Kinetic simulation of the sheath dynamics in the intermediate radio frequency regime

M Shihab\textsuperscript{1,4}, A T Elgendy\textsuperscript{1}, I Korolov\textsuperscript{2}, A Derzsi\textsuperscript{2}, J Schulze\textsuperscript{3}, D Eremin\textsuperscript{1}, T Mussenbrock\textsuperscript{1}, Z Donkó\textsuperscript{2} and R P Brinkmann\textsuperscript{1}

\textsuperscript{1} Ruhr-University Bochum, Institute for Theoretical Electrical Engineering, D-44801 Bochum, Germany
\textsuperscript{2} Institute for Solid State Physics and Optics, Wigner Research Centre for Physics, Hungarian Academy of Sciences, Budapest, Hungary
\textsuperscript{3} Institute for Plasma and Atomic Physics, Ruhr-University Bochum, Germany
\textsuperscript{4} Department of Physics, Faculty of Science, Tanta University, Tanta 31527, Egypt

E-mail: Mohammed.Shihab@ruhr-uni-bochum.de

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Abstract

The dynamics of temporally modulated plasma boundary sheaths is studied in the intermediate radio frequency regime where the applied radio frequency and the ion plasma frequency (or the reciprocal of the ion transit time) are comparable. Two fully kinetic simulation algorithms are employed and their results are compared. The first is a realization of the well-known particle-in-cell technique with Monte Carlo collisions and simulates the entire discharge, a planar radio frequency capacitively coupled plasma with an additional ionization source. The second code is based on the recently published scheme Ensemble-in-Spacetime (EST); it resolves only the sheath and requires the time-resolved voltage across and the ion flux into the sheath as input. Ion inertia causes a temporal asymmetry (hysteresis) of the charge–voltage relation; other ion transit time effects are also found. The two algorithms are in good agreement, both with respect to the spatial and temporal dynamics of the sheath and with respect to the ion energy distributions at the electrodes. It is concluded that the EST scheme may serve as an efficient post-processor for fluid or global simulations and for measurements: it can rapidly and accurately calculate ion distribution functions even when no genuine kinetic information is available.

(Some figures may appear in colour only in the online journal)

1. Introduction

Plasma-enhanced surface modification employs the energy of plasma-generated particles. For industrial applications, computer-aided design of such processes, e.g. the calculation of the flux, energy distribution, and angular distribution of the surface-incident particles, is therefore of great importance [1, 2]. In theory, this poses no problems, as the basis of plasma simulation is sound [3–5]. All species of a discharge—electrons, ions, neutrals—can be described kinetically, i.e. by time evolution equations for the six-dimensional distribution functions \( f_s(\vec{r}, \vec{v}) \) in the domain \( V \times \mathbb{R}^3 \) (\( V \subset \mathbb{R}^3 \) is the spatial domain, i.e. the discharge chamber; \( \mathbb{R}^3 \) is the set of all velocities, and \( s \) is a species index).

In practice, however, the problems are considerable. The numerical effort of directly solving kinetic plasma models is definitely daunting, at least today and in 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 with Monte Carlo collisions (PIC/MCC), are then feasible and indeed yield a lot of insight [6, 7]. However, this approach cannot capture the often complicated geometries of real discharges.

If the latter requirement is an issue, alternatives to kinetic models must be sought. One possibility is the fluid
approach where 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, often kinetic aspects of the problem are significant and must be retained. This is the realm of ‘hybrid schemes’ which combine fluid and kinetic arguments 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 [8–10].

Our study is motivated by a particular type of hybrid scheme, namely one that couples a fluid description of the plasma bulk with a kinetic model of the plasma sheath. From the following ordering which is typical for processing (where $L$ is the discharge size, $l_{\text{ion}}$ is the ionization length, $\lambda$ is the ion mean free path and $s$ is the sheath thickness), one can infer that it is solely the plasma bulk that determines the fluxes of the energetic species to the wall (fluid quantities), while it is solely the sheath that determines the energy and angular distribution functions at the surface (kinetic quantities):

$$L \approx l_{\text{ion}} \gg s \approx \lambda.$$  

To determine the energy and angular distribution of the surface-incident particles, a hybrid scheme should therefore suffice. Most simply, it may be implemented as follows: a fluid code (with respect to the ions) is run to convergence. The fields of the final state (or, in the radio frequency (RF) case, of the final RF 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 distribution 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 the Kushner group [10, 11] or the commercial codes COMSOL and CFD-ACE+ [12, 13]. The scheme has, however, one essential drawback: it is not self-consistent on the kinetic level. To correct this deficiency, we have recently proposed a novel scheme, the Ensemble-in-Spacetime (EST), which constructs a uniform solution of the kinetic equations for the particles and Poisson’s equation for the field, i.e. it provides a fully self-consistent kinetic simulation of the sheath.

The set-up of our study is shown in figure 1. It is a single-frequency capacitively coupled plasma driven at a voltage of $V_0 = 100 \, \text{V}$ and radio frequency of $\omega_{\text{RF}} = 2\pi \times 0.5 \, \text{MHz}$. The electrode gap is $d = 2 \, \text{cm}$, the gas is argon at a pressure of $p = 1 \, \text{Pa}$ and a fixed temperature of $T = 300 \, \text{K}$. All particles that 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 need for this additional source is purely technical: single-frequency capacitively coupled plasmas (CCPs) with gaps in the cm range cannot be sustained at the assumed pressure and frequency. An alternative would have been to simulate 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 [18]. It is a one-dimensional planar (1d3v) bounded implementation, which incorporates a Monte Carlo treatment of collision processes with the cross sections taken from [19, 20]. The additional bulk ionization source $S^*$ is modeled by electron–ion pair creation in a Gaussian-shaped 2.5 mm-wide region at the center of the discharge; the average energy of the new electrons is $3 \, \text{eV}$. Figure 2 shows the obtained time-averaged density profiles. Figure 3 shows the time-resolved electrical potential, where the characteristics of a quasineutral bulk with a relatively weak electrical field and of electron-depleted sheaths with much higher voltage drop can be observed. The plasma bulk has a positive potential relative to the electrodes, except for brief moments in the RF cycle,
where electrons can reach the electrodes to balance the ion flux \[\bar{n}_i(x)\].

The second algorithm studied in this paper, EST, was specially 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 \(\Phi\) as shown in figure 4, the ion flux is \(\bar{n}_i(x)\), the dashed line denotes the phase-averaged electron density \(\bar{n}_e(x)\).

![Figure 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 denotes the phase-averaged electron density \(\bar{n}_e(x)\).](image1)

![Figure 3. The potential along the discharge at different phases within the RF cycle (dashed \(\phi = 0\), solid \(\phi = \pi/2\), and short-dashed \(\phi = \pi\)).](image2)

above the sheath edge, and \(\tau_{RF} = 2\pi/\omega_{RF}\) is 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 consistent fluid sheath model [22–24]. Then, three modules are iterated.

(i) A Monte Carlo module finds the trajectories of the 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 collisions with isotropic and backscattering components with a spatial uniform background gas density are performed in the same way as in the PIC code. The set of all trajectories represents the response of the ions to the field.

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

(iii) 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 voltage.

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.

3. Results and discussion

In spite of their different mathematical structures, 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.

Figure 5 shows the comparison of the phase-averaged ion densities provided by the two simulation methods. The largest deviation is seen in the bulk, increasing toward the discharge center. We believe that this deviation is due to the relatively low argon pressure \((p = 1\ Pa, \text{ where } \lambda \approx 0.7\ cm)\), and it illustrates
one of the limitations of EST. The ions in the PIC simulation are mostly born with room temperature in the ionization zone shaded in figure 5; they are initially not in drift equilibrium (which takes about one ion mean free path \( \lambda \) to establish). The ions in the EST model, on the other hand, are already started in full drift equilibrium. (Any more specific initial distribution would constitute the use of non-fluid dynamic information which the EST scheme tries to avoid.) In other words: for \( \lambda \approx L \), where (1) is marginally violated, also the bulk exhibits kinetic effects which are not captured by EST.

In the sheath itself, however, the named effect does not play a role, as the electric field is much larger and the ions in both PIC and EST 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 ion–neutral collisions, and due to the fact that the EST scheme assumes the electrons in Boltzmann equilibrium, while PIC treats them kinetically. (We checked a third possible cause for differences: EST uses a noise-reducing Fourier scheme to reconstruct the ion densities from the trajectories in space–time; PIC has only access to the instantaneous state and cannot remove any noise. However, this effect turned out not to be influential.) The good agreement carries over to other sheath quantities: figure 6 shows the ion energy distributions at the electrode, figure 7 the phase-

resolved sheath thickness \( s(t) \), calculated using the definition of [23] and figure 8 the charge–voltage characteristics. The results of EST (solid) and of PIC/MCC (short-dashed) are nearly identical.

As discussed in the introduction, it is in particular the faithful representation of the ion energy distribution at the electrode that legitimizes EST as a post-processor. Physically, however, the charge voltage characteristics of figure 8 and its pronounced hysteresis are even more interesting: these curves demonstrate that both models capture ion kinetic effects correctly.

To understand the argument, we consider the space–time trajectories of an ensemble of ions as displayed in figure 9. Here, for simplicity, thermal spread—ion temperature—is neglected and ion–neutral collisions are turned off. The ions enter the interval with Bohm speed and with uniform phase distribution. As long as they are outside the sheath, they experience no electric field and are not accelerated. This motion translates into a temporally constant ion density, see figure 10. However, once the ions cross the momentaneous sheath edge \( s(t) \), they get accelerated and quickly drawn to the electrode. For \( \omega_{\text{RF}} \approx \omega_{\text{pi}} \), this effect is not temporally symmetric; more ions are collected when the sheath expands than when it retracts. The ion flux in the sheath is thus modulated, and so is the ion density \( n_i(x, t) \), see figure 10.
Clearly, the hysteresis vanishes both for \( \omega_{\text{RF}} t/\tau \) and \( \omega_{\text{RF}} \). The phenomenon will surely have an impact on the overall behavior of the discharges. It will be interesting to reconsider the studies on the plasma series resonance [25–28] and stochastic heating [29–31] in the regime \( \omega_{\text{RF}} \approx \omega_{\text{pi}} \).

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### References

[1] Hamaguchi S 1999 *IBM J. Res. Dev.* **43** 199
[2] Makabe T and Petrović Z 2006 *Plasma Electronics: Applications in Microelectronic Device Fabrication* (London: Taylor and Francis) chapter 12
[3] Mussenbrock T 2012 *Contrib. Plasma Phys.* **52** S571
[4] Donkó Z 2011 *Plasma Sources Sci. Technol.* **20** 024001
[5] Kim H C, Iza F, Yang S S, Radmilović-Radjenović M and Lee J K 2005 *J. Phys. D: Appl. Phys.* **38** R283
[6] Birdsall C K and Langdon A B 1991 *Plasma Physics via Computer Simulation* (London: Institute of Physics Publishing)
[7] Birdsall C K 1991 *IEEE Trans. Plasma Sci.* **19** 65
[8] Kratzer M, Brinkmann R P, Sabisch W and Schmidt H 2001 *J. Appl. Phys.* **90** 2169
[9] Salabas A and Brinkmann R P 2005 *Plasma Sources Sci. Technol.* **14** S53
[10] Kushner M J 2005
[11] Kushner M J 2005 *J. Phys. D: Appl. Phys.* **38** 1633
[12] www.comsol.com/showroom/gallery/11400/
[13] www.esi-cfd.com/content/view/524/
[14] Shihab M, Ziegler D and Brinkmann R P 2012 *J. Phys. D: Appl. Phys.* **45** 185202
[15] Kawamura E, Vahedi V, Lieberman M A and Birdsall C K 1999 *Plasma Sources Sci. Technol.* **8** R45
[16] Fadlallah M, Boothe J-P, Derouard J and Sadeghi N 1996 *J. Appl. Phys.* **79** 8976
[17] Jacobs B, Gekelman W, Pibility P and Barnes M 2011 *Phys. Plasmas* **18** 053503
[18] Turner M M, Deresi A, Donkó Z, Eremin D, Kelly S J, Lafleur T and Mussenbrock T 2013 *Phys. Plasmas* **20** 013507
[19] Phelps A V 1994 *J. Appl. Phys.* **76** 747
[20] Phelps A V and Petrović Z Lj 1999 *Plasma Sources Sci. Technol.* **8** R21
[21] Schulze J, Schungel E, Donkó Z and Czarnetzki U 2010 *J. Phys. D: Appl. Phys.* **43** 225201
[22] Brinkmann R P 2007 *J. Appl. Phys.* **102** 093303
[23] Brinkmann R P 2009 *J. Phys. D: Appl. Phys.* **42** 194009
[24] Brinkmann R P 2011 *J. Phys. D: Appl. Phys.* **44** 042002
[25] Mussenbrock T and Brinkmann R P 2006 *Appl. Phys. Lett.* **88** 151503
[26] Czarnetzki U, Mussenbrock T and Brinkmann R P 2006 *Phys. Plasmas* **13** 123503
[27] Bora B, Bhuyan H, Favre M, Wyndham E and Chuaqui H 2012 *Appl. Phys. Lett.* **100** 094103
[28] Schulze J, Kampschulte T, Luggenhölscher D and Czarnetzki U 2007 *J. Phys. D:* **Conf. Ser.** 86 012010
[29] Mussenbrock T and Brinkmann R P 2007 *Plasma Sources Sci. Technol.* **16** 377
[30] Schulze J, Heil B G, Luggenhölscher D, Brinkmann R P and Czarnetzki U 2008 *J. Phys. D: Appl. Phys.* **41** 195212
[31] Ziegler D, Mussenbrock T and Brinkmann R P 2009 *Phys. Plasmas* **16** 023503

![Figure 9](image-url)  
**Figure 9.** The space–time trajectories \( x(t) \) of an ensemble of ions (colored lines). The sheath edge is represented as a black line.

![Figure 10](image-url)  
**Figure 10.** The ion density \( n_i \) as a function of the normalized time \( t/\tau_{\text{RF}} \) at different positions \( x \) in the simulation domain.

The resulting global effect is the phase shift between the sheath voltage \( V(t) \) and the sheath charge \( Q(t) \), shown in figure 8. Clearly, the hysteresis vanishes both for \( \omega_{\text{RF}} \ll \omega_{\text{pi}} \) and \( \omega_{\text{RF}} \gg \omega_{\text{pi}} \); in the first case, the sheath expansion is slow compared with the ion Bohm 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.

### 4. Summary and conclusion

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 average electron temperature are known. It can, therefore, be used as a post-processor to restore kinetic information from the limited information provided by fluid plasma models. The gain in efficiency is considerable. However, the PIC/MCC method remains the standard against which all other schemes must be compared.

The physically most interesting result is the pronounced hysteresis of the sheath charge–voltage relation \( V(Q) \) which arises in the intermediate radio frequency regime. This phenomenon will surely have an impact on the overall behavior of the discharges.