_{RF}}{\omega_{pi}} \frac{\partial u_i}{\partial t} + u_i \frac{\partial u_i}{\partial x} &= E, \quad (4.2) \\
\frac{\partial E}{\partial x} &= n_i - \exp(\Phi), \quad (4.3) \\
\frac{\partial \Phi}{\partial x} &= -E. \quad (4.4)
\end{align*}
\]

\( \omega_{RF} \) and \( \omega_{pi} = \sqrt{\frac{n_e^2}{\varepsilon_0 m_i}} \) are the RF frequency and the ion plasma frequency, respectively. The full and non-restricted analytical solution of 4.1-4.4 is quite difficult. For example: The smallness parameter \( \epsilon = \omega_{pi}/\omega_{RF} \) has been used to find an analytical solution of 4.1-4.4 (103). However, this analysis is restricted to the high frequency regime because \( \epsilon \) is not small enough in the intermediate regime.

In this section we introduce a semi-analytical model which is not restricted to the ratio of the ion plasma frequency to the RF frequency but to small amplitude perturbations of dynamic quantities. Thus, the model gives us the opportunity to check the temporal asymmetry of the plasma sheath in the intermediate regime. We let

\[
\begin{align*}
n_i &= \bar{n}_i + \delta n_i, \quad (4.5) \\
u_i &= \bar{u}_i + \delta u_i, \quad (4.6) \\
E &= \bar{E} + \delta E, \quad (4.7) \\
\Phi &= \bar{\Phi} + \delta \Phi, \quad (4.8)
\end{align*}
\]

where \( \bar{n}_i, \bar{u}_i, \bar{E}, \bar{\Phi} \) are the zero-order quantities, and \( \delta n_i, \delta u_i, \delta E, \) and \( \delta \Phi \) are small first-order perturbations. One can write the zero order parts of 4.1-4.4
as
\[ \bar{n}_i \frac{\partial \bar{u}_i}{\partial x} + \bar{u}_i \frac{\partial \bar{n}_i}{\partial x} = 0, \] (4.9)
\[ \bar{u}_i \frac{\partial \bar{u}_i}{\partial x} = \bar{E}, \] (4.10)
\[ \frac{\partial \bar{E}}{\partial x} = \bar{n}_i - \exp(\bar{\Phi}), \] (4.11)
\[ -\frac{\partial \bar{\Phi}}{\partial x} = \bar{E}, \] (4.12)

whereas the first-order parts are
\[ \eta \frac{\partial \delta n_i}{\partial t} + \frac{\partial}{\partial x} (\bar{u}_i \delta n_i + \bar{n}_i \delta u_i) = 0, \] (4.13)
\[ \eta \frac{\partial \delta u_i}{\partial t} + \frac{\partial (\bar{u}_i \delta u_i)}{\partial x} = \delta E, \] (4.14)
\[ \frac{\partial \delta E}{\partial x} = \delta n_i - \exp(\bar{\Phi}) \delta \Phi, \] (4.15)
\[ \frac{\partial \delta \Phi}{\partial x} = -\delta E. \] (4.16)

For obtaining 4.13-4.16 we have neglected nonlinear terms in the first-order quantities because the perturbations are assumed to have small amplitudes. The scenario of getting a solution, as well known in such cases, is first the calculation of the zero order quantities, then the evaluation of the first order quantities, and finally the construction of the solution by adding the both orders. For simplicity and because of the periodic dynamics of the RF sheath, a time dependence of \( \exp(-i\omega t) \) is introduced:

\[ -i\eta \omega \delta n_i + \frac{\partial}{\partial x} (\bar{u}_i \delta n_i + \bar{n}_i \delta u_i) = 0, \] (4.17)
\[ -i\eta \omega \delta u_i + \frac{\partial (\bar{u}_i \delta u_i)}{\partial x} = \delta E, \] (4.18)
\[ \frac{\partial \delta E}{\partial x} = \delta n_i - \exp(\bar{\Phi}) \delta \Phi, \] (4.19)
\[ \frac{\partial \delta \Phi}{\partial x} = -\delta E. \] (4.20)

Combining 4.17-4.20 in one differential equation gives
\[ \frac{\partial^4 \delta u_i(x)}{\partial x^4} = f\left( \frac{\partial^{1-3} \delta u_i(x)}{\partial x^{1-3}}, \frac{\partial^{1-4} \bar{u}_i(x)}{\partial x^{1-4}}, \ldots \right), \] (4.21)
we used the expression \( f(\ldots) \) instead of writing a lengthy equation. The subsequent analysis is restricted to the wavelengths that are long compared with the local Debye length. Hence one can assume a homogeneous plasma with zero order quasineutrality in \( 4.21 \) and consequently it reads as

\[
\eta^2 \omega^2 \bar{n}_i \delta u_i - 2i \eta \omega \frac{\partial \delta u_i}{\partial x} + (\bar{n}_i + \bar{u}_i - \eta^2 \omega^2) \frac{\partial^2 \delta u_i}{\partial x^2} + \\
-2i \eta \omega \bar{u}_i \frac{\partial^3 \delta u_i}{\partial x^3} + \bar{u}_i^2 \frac{\partial^4 \delta u_i}{\partial x^4} = 0. \tag{4.22}
\]

Letting \( \delta u_i \sim \exp(ikx) \), we obtain the dispersion relation

\[
(\eta \omega - k \bar{u}_i)^2 (1 + k^2 / \bar{n}_i) - k^2 = 0. \tag{4.23}
\]

Although this dispersion relation is for a homogeneous plasma, the roots can be considered to be the local wavenumbers in the WKB sense in a weakly inhomogeneous plasma. This dispersion relation is still valid in the plasma bulk including the transition region. A similar expression of \( 4.23 \) has been derived \((104, 105)\). It could also be derived by a kinetic approach if the Landau damping contribution is neglected \((106)\); this is a valid approximation in the context of low temperature plasmas where \( T_e \gg T_i \). In general, the solution of the dispersion relation \( 4.23 \) gives 4 modes for \( k \), two modes are real and two modes are complex and conjugates. It is clear that no complex \( \omega \) is obtained for real \( k \) so that all modes described by the dispersion relation are stable. The stable waves have the following properties \((107)\):

1. if a wave is purely propagating (real \( k \) for real \( \omega \)), then the sign of the group velocity correctly gives the direction of the energy flow.

2. if a wave has a complex \( k \) for real \( \omega \), then the wave is evanescent, i.e., it decays away from the source.

The dispersion relation of the two real roots could be approximated by the omission of the charge separation term \( k^2 / \bar{n}_i \) in \( 4.23 \) and reads as

\[
k \approx \frac{\eta \omega}{(\bar{u}_i \pm 1)}. \tag{4.24}
\]

Which represent Doppler-shifted ion acoustic waves, for more details, the reader is referred to appendix C in \((104)\). The ion acoustic modes are purely propagating and their group velocities are given by

\[
v_g = \frac{\partial \omega}{\partial k} = \frac{(\bar{u}_i \pm 1)}{\eta}. \tag{4.25}
\]
We can conclude from equation 4.25 that the direction of the energy propagation is independent of the direction of wave propagation. Whereas within the sheath (i.e., $|\bar{u}_i| > 1$), the two modes carry energy in the direction of the ion drift velocity (i.e., toward the electrode). This simple conclusion agrees with the interpretation of equation 18 in (108). Moreover, the other two modes have a complex $k$ for real $\omega$. Because they are stable, both modes must be evanescent.

If we visit again the equations of the first order 4.13-4.16 assuming a time dependence of $\exp(-i\omega t)$ and assuming that the potential and the electric field asymptotically vanish in the plasma bulk (i.e. $\Phi|_{x \to \infty} = 0$ and $\Phi'|_{x \to \infty} = 0$), the asymptotic bulk dynamics could be described as

$$-i\eta \omega \delta n_i - \frac{\partial \delta n_i}{\partial x} + \frac{\partial \delta u_i}{\partial x} = 0,$$

(4.26)

$$-i\eta \omega \delta u_i - \frac{\delta u_i}{\partial x} + \frac{\delta \Phi}{\partial x} = 0,$$

(4.27)

$$-\delta n_i + \delta \Phi - \frac{\partial^2 \delta \Phi}{\partial x^2} = 0.$$  

(4.28)

Supposing a spatial dependence of the first order ion density as $\delta n_i \sim \exp(ikx)$ in 4.28 yields the relations $\delta u_i \sim (k + \omega)/k \exp(ikx)$ and $\delta \Phi \sim 1/(1 + k^2) \exp(ikx)$, respectively. This gives us the dispersion relation:

$$k^4 + 2k \eta \omega + 2k^3 \eta \omega + k^2 \eta^2 \omega^2 + k^2 \eta^2 \omega^2 = 0,$$

(4.29)

which gives two conjugate roots and two real roots. All the modes are purely propagating, i.e., have a real $k$ for real $\omega$. One can observe that the dispersion relation 4.23 equals to the bulk limit dispersion relation 4.29 when $\tilde{n}_i|_{x \to \infty} \to 1$ and $\tilde{u}_i|_{x \to \infty} \to -1$. As a numerical example to prove that all the modes are stable, we set $\omega = 1$ and $\eta = 1$ and solve 4.29 for the possible modes. The result is $k_1 \approx -1.88$, and $k_2 \approx -0.53101$, $k_3 \approx 0.207 + 0.978i$, and $k_4 \approx 0.207 - 0.978i$. The two real modes ($k_1$ and $k_2$) are corresponding to Doppler shifted ion acoustic waves. The imaginary mode $k_3$ attenuates with increasing $x$. To examine the mode $k_4$, we solve the set of equations 4.9-4.12 and 4.17-4.20 using an arbitrary perturbation of all the quantities at the electrode. Then we divide the spatial dependent first order quantities by the mode $\exp(ik_4x)$. We find that all the first order perturbations are damped and vanish in the plasma bulk. The wave amplitude cancels the resultant amplification of the mode $k_4$. As a simple conclusion, all the modes are stable and the first order part can be solved using the boundary conditions $\delta \Phi|_{x \to x_E} = \tilde{V}_{sh}$ (where $\tilde{V}_{sh}$ is the time dependent sheath voltage amplitude which of course is smaller than the DC sheath voltage component), $\delta n_i|_{x \to \infty} = 0$, $\delta u_i|_{x \to \infty} = 0$, and $\delta E|_{x \to \infty} = 0$. 


Figure 4.7: The ion density at different moments (dashed at $\phi = 0$, dotted at $\phi = \pi/2$, and solid at $\phi = \pi$) during the RF period.

Figure 4.8: The sheath charge-voltage relation as calculated by the semi-analytical model.
4.3 Semi-analytical approach

Figure 4.9: The IED as calculated by the semi-analytical model.

Figure 4.10: The ion flux as calculated by the semi-analytical model at different moments (dashed at $\phi = 0$, dotted at $\phi = \pi/2$, and solid at $\phi = \pi$) during the RF cycle.
Figure 4.11: The sheath charge-voltage relation as calculated by the semi-analytical model employing a normalized RF potential amplitudes of 30, 60, 90 Volt.

Figure 4.12: The sheath charge-voltage relation as calculated by the analytical model (solid line) and EST model (dashed line).
4.3 Semi-analytical approach

Figure 4.13: The ion flux as calculated by the semi-analytical model (black) and EST model (gray) at different moments (dashed at $\phi = 0$, dotted at $\phi = \pi/2$, and solid at $\phi = \pi$) during the RF period.

4.3.2 Results

The semi-analytical model has been evaluated assuming a reference density of $10^{10}$ cm$^{-3}$, an electron temperature of 3 eV, a sheath potential of $V_{\text{sh}}(t) = 300(1 - 0.2 \cos(\omega_{\text{RF}} t))$ Volt, and a driving frequency of $2\pi \times 3.3$ MHz. The aforementioned boundary conditions were applied. The ion density profile, the sheath charge-voltage relation, and energy distribution of ions bombarding the electrode are displayed in Figure 4.7, Figure 4.8, and Figure 4.9, respectively. The model predicts qualitatively the effects of the ion inertia on the sheath dynamics as the EST model does. The IED has been calculated via

$$g(\varepsilon) = \frac{1}{2\pi} \int_0^{2\pi} \int_u \delta\left(\frac{1}{2}u^2 - \varepsilon\right) u f(x_E, u) \, du \, dt,$$

assuming an ion distribution function at the electrode of

$$f(x_E, u) = \frac{n_i(x_E, t)}{\delta(u - u_i)}$$

which corresponds to cold ions. $\varepsilon = \frac{1}{2}u_i^2$ is the ion energy. As another ion transit time effect, the modulation of the ion flux as displayed in Figure 4.10. Due to the quasineutrality and the weak time dependence of the electric field within the plasma bulk, the modulation of the ion flux is trivial. On the other hand, the ion flux modulation within the sheath is significant due to the partial interaction with the RF sheath field. As a test of the validity of the first order approximation, Figure 4.11 displays the sheath charge-voltage relation as calculated by the analytical model employing RF potential amplitudes of
30, 60, and 90 volts. The approximation is still valid until $\delta \Phi \approx 0.4 \Phi$. It is clearly seen from Figure 4.11 that the ion energy spread increases with increasing the RF amplitude.

### 4.3.3 Comparison with EST

To verify the outcomes of the semi-analytical model, the charge-voltage relation in Figure 4.12 is compared to the calculated one using the EST model under the same plasma parameters. Also, the spatiotemporal ion flux is compared as shown in Figure 4.13. The two models exhibit a good agreement. However, one can speculate on the discrepancy between the two models. First, the semi-analytical model is not kinetically self consistent as the EST model. Second, the semi-analytical model ignores the higher non-linear orders which would be of great impact at large RF amplitudes.
5

Verification of the EST Model

5.1 Introduction

The numerical effort of directly solving the equation of motion of all particles in plasmas is undoubtedly prohibitive, at least for the foreseeable future. Mathematical simplifications must be employed, and, therefore, compromises made. One option is to reduce the spatial dimension of the problem by assuming a planar or axisymmetric discharge geometry. Stochastic solutions of the kinetic equations, based on the particle-in-cell approach (PIC) merged with Monte Carlo collision calculation, are then feasible and indeed yield a lot of insight (37, 109). However, this approach keeps only the physics of the discharge intact; a realistic description of the often complicated discharge geometry is not frequently possible. If the latter requirement is an issue, alternatives to kinetic models must be sought. One possibility is the fluid approach, in which particles are not represented by distribution functions but only by fluid variables, typically by the number density and the flux, and, in the case of the electrons, the temperature. However, the kinetic information must be restored somehow. This is the realm of ”hybrid schemes” which combine fluid and kinetic arguments in some clever way, with the intent to achieve physically sound kinetic information without paying the price of a full kinetic simulation. Of course, the validity of such approaches is always an issue (26, 28).
To determine the energy and angular distribution of the surface-incident particles, such a hybrid scheme should thus suffice. Most simply, it may be implemented as follows: A fluid code (which may be a hybrid code in other respects) is run to its convergence. The fields of the final state (or, in the RF case, of the final period) are transferred to a Monte Carlo module which simulates the trajectories of a sufficiently large set of test particles and calculates the normalized distribution functions at the selected surface. The absolute distribution functions are finally found as products of the normalized functions with the respective fluxes (to be determined from the fluid code).

This approach has been implemented into many simulation codes, for example into the codes HPEM and nonPDPsim by Kushner and co-workers or the commercial codes COMSOL and CFD-ACE (28, 110, 111, 112). The scheme has, however, one essential deficiency: It is, on the kinetic level, not self-consistent. To correct this deficiency, we have proposed the novel scheme Ensemble in Space-Time (EST) –see chapter 3– which constructs a uniform solution of the kinetic equations for the particles and Poisson’s equation for the field, i.e., provides a fully self-consistent kinetic simulation of the sheath. Its input parameters are still fluid-dynamic: the fluxes of the energetic species, the voltage across the sheath, the electron temperature, and the composition of the neutral background (93).

Of course, this raises a question: How does EST compare to a fully kinetic scheme and experimental results? Is the energy distribution of the surface incident ions given by the EST scheme the same as the one calculated from a complete PIC or the actual experimental results? We will show that the answer to this question is yes. As a test ground, we will study the sheath dynamics in the regime where kinetic effects are most pronounced, namely in the intermediate regime where the applied radio frequency \( \omega_{RF} \) is comparable to the ion plasma frequency \( \omega_{pi} \) and the inverse ion sheath transit time \( \tau_i \). Here, the ions can only partially respond to the time varying field in the sheath, and interesting effects can be observed like temporal asymmetries and phase shifts between applied voltage and ion energy (32, 113, 114). A paper based on the content of this chapter has been submitted for publication (115).

5.2 Setup and kinetic simulation

The set-up of our study is shown in Figure 5.1. It is a single frequency capacitively coupled plasma driven at a voltage of \( V_0 = 100 \) V. The electrode gap is \( d = 2 \) cm, the RF frequency of \( \omega_{RF} = 2\pi \times 0.5 \) MHz, the gas is Argon at a pressure of \( p = 1 \) Pa and a fixed temperature of \( T = 300 \) K.
Figure 5.1: Schematic of situation studied in this chapter. The discharge is a planar capacitively coupled plasma, run with Argon at $p = 1$ Pa. The electrode gap is 2 cm. The shaded area represents the assumed additional Gaussian ionization source $S^*$. 

All particles which impact the two electrodes are absorbed, the emission of secondary electrons is neglected. A particular feature of the model is an additional ionization source $S^*$ assumed in the plasma bulk. The motivation for this additional source is purely technical: Single-frequency CCPs with gaps in the cm range cannot be sustained at the assumed pressure and frequency. Of course, we could have simulated a double frequency CCP or other hybrid discharge, but this would have made our results below more difficult to interpret.

The first scheme used in this study is a benchmarked realization of PIC/MCC (116). It is a one-dimensional planar (1d3v) bounded implementation, which incorporates a Monte Carlo treatment of collision processes with the cross sections taken from (117, 118). The additional bulk ionization source $S^*$ is modeled by electron-ion pair creation in a Gaussian-shaped 2.5 mm-wide region in the center of the discharge; the average energy of the new electrons is 3 eV. Figure 5.2 displays the obtained time averaged density profiles, Figure 5.3 shows the time resolved electrical potential. All expected features are visible, in particular the emergence of a quasineutral bulk with a relatively weak electrical field and of electron-depleted sheaths with much higher voltage drop. The plasma bulk has a positive potential relative to the elec-
Figure 5.2: Distribution of charged particle densities calculated by the PIC simulation. The solid line represents the phase-averaged ion density $\bar{n}_i(x)$, the dashed line represents the phase-averaged electron density $\bar{n}_e(x)$.

Figure 5.3: The potential along the discharge at different phases (dashed $\phi = 0$, solid $\phi = \pi/2$, and dotted $\phi = \pi$) within the RF cycle.
5.2 Setup and kinetic simulation

![Graph](image)

**Figure 5.4:** The sheath voltage – the difference between the plasma potential and the electrode potential – as a function of the normalized time $t/\tau_{RF}$ as calculated by PIC.

...troses, except for brief moments in the RF cycle where electrons can reach the electrodes to balance the ion flux.

The second algorithm is our *Ensemble-in-Spacetime*. EST was designed as a tool for technology-oriented computer aided design (TCAD); it provides a fast, kinetically self-consistent simulation of a DC or RF plasma boundary sheath and the resulting ion energy distribution. EST differs from PIC in several aspects. It does not simulate an entire discharge but only the sheath; the ’operation parameters’ – sheath voltage, ion fluxes, electron temperature – must be provided as an input. (In our example, the sheath voltage is deduced from the potential as shown in Figure 5.4, the ion flux is $\Psi_i = 6.25 \times 10^{14} \text{cm}^{-2}\text{s}^{-1}$, and the electron temperature is $T_e = 1.0 \text{eV}$.) Like PIC, EST has kinetic equations for the ions which are solved in a stochastic sense. However, the electrons are not treated kinetically but assumed to follow a Boltzmann relation with given temperature, $n_e = n_{e0} \exp(e\Phi/T_e)$.

Finally, EST has a different solution strategy than PIC. It does not follow the transient evolution until it reaches a ”converged” (= periodic) state, but rather seeks within the space of all periodic sheath states for a solution of the equations of motion. The central structure of the algorithm is the name-giving *spacetime*, a discretized (grid) representation of the domain $[x_E, x_B] \times [0, \tau_{RF}]$. (Here, $x_E$ is the location of the electrode, $x_B$ an arbitrary point deep in the plasma bulk, and $\tau_{RF} = 2\pi/\omega_{RF}$ the RF period.) The algorithm starts by assigning to each node $(x_k, t_l)$ of the spacetime a potential $\Phi_{kl}$; calculated with a fluid sheath model (65, 72, 73). Then, three modules are iterated:
1. A Monte Carlo module finds the trajectories of the \textit{ensemble}, a large set of test ions. The ions are started at \(x_B\) with their drift speed and uniform distribution in phase and are followed until they leave the system at \(x_E\). Elastic scattering and charge exchange collisions with a spatial uniform background gas are performed in the same way as in the PIC code. The set of all trajectories – illustrated in Figure 5.9 below – represent the response of the ions to the field.

2. A harmonic analysis module reconstructs from the calculated trajectories and from the prescribed flux the ion density \(n_{ikl}\) for each node \((x_k, t_l)\) of the spacetime grid. By construction, it is a periodic quantity.

3. A field module solves for each phase point \(t_l\) the Boltzmann-Poisson equation with the calculated ion densities to update the potential. The electron density is obtained by Boltzmann’s relation, with the electron temperature as specified; the boundary condition is derived from the prescribed sheath potential.

The iteration is terminated when the updates of the potential are sufficiently small. Owing to the stochastic nature of the Monte Carlo module, the algorithm exhibits no convergence in the strict sense, but reasonable accuracy (on the \(10^{-3}\) level) can typically be achieved in less than five iterations.

### 5.3 Results and discussion

In spite of their different mathematical structure, both models yield very similar results. In the following, we will compare several quantities and comment on the likely causes of the remaining differences between EST and PIC. (To give a fair assessment, we did not ”fine-tune” the models for agreement, instead both schemes are used ”as published”. Of course, the same discharge parameters and cross sections (117) are employed.)

Our first comparison – see Figure 5.5 – is between the phase averaged ion densities. The largest deviation can be seen in the bulk, increasing towards the discharge center. We believe that this deviation is unavoidable, and it provides in fact an opportunity to illustrate one of the limitations of the EST model: At the low gas pressure of \(p = 1\) Pa, the ion mean free path is approximately \(1\) cm. The PIC ions, which are mostly born cold in the shaded ionization zone, travel only a distance of about \(0.5\) cm to the sheath edge. This does not suffice to establish drift equilibrium. The EST ions, on the other hand, are started at \(x_B\) already with drift speed – in the logic of the EST scheme, more specific (= non-fluiddynamic) information is not available. In
5.3 Results and discussion

Figure 5.5: The time averaged ion density along the discharge. The dotted line presents the PIC simulation. The solid line presents the EST results. The shaded area displays the heating source area. The rectangle in the left bottom of the figure denotes the maximum extension of the RF modulated sheath.

Other words: EST focuses solely on the sheath—it cannot reconstruct kinetic phenomena of the bulk.

In the sheath itself, however, the named effect does not play a role, at the potential differences are much larger and both PIC and EST ions are far from drift equilibrium. The initial conditions matter less, and the agreement of the ion densities is much better. Any remaining discrepancies are probably due to small differences in the treatment of collisions, and due to the fact that EST assumes the electrons in Boltzmann equilibrium, while PIC treats them kinetically. (There may be a third possible cause for differences: EST uses a noise-reducing Fourier scheme to reconstruct the ion densities from the trajectories through the whole space-time, while PIC has only access to the instantaneous state and cannot not remove any noise. However, this effect is not very influential.) The good agreement carries over to other sheath quantities: Figure 5.6 shows the ion energy distributions at the electrode, Figure 5.7 the phased resolve sheath thickness $s(t)$, calculated using the definition of (73), and Figure 5.8 the charge-voltage characteristics. The results of EST (solid) and of PIC/MCC (dotted) are nearly identical.

As discussed in the introduction, it is in particular the faithful representation of the ion energy distribution at the electrode that legitimates EST as a post-processor. Physically, however, the charge voltage characteristics of Figure 5.8 and its hysteresis is even more interesting: It demonstrates that both models capture kinetic effects correctly. To understand the argument, consider the space-time trajectories of an ensemble of ions as displayed in Figure 5.9. (For simplicity, thermal spread—ion temperature—is neglected
**Figure 5.6:** IED as a function of ion energy. The dotted line presents the PIC simulation results. The solid line presents the EST results.

**Figure 5.7:** Sheath width as a function of normalized time $t/\tau_{RF}$. The dotted line presents the PIC simulation. The solid line presents the EST results.
Figure 5.8: Parametric plot of the sheath potential $V_{sh}$ and the sheath charge per area $Q$ as a function of time $t$. The arrows indicate the orientation. The dotted line represents the PIC/MCC results; the solid line those of EST.

Figure 5.9: The space-time trajectories of an ensemble of ions (colored lines). The sheath edge is represented as black line.
and ion-neutral collisions are turned off.) The ions enter the depicted interval with Bohm speed and with a uniform phase distribution. As long as they are outside the sheath, they experience no electrical field and are not accelerated. This motion translates into a temporally constant ion density, see Figure 5.10. However, once the ions cross the momentaneous sheath edge $s(t)$, they get accelerated and quickly drawn to the electrode. For $\omega_{RF} \approx \omega_{pi}$, this is clearly not a temporally symmetric effect, as more ions are collected when the sheath expands, as when it retracts. The ion flux is thus temporally modulated, and so is the ion density $n_i$ in the sheath. (See again Figure 5.10.) The resulting global effect is the phase shift between the sheath voltage $V_{sh}(t)$ and the sheath charge $Q(t)$ that was depicted in Figure 5.10. Clearly, the effect vanishes both for $\omega_{RF} \ll \omega_{pi}$ and $\omega_{RF} \gg \omega_{pi}$: In the first case, the sheath expansion is slow compared to the ion speed and the sheath-entering flux is independent of phase. In the second regime, the field changes too quickly for the ions to follow, and only the phase-averaged field determines the motion.

The temporal asymmetry of the sheath charge-voltage relation surely impacts the overall behavior of a discharge. It will be interesting to reconsider the studies on the plasma series resonance \cite{49, 54} and stochastic heating \cite{47, 50, 75} in the regime $\omega_{RF} \approx \omega_{pi}$ where a full $V$-$Q$-hysteresis is present.

As a simple conclusion of this section, two kinetic models have been compared in the intermediate radio frequency regime, Particle-in-Cell/Monte Carlo collisions (PIC/MCC) and Ensemble-in-Spacetime (EST). The EST model, resolving the sheath alone, yields ion energy distributions and charge-voltage-relations close to those obtained by the fully kinetic PIC-MCC simulations, provided that the flux of the ions into the sheath, the sheath voltage,
and the electron temperature are known. It can thus be used as an efficient post-processor to restore kinetic information from the restricted information provided by fluid plasma models. The gain in efficiency is considerable. However, PIC-MCC remains indispensable, as it is the standard against which all other schemes must be compared.

5.4 Validation of the EST model via Experiment

Measurements were performed by Baloniak et al (21, 22) in an inductively coupled plasma reactor. The height and the diameter of the chamber were 145 mm and 200 mm, respectively. A pure Argon discharge at a pressure of 1 Pa and a power of 100 W was used. An Argon gas flow of 3 sccm was applied. The ICP coil was driven by a frequency of 13.56 MHz. A stainless steel electrode with an embedded retarding field analyzer (RFA) was inserted into the chamber face to face with ICP coil. The electrode diameter was 140 mm. An external DC bias of $-29.9 \text{ V}$ and rectangular RF bias voltage of $20 \text{ V}_{\text{pp}}$ and with a duty cycle of 50% were applied to the electrode. The RF frequency was 1 MHz. The electron density and electron temperature close to the sheath edge were determined as $n_e = 1.18 \times 10^{10} \text{cm}^{-3}$ and $T_e = 4.67 \text{eV}$. The gas temperature assumed to be 300 K. The bias voltage was monitored by a voltage probe close to the electrode and consequently an accurate sheath potential was calculated. The ion velocity distribution was measured after a quantitative calibration was done. Because the energy resolution in RFA is limited, the modeled ion velocity distribution cannot directly be compared with the measured data. The energy resolution of an RFA must be described by an instrument function $h(E)$, for which a Gaussian distribution can be assumed:

$$h_{\varepsilon'}(\varepsilon) = \frac{1}{\delta \sqrt{2\pi}} \exp\left(-\frac{(\varepsilon - \varepsilon')^2}{2\delta^2}\right) \quad (5.1)$$

where $\delta$ denotes the standard deviation of the Gaussian distribution,

$$\delta = \frac{\Delta \varepsilon}{2 \sqrt{2 \ln 2}}. \quad (5.2)$$

Therefore the measured IVDF is always a convolution of the actual distribution $f_c$ and the instrument functions $h_{\varepsilon'}$:

$$f_m \approx f_c \ h_{\varepsilon'}. \quad (5.3)$$
Using the measured sheath parameters, the ion velocity distribution was calculated employing EST model. In collisionless plasmas, the relation between the ion velocity distribution $f(v) = f(\varepsilon(v))$ and the ion energy distribution $g(\varepsilon)$ could be simply written as

$$f(\varepsilon(v)) = \sqrt{2m_i\varepsilon} \ g(\varepsilon).$$  \hfill (5.4)

Here $m_i$ stands for the ion mass; $v$ and $\varepsilon$ are the ion speed and the ion energy, respectively. The measured and the calculated IVD are compared in Figure 5.11 where an energy resolution of 3.2 eV assumed in the simulation as well as in the experiment, i.e., the linewidth of the IVDF is $\Delta \varepsilon = 3.2\text{eV}$. The solid line shows the experimental results and the dotted line shows the simulation. The data shows excellent agreement concerning the shape of the IVD as well as the position of the peaks. The toggling of the sheath potential between two values yields the two major peaks in the IVDF. The two peaks correspond to the two alternating sheath voltages. However, the asymmetry of the height of the IVD is mostly a result of ion inertia effects. If we keep a close eye again to Figure 5.11, we will observe an ion energy offset between the two peaks due to the ion inertia.
6

Magnetized and Non-Magnetized Plasma Sheaths

6.1 Introduction

As discussed before, the asymmetry of the discharge could be produced geometrically and/or electrically. In this chapter we will explain how an external magnetic field could also be used to enhance or compensate the asymmetry of the discharge (119). Our tools are a 1d3v PIC/MCC code and EST model. The influence of the magnetic field on the discharge and on the IED is demonstrated through a comparison between simulation cases with and without magnetic field. So far, we said that in magnetized discharges, a slowly rotating static and usually spatially homogeneous magnetic field is applied (34, 120, 121), with the goal to obtain a high electron density at very low gas pressures in the Pascal (or even in the sub-Pascal) range. The spatial dimensions of such discharges are typically of the order of a few centimeters, and the magnetic field strength can vary up to tens of mTorr. Under such conditions the electrons are magnetically confined while ions are not. This could be easily proved by studying the ion equation of motion. The normalization of dynamical quantities $\tilde{r} \rightarrow \lambda_D \tilde{r}$, $t \rightarrow t/\omega_{RF}$, $\tilde{v} \rightarrow \tilde{v}\sqrt{T_e/m_i}$, and

...
\( \vec{B} = \hat{B} \vec{B} \) in the ion equation of motion,

\[
m_i \frac{\partial^2 \vec{r}}{\partial t^2} = e \vec{E} + e \vec{v} \times \vec{B},
\]

yields:

\[
\frac{\partial^2 \vec{r}}{\partial t^2} = \left( \frac{\omega_{pi}}{\omega_{RF}} \right)^2 \vec{E} + \frac{\Omega_i \omega_{pi}}{\omega_{RF}^2} \vec{v} \times \vec{B}.
\]

Where \( \vec{r} \) is the position vector, \( \vec{E} \) is the electric field, \( \vec{B} \) is the magnetic field. \( \omega_{pi} \) and \( \Omega_i \) are the ion plasma frequency and the ion cyclotron frequency, respectively. The ratio of \( c_1 \) to \( c_2 \) is \( R_i = \frac{\omega_{pi}}{\Omega_i} \). The constant \( R_i \) could be derived by dividing the Larmor radius \( r_L \) and the sheath width \( s \), i.e., \( R_i = \frac{\omega_{pi}}{\Omega_i} \approx r_L/s \). The effect of magnetic field on ion dynamics could be revealed from the value of the ratio \( R_i \). When \( R_i \to \infty \), the ions are not magnetized. When \( R_i \to 0 \), in contrast, ions are fully magnetized. In typical Argon RF discharges with a bulk density of \( 10^{10} \) cm\(^{-3} \) and a magnetic field of 6 mTesla, the constant \( R_i \approx 1440 \). So the ions are not magnetized. However the electrons are magnetized due to their small mass where the plasma parameters give \( R_e = 5.3 \). Consequently, the applied magnetic field predominantly influences the electron dynamics.

The magnetic field introduces a number of effects that act to increase the density and reduce the sheath voltage at a fixed absorbed power:

1. The electron motion is strongly inhibited across the field. Moreover, the magnetic field can confine the energetic (ionizing) electrons to a small volume near the electrode. Both lead to increase the rate of ionization and consequently increase the plasma density and the ion flux to the sheath as well.

2. A transverse field can increase the efficiency of stochastic heating due to multiple correlated collisions of electrons with the oscillating sheaths.

3. There is an increase in the efficiency of ohmic heating due to higher electric fields in the plasma.

Is magnetized plasma could be studied as 1D problem? To answer this question let consider Maxwell equations in case of magneto-static approxima-
6.1 Introduction

Figure 6.1: Schematic of the discharge configuration.

\[ \vec{\nabla} \cdot \vec{E} = \rho / \epsilon, \]
\[ \vec{\nabla} \times \vec{E} = 0, \]
\[ \vec{\nabla} \cdot \vec{B} = 0, \]
\[ \vec{\nabla} \times \vec{B} = \mu J. \]  
(6.3)

Then, the rotation of the magnetic field produces a current in a direction perpendicular to the direction of the magnetic field. Therefore, 1D model would be not sufficient to study such plasmas. However, an arbitrary magnetic field could be applied to cancel the current \( J \). In this case, the magneto-static approximation is simplified to be an electro-static approximation and the plasma dynamics can be studied as 1D problem. However, to include the particle drifts (as \( E \times B, \nabla B \times \vec{B} \)), 1D model is not sufficient.

In this work we investigate the discharge configuration depicted in Figure 6.1. In a geometrically symmetric plane electrode setup, with an electrode gap of \( L = 5 \text{ cm} \), an external magnetic field is applied near the left electrode. Within a small corridor (indicated by the dashed lines) the magnetic field can be considered parallel to the electrodes along the \( x \)-direction. It is here assumed to decay from the left electrode into the plasma as

\[ \vec{B} = \frac{B_0 \hat{a}_x}{1 + (z/L)^2}. \]  
(6.4)
The magnetic field amplitude is chosen to be $B_0 = 6$ mT and the decay length is $l = 5$ mm. It is worth noting that the configuration of the magnetic field in $(x, z)$ coordinates satisfies

$$\frac{\partial B_x}{\partial z} = \frac{\partial B_z}{\partial x}. \quad (6.5)$$

This necessarily leads to a physical magnetic field configuration as shown in Figure 6.1. This two-dimensional field configuration is similar to that of a half magnetron. This magnetic field configuration cancel the current $J$ in Maxwell equations. Therefore, the 2D problem can be simplified to be 1D problem. Such a simplified approach captures the essential physics because within the small corridor (indicated by the dashed line in Figure 6.1) the magnetic field lines are almost perfectly parallel to the electrodes. The steady state current in this configuration is therefore almost completely perpendicular to the electrodes so that the one-dimensional approach is justified.

\section*{6.2 Numerical analysis}

In order to allow for the electrical asymmetry effect (EAE) a driving voltage $V(t)$ of two consecutive harmonics, i.e.,

$$V(t) = 250V[\sin(2\pi \times 13.56 \text{ MHz } t + \Delta \phi) + \sin(4\pi \times 13.56 \text{ MHz } t)],$$

is connected through a blocking capacitor of $C_B = 1.5$ nF to the left electrode. The relative phase $\Delta \phi$ between the two harmonics is the control parameter for the EAE and varies between $\Delta \phi = [-\pi/4, \pi/4]$. The right electrode is grounded. The pressure of the background Argon gas is fixed at $p = 1$ Pa.

To analyze the magnetized (and non-magnetized) CCRFD described above, we employ two conceptually different kinetic approaches. The first is an explicit electrostatic 1d3v PIC code. Within a single PIC cycle (as shown in chapter 2) the Poisson solver, the particle pusher and the Monte-Carlo collision module are successively iterated until convergence of the overall simulation is achieved. To couple the different modules, a first order field interpolation and charge assignment is performed. The particles are moved according to the Lorentz force using a push algorithm based on Boris approach (37). Collisions of electrons and ions with the fixed background gas are performed in the frame of a slightly modified null-collisions method [16], with respect to the classical method proposed by (40). Since Argon is used as the background gas the collision processes include elastic scattering of both electrons and ions, ionization and excitation due to electron collisions, and backward scattering (i.e., charge exchange collisions) of ions. Cross-section data are taken from Phelps (117) and the LXCat Database website.
“http://www.lxcat.laplace.univ-tlse.fr”. The second approach relies on EST which resolves only the sheath, unlike PIC which treats the entire discharge. EST is an iterative algorithm based on the solution of a set of kinetic equations for the ions, Boltzmann’s relation for the electrons, and Poisson’s equation for the electric field. Similar to PIC, EST employs the null-collision method to account for elastic and charge exchange collisions of ions, but does not take into account ionization and excitation processes. EST is fed by two input parameters: the first is the ion flux given at the sheath edge. The second is the sheath voltage which can have an arbitrary periodic waveform. Because of its efficient convergence behavior, EST allows for fast and kinetically self-consistent simulations of sheaths of CCRFDs. More details concerning the mathematical description and the validation of the model can be found in chapter 3. In order to compare the results of PIC and EST model, we extract the ion flux and the sheath voltage from the PIC simulations at the formal sheath edge defined as $s(t)$ using the definition (72)

$$\int_0^{s(t)} n_e \, dx = \int_{s(t)}^{\infty} (n_i - n_e) \, dx.$$  \hspace{1cm} (6.6)

Ion flux and sheath voltage are then used as input parameters for EST.

## 6.3 Results

### 6.3.1 Magnetized CCRFD

We start our analysis by comparing two different discharge scenarios, without (case I) and with (case II) an applied static magnetic field at the left electrode by means of PIC simulations. The simulation parameters are specified in section II. The relative phase between the two driving voltages is set to $\Delta \phi = 0$. Figure 6.2 shows the space time dynamics of the electron densities for the two different cases. For case I we obtain, as expected, a symmetric density profile and a symmetric sheath dynamics. In contrast, in case II the discharge shows a significant asymmetry due to the applied magnetic field. One can observe that the magnetized sheath in front of the left driven electrode is much smaller than the non-magnetized sheath in front of the right grounded electrode. This asymmetry can also be seen in Figure 6.3 where the (time-averaged) ion density profiles are shown. It is clearly visible that the maximum is shifted and the slope is steeper towards the magnetized region in front of the left electrode. This asymmetry in the ion density (and the ion flux) is the reason of the different sheath widths. Figure 6.4 compares the temporal
Figure 6.2: Electron density as obtained in the PIC simulation results without (top, case I) and with (bottom, case II) an external magnetic field.

Figure 6.3: Average ion density without (dashed, case I) and with (solid, case II) magnetic field.
behavior of the sheath widths for the two different cases of both the left (driven) and right (grounded) sheath. While in case I both sheaths show very similar oscillations, in case II the magnetized sheath is on average significantly smaller than the non-magnetized sheath (right). This magnetically induced asymmetry results in a substantial DC self bias voltage (of about 130 V). Due to the high ion mass and a relatively low ion velocity, the magnetic field in front of the left electrode is not able to significantly confine the ions. The main effect introduced by the external magnetic field is the efficient electron heating near the left sheath. One can observe a locally increased plasma density and an increased ion flux to the electrode. Therefore, despite the magnetic field, we attribute the differences in the energy distribution functions of ions impinging the electrodes (Figure 6.5) mainly to the different plasma density profiles (thus the ion flux towards the electrodes) and to the sheath voltages accelerating the ions.

### 6.3.2 Magnetized CCRFD with electrical asymmetry

The parameter which controls the EAE is the relative phase between the two driving voltages. By introducing a spatially inhomogeneous magnetic field, which itself produces an asymmetry, one is able to amplify or (over-) compensate the EAE. As an example, we compare the previously discussed PIC
results for case II with the results obtained for the same discharge conditions, but for relative phases of $\Delta \phi = \pm \pi/4$ instead of $\Delta \phi = 0$. Figure 6.6 shows the IEDFs for this scenario. Comparing the cases (cf. also Figure 6.5) one can observe that for $\Delta \phi = -\pi/4$ the EAE can be used to amplify the magnetically induced asymmetry (in terms of the ion bombardment energy). Using a relative phase of $\Delta \phi = \pi/4$ leads to an over-compensation of the magnetically induced asymmetry.

### 6.3.3 Magnetic field influence

We have claimed above that the magnetic field does not directly influence the dynamics of the sheaths and thus plays only a minor role when calculating IEDFs. To justify this hypothesis we analyze the sheath dynamics using EST model, which does not include any magnetic field effects. For this purpose we compare the IEDFs for the two cases without magnetic field (case I) and with magnetic field (case II) obtained by PIC with the results from EST. As described in section III we extract the ion flux and the sheath voltage from the PIC simulation and use them as input parameters for EST. Figure 6.7 shows the space time dynamics of the electron density for both simulation approaches for case II (magnetized case for $\Delta \phi = 0$). The sheath dynamics is
Figure 6.6: IEDFs for the magnetized case with and without electrical asymmetry.
Figure 6.7: Electron density in the left sheath region obtained with the PIC method (top) and the EST model (bottom) for the magnetized case (case II).

in very good qualitative agreement. Deviations can be observed particularly only near the sheath edge. This deviation is attributed to the different collision models used in PIC and EST: The latter does not account for ionization and excitation processes, while the PIC simulations allows for both ionization and excitation, and therefore efficient plasma generation.

A second reason (which has to be proofed) is nonlinear electron resonance heating due to the excitation of higher harmonics in the plasma current and thus the plasma series resonance \((49, 50)\). The higher harmonics are clearly visible in the sheath dynamics. Of course, this nonlinear sheath bulk interaction is not included in EST results, since EST resolves one sheath only. Figure 6.8 compares the IEDFs for the unmagnetized case I obtained using both PIC and EST. It is evident that both models are in excellent agreement. We can therefore conclude that it is well justified to use the non-magnetic EST for studying the effect of the magnetic field on IEDFs. Figure 6.9 shows the IEDF for the magnetized case II. The results are again obtained using both PIC and EST. Also for this case the results are in excellent agreement. Only minor deviations can be observed when comparing the magnitudes of the double peak structure. It is important to note that EST yields the same IEDFs as PIC, regardless of whether the discharge is magnetized or non-magnetized.
Figure 6.8: IEDFs obtained for the non-magnetized case (case I) and $\Delta \phi = 0$ using the PIC (top) and the EST model (bottom).

Figure 6.9: IEDFs for the magnetized (case II) and $\Delta \phi = 0$; PIC results are top and EST results are bottom.
The reason is, that both the sheath dynamics and the ion motion in the sheath are mainly driven by the electric field, and only weakly affected by the magnetic field.

6.4 Conclusion

In this work we analyze the effect of a spatially inhomogeneous static magnetic field on the characteristics of capacitively coupled radio frequency discharges, with a focus on the energy distribution function (IEDF) of the ions impinging the electrodes and the sheath dynamics. By means of a one-dimensional Particle-in-Cell algorithm using Monte-Carlo collisions (PIC) we show that geometrically symmetric capacitively coupled radio frequency discharges can be asymmetrized by applying a spatially inhomogeneous static magnetic field. This magnetically induced effect is similar to the electrical asymmetry effect (EAE). We further show that the EAE can be amplified or (over-)compensated. We find that the novel Ensemble-in-Space time scheme (EST) which resolves the sheath only, is able to provide IEDFs almost equal to the ones obtained from the fully self-consistent PIC simulation. Furthermore, we find an excellent agreement of the two models for both cases, the non-magnetized and the magnetized case. We conclude that EST may be used as an efficient post-processing tool to obtain the IEDFs in cases where only simplified (i.e., not kinetic) information is available.
7

EST and Plasma Processing

7.1 Introduction

Capacitive discharges have been widely used for low-pressure materials processing. However, they suffer from a deficiency in their performance; i.e., operate at high sheath voltages with low ion flux and high ion-bombarding energy at a given power level. To circumvent these disadvantages, other sources have been employed and various attempts have been made to improve the performance of the capacitive discharge, for example, reactive magnetron sputtering (RMS). It is a widely used technique to deposit many thin film materials as for examples oxides and nitrides using metal targets and the addition of oxygen and/or nitrogen as a reactive component to the argon plasma gas.

The film stoichiometry and its structure has been found to be a function of the energy per deposited atom $\bar{\varepsilon}$. This energy depends on the ion energy $\varepsilon_i$ and on the ion-to-neutral ratio $\Psi_i/\Psi_{\text{growth}}$ with $\bar{\varepsilon} = \varepsilon_i \cdot \Psi_i/\Psi_{\text{growth}}$ where $\Psi_i$ is the flux of incident ions and $\Psi_{\text{growth}}$ is the total flux of incorporated atoms in the film. The dependence of the crystal orientation of titanium nitride (TiN) on the energy per deposited atom has been proved experimentally by Petrov et al. (122). They varied the ion energy and kept the ion-to-neutral ratio constant. In a second experiment, the ion energy was kept constant and only $\Psi_i/\Psi_{\text{growth}}$ was changed. Different values of the ion energy $\varepsilon_i$ and the ion-to-neutral ratio $\Psi_i/\Psi_{\text{growth}}$ which give the same values of the energy
per deposited atom $\bar{\varepsilon}$ yield quite different thin film properties. Therefore one can conclude that the average energy per atom is not a universal growth parameter.

The exact details of ion-induced surface processes, however, remain still unclear. Although, one can devise an experiment providing a unique energy per deposited atom $\bar{\varepsilon}$ and a constant ion-to-neutral ratio, film growth may still depend on more subtle effects such as the particular IED and/or the temporal sequence of the incident species. In the case of pulsed plasmas and/or pulsed bias concepts, the sequence of adsorption of neutral species and of the ion-induced densification of the film may occur simultaneously or sequentially which may affect structure evolution. The most prominent example in that respect is high power pulsed magnetron sputtering (HPPMS) plasmas, where a high power plasma pulse interacts with a surface for a short time, followed by a long off period, where relaxation may occur and/or neutral species may still adsorb.

In this chapter, we resolve this ambiguity in the interpretation of the experiments by devising an approach, where we keep the average energy per deposited atom $\bar{\varepsilon}$, the average energy of the incident ions $\bar{\varepsilon}_i$, the total ion flux $\Psi_i$ and the ion-to-neutral ratio $\Psi_i/\Psi_{\text{growth}}$ constant, but changing only the IED. This is realized using an RF-magnetron to maintain a constant plasma background assuring a constant stream of neutrals $\Psi_{\text{growth}}$ and ions $\Psi_i$ towards the substrate. At substrate level, variable biasing is applied to modulate the energy and temporal distribution of the ions impinging on the substrate (IED). The measurements are carried out by M. Prenzel and A. von Keudell (123).

We selected aluminum oxide (Al$_2$O$_3$) because of its applications ranging from microelectronics, wear resistant coatings to catalytic surfaces (124). The most common phases of Al$_2$O$_3$ are the $\gamma$- and $\alpha$-phase. $\alpha$ – Al$_2$O$_3$ with its hexagonal closed package (125) structure is often used as hard coating on machining tools. The metastable $\gamma$ – Al$_2$O$_3$ phase exhibits high porosity and low surface energy making it most suitable as absorbent or as catalyst.

With increasing temperature Al$_2$O$_3$ undergoes various phase transitions from an x-ray amorphous phase to $\gamma$ – Al$_2$O$_3$ at 600°C, to $\delta$- or $\theta$-phase at 700-800°C and finally to the stable $\alpha$ phase at 900-1050°C (124). It is known that these thermodynamical transition temperatures can be lowered by applying an additional ion bombardment. Today, more and more applications desire lower deposition temperatures to be able to also coat heat sensitive materials. This dependence of the transition temperatures on the energy distribution of the bombarding ions is exploited in this chapter, by monitoring the transition
from x-ray amorphous to γ-alumina in our experiment to assess the influence of the ion energy distribution on thin film growth.

7.2 Experimental setup

7.2.1 Film deposition
Deposition is performed using a two frequency RF-magnetron sputter experiment. A schematic for the reactive magnetron sputtering (RMS) is shown in Figure 7.1. The inner magnetic field density is higher than the outer magnets; this magnetic configuration refers to a magnetic configuration from type I (126). The target consists of 99.99 pure aluminium and has a diameter of 140 mm. The plasma is powered with 300 W at 71 MHz and with 200 W at 13.56 MHz. The high frequency at 71 MHz supports a high plasma density, whereas the low frequency of 13.56 MHz adjusts the bias potential at the target and thereby the energy of the sputtering ions. An Argon discharge is generated at a pressure of 0.1 Pa using a constant Argon flow rate of 9 sccm. A feedback loop regulates the oxygen flow into the chamber avoiding target poisoning. The feedback loop is based on maintaining a constant intensity of the AlI emission line at 396.2 nm measured by a photomultiplier. Consequently, stoichiometric correct Al₂O₃ coating are prepared. The distance between the target and the Si(100) p-doped substrate is 50 mm. The substrate is heated during the deposition process to a temperature of 500° C, 550° C and 600° C, respectively.

7.2.2 Substrate biasing
The substrate electrode is biased with rectangular waveforms with varying amplitudes $V_{\text{max}}$ and frequencies $f$ between 1 MHz and 1.4 MHz. At this frequency, the ions respond partially to the instantaneous electric field. The bias signal at the substrate electrode is recorded by a VI probe in the RF-feed line. In addition, the DC self-bias $V_{\text{DC}}$ is recorded by low-pass filtering of the RF-signal at the substrate electrode. The deposition is performed using three different rectangular bias waveforms. By adjusting the power of the substrate bias, the DC-self bias varies as well. The parameters are adjusted in a manner so that the average energy of the incident ions $\varepsilon_i$ remains identical irrespective of the chosen frequency $f$. The biasing parameters are directly defining the energy distribution of the incident ions and thereby also the average energy of the ions. This ion energy distribution (IED) has been measured by a retarding field analyzer (RFA) in a pure argon discharge and is being compared to the
Figure 7.1: A schematic of reactive magnetron sputtering set up.
EST model results (more details are present in chapter 5). Perfect agreement has been found.

Due to the low pressures, all sheaths can be considered collisionless, and the bias waveform can be directly converted into an IED. This direct conversion of the bias waveform into an IED has the great advantage that an IED can be also estimated for experimental conditions, where a direct RFA measurement is not possible. In the case of $\text{Al}_2\text{O}_3$ deposition, the substrate is heated and any $\text{Al}_2\text{O}_3$ coating will also create insulating films inside the RFA, making a direct measurement impossible. Since the majority of the ions in our plasma consists of Argon, the estimate of the IED from the data of a pure Argon plasma is reasonable.

The sheath voltage is calculated from the oscilloscope data representing the bias signal at the substrate electrode, the measured floating potential, and the measured plasma potential. Because most of the produced ions in the plasma are Argon ions, we assume $m_i = 40$ amu. Semmler et al. (127) measured the electron temperature with an Automated Probe System (APS3) fully automated Langmuir probe using this setup under similar conditions. Regarding these measurements, we expect an electron temperature of $T_e = 4$ eV. The absolute ion fluxes are determined by measuring the electron density in front of the substrate under deposition conditions, using a Plasma Absorption Probe (PAP) (21). The tip of the probe is mounted in a distance of 23 mm to the target. In the center above the target an electron density of $n_e = 5.8 \times 10^{16}$ m$^{-3}$ is determined. By applying the Bohm criterion an absolute ion flux of $\Psi_i = 13.5 \times 10^{18}$ m$^{-2}$s$^{-1}$ is determined. This is compared to the flux of incorporated species as derived from the growth rate. This yields a constant ion-to neutral flux ratio in our experiment of $\Psi_i/\Psi_{\text{growth}} = 0.24$. The mean energy $\varepsilon_i$ is calculated from the modeled IED.

### 7.3 Thin film analysis

The aluminum oxide films are analyzed by Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD) and profilometry. Ex-situ FTIR transmission measurements of the $s$ component are performed at $70^\circ$ angle of incidence using a Bruker IFS 66/S spectrometer in the wavenumber range between 400 cm$^{-1}$ and 6000 cm$^{-1}$. Non-coated silicon wafers are used to take the background spectra. The infrared spectra are compared to a model for the optical constants established by Chu et al. (128). According to that model,
the optical dielectric function of aluminum oxide is composed of a series of classical Lorentz oscillators:

\[ \epsilon_2(\omega) = \epsilon_\infty + \sum_n \frac{S_n \omega_{l,n}^2}{\omega_{l,n}^2 - \omega^2 - i \omega \gamma_n}, \]  

(7.1)

\( S_n \) is the strength of the oscillation, \( \omega_{l,n} \) the wavenumber of the transversal oscillation, \( \omega_{l,n} \) the wavenumber for the longitudinal oscillation, and \( \gamma \) the damping factor. \( \epsilon_\infty \) describes the contribution of the optical transitions outside the infrared spectral range. Each oscillator consists of a longitudinal and a transversal oscillation. According to this description, crystalline \( \gamma \)-alumina can be identified by a pronounced peak at 950 cm\(^{-1} \) corresponding to the longitudinal (LO) optical mode (129). Amorphous alumina layers exhibit a rather featureless absorption spectrum.

XRD spectra are taken at grazing incidence of 15° using Cu K\( \alpha \) radiation employing a Bruker D8 General Area Detection Diffraction System (GADDS). Voltage and current were adjusted to 40 kV and 40mA, respectively. The scattering angle is varied between 20° and 70°.

Finally, film thickness has been measured by profilometry. From the known density of alumina, the flux \( \Psi_{\text{growth}} \) of incorporated atoms during the deposition time can be calculated.

### 7.4 Results

#### 7.4.1 Variation of the IVDF

Film deposition has been performed at constant plasma parameters of 200 W with 13.56 MHz and 300 W with 71 MHz, but varying bias signal at the substrate electrode. This variation is performed using a rectangular pulsed bias with different maximum voltages \( V_{\text{max}} \) and frequencies \( f \). The rectangular bias consists of two phases, the time \( \tau_{\text{on}} \) when the bias is applied and \( \tau_{\text{off}} \) when the substrate is at floating potential. The duty cycle d.c. corresponds to \( \tau_{\text{on}}/(\tau_{\text{on}} + \tau_{\text{off}}) \) and the frequency \( f \) of the biasing to \( 1/(\tau_{\text{on}} + \tau_{\text{off}}) \). Since the total flux of ions and neutrals remains constant, the average energy per deposited atom can be calculated as:

\[ \bar{\varepsilon} = \varepsilon_{i,\text{max}} \frac{\Psi_i \tau_{\text{on}}}{\Psi_{\text{growth}}(\tau_{\text{on}} + \tau_{\text{off}})} = \varepsilon_{i,\text{max}} \frac{\Psi_i \tau_{\text{on}} f}{\Psi_{\text{growth}}}. \]  

(7.2)

The growth flux \( \Psi_{\text{growth}} \) is calculated from the growth rate of the films. Averaging the IVD tailored by the biasing signal yields the term \( \varepsilon_{i,\text{max}} \tau_{\text{on}} f \).
Equation 7.2 indicates that an identical parameter \( \bar{\varepsilon} \) can be obtained for identical values of \( \Psi_i / \Psi_{\text{growth}} \) and \( \tau_{\text{on}} \) in the growth flux by simultaneously changing \( \varepsilon_{i,\max} \) and \( f \). Then the average energy per incorporated atom \( \bar{\varepsilon} \) is calculated by weighting the average ion energy with the ratio of ion fluxes to growth flux.

The corresponding variation of the substrate bias signal for three frequencies \( f \) are shown Figure 7.2. It can be seen that the maximum negative voltage \( V_{\text{max}} \) at the substrate electrode is simultaneously changed by the \( \tau_{\text{off}} \) period, by adjusting the power of the biasing system for a given frequency \( f \). The different biasing concepts lead to different ion energy distributions, as being modeled using the very same \( VI \) curves as shown in Figure 7.3. One can clearly identify the bimodal structure of the distribution functions. Due to the low pressure any enhancement of the low energy part is almost negligible, and due to the low frequency of the bias in the range of only \( f = 1..1.4 \) MHz, the actual applied voltage almost directly transfers to the energy of the incident ion. The inertia of the ions leading to an averaging of the sheath energies becomes only significant for higher frequencies. One can clearly see, that the IED changes in a very characteristic manner. For low duty cycles or low frequencies, the maximum energy of the ions is higher, but their contribution to the total ion flux is smaller as indicated by the peak height. At higher duty cycles it is the reverse. If one averages the ion energy distribution functions, one obtains in each case an energy around \( \varepsilon_i = 55 \) eV. Together with the growth flux, this results in an average energy per deposited atom of approximately \( \bar{\varepsilon} = 10 \rightarrow 15 \) eV, as calculated from 7.2.

### 7.4.2 Connection between IVDF and thin film properties

Alumina films were deposited using three different schemes with identical \( \bar{\varepsilon} \approx 11 \) eV and \( \varepsilon_i \approx 55 \) eV, but different maximum ion energies \( \varepsilon_{i,\max} \) and duty cycles d.c. or frequencies \( f \). In addition, three different substrate temperatures have been chosen, namely \( 500^\circ \) C, \( 550^\circ \) C and \( 600^\circ \) C. Thereby, the influence of the ion bombardment on the formation of the crystalline \( \gamma \)-phase may become visible by a lower transition temperature from the x-ray amorphous to the \( \gamma \)-phase. It is well known that Alumina film deposition and especially the formation of the different crystalline phases is very sensitive to the water vapor partial pressure during the deposition process (130). Consequently, any poor base pressure may lead to the suppression of the nucleation of certain crystalline phases. Therefore, all experiments have been
Figure 7.2: Time dependent voltage at the substrate electrode for three parameter sets: (a) $f = 1.01$ MHz and $V_{\text{max}} = 110$ eV; (b) $f = 1.2$ MHz and $V_{\text{max}} = 100$ eV; (c) $f = 1.4$ MHz and $V_{\text{max}} = 75$ eV.
Figure 7.3: Ion velocity distribution function for three parameter sets: (a) $f = 1.01$ MHz and $\varepsilon_{i,\text{max}} = 120$ eV; (b) $f = 1.2$ MHz and $\varepsilon_{i,\text{max}} = 100$ eV; (c) $f = 1.4$ MHz and $\varepsilon_{i,\text{max}} = 80$ eV.
performed by properly baking the setup prior to deposition; the base pressure after baking was $2 \times 10^{-5}$ Pa and we expect a water condensation rate below 0.01nms$^{-1}$. In addition, for each set of deposition parameters, three samples were prepared to control the reproducibility of the experiments. The deposited films were analyzed by Fourier Transform Infrared Spectroscopy (FTIR), as shown in Figure 7.4. All films, deposited at 600$^\circ$ C show a pronounced feature at 950cm$^{-1}$ indicating the LO absorption of the $\gamma - \text{Al}_2\text{O}_3$, being characteristic for $\gamma$-crystalline films showing that the alumina films grow in the thermodynamically preferred $\gamma$-phase, irrespective of the biasing scheme. The sharp peak at 950 cm$^{-1}$ is much less pronounced in the spectra of all other films, except the films deposited at a frequency of $f = 1.2$ MHz at 550$^\circ$ C. All FTIR spectra are modelled using four oscillators described by 7.1. A good agreement between model and measured spectrum can be found, shown for a sample with 1.01MHz bias and 600$^\circ$ in Figure 7.4.

The films were also analyzed by XRD as shown in Figure 7.5. The (400) peak at 45.86$^\circ$ and the (440) peak characteristic at 67.03$^\circ$ are identified as $\gamma$-alumina [20]. The results of XRD are consistent with the FTIR results. These peaks are absent in the films deposited at 500$^\circ$ C indicating their x-ray amorphous structure. However, one may already identify small peaks at the positions of the $\gamma$-peaks in the sample with 1.2MHz bias and 500$^\circ$ C in Figure 7.5. Only, the films deposited at 550$^\circ$ C and at $f = 1.2$ MHz show the appearance of the crystalline peaks below the thermodynamics transition temperature from amorphous to crystalline alumina. Finally, All samples exhibit the $\gamma$-alumina peaks at 600$^\circ$ C.

7.4.3 Discussion

The thin film analysis revealed that the biasing scheme at $f = 1.2$ MHz leads to the growth of $\gamma$-alumina already at 500$^\circ$ C or slightly above, which is significantly lower than any temperature for $f = 1.01$ MHz or $f = 1.4$ MHz. This leads to the question about the peculiar nature of the deposition parameters $f = 1.2$ MHz and $\varepsilon_{i,\text{max}} = 100$ eV. The three biasing scheme are different in two aspects, namely the different off times $\tau_{\text{off}}$ and the different maximum ion energies $\varepsilon_{i,\text{max}}$. The flux of neutrals and of ions is $\Psi_{\text{growth}} = 56.1 \times 10^{18}\text{m}^{-2}\text{s}^{-1}$ and $\Psi_{i} = 13.5 \times 10^{18}\text{m}^{-2}\text{s}^{-1}$, respectively.

The possible influence of both parameters on the growth process is discussed in the following:

1. Influence of $\tau_{\text{off}}$: The different off times $\tau_{\text{off}}$ may correspond to different time spans for the material to relax into a crystalline structure in between the periods during $\tau_{\text{on}}$ when energetic ions impact on the surface.
Figure 7.4: Infrared spectra for films deposited at three different substrate temperatures and different biasing schemes: (a) $f = 1.01$ MHz and $\varepsilon_{i,\text{max}} = 120$ eV; (b) $f = 1.2$ MHz and $\varepsilon_{i,\text{max}} = 100$ eV; (c) $f = 1.4$ MHz and $\varepsilon_{i,\text{max}} = 80$ eV. The average energy per incorporated atom $\bar{E} = \langle E \rangle$ is equivalent to 11 eV. The average ion energy $\varepsilon_i$ is 55 eV. A clear correlation between the modelled peak at 950 cm$^{-1}$ in 7.1 and the observed peaks in the spectra has been found. As an exception, here and in Figure 7.5 $\langle E \rangle$ is used to represent the energy per deposited atom.
Figure 7.5: X-ray diffraction spectra for films deposited at three different substrate temperatures and different biasing schemes (a) \( f = 1.01 \text{ MHz} \) and \( \varepsilon_{\text{i,max}} = 120 \text{ eV} \); (b) \( f = 1.2 \text{ MHz} \) and \( \varepsilon_{\text{i,max}} = 100 \text{ eV} \); (c) \( f = 1.4 \text{ MHz} \) and \( \varepsilon_{\text{i,max}} = 80 \text{ eV} \). The average energy per incorporated atom \( \bar{E} \) is equivalent to 11 eV. The average ion energy \( \varepsilon_{i} \) is 55 eV.
One may speculate that the adsorbing neutrals require an appropriate time to reach a preferred crystalline adsorption site. If an incident ion impacts on the surface, the motion of the neutral adsorbing atoms may be disturbed and crystal formation is thus prevented. The total time span of a pulse is of the order of $1\mu s$, resulting in a fluence per pulse of neutral and of ionic species of $10^9 \text{cm}^{-2}$. If one compares this fluence with the typical number of surface sites of $10^{15} \text{cm}^{-2}$, one can conclude that the ions and neutrals impact the surface well apart from each other of the order of 1000 lattice sites equivalent to 30 nm. Any mutual and simultaneous reaction of neutrals and ions might only occur, if either the lateral size of the collision cascade or the travel distance of the neutrals within $1\mu s$ via surface diffusion is larger than these 1000 lattice sites as the average distance in between the impacting species. The adsorbed neutrals may diffuse on the growing film surface during the time span of a $1\mu s$ pulse.

If one assumes a minimum activation barrier for surface diffusion of 1.6 eV as reported for $\alpha - \text{Al}_2\text{O}_3$, the adsorbed neutrals are estimated to diffuse a few nanometers within a $1\mu s$ pulse. This may be compared with the surface regions that are affected by the collision cascades of the incident ions. TRIM simulations reveal that the lateral size of the collision cascade for argon ions with an energy of a few 100 eV is also only of the order of a few nanometers. Consequently, the impact of neutrals and of ions during one d.c. are isolated events well apart at the growing film surface. One may consider these events, therefore, uncorrelated and any detailed influence of the timing of the off time can thereby be excluded.

2. Influence of $\varepsilon_{\text{max},i}$: The different maximum ion energies result also in different collision effects upon impact at the growing film surface. To quantify the ion-induced effects, we performed TRIM calculations to assess the energy dissipation of the incident Argon ions during thin film growth in the energy range between 30 and 100 eV. In Figure 7.6, one can see that the displacements per ions increase with energy and reach a value of 0.8 per incident ion and 0.2 nm depth interval at the film surface at an energy of 100 eV. In total, one surface atom is displaced by an incident argon ion at 100 eV within the penetration depth of the incoming ions. The ion-to-neutral during film growth is 1/5. One may argue that the displacement of one surface bonded atom causes the rearrangement of this atom including also its next neighbors. Additionally, an optimum ratio between displacements per ion and the activation of surface atoms appears to be reached at an Ar ion energy of 100 eV.
**Figure 7.6**: Displacements per incident ions and depth interval of 0.2 nm for Argon bombardment of $\text{Al}_2\text{O}_3$ (solid symbol). Implantation of 100 eV argon ions in $\text{Al}_2\text{O}_3$ (open symbols).
Larger ion energies may result in re-sputtering while lower energies result in a reduced number of displacements per ions and hence a reduced adatom mobility. Thereby, the displacement events at the growing film surface being proportional to $\Psi_i$ are able to affect all incorporated atoms during film growth given by $\Psi_{\text{growth}}$.

### 7.5 Conclusion

$\text{Al}_2\text{O}_3$ films were deposited by reactive magnetron sputtering using arbitrary substrate biasing. By choosing a rectangular bias with varying frequency and varying maximum voltage an experiment is devised where the average ion energy, the average energy per incorporated atom and the fluxes of ions and neutrals remain constant. Only the ion velocity distribution (IVD) is varied. We observed a reduction in the substrate temperature necessary for the formation of $\gamma$-alumina films from 600° C to almost 500° C for a frequency of the rectangular biasing of 1.2 MHz, with a resulting rather mono-energetic ion energy distribution of the ions impacting on the surface with $\varepsilon_{i,\text{max}} = 100$ eV. Based on TRIM calculations, this corresponds to a situation, where each surface bond atom is rearranged exactly one time during film growth which apparently promotes the formation of $\gamma$-alumina films.
Conclusion and Outlook

8.1 Conclusion

The kinetic calculation of the sheath dynamics and the ion bombarding energy and the ion angular distribution have been the scope of this thesis. Although the modeling of the sheath dynamics and the calculation of the ion energy distribution functions (IEDFs) and the ion angular distribution functions (IADFs) began several decades ago, we have seen that there is no simulation tool provides a kinetically self-consistent solution that is at the same time computationally efficient. Thus, in this thesis we have presented a mathematical model that enables the efficient, kinetically self-consistent simulation of radio-frequency (RF) modulated plasma boundary sheaths in all technically relevant discharge regimes.

This model treats the ions kinetically and assumes Boltzmann’s relation for the electrons in addition to Poisson’s equation for the electrical field. Boundary conditions specify the ion flux at a point deep in the plasma bulk and a periodically, though not necessarily harmonically, modulated sheath voltage $V_{sh}$ or sheath charge $Q(t)$. These boundary conditions could be imported from fluid models (such as HPEM (28), nonPDPsim (110), Comsol (111), CFD-ACE+ (112)), global simulations, or experiments. The model equations are solved in a statistical sense using the ensemble in space-time (EST) iterative algorithm. The self-consistency achieved via three modules, termed sequentially the ion Monte Carlo module, the ion density harmonic analysis module, and the field module. The third module, the field module, finally solves the Poisson-Boltzmann equation with the calculated ion densities to
generate an updated set of potential values for the spatio-temporal grid. The iteration is started with the potential values of a self-consistent fluid model and terminates when the updates become sufficiently small. A subsequent post-processing determines important quantities, in particular the phase-resolved and phase-averaged values of the ion energy and angular distributions and the total energy flux at the electrode. A drastic reduction of the computational effort compared with PIC calculations is therefore achieved. The model is applicable for virtually all important parameter regimes and has no practically relevant restrictions regarding the amplitude, waveform, and frequency of the applied RF, the number of the ion species, and the pressure and composition of the neutral background.

A comprehensive article describing the EST model has been published (93). To ensure the validity of the results of EST model, we compared the model results with analytical models (93, 101, 102), PIC simulations (115, 119), and experimental results (100). As a post-processing tool: The EST model has been used to reveal the effect of arbitrary RF waveforms on the characteristic of deposited thin films in reactive magnetron sputtering. The tailored IEDFs have been found to affect on the temperature required to promote the formation of γ-Alumina phase (123).
8.2 Outlook

When compared to experiments and another valid theoretical models, the EST model shows a good agreement. The EST model resolves the ion kinetics and covers the sheath and the presheath. However, the results of the research suggest that there are opportunities for additional research and for improvement of the model to extend the applicability of the model and make it more realistic. The current EST model has several limitations. First, it neglects ionization and excitation in the sheath and the presheath. This limits the model to cases where the sheath is small in comparison to the electrode spacing. This is a crucial point, because several technologically interesting plasmas, such as those used in industry, are at low pressure and exhibit narrow electrode spacing. The sheath regions of these plasmas are of the same order as the electrode spacing.

A second limitation of the EST model is the assumption of Boltzmann equilibrium for electrons. This is a particularly crucial point, because the electron kinetics is mandatory in understanding different phenomena such as plasma heating. Moreover, the assumption of Boltzmann equilibrium for electrons limits the EST model to those frequencies lower than the electron plasma frequency.

A third area for improvement in the EST model is to extend it in order to simulate the whole discharge. The ability of the EST model to calculate the sheath dynamics without the necessity of simulating the whole discharge is a great advantage, and one that is required in the context of plasma technology. However, covering the whole discharge in an efficient manner is a fundamental topic for future research. The strategy of the EST model can be applied to enable fast kinetic simulation for the whole discharge.

A fourth improvement would be the generalization of the model to two dimension. This would be very important for different technologically interesting plasmas. When the equipotential surfaces over the substrate is curved and not parallel to the surface of the substrate, the ion flux and the sheath held are 2D spatially dependent. In such cases to calculate real IEDFs and IADFs, the EST model must be two dimensional model.
## Symbols and Abbreviations

### Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>$A$</td>
<td>represents an area</td>
</tr>
<tr>
<td>$\vec{B}$</td>
<td>magnetic flux vector</td>
</tr>
<tr>
<td>$B$</td>
<td>magnitude of magnetic flux</td>
</tr>
<tr>
<td>$d_0$ and $d_1$</td>
<td>nonlinear eigenvalues in Brinkmann AAA model</td>
</tr>
<tr>
<td>$\vec{E}$</td>
<td>electric field vector</td>
</tr>
<tr>
<td>$E$</td>
<td>electric field magnitude</td>
</tr>
<tr>
<td>$\bar{E}$</td>
<td>time averaged electric field</td>
</tr>
<tr>
<td>$e$</td>
<td>electron charge</td>
</tr>
<tr>
<td>$F$</td>
<td>Force</td>
</tr>
<tr>
<td>$f$</td>
<td>frequency</td>
</tr>
<tr>
<td>$f(\vec{r}, \vec{v}, t)$, $f_s(\vec{r}, \vec{v}, t)$, or $f_s$</td>
<td>distribution function in phase space</td>
</tr>
<tr>
<td>$f(v)$</td>
<td>ion velocity distribution function</td>
</tr>
<tr>
<td>$g$ and $g'$</td>
<td>pre-collision and post-collision relative velocity</td>
</tr>
<tr>
<td>$g(\varepsilon)$</td>
<td>ion energy distribution function</td>
</tr>
<tr>
<td>$J$</td>
<td>surface current</td>
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<tr>
<td>$J_i$</td>
<td>ion surface current</td>
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<tr>
<td>$m$</td>
<td>particle mass</td>
</tr>
<tr>
<td>$m_e$</td>
<td>electron mass</td>
</tr>
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</table>
$m_i$ ion mass
$N_g$ gas density
$\bar{n}$ a reference density
$n_e$ electron density
$n_i$ ion density
$P_c$ represents a collision probability
$P_k$ represents a collision probability of event $k$
$P_{null}$ represents a collision probability
$p$ pressure
$Q$ unbalanced charge in the sheath
$q$ charge coordinate
$r_L$ Larmor radius
$R$ random number
$r$ position vector ($x\hat{a}_x + y\hat{a}_y + z\hat{a}_z$)
$S^*$ artificial ionization source
$s(t)$ the temporal sheath width
$\bar{s}$ the time averaged sheath width
$T$ period of a periodic function
$T_e$ electron temperature
$T_i$ ion temperature
$t$ time
$u$ fluid velocity (average velocity)
$u_d$ drift velocity
$V_{sh}$ sheath voltage
$\bar{V}_{sh}$ the DC component of the sheath voltage
$\tilde{V}_{sh}$ the amplitude of the sheath voltage
$\vec{v}$ velocity vector
$v$ or $V$ the magnitude of the velocity vector
$v'$ or $V'$ the magnitude of the post-collision velocity vector
$x$ one dimension spatial space
$x_B$ position of a point in the bulk
$x_E$ position of electrode or substrate
$\alpha$, $\gamma$, $\delta$, and $\theta$ refer to $Al_2O_3$ phases and they also used to represent other quantities as shown below.
\( \Gamma_0 \) RF period averaged value of \( \Xi_0 \)
\( \Gamma_1 \) RF period averaged value of \( \Xi_1 \)
\( \gamma \) the ratio of the specific heat at constant pressure to that at constant volume
\( \Delta \varepsilon \) width of the ion energy distribution
\( \delta n_i \) first order perturbation of ion density
\( \delta E \) first order perturbation of electric field
\( \delta u_i \) first order perturbation of the ion average velocity
\( \delta \Phi \) first order perturbation of potential
\( \delta(x) \) Dirac delta function of \( x \)
\( \epsilon_0 \) dielectric constant or vacuum permittivity
\( \varepsilon \) energy
\( \bar{\varepsilon} \) energy per deposited atom
\( \varepsilon_i \) ion energy
\( \vartheta \) scattering angle
\( \lambda_D \) Debye length
\( \mu_0 \) vacuum permeability
\( \mu \) mobility or vacuum permeability
\( \nu \) collision frequency
\( \nu' \) maximum collision frequency
\( \nu_i \) ion collision frequency
\( \nu_k \) collision frequency of event \( k \)
\( \Xi_0(\xi) \) special function constructed from \( \Psi_0'(\xi) \)
\( \Xi_1(\xi) \) special function constructed from \( \Psi_1'(\xi) \)
\( \xi \) a dimensionless parameter in Brinkmann AAA model
\( \Pi \) stress tensor
\( \rho \) volume charge density
\( \sigma \) cross section
\( \tau_i \) ion transit time
\( \tau_{RF} \) radio-frequency period
\( \Phi \) potential
\( \phi \) phase difference
\( \varphi \) azimuthal angle
\( \chi \) deflection angle
\( \Psi_i \) \hspace{1em} \text{ion flux}

\( \Psi_{\text{growth}} \) \hspace{1em} \text{growth flux}

\( \Psi_0(\xi) \) \hspace{1em} \text{special function used by Brinkmann sheath model}

\( \Psi'_0(\xi) \) \hspace{1em} \text{derivative of } \Psi_0(\xi)

\( \Psi_1(\xi) \) \hspace{1em} \text{special function used by Brinkmann sheath model}

\( \Psi'_1(\xi) \) \hspace{1em} \text{derivative of } \Psi_1(\xi)

\( \Omega_i \) \hspace{1em} \text{ion cyclotron frequency}

\( \omega_{\text{ion}} \) \hspace{1em} \text{ion transit frequency}

\( \omega_{\text{pe}} \) \hspace{1em} \text{electron plasma frequency}

\( \omega_{\text{pi}} \) \hspace{1em} \text{ion plasma frequency}

\( \omega_{\text{RF}} \) \hspace{1em} \text{radio frequency in radians/second}

\( \bar{x} \) \hspace{1em} \text{\( x \) is a vector}

\( \bar{x} \) \hspace{1em} \text{time averaged of } x \text{ or the zero order of } x

\( \hat{x} \) \hspace{1em} \text{\( x \) is a reference}

\( \Xi \) \hspace{1em} \text{\( x \) is a tensor}

**Subscripts** (in most cases)

<table>
<thead>
<tr>
<th>Subscript</th>
<th>Description</th>
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</thead>
<tbody>
<tr>
<td>B</td>
<td>related to the plasma bulk</td>
</tr>
<tr>
<td>c</td>
<td>related to collision</td>
</tr>
<tr>
<td>E</td>
<td>related to the electrode</td>
</tr>
<tr>
<td>e</td>
<td>related to electron</td>
</tr>
<tr>
<td>i</td>
<td>related to ion</td>
</tr>
<tr>
<td>max</td>
<td>related to a maximum of a function</td>
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<tr>
<td>RF</td>
<td>related to radio frequency</td>
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<tr>
<td>SB</td>
<td>related to self bias</td>
</tr>
<tr>
<td>s</td>
<td>related to species</td>
</tr>
<tr>
<td>Abbreviations</td>
<td>Description</td>
</tr>
<tr>
<td>--------------------------</td>
<td>-----------------------------------------------------------------------------</td>
</tr>
<tr>
<td>AAA</td>
<td>the Advanced Algebraic Approximation model</td>
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<tr>
<td>CCP</td>
<td>capacitive coupled plasma</td>
</tr>
<tr>
<td>CFD-ACE</td>
<td>a commercial plasma simulation tool</td>
</tr>
<tr>
<td>Comsol</td>
<td>a commercial plasma simulation tool</td>
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<td>DC</td>
<td>direct current</td>
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<tr>
<td>EST</td>
<td>Ensemble-in-Spacetime</td>
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<td>FTIR</td>
<td>Fourier transform infrared</td>
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<td>GPU</td>
<td>graphics processing unit</td>
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<td>HPEM</td>
<td>Hybrid plasma equipment model via M. Kushner and co-workers, University of Michigan</td>
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<tr>
<td>HPPMs</td>
<td>high power pulsed magnetron sputtering</td>
</tr>
<tr>
<td>IADF</td>
<td>ion angular distribution</td>
</tr>
<tr>
<td>IEDF</td>
<td>ion energy distribution</td>
</tr>
<tr>
<td>MERIE</td>
<td>magnetically enhanced reactive ion etchers</td>
</tr>
<tr>
<td>MCC</td>
<td>Monte Carlo simulation</td>
</tr>
<tr>
<td>nonPDPsim</td>
<td>plasma simulation tool developed via M. Kushner and co-workers, University of Michigan</td>
</tr>
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<td>PIC</td>
<td>Particle-in-Cell</td>
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<td>RFA</td>
<td>retarding field analyzer</td>
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<tr>
<td>RF</td>
<td>radio frequency</td>
</tr>
<tr>
<td>RMS</td>
<td>reactive magnetron sputtering</td>
</tr>
<tr>
<td>TRIM</td>
<td>Monte Carlo simulation code</td>
</tr>
<tr>
<td>XRD</td>
<td>X-ray diffraction</td>
</tr>
<tr>
<td>1D, 2D, and 3D</td>
<td>one, two, and three dimension</td>
</tr>
<tr>
<td>1d3v</td>
<td>one spatial dimension and three velocity dimension</td>
</tr>
</tbody>
</table>
Collision Processes in EST

Typically, in low temperature plasmas, the neutral particle density is approximately $10^6$ times the ion density and the electron density. Moreover, the ion-neutral cross sections are greater than the electron-neutral cross sections, as has been shown by Vahedi et al (38). Therefore, it is safe to neglect the electron-ion, electron-electron, ion-ion (i.e., coulomb collisions) and elastic electron-neutral collisions in the sheath compared to the ion-neutral collisions. In addition, the excitation, de-excitation, ionization, recombination processes would be neglected in the plasma sheath. Only ion-neutral elastic scattering and charge exchanges collisions are significant. Let assume an elastic collision between an ion and neutral particle. The ion mass is $m$ and its pre-collision velocity is $v$. The neutral particle mass and its pre-collision velocity are given as $M$ and $V$, respectively. The relative velocity is given by $g = v - V$. The post-collision velocities and relative velocity are $v'$, $V'$, and $g' = v' - V'$, respectively. Note that $g = g'$. It is convenient to carry out the simulation in the center of mass system where the determination of the post-collision velocities reduces to the calculation of the change in direction of the relative velocity, i.e., the deflection angle of the relative velocity $\chi$. The post-collision velocities $(v', V')$ of the ion and the neutral particle without the charge-exchange can be read as (131):

$$ v' = v - \frac{m}{m + M} [g(1 - \cos \chi') + h \sin \chi'], \quad (10.1) $$

$$ V' = V + \frac{m}{m + M} [g(1 - \cos \chi') + h \sin \chi']. \quad (10.2) $$
The Cartesian components of $h$ are

\[ h_x = (g^2 - g_x^2)^{1/2} \cos \varphi, \] (10.3)

\[ h_y = -\frac{g_x g_y \cos \varphi + gg_z \sin \varphi}{(g^2 - g_x^2)^{1/2}}, \] (10.4)

\[ h_z = -\frac{g_x g_z \cos \varphi - gg_y \sin \varphi}{(g^2 - g_x^2)^{1/2}}, \] (10.5)

where $\varphi$ denotes the azimuthal angle of the collision plane. In a charge exchange, an electron is assumed to hop from the neutral particle onto the ion, causing the neutral particle to become an ion with zero velocity in the neutral particle frame. After transferring back to the laboratory frame, the ion leaves the collision with the velocity of the incident neutral particle, and the new neutral takes the velocity of the incident ion. When a charge exchange occurs, $V'$ is the ion velocity and $v'$ is the neutral particle velocity. In the case of isotropic scattering (i.e., the post-collision velocities take random directions), the deflection angle which is in the interval $[0, \pi]$ can be randomly sampled by:

\[ \cos \chi = 1 - 2R. \] (10.6)

Also the probability distribution of $\varphi$ is uniformly distributed on $[0, 2\pi]$ and can be sampled regarding to a random number $R$:

\[ \varphi = 2\pi R. \] (10.7)

Nevertheless, for ions at low energy scattering against neutrals, the dominant process is relatively short-range polarization scattering. The scattering becomes more Coulomb-like, but with the impact parameter at an atomic radius. An incoming ion can polarize the atom by repelling or attracting the charge cloud quasistatically. More details could be found elsewhere (34, 132).

The ion collision frequency term $\nu_i$ in the fluid model must be chosen compatible with the kinetic representation of the collisional interactions in the ion Monte Carlo module. We require the drift velocity of the expression

\[ \frac{q_i}{m_i} E = \nu_i(u_i)u_i, \] (10.8)

to coincide with the results of a zero-dimensional Monte Carlo calculation. In general we find that $\nu_i(u_i)$ is represented well by

\[ \nu(u_i) = \frac{1}{\lambda_i} \sqrt{u_0^2 + u_i^2}, \] (10.9)
where \( u_0 \) represents Langevin interaction (polarization scattering) which is dominant for small reduced electric field, i.e., the ratio of the electric field to the plasma density. \( u_i \) and \( \lambda_i \) are the ion speed and the ion mean free path, respectively. The given representation of the friction term provides not only a agreeable choice regarding the consistency of the fluid dynamic description and the Monte Carlo description, it also leads to the experimentally observed (133) and theoretically calculated (134, 135) behavior of the drift velocity,

\[
    u_d \propto E/N_g \quad \text{when } E/N_g \to 0, \\
    u_d \propto \sqrt{E/N_g} \quad \text{when } E/N_g \to \infty. \tag{10.10}
\]

In Figure 10.1, the ion drift velocity as a function of reduced field is shown graphically for Argon ions in Argon. The red solid line represents the results of 10.8 and the blue solid line represents the results of the 0D Monte Carlo simulation. In the Monte Carlo module, the ion drift velocity is calculated in such a way similar to real drift tubes, in which a uniform electric field is assumed along a tube filled with Argon gas. The gas pressure is 20 mTorr and the gas temperature 300 K. The cross sections of elastic scattering and the charge exchange are taken from (117). A number of ions are released at the side of the tube to which the electric field points. The ions start the motion assuming Maxwell distribution for the ion velocity. The ions drift in the gas towards the other end of the tube. Ions that reflected back to the entrance of the tube are restarted in order to keep the ion flux constant. The drift velocity is simply calculated via \( u_d(E/N_g) = NL/\sum_{k=1}^{N} t_k \), where \( L \)
Figure 10.2: The ion energy distributions at different pressures.

Figure 10.3: The ion angular distributions at different pressures.
is the tube length and holds $L \gg \lambda_\text{i}$, $t_k$ is the ion travel time along the tube, and $N$ the number of ions. In the fluid module, $u_0 = 195\text{ms}^{-1}$ and $\lambda_\text{i} = 1/1.5 \times 10^{-18} N_\text{g}$ m have been found as a good fitting parameters with the kinetically calculated ion drift velocity, where $N_\text{g}$ is the gas density.

In order to demonstrate the impact of ion-neutral collisions, we display here the calculated IED and IAD as a function of the gas pressure. The other parameters are kept constant; $\omega_{\text{RF}} = 1 \text{ MHz}$, $T_e = 3 \text{ eV}$, the ion flux is $1.2 \text{ Am}^{-2}$, and the same sheath potential. Figure 10.2 shows the IED from collisionless plasma sheaths with pressure of 5 mTorr to collisional plasma sheaths. The bimodal shape disappears as a consequence of the ion collisions. More peaks at lower energies are expected at higher pressures due to the charge exchange collision. In addition, the anisotropy of the ions hits the electrode decreases by increasing the gas pressure. This is clearly seen in the spread of the IAD in Figure 10.3 by increasing the gas pressure.
An Overview of the EST Algorithm

The final version of the EST model has been written in C++. The code is written in approximately 5000 lines and totals 170 pages. Therefore, it is not practical to include it in this thesis. However, here we will simply focus on the structure and the features of the code. The code is compiled using a Makefile with the operating system Ubuntu and a g++ compiler. The Makefile calls three cpp files named main.cpp, mu_bsfunctions.cpp, and TrajectorySolver.cpp, as well as a set of header files. It is worth noting that the cpp files and header files could be gathered in one c++ file. However, separate executable files are preferred to facilitate the handling of the code. The file main.cpp exists mainly to set up the simulation parameters such as the ion flux, the RF frequency, the gas pressure, the electron temperature, and the sheath potential or the sheath current. Also to define whether the simulation parameters are manually entered or if a tables should be used. Moreover, main.cpp is the main executable file which call the functions, structures, and templates. The file mu_bsfunctions.cpp mainly contains the definition of the Advanced Algebraic Approximation model functions. Finally, the file TrajectorySolver.cpp contains the Monte Carlo module, the ion Lagrangian tracker, and the ion density and the ion flux mapping functions. The output of the code is written in several *.dat files using the SI unites such as the ion density, the electron density, electric field, potential and so forth. The output structure can be easily reformatted to suit 2D plot, Splot, and 3D plot. There are different examples to explain the formatting of the output structure. The
data can be plotted utilizing Gnuplot and Python (Free plotting softwares) or commercial tools as MATLAB or Mathematica. A set of Gnuplot and Python manuscripts are already in use. The code contains various numerical tools such as the damped Newton method, which is a globally convergent method for nonlinear systems of equations, the Range-Kutta scheme, the hybrid Newton-Raphson Bisection method, and so forth. To reduce the computational cost of the model various discretization levels were used. Hence, one can independently control the smoothness and the resolution of the solution of each part of the code.
12

Publications

12.1 Journal publications

12.1.1 Refereed journal publications


12.1.2 Submitted journal publications


12.2 Papers in preparation


12.3 Conference proceedings

12.4 Conferences

12.4.1 Oral contributions

(short list)


12.4.2 Poster contributions

(short list)


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Declaration

I herewith declare that I have produced this paper without the prohibited assistance of third parties and without making use of aids other than those specified; notions taken over directly or indirectly from other sources have been identified. The chapters (3-7) base essentially on my publications and my scientific cooperations. This thesis has not previously been presented in identical or similar form to any other German or foreign examination board. The thesis work was conducted from 2009 to 2013 under the supervision of Prof. Dr. rer. nat Ralf Peter Brinkmann at the Institute of Theoretical Electrical Engineering, Ruhr-Universität Bochum.

Bochum, Jun 18th, 2013
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Praise be to Allah, Lord of the Worlds.