

A version of the particle-in-cell method with adaptive mass alteration for the silane plasma simulation*

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Abstract. A new method is proposed to reduce non-physical effects in the PIC method. The method is demonstrated on an example of simulation of recombination of positive and negative ions in the radio frequency (RF) discharge.

In discharges in electro-negative gases, the negative ions are generated by dissociative attachment and lost by recombination with the positive ions. The forward modeling of the recombination in Particle-in-Cell simulation by removing a positive and a negative super-particles gives rise to essential numerical fluctuations. These fluctuations lead to non-physical electric fields and heating of electrons. In this paper, a method is proposed that enables the simulation of the recombination of positive and negative ions in an accurate way, even at low densities. The recombination in a cell is done by changing the mass of super-particles in this cell. Thus, it is possible to recombine any desired amount of real particles even if it appears to be less than one super-particle. Moreover, the number of super-particles in each cell is kept nearly constant by adding zero-mass super-particles (if a cell contains too few particles) or by removing a super-particle while increasing the mass for each of the rest (if a cell contains too many). It is shown that this simulation of ion recombination results in a lower level of non-physical fluctuations. The lower level of computational noise is shown to result in a lower ionization rate and lower ion fluxes as compared to the PIC/MCC simulations without adaptive mass alteration.

1. Introduction

The interest in silane (SiH_4) glow discharges is stimulated by the widespread use of amorphous silicon films in microelectronics. An electric glow discharge is a type of plasma formed by passing a current at 100 V to several kV through a gas. In a RF glow discharge, the applied voltage is oscillating with a frequency from radio frequency band (1–100 MHz). The glow discharge under consideration is created in plasma chemical reactors operating at low pressures in the course of the chemical vapor deposition of hydrogenated amorphous silicon (a-Si:H) films. The planar reactors with two parallel electrodes are often used. Due to the non-Maxwell character

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of the electron and the ion energy distribution, the Particle-in-Cell plus Monte Carlo (PIC/MCC) method is frequently used to simulate these discharges [1–3]. An important process that should be taken in electro-negative discharges is a recombination between positive and negative ions, generated by (dissociative) ionization and attachment, respectively.

The major challenge in the simulation of recombination in the PIC/MCC method is the transition from real physical particles to super-particles used in the numerical simulation. Consider a cell of the computational grid that has some super-particles for both positive and negative ions to be recombined. The number of super-particles in a cell is proportional to the number of real particles, for example, M real particles for one super-particle. Recombination implies that some particles should be removed, according to the recombination rate of real particles. A problem arises when the number of real particles that have to disappear during one time step is less than M . Thus, removing one super-particle at one time step is too much. In this case, recombination should be done by removing either one super-particle for a number of cells at each time step or once for a number of time steps in each cell. In such a way in one cell only a super-particle is removed, which results in a dramatic density alteration for this cell, while the density in the neighboring cells remains unchanged. This means that more non-physical fluctuations are introduced, because from the physical stand-point, the rates of recombination in the neighboring cells should be nearly equal.

One should consider the novelty of the proposed adaptive mass alteration procedure. It is common in the particle simulation [4, 5] that one superparticle represents a large group of real particles (e.g. ions in plasma, stars in galaxy, etc.) and the size (or mass) of the group remains the same in the course of simulation. This provides automatical fulfillment of the charge/mass conservation law and makes computation more convenient. From another point of view, the constant mass of a superparticle makes it impossible to simulate processes with a mass change less than the mass of one superparticle. And since such processes with a small mass change are impossible, the simulation often becomes unrealistic.

There are particle methods with a variable superparticle mass. One important example is the SPH (Smoothed Particle Hydrodynamics) method (e.g. [6]). The SPH, like all other Lagrangian methods considers a superparticle as a tracer, or a probe, but not as a group of real particles. The mass of this tracer could be changed with the size of the SPH softening kernel. The kernel size changes due to the physical conditions in order to provide a necessary precision.

The idea of the adaptive mass alteration is nearly the same: the number of superparticles in a cell (and, consequently, the masses of superparticles) is changed due to the physical conditions (in our case, too low or too high density) to provide a relatively low noise level. But since this idea is applied

not to a Lagrangian, but to an Eulerian particle method, to the classical PIC method, one could say that the adaptive mass alteration is a novel numerical technique.

2. The adaptive mass alteration algorithm in the PIC/MCC method

Let us introduce a new adaptive particle mass modification of the PIC/MCC method.

In the beginning of computation, the superparticles of each sort (electrons and various ions) are distributed in the following way. There are N superparticles in each cell, their total mass and charge being proportional to the density in the cell. The ratio $p = q/m$ is constant for each sort of particles. A particle stores either the mass m , or the charge q . Consider a cell that lacks superparticles of some sort (the density of some species is equal to zero in this cell). In this case, void superparticles are created. The charge and mass of void superparticles are equal to zero. The velocity distribution of void superparticles must correlate with that of real particles of the same sort, their average velocity being zero. That is, if for a sort of ions initially there is some temperature T_i (in the whole domain), then the velocity of super-particles of this sort are initially set with a Maxwell distribution based on the value T_i . The velocities of void super-particles of the same sort are set with the same Maxwell distribution, but since their mass is zero, the void superparticles carry no energy.

Void superparticles move by inertia with the mirror reflection from boundaries. Usually, void superparticles exist only in the beginning of simulation, because their masses are immediately adapted when non-zero mass superparticles appears with the void ones in this cell. If the number of superparticles in a cell is too small at the time t , $N(t) < aN$, super-particles must be added. Here $N(t)$ is the current number of particles in the cell, and a is a model parameter. Super-particles must be added with no changes to the total mass (charge) and an average velocity in the cell. Let us consider the algorithm step-by-step:

1. The total mass in the cell is computed;
2. The average velocity is computed;
3. $aN - N(t)$ new super-particles are added with a necessary velocity distribution (the average velocity of the added superparticles is 0);
4. The new mass of a superparticle in this cell is computed (the total mass is divided by the new number of superparticles). This mass is assigned to each superparticle in this cell;
5. The average velocity is added to the velocities of the new superparticles.

On the other hand, at some instant of time there could be too many superparticles in a cell: $N(t) > dN$, where d is another model parameter. In this case, $N(t) - dN$ superparticles in this cell are removed at Step 3.

The algorithm thus has two model parameters, a and d , that control adding and removing superparticles. These parameters are selected by hand in order to minimize the number of adding or removing operations. On the other hand, they should not differ too much from unity. If $a \ll 1$, the number of superparticles in a cell might be much less than N . If $d \gg 1$, the number of superparticles per cell might be too high, resulting in an increase of computation time. In our computations with 100 superparticles per cell, $a = 0.5$, $d = 2$; on average in 9% of the cells superparticles are added at each time step, and on average in 3% of the cells superparticles are removed. Figure 1 shows the statistics for adding and removing with other values of the parameters. The above algorithm is performed in every cell at the end of every time step, when all the physical processes (motion, various collisions, recombination, etc.) have been already computed. It should be noticed that the mass alteration does not affect the movement of superparticles, because it depends on the charge-to-mass ratio. The average velocity of particles in a cell remains the same through the mass alteration.

A superparticle could be considered in two ways: a cloud of particles, or one single “probe” particle. In the movement of particles, the superparticle is considered to be a probe with a constant mass, and in the course of the density evaluation to be a cloud with a variable mass. In the process of collision we also consider a superparticle as a probe particle with an unchangeable mass and charge. Because the rate of the Coulomb collisions depends on a charge, the rate of collision processes should be the same.

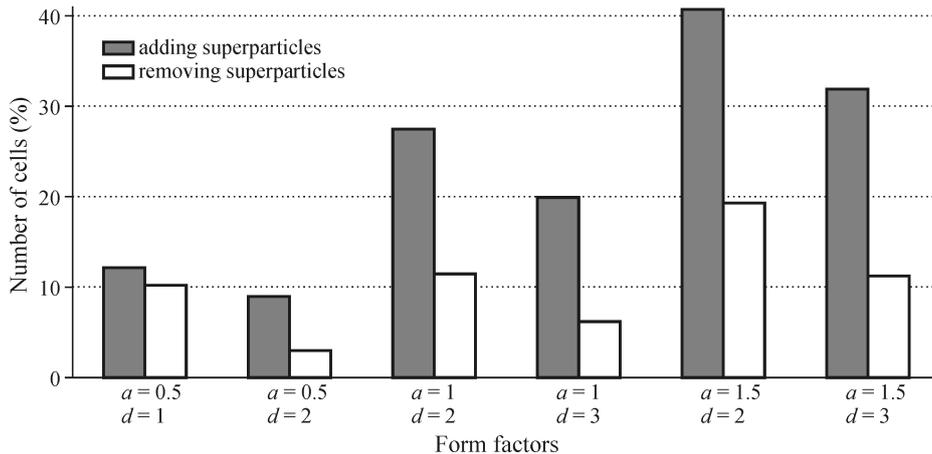


Figure 1. The time-averaged number of cells (in percent), where the superparticles are either added or removed for various values of parameters

Moreover, the mass alteration does not affect the probability of recombination (whether it is possible to compute a recombination in a cell or not), because the number of superparticles in a cell remains the same, only their masses altering. Such a recombination implies that the densities of both the positive and the negative ions must slightly decrease. The amount of density that is removed due to the recombination is much smaller than that in a cell. Because of the adaptive masses it is possible to remove any desired small amount of density. Thus, with an adaptive mass alteration we can compute a recombination in any cell, provided the densities of both the positive and the negative ions are above zero. Generally, we can say that recombination is handled in the same way as it is with real particles.

The main feature of the algorithm in question is thus the superparticle mass alteration. It provides a constant number of superparticles in a cell and a more natural way of simulation of some physical processes such as recombination.

3. Computational model

3.1. The basic equations. Let us consider the Boltzmann kinetic equation together with the Poisson equation in a cylindrical coordinate system:

$$\frac{\partial f_\alpha}{\partial t} + \vec{v} \cdot \vec{\nabla} f_\alpha + \frac{qE}{m} \frac{\partial f_\alpha}{\partial v} = \text{St}\{f_\alpha\}, \quad (1)$$

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \Phi}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \Phi}{\partial \varphi^2} + \frac{\partial^2 \Phi}{\partial z^2} = 4\pi\rho, \quad (2)$$

where ρ is a charge density, v is velocity, $E = -\vec{\nabla}\Phi$ is an electric field, Φ is potential, f_α is a distribution function for a particle species α , and $\text{St}\{f_\alpha\}$ describes the effect of collisions. Both equations are written down in the non-dimensional form, thus for the computation all the quantities should be used in the non-dimensional units. The transition to non-dimensional values is performed as follows. The basic characteristic quantities are:

- charge $q_0 = 1.6 \cdot 10^{19}$ C;
- length $L_0 = 0.01$ m;
- mass $M_0 = 9.1 \cdot 10^{-31}$ kg;
- energy $E_0 = 1$ eV = $1.6 \cdot 10^{-19}$ J.

3.2. Computational methods. Figure 2 displays the computational domain. The RF electrode is above, the grounded electrode is below. The size of the domain is R_M along the radius, Z_M along the z -coordinate. For the computation, the following values were taken: $R_M = 0.11$ m, $Z_M = 0.025$ m.

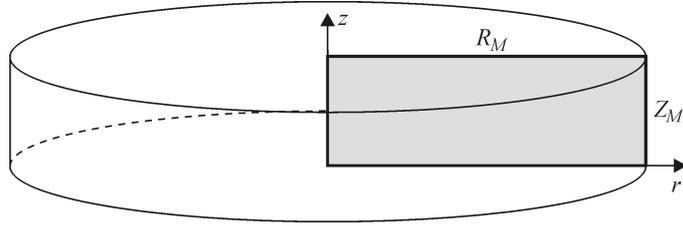


Figure 2. The shape of the computational domain

As was stated in Section 3.1, the main equations employed are the Poisson equation and the Boltzmann equation. Let us briefly consider the computational methods used to solve them in the present paper.

The Poisson equation is solved with the Fourier Transform method along z direction as well as along the angular direction. Along the radial direction, the sweeping method is applied.

A collisionless part of the Boltzmann equation (that is, the Vlasov equation, with zero right-hand side) is solved with the classical PIC method [4]. The Leapfrog scheme is applied to solve the equations of motion for each particle. It should be noted that the masses of superparticles being changed, do not affect the equations of motion since the equations refer to the charge-to-mass ratio.

The main effort in the present paper is being given to the collisional part of the Boltzmann equation. The mass alteration procedure is introduced to reduce non-physical effects related to collisions (particularly, to the ion-ion recombination). Two types of collisions are taken into account: the collisions of electrons with gas molecules and ion-ion collisions resulting in recombination. A more detailed description of the physical model is given in Section 4.

The collisions of electrons with gas molecules are treated with the null collision technique [1]. First, for the given electron, the gas is defined (considering two gases with the densities n_1 and n_2) by means of the random number t , $0 < t < 1$. If the condition

$$t < \frac{n_1}{n_1 + n_2}$$

is valid, then the first gas is taken for collision, otherwise the second.

In the same way, one of the possible collisional processes is selected (the processes are listed in Section 4). Since the type of the process is known, it is possible to find its cross-section σ . The cross-section is used to compute the collision probability

$$p = 1 - \exp(-v\tau\sigma n).$$

Here v is the electron velocity, τ is the timestep and n is the gas density. By means of a random number, the collision probability defines either this

electron superparticles suffer a collision or not. If the collision occurs, then the scattering angle is defined as

$$\theta = \sqrt{1 - \tau \ln(1 - x)}.$$

Here x is a random number, $0 < x < 1$. Finally, the new components of the velocity vector are computed.

The described combination of the PIC method with a null collision technique is often referred to as PIC/MCC method (Particle-in-Cell plus Monte Carlo Collision), [2].

To compute the second type of collisions, the ion-ion recombination, we just alter the masses of superparticles representing the corresponding ions. The mass alteration for the superparticles follows the equation

$$\frac{\partial n^-}{\partial t} = -k_{\text{rec}} n^+ n^-.$$

Here n^+ is the density of positive ions, n^- is the density of negative ions and k_{rec} is the recombination coefficient. Section 2 gives a detailed description of the mass alteration procedure, and Section 5 gives an example of mass alteration applied to the ion-ion recombination.

In this domain, a cylindrical grid is introduced with the size $N_R \times N_\varphi \times N_Z = 50 \times 32 \times 30$; 100 particles of each sort are inserted in each grid cell, 19.2 million particles in total. The computations were performed with the NKS-160 cluster at the Siberian Supercomputer Centre, Novosibirsk, Russia, as well as with the LISA cluster, at the SARA Super-Computing Centre, the Netherlands.

4. The physical model of the RF glow discharge silane plasma

The employed physical model of the RF glow discharge in silane plasma was designed in [7,8]. A more detailed description of physico-chemical processes in the silane plasma can be found, for example, in [9]. The model under study involves ions SiH_2^+ , SiH_3^- , H_2^+ and electrons. They move in the background of the neutral gas, which is a mixture of silane SiH_4 and hydrogen H_2 . Only collisions of electrons with gas molecules are taken into account, because the density of charged particles is six orders of magnitude lower than the neutral gas density. The model includes the following processes:

- Elastic scattering $e + \text{SiH}_4 \rightarrow \text{SiH}_4 + e$;
- Ionization $e + \text{SiH}_4 \rightarrow \text{SiH}_2^+ + 2\text{H} + 2e$;
- Vibrational excitation $e + \text{SiH}_4 \rightarrow \text{SiH}_4^* + e$;
- Attachment $e + \text{SiH}_4 \rightarrow \text{SiH}_3^- + \text{H}$;

- Dissociation $e + \text{SiH}_4 \rightarrow \text{SiH}_2 + 2\text{H} + e$;
- Elastic scattering $e + \text{H}_2 \rightarrow \text{H}_2 + e$;
- Ionization $e + \text{H}_2 \rightarrow \text{H}_2^+ + 2e$;
- Vibrational excitation $e + \text{H}_2 \rightarrow \text{H}_2^* + e$;
- Dissociation $e + \text{H}_2 \rightarrow 2\text{H} + e$.

There are two vibration excitation channels for silane and three for hydrogen. The channels differ by the energy threshold and also by the loss of energy as a result of reaction. The radicals produced in the chemistry are not considered because in real discharges they are captured at the reactor walls. Thus, new particles appear only as a result of ionization (a new electron and a positive ion) and of attachment (a negative ion with a loss of an electron). In all processes, except elastic scattering, the electron loses an amount of energy equal to the reaction threshold. The modeling of electron-neutral collisions includes the null collision technique to make the total cross-section independent of the electron energy [1, 2]. Moreover, the model includes the recombination of positive and negative ions:

- $\text{SiH}_2^+ + \text{SiH}_3^- \rightarrow \text{SiH}_2 + \text{SiH}_3$;
- $\text{H}_2^+ + \text{SiH}_3^- \rightarrow \text{H}_2 + \text{SiH}_3$.

Charged particles (positive ions and electrons) are captured at the electrodes. Negative ions cannot approach the electrodes because of the electrode space charge sheaths. Thus recombination is the only way to decrease the negative ion density (which is constantly increased by means of attachment). It is clear that for simulation of a stationary discharge, recombination should be taken into account. Four kinds of superparticles must therefore be present in the model: electrons, and three sorts of ions (SiH_2^+ , SiH_3^- and H_2^+).

5. Simulation of recombination in the RF glow discharge silane plasma

Recombination of positive ions with a density n^+ and negative ions with density n^- is described by the following equation:

$$\frac{\partial n^-}{\partial t} = -k_{\text{rec}} n^+ n^-. \quad (3)$$

Here k_{rec} is a recombination coefficient (for the reaction $\text{SiH}_2^+ + \text{SiH}_3^-$, $k_{\text{rec}} = 1.2 \cdot 10^{-7} \text{ cm}^3/\text{s}$). The number of ions ΔN^+ that disappear from a small volume ΔV in a short time interval Δt due to recombination computed from this equation is:

$$\Delta n^+ = k_{\text{rec}} n^+ n^- \Delta t; \quad \Delta N^+ = \Delta n^+ \Delta V. \quad (4)$$

Within the physical model under study, the recombination of negative ions SiH_3^- with both positive ions, SiH_2^+ and H_2^+ , is possible.

In this section, we restrict the discussion to the recombination of the ions SiH_3^- and SiH_2^+ . The difficulty of such a simulation is the description of recombination with super-particles. This can be illustrated in the following way. If the time interval in formula (4) is the simulation time step $\Delta t = \tau = 1.68 \cdot 10^{-10}$ s, and a small volume is the cell volume $\Delta V = r h_r h_\phi h_z \approx 4 \cdot 10^{-3}$ cm³, then, with a characteristic ion density $n^+ \approx n^- = 10^{10}$ cm⁻³, the number of recombining ions in one cell is ≈ 10 . It is obviously lower than the number of real ions corresponding to one superparticle (with the parameters given in Section 3 is $\approx 10^6$ ions). Thus, if a cell would contain 100,000 superparticles (that is unrealistic), then recombination is simulated by removing one of them at each time step. If the number of superparticles per cell is smaller, then recombination must be computed in a number of cells together (by removing one superparticle from a hundred of cells, for example), or once for a number of time steps (by removing one superparticle from each cell once for a hundred of time steps, for example). With the given computation parameters, 48 thousand cells, 10 real recombining particles, and 10^6 real ions for a superparticle, the simulation of recombination requires one superparticle to be removed in the whole computational domain every two time steps. Thus, if in a cell a super-particle is removed, it will result in a dramatic density alteration in this cell, the density in surrounding cells being unchanged. This implies that significant non-physical fluctuations of the density will be introduced (from the physical standpoint the change of the density as a result of recombination must be comparable in the neighboring cells). Also, if there are different densities in two cells, the recombination rates should also be different. Nevertheless, the ratio $\Delta n^+ / n^+$ will be made equal in these two cells. Here we simulate recombination in a stationary RF discharge, with nearly equal positive and negative ion densities, $n^+ \approx n^- \approx 10^{10}$ cm⁻³. If one of a hundred superparticles in a cell is removed, the density change will be about 1 %, and the net charge will not be changed at all, because a superparticle with the opposite charge is also removed. The statistical fluctuations are about 10 % in this case, so recombination should not essentially increase fluctuations. But the problem is that only one cell in the whole domain loses a superparticle, and the choice of the cell is arbitrary. Moreover, the alteration of density in the cell that loses a super-particle will be greater (in the given example, by 10^5 times) than it should be as a result of recombination in reality.

The method described in Section 2 enables us to simulate a recombination in a more natural way. It is possible to remove any desired small number of real particles, even if it is less than one superparticle. This is

done in the following way. The density of ions in a cell decreases by Δn^+ . This means that the mass of each SiH_2^+ superparticle in this cell is multiplied by the factor

$$k^+ = 1 - \frac{\Delta n^+}{n^+}.$$

It should be noted that k^+ will be different for another sort of positive ions (if the reaction between SiH_3^- and H_2^+ is considered). In the same way, the factor k_- is computed for negative ions SiH_3^- :

$$k^- = 1 - \frac{\Delta n^-}{n^-}.$$

With the above parameters $n^+ = n^- = 10^{10} \text{ cm}^{-3}$, $k_{\text{rec}} = 1.2 \cdot 10^{-7}$, 48 thousand cells, 10^6 real ions for a superparticle and 10 real recombining particles of both sorts in a cell), the recombination could be simulated by removing one superparticle in each of five randomly selected cells once per 10 time-steps (instead of one superparticle in the whole domain for two time-steps, as mentioned above).

Figure 3 shows a maximum value of the quantity that measures the nonphysical density alteration as a result of recombination. It is defined as:

$$x(t_n) = \max_{i,p,k} \frac{(\Delta n^+)_{i,p,k}}{n_{i,p,k}^+}, \quad 1 = 1, \dots, N_R, \quad p = 1, \dots, N_\phi, \quad k = 1, \dots, N_Z, \quad (5)$$

where $t_n = n\tau$ is the time, τ is the time step. The alteration of the density in (5) is a result of recombination only, other processes such as ionization and transport of ions, are not taken into account. Thus, if removing one superparticle in a cell corresponds to recombination in a number of cells at several time steps, then the quantity x will be much greater than the real physical value of density alteration divided by density, $\Delta n^+/n^+ = k_{\text{rec}}\Delta t n^- \approx 10^{-7}$,

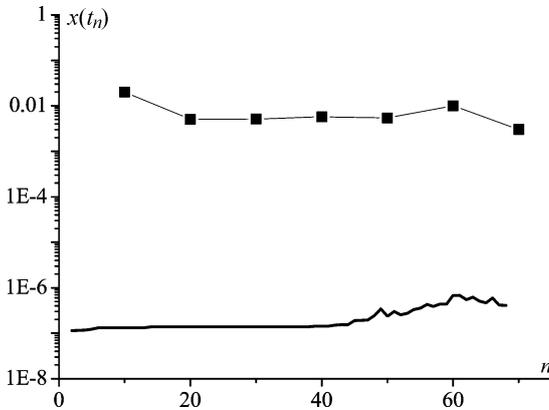


Figure 3. Maximum ratio of density alteration to the value of density depending on the number of time step, $x(t_n)$: (1) the line with squares represents the classical PIC method, all the superparticles have equal masses; (2) the solid line represents the method with adaptive mass alteration. In the case (1), the recombination is performed once during 10 time-steps

as determined from (3), as shown by the line with squares in Figure 3. On the contrary, if recombination is computed separately in each cell, then x is nearly equal to the physical value of density alteration divided by density, as shown by the smooth line in Figure 3.

6. Results

In Figure 4, the time and space averaged electron energy distribution function is displayed. The electron energy distribution approximately corresponds to the results of a hydrodynamical simulation [8]. Most of the electrons are in the interval 1.5–4 eV, the energy of the electrons in the maximum of electron energy distribution function (EEDF) is 2 eV.

The dependence on the power of the ion flux to the grounded electrode was also computed. The results are displayed in Figure 5 and compared with experimental results [10], showing good agreement. The differences are probably explained by the differences in geometry between the computational domain and the experimental device.

Finally, we want to show that the proposed method not only decreases the level of computational noise but consequently improves the simulation

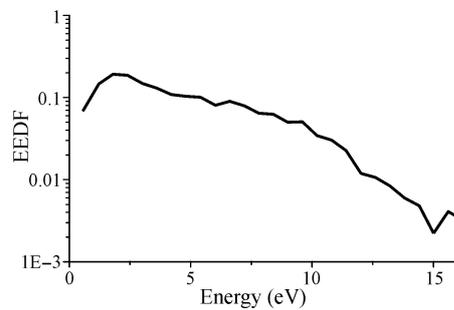


Figure 4. Electron energy distribution function, time average over the whole discharge

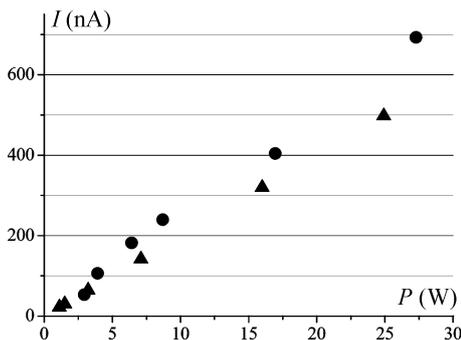


Figure 5. A calculated ion flux (in nano-amperes) towards the grounded electrode plotted against power (in Watts), for an RF frequency of 58 MHz (circles) as compared to the experiment (triangles) [7]

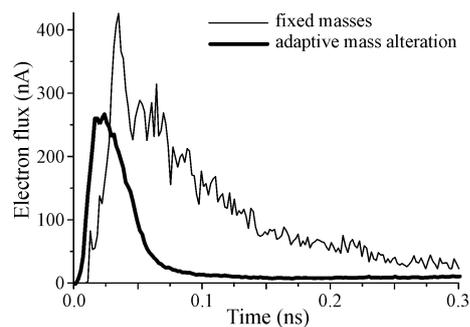


Figure 6. An electron flux to the grounded electrode in the beginning of the simulation, for the RF voltage of 60 V, corresponding to a power of 17 W in Figure 5.

results. For example, Figure 6 shows an electron flux towards the grounded electrode versus time, in the beginning of simulation. Here we show that the adaptive mass alteration results in reduction of noise of the physical quantities. The beginning of the simulation was chosen because at this time instant we have the noisiest time dependence of the current for both cases—adaptive masses and fixed masses. Since the discharge is not yet in the periodic steady state, one cannot expect the fluxes to be equal, but the flux computed with the adaptive mass alteration is significantly less noisy.

Let us consider the simulation without adaptive mass alteration. Initially, we set 100 superparticles per cell, and in the process of simulation of the average number of superparticles per cell remains close to 100. The fact is, there are cells with higher and lower values of density, because the masses of superparticles are fixed. In our simulation, the ratio of densities of higher and lower density cells can become as high as 10, lower density cells being located near the boundary and higher density ones near the center of the domain. If the average number of superparticles per cell is still 100, we can estimate the number of superparticles in a lower density cell of about 20 and in a higher density one of about 200. Thus, the general level of computational noise will be defined by the cells having 20 superparticles. The estimate of computational noise in the PIC method is given by the expression $N^{-1/2}$, where N is the number of superparticles per cell. Then, instead of the desired 10 % of computational noise there will be more than 20 %. On the other hand, in the simulation with the adaptive mass alteration, the number of superparticles per cell is kept nearly constant, about the initial 100 in all cells. Thus, the level of computational noise remains close to 10 %.

For this reason, there is a difference between the simulation results with and without adaptive mass alteration. First of all, as Figure 7 shows, the electrons become hotter. In case of no mass alteration, a maximum of the EEDF is near to 8 eV, so much higher than in Figure 4, where it is 2 eV, with the mass alteration. The hotter electrons increase the rate of ionization

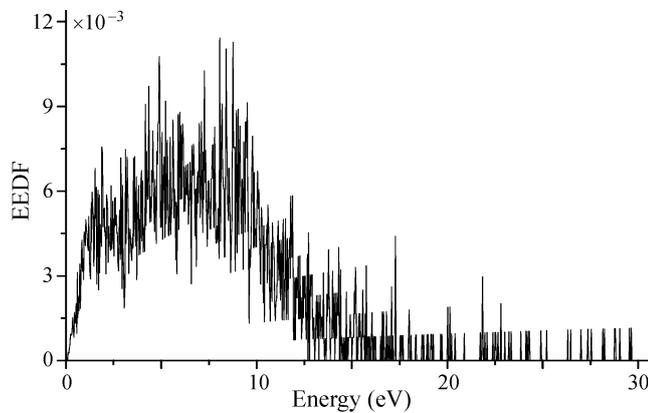


Figure 7. An electron energy distribution function without adaptive mass alteration, time-average over the whole discharge

in the simulation without adaptive mass alteration, raising the number of model ionization events per one RF cycle with a factor of two (33.449 events with adaptive mass alteration against 68661 without it). The higher ionization rate results in a higher ion flux at the grounded electrode, as shown in Figure 8. Moreover, with fixed masses, the number of superparticles near to the electrode (where the flux is measured) could be very low (5–10 superparticles per cell or even less). Thus, a standard (fixed-mass) PIC method might bring about a large error near to the electrode.

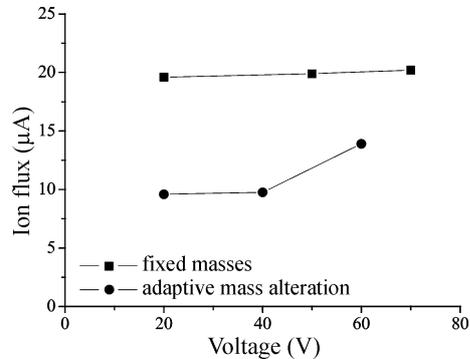


Figure 8. An ion flux at the grounded electrode for the RF voltage of 60 V corresponding to the power of 17 W in Figure 5. The fluxes of ions differ nearly by a factor of two

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