# Hybrid Monte Carlo-fluid model of a direct current glow discharge

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A self-consistent hybrid Monte Carlo-fluid model for a direct current glow discharge is presented. The Monte Carlo part simulates the fast electrons while the fluid part describes the ions and slow electrons. Typical results of the model include collision rates of the fast electrons, energy distributions of these electrons, fluxes and densities of the different plasma species, the electric field and the potential distribution, all as a function of position from the cathode. The influence of the negative glow on the calculations in the cathode dark space is studied. Moreover the influence of three-dimensional scattering instead of forward scattering and the incorporation of side wall effects is investigated. Calculations are carried out for a range of voltages and pressures in order to study their influence on the calculated quantities. Comparison was made between total electrical currents calculated in the model and experimentally measured ones to check the validity of the model. © 1995 American Institute of Physics.

# **I. INTRODUCTION**

Glow discharges are being used in many fields of application. They serve extensively as plasma processing devices in microelectronics, e.g., for ion etching, thin film deposition, and plasma treating of surfaces,<sup>1-3</sup> and they also find application as atomization-excitation-ionization sources in analytical chemistry.<sup>4,5</sup> To attain better results in these application fields, a quantitative understanding of the glow discharge is required. We try to obtain this by mathematical modeling. Three major approaches of modeling can be found. The first one is to deal with the glow discharge plasma as a fluid. $^{6-12}$  It is assumed that the species in the discharge are not far from hydrodynamic equilibrium. This assumption is not really valid for the fast electrons, so that the fluid model is only an approximation. The second approach is a kinetic (Boltzmann) model,  $^{13-15}$  which copes with the nonequilibrium situation of the electrons. The third way is via Monte Carlo simulations.<sup>16,17</sup> This is the most accurate one, because it deals with particles on the lowest microscopical level. However, the Monte Carlo model on its own is not a self-consistent method. Recently some hybrid models of the glow discharge have been developed.<sup>18-24</sup> These account for the nonequilibrium nature of the fast electrons by treating them with Monte Carlo or kinetic models whereas self-consistent results can be achieved by describing the slow electrons and the ions with a fluid model.

In this work a hybrid self-consistent model (combined Monte Carlo fluid) is developed for the entire direct current glow discharge. The electrons are split up in a fast and a slow group. The fast electrons are modeled with Monte Carlo simulations whereas the slow electrons and ions are described with a fluid approach. Almost all models described in the literature refer to glow discharges used for plasma etching and deposition and are performed for discharge voltages of 100-500 V. This model, however, is meant to describe a glow discharge used as ion source for mass spectrometry in which voltages of about 1 kV are common. Hence in this work emphasis is placed on these higher voltages although the results of lower voltages are also mentioned. Typical results of the model include collision rates and energy distributions of the fast electrons, fluxes and densities of the plasma species, the electric field and the potential distribution, all as a function of position from the cathode. Comparison will be made between the actual Monte Carlo simulations and

- Monte Carlo calculations in the cathode dark space (CDS) only to study the influence of the negative glow (NG),
- (ii) one-dimensional Monte Carlo calculations to study the influence of three-dimensional scattering, and
- (iii) three-dimensional calculations as if no side walls are present to study the influence of absorption, reflection, and secondary electron emission at the walls.

Furthermore the influence of voltage and pressure on the calculated quantities will be investigated. Comparison of the calculated electrical currents with experimentally measured ones will be used to verify the validity of the model.

#### **II. DESCRIPTION OF THE MODEL**

#### A. Model assumptions

The fluid model is one dimensional, i.e., it applies to a discharge between two infinitely wide electrodes (anode and cathode) so that quantities vary only with distance from the electrodes. In the Monte Carlo simulations however, three-dimensional motion of the electrons is already incorporated, although the electrons feel only the influence of an axial electric field since this follows from the fluid model (see further). The output results of the Monte Carlo model are

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FIG. 1. Standard glow discharge cell in the Fisons VG 9000 glow discharge mass spectrometer. (1) sample (cathode), (2) mask, (3) insulator, (4) cell body (anode, made of tantalum), (5) gas inlet (argon), (6) exit slit.

only taken in one dimension, so that combined with the onedimensional fluid model no problems of current and charge conservation will arise. The discharge geometry to which the Monte Carlo simulations are applied is that of a standard discharge cell of the Fisons VG9000 mass spectrometer for analyzing flat samples (see Fig. 1). The plasma is assumed to consist of four species: neutral ground state argon atoms at rest and uniformly distributed throughout the discharge ( $Ar^0$ ), singly charged positive argon ions ( $Ar^+$ ), and fast and slow electrons. Collision processes of the fast electrons taken into account in the Monte Carlo model are excitation, ionization, and elastic collisions. The motion of ions and slow electrons in the fluid model is assumed to be collisionally dominated and described by diffusion and migration.

#### **B. Monte Carlo model**

The Monte Carlo model is similar to the one described for the electron transport in the CDS only,<sup>17</sup> which is a onedimensional Monte Carlo model but with already the incorporation of three-dimensional scattering. A few modifications had to be carried out due to the extension to the NG and the complete three-dimensional motion.

The complete three-dimensional motion is determined by the axial position z, the radial position r  $(r^2 = x^2 + y^2)$ , and the axial and azimuthal angles  $\theta$  and  $\phi$ . The electrons start at the cathode with all radial positions being equally probable. Their energy is assumed to be 4 eV<sup>17</sup> and their three-dimensional direction is randomly chosen:

$$\cos \theta = rn, \quad \phi = 2\pi(rn), \tag{1}$$

where rn is a random number in the interval [0-1]. The initial three-dimensional velocities are determined by the energy E and the angles  $\theta$  and  $\phi$ :

$$\nu_{z_0} = \cos \theta \sqrt{\frac{2E}{m}}, \quad \nu_{x_0} = \sin \theta \cos \phi \sqrt{\frac{2E}{m}},$$

$$\nu_{y_0} = \sin \theta \sin \phi \sqrt{\frac{2E}{m}}.$$
(2)

Their three-dimensional motion under the influence of the axial electric field is determined by Newton's laws:

$$z = z_0 + \nu_{z_0} \Delta t + \frac{q \mathscr{E}}{2m} (\Delta t)^2, \quad x = x_0 + \nu_{x_0} \Delta t,$$
  

$$y = y_0 + \nu_{y_0} \Delta t,$$
  

$$\nu_z = \nu_{z_0} + \frac{q \mathscr{E}}{m} \Delta t, \quad \nu_x = \nu_{x_0}, \quad \nu_y = \nu_{y_0},$$
 (3)

where  $x_0, y_0, z_0$  and x, y, z are the position coordinates before and after the timestep  $\Delta t, v_{x0}, v_{y0}, v_{z0}$ , and  $v_x, v_y, v_z$  are the velocities before and after the timestep,  $\mathscr{E}$  is the axial electric field, and *m* is the electron mass. After each timestep the probability of collision is calculated. If a collision takes place, the kind of collision is determined. Depending on the kind of collision the new energy and direction are calculated. A detailed description of this procedure can be found in Ref. 17. In Ref. 17, however, calculation of the new direction after collision was restricted to the axial angle  $\theta$ . In the present complete three-dimensional model also the azimuthal angle  $\phi$  has to be calculated. For a given axial and azimuthal scattering angle,  $\chi$  and  $\psi$  (for their meaning, see Ref. 17), the new axial and azimuthal angles  $\theta$  and  $\phi$  are determined by<sup>25</sup>

$$\begin{aligned} \sin \theta \cos \phi \\ \sin \theta \sin \phi \\ \cos \theta \end{aligned}
= \begin{pmatrix} \cos \theta_0 \cos \phi_0 & -\sin \phi_0 & \sin \theta_0 \cos \phi_0 \\ \cos \theta_0 \sin \phi_0 & \cos \phi_0 & \sin \theta_0 \sin \phi_0 \\ -\sin \theta_0 & 0 & \cos \theta_0 \end{aligned}
\times \begin{pmatrix} \sin \chi \cos \psi \\ \sin \chi \sin \psi \\ \cos \chi \end{pmatrix}.$$
(4)

When the new energy and three-dimensional direction are calculated, the electron trajectory is again defined by Newton's laws during the next timestep, etc. This procedure is repeated until the electrons collide at the walls or until they reach energies lower than the excitation threshold of Ar. Indeed, when electrons in the NG have energies lower than the excitation threshold of Ar, they are transferred to the slow electron group (described in the fluid model), because they cannot produce inelastic collisions anymore and are therefore no longer important as "fast electrons." Their only role is to carry electrical current and to provide negative charge density. This transfer to the slow electron group is assumed to occur at a collisional rate based on elastic collisions ( $\sigma$ ~10<sup>-15</sup> cm<sup>2</sup>).<sup>18</sup> The assumption is not so critical because the splitting up in slow and fast electrons is only artificial. In

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the CDS such a transfer is not included since slow electrons do not remain slow due to rapid acceleration and are therefore not assumed to be created.

When the electrons collide at the walls of the cell, they can be absorbed, reflected, or cause secondary electron emission. The secondary electron emission coefficient for Ta as a function of the electron energy,  $\delta$ , is taken from Ref. 26 and is rather high (maximum 1.3 at 600 eV) which means that electrons cause easily the emission of a secondary electron. When  $\delta$  is higher than 1, at least one secondary electron is emitted. The calculated value of  $\delta$  (if  $\delta < 1$ ) or of  $\delta - 1$  (if  $\delta > 1$ ) is compared with a random number (*rn*) between 0 and 1. If  $\delta < rn$ , no secondary electron emission took place and the electron is simply absorbed. If  $\delta > rn$ , secondary electron emission or reflection has taken place. We assumed that about 10% of the electrons are reflected back with no change in energy and in a direction symmetrical to the normal of the wall, whereas the remaining 90% caused secondary electron emission, resulting in a slow electron of about 4 eV and with a direction randomly chosen with respect to the normal of the wall.27

#### C. Fluid model

The fluid model for the ions and slow electrons described here is a single moment approach of the Boltzmann equation. The relevant coupled equations are the continuity equations of ions and electrons [Eqs. (5) and (6), respectively], the flux equations based on diffusion and migration of ions and electrons [Eqs. (8) and (9), respectively], and the Poisson equation [Eq. (7)]:

$$\frac{\delta n_i}{\delta t} + \frac{\delta j_i}{\delta x} = r_i, \tag{5}$$

$$\frac{\delta n_e}{\delta t} + \frac{\delta j_e}{\delta x} = r_e, \tag{6}$$

$$\frac{\delta^2 V}{\delta x^2} + \frac{e}{\epsilon_0} \left( n_i - n_e - n_{e, \text{fast}} \right) = 0, \tag{7}$$

$$j_i = -\mu_i n_i \frac{\delta V}{\delta x} - D_i \frac{\delta n_i}{\delta x},\tag{8}$$

$$j_e = \mu_e n_e \frac{\delta V}{\delta x} - D_e \frac{\delta n_e}{\delta x}, \qquad (9)$$

where  $n_i$  and  $n_e$  are the ion and slow electron densities,  $j_i$ and  $j_e$  are the corresponding fluxes, V is the electrical potential,  $n_{e,\text{fast}}$  is the fast electron density which results from the Monte Carlo model, and  $r_i$  and  $r_e$  are the creation rates of ions and slow electrons which also result from the Monte Carlo model. Finally  $\mu_i$ ,  $\mu_e$ ,  $D_i$ , and  $D_e$  are the ion and electron mobilities and diffusion coefficients, respectively. These transport coefficients are taken from Ref. 18:  $D_e=2.10^5$  cm<sup>2</sup> s<sup>-1</sup>,  $\mu_e=2.10^5$  cm<sup>2</sup> s<sup>-1</sup> V<sup>-1</sup>,  $D_i=400$ cm<sup>2</sup> s<sup>-1</sup>, all at 1 Torr, and  $\mu_i$  is given by the Frost formula:<sup>28</sup>

$$\mu_i = \frac{\mu_{i0}}{\sqrt{1+a|\mathscr{B}/n|}},\tag{10}$$

where a is  $7.36 \times 10^{14}$  cm<sup>-2</sup> V<sup>-1</sup>,  $\mu_{i0}$  is 1420 cm<sup>2</sup> s<sup>-1</sup> V<sup>-1</sup> at 1 Torr,  $\mathscr{E}$  is the electric field, and n is the argon gas atom density.

These five equations can be reduced to three equations by inserting Eqs. (8) and (9) in Eqs. (5) and (6), respectively. The resulting three equations are second order in space, so six boundary conditions are required. We took:  $V = -V_c$ ,  $n_e=0, \ \delta n_i/\delta x=0$  at the cathode and  $V=0, \ n_e=0, \ \delta n_i/\delta x=0$ at the anode. Due to the severe nonlinearity and strong coupling of these three equations, solving the fluid model is a difficult numerical problem. The method we used was developed by Passchier et al.<sup>6,7</sup> It is a fully implicit method based on the Scharfetter-Gummel exponential scheme for the transport equations. $^{6-9,29,30}$  The advantage of this scheme is its ability to switch between situations where either the drift component or the diffusion component of the particle flux is dominant (i.e., high and low electric field, CDS and NG). The basic idea is that the particle flux is assumed constant between mesh points, instead of the densities. A more detailed derivation of the Scharfetter-Gummel scheme is given in the Appendix. The three discretized equations are solved in the following sequence. Going from time  $k\Delta t$  to (k  $(+1)\Delta t$ , Eqs. (6)  $(n_e)$  and (7) (V) are solved together with known  $n_i$  at time  $k\Delta t$ . After discretization Eq. (6) can be written as a function of the following unknowns (see Appendix):

$$\Psi_{n_e}[V_{i-1,k+1}, V_{i,k+1}, V_{i+1,k+1}, (n_e)_{i-1,k+1}, (n_e)_{i,k+1}, (n_e)_{i+1,k+1}, C_{n_e}] = 0,$$
(11)

where  $C_{ne}$  contains the known terms. After discretization Eq. (7) becomes a function of the following unknowns:

$$\Psi_{V}[V_{i-1,k+1}, V_{i,k+1}, V_{i+1,k+1}, (n_{e})_{i,k+1}, C_{V}] = 0, \quad (12)$$

where  $C_V$  again contains the known terms. The system of nonlinear equations [Eq. (11)] and linear equations [Eq. (12)] for each gridpoint is solved simultaneously by means of the Newton-Raphson method. When  $n_e$  and V are known at time  $(k+1)\Delta t$ , Eq. (5) is solved for  $n_i$  at time  $(k+1)\Delta t$ . After discretization Eq. (5) yields a tridiagonal system of linear equations, because V at time  $(k+1)\Delta t$  is known:

$$\Psi_{n_i}[(n_i)_{i-1,k+1},(n_i)_{i,k+1},(n_i)_{i+1,k+1}] = C_{n_i}, \quad (13)$$

where  $C_{ni}$  contains the known terms. This is solved by the Thomas algorithm.<sup>31</sup>

# D. Combined Monte Carlo-fluid model

The combined model is solved by the iterative procedure described in Ref. 18. We start with the Monte Carlo model using an initial guess for the fast electron flux at the cathode and for the electric field distribution throughout the discharge (i.e., linear in the CDS and zero in the NG). Results of the Monte Carlo model are the creation rates of ions and slow electrons, as a function of position from the cathode. These creation rates are used as input in the fluid model. The fluid model yields a new electric field distribution. Also from the ion flux at the cathode calculated in the fluid model, the new fast electron flux at the cathode is calculated by  $j_{e, \text{fast}}(0) = -\gamma j_i(0)$ , where  $\gamma$  is the ion induced secondary electron

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FIG. 2. Rate profiles of the fast electrons as a function of position from the cathode (100 Pa-1000 V).

emission coefficient. The new electric field distribution and the new fast electron flux are introduced in the Monte Carlo model and the procedure is repeated until convergence is reached.

## **III. RESULTS AND DISCUSSION**

Results are shown for an argon discharge with a molybdenum cathode (secondary electron emission coefficient is taken to be  $0.12^1$ ) at 100 Pa gas pressure and 1000 V discharge voltage. Results at lower voltages (for example, 300 V) are qualitatively the same. The differences that were observed will be mentioned.

Figure 2 shows the collision rates of the fast electrons throughout the discharge. The excitation rate profile explains the different bright and dark layers in the discharge. Relatively close to the cathode the excitation rate is rather low since the electrons have already achieved energies too high for efficient excitation (the excitation cross section goes through a maximum at about 25 eV). This relatively dark zone corresponds to the CDS. Further in the plasma where the electric field is low, the electrons move back and forth by scattering and can be found many times at the same place, leading to a high degree of excitation in that region. This high excitation rate corresponds to the very bright NG. The ionization rate shows nearly the same behavior but is approximately twice as high. Most of the Ar ions are created in the beginning of the NG. The rate of electron "transfer" from the fast to the slow electron group is zero in the CDS as explained before, but is nearly equal to the ionization rate in the NG. At 300 V the rate profiles are qualitatively the same, but they drop faster to zero towards the anode backplate; at 1 cm from the cathode there is almost no excitation, ionization, and electron transfer, since there are not many fast electrons anymore.

In Fig. 3 the fluxes of the different species are presented. Due to the combination of the three-dimensional Monte Carlo model with the one-dimensional fluid model, care has to be taken that current conservation is preserved. Therefore, the fast electron flux is not taken from the Monte Carlo model but is calculated explicitly by



FIG. 3. Fluxes of the plasma species as a function of position from the cathode (100 Pa-1000 V).

$$j_{e,\text{fast}}(x+1) = j_{e,\text{fast}}(x) + r_{e,\text{fast}}(x)\Delta x, \qquad (14)$$

where  $r_{e,\text{fast}} = r_i - r_e$ . The values resulting from the Monte Carlo model deviate slightly from the explicitly calculated values due to the three-dimensional motion and the wall effects. However, this deviation is only small since most of the electrons that collide at the walls cause secondary electron emission (or are reflected) and only a small fraction is absorbed. Hence the walls act rather as an energy sink (electrons coming in with high energies leave the walls with only 4 eV) than as a particle sink, since the electrons are not completely lost, they are only transferred to the slow group. In the CDS the flux of the slow electrons is still zero since slow electrons are not created in this region. The flux of the fast electrons is directed away from the cathode and increases towards the CDS-NG interface whereas the ion flux is directed towards the cathode and increases to the same extent as the electron flux but in the opposite direction, resulting in a constant total current. In the NG the flux of the fast electrons remains nearly constant due to the nearly equal ionization and electron transfer rates (Fig. 2). The flux of the slow electrons shows a rather large increase and the ion flux varies by the same amount. Moreover the ion flux goes through zero at about 0.7 cm from the cathode. In the CDS most of the total electrical current is carried by the ions whereas in the NG the (slow) electrons are the dominant current carriers. From Fig. 3 it is seen that the total current is  $2.8 \times 10^{16}$  cm<sup>-2</sup> s<sup>-1</sup>, which corresponds to an electrical current of 4.5 mA/cm<sup>2</sup>. Experimentally a value of 5.29 mA/cm<sup>2</sup> was obtained for a Mo cathode in Ar at 100 Pa and 962 V. The two values agree reasonably well which clearly validates the present model. At 300 V the fluxes show qualitatively the same behavior, but the variation in slow electron and ion flux is steeper in the beginning of the NG and much flatter further in the NG where the creation rates are nearly zero. The ion flux goes through zero at about 0.5 cm from the cathode.

Figure 4 shows the densities of the different species in the discharge. The ion density is nearly constant in the CDS and increases rapidly in the NG, reaching a maximum halfway the discharge. The slow electron density is zero in the CDS and nearly equal to the ion density in the NG. This

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FIG. 4. Densities of the plasma species as a function of position from the cathode (100 Pa-1000 V).

results in a net positive space charge in the CDS and nearly charge neutrality in the NG. Close to the anode the slow electron density goes to zero while the ion density remains at a constant value comparable to the values in the CDS. The fast electron density is nearly five orders of magnitude lower than the ion and slow electron densities and hence does not contribute to the space charge. At 300 V the ion and slow electron densities show a more asymmetrical behavior, peaking closer to the cathode, following from the creation rate profiles. The fast electron density has nearly dropped to zero at 1 cm from the cathode, since the majority of the fast electrons is already transferred to the slow group.

The densities result in the electric field and potential distribution illustrated in Fig. 5. The potential distribution shows a large variation in the CDS. The position where the potential goes through zero is defined as the CDS-NG interface and is calculated to be at 0.155 cm from the cathode at these discharge conditions. The potential is slightly positive (about 8.4 V) in the NG, which is called the plasma potential. Close to the anode the potential returns again to zero. The electric field shows a large, almost linear increase in the CDS. It does not cross the zero line at the CDS-NG interface but bends off to a small negative value in the NG. It goes through zero at about 0.7 cm and then takes small positive



FIG. 5. Potential and electric field as a function of position from the cathode (100 Pa-1000 V).

![](_page_4_Figure_6.jpeg)

FIG. 6. Energy distribution of the fast electrons at the anode backplate (100 Pa-1000 V).

values. Close to the anode it rises to about 500 V/cm. This region close to the anode where the electric field rises again, where a net positive space charge is observed, and where the potential becomes zero is called the anode zone. At 300 V the electric field and potential distribution show the same behavior.

Figure 6 represents the energy distribution of the fast electrons bombarding the anode. It should be noticed that only the fast electron group is shown, constituting about 23% of the total number of electrons only (see further). The remaining 77% belongs to the slow electron group. When these electrons, which have energies lower than 12 eV, would be included in the figure, the part of the energy distribution below 12 eV would increase to a large extent, and the shape of the energy distribution of higher energies would not be so easily distinguished. Hence, for the sake of clarity, only the energy distribution belonging to the fast electron part is shown. The distribution has a maximum at low energies (below 50 eV), it is low in the middle-energy range (50-800)eV) and rises again slightly at energies of about 900-950 eV. It appears that the electrons have either very low or rather high energies. The explanation for this can be found in the cross sections for the relevant electron collisions; the cross sections of electron impact excitation and ionization reach, indeed, a maximum at about 25 (Ref. 32) and 100 eV (Refs. 15 and 33), respectively. Hence, the low-energy electrons, which possess energies at which the collision cross sections are high, are subject to many collisions thereby losing more energy while the high-energy electrons are too fast for efficient collisions and maintain their energy. A peak is also observed at maximum energy. It means that about 0.6% of the fast electrons have traversed the entire discharge without any collisions. The calculations also yielded information about the total electron multiplication throughout the discharge. Starting from one electron at the cathode, nearly 12 secondary electrons bombard the anode. About 77% of these electrons belong to the slow electron group whereas the remaining 23% still belong to the fast electron group. At 300 V the distribution is shifted to lower energies; a peak at maximum energy is hardly observed and relatively more slow electrons are formed.

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![](_page_5_Figure_0.jpeg)

FIG. 7. Fraction of fast electrons that reenter the CDS after scattering in the NG, as a function of distance to the cathode (100 Pa-1000 V).

We also investigated the influence of the NG on the calculations in the CDS, i.e., the number of electrons that return back to the CDS from the NG. This number appears to be rather high. One electron starting at the cathode returns on the average seven times back to the CDS. Figure 7 shows the position which the electrons can reach in the CDS before they are turned back under the influence of the electric field. This "range" is an almost exponentially decreasing curve. About 70% can reach 0.1 mm into the CDS (z=0.145 cm), about 50% can penetrate 0.2 mm (z=0.135 cm), about 10% can reach 0.8 mm (z=0.075 cm) and less then 5% can go further back than 1 mm (z=0.055 cm). Figure 8 shows the electron density in the CDS, calculated considering the CDS only<sup>17</sup> and also when the NG is incorporated in the model. Due to the backscattering from the NG into the CDS, the density near the CDS-NG interface is 4-5 times higher than when the calculations stop at the interface. This also yields higher excitation and ionization rates. Moreover, the mean energy of the electrons is lowered when the NG is incorporated in the calculations. At the CDS-NG interface the mean energy is halved while the maximum mean energy (at about

![](_page_5_Figure_3.jpeg)

FIG. 9. Fast electron mean energy in the CDS as a function of position from the cathode, calculated considering (1) the entire discharge and (2) the CDS only (100 Pa-1000 V).

0.1 cm from the cathode) is reduced from 700 to 600 eV, as can be seen in Fig. 9. The incorporation of the NG in the calculations hence influences the results to a large extent.

The influence of three-dimensional scattering on the calculations was also investigated: due to the back and forth scattering, the electron density in the beginning of the NG was higher than derived from a model where only forward scattering is considered. This results in a higher ionization rate, as is illustrated in Fig. 10. However the number of fast electrons that can reach the backplate of the cell is less, compared to the case where the electrons all move forward and are not affected by the side walls. Therefore the ionization rate at the end of the NG is lower with threedimensional scattering than with forward scattering. The excitation rate shows the same behavior. Since it is known from optical emission profiles<sup>18</sup> that the excitation rate is characterized by a peak at the beginning of the NG, the calculations with three-dimensional scattering yield more correct results. The total amount of ionization is slightly higher with threedimensional scattering, which yields somewhat higher par-

![](_page_5_Figure_7.jpeg)

FIG. 8. Fast electron density in the CDS as a function of position from the cathode, calculated considering (1) the entire discharge and (2) the CDS only (100 Pa-1000 V).

![](_page_5_Figure_9.jpeg)

FIG. 10. Ionization rate as a function of position from the cathode, calculated (1) with three-dimensional and (2) with forward scattering (100 Pa-1000 V).

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![](_page_6_Figure_0.jpeg)

FIG. 11. Comparison of calculated and experimentally measured electrical currents for different voltages and pressures.

ticle densities and fluxes. The total electrical current is about 30% higher with three-dimensional scattering (4.5 mA/cm<sup>2</sup> compared to  $3.36 \text{ mA/cm}^2$  when forward scattering is considered).

As stated before, the calculations were carried out in the discharge cell represented by Fig. 1, and absorption, reflection and secondary electron emission on the side walls were explicitly taken into account. To demonstrate that this explicit incorporation of the side walls is really necessary, the calculations were carried out in three dimensions as if no side walls existed. The electrons stay then in the plasma for a very long time and can cause a large number of collisions, resulting in high ionization rates, high electron multiplication rates, high densities, etc. The calculated quantities are all about 50 times higher than when the side walls were taken into account. The total electrical current was calculated to be 230 instead of  $4.5 \text{ mA/cm}^2$ . This indicates that the results are unrealistic and that the side walls indeed have to be taken into account.

In order to investigate the influence of Ar gas pressure and discharge voltage on the calculated quantities, the simulations were carried out at different voltages and pressures. Figure 11 shows the calculated total currents as a function of voltage at two pressures. Comparing these calculated values with the ones experimentally measured with the VG 9000 mass spectrometer for a Mo cathode in an Ar discharge,<sup>34</sup> we conclude that the model does not allow to describe the real situation in a quantitative way, although the calculated values are already in the right order of magnitude. The influence of pressure seems somewhat too large. The influence of the voltage seems reasonably well described at low voltages (<600 V) but not yet at higher voltages. In the model, the magnitude of the flux results from the magnitude of the ionization rate which in turn depends on the energy of the electrons. The energy of the electrons follows from the discharge voltage. In this manner the discharge voltage determines the electrical current. At low voltages (<600 V) the mean energy of the electrons is about 100 eV or less, so lying before the maximum in the ionization cross section.<sup>15,33</sup> When increasing the voltage the corresponding electron energy also in-

creases, which yields higher ionization rates and higher electrical currents. However, at voltages higher than 600 V, the mean energy of the electrons is rather high (about 200 eV for 600 V to about 500 eV for 1400 V discharge voltage) and lies in the energy range where the ionization cross section is over its maximum and is nearly constant. The variations in discharge voltage are hence not reflected in variations of ionization rates and electrical currents at these voltages. This feature of constant or even slightly decreasing ionization rates and electron multiplication factors with increasing voltages is also found in other simulation work<sup>15,35-37</sup> at higher voltages. Meyyappan and Kreskovsky<sup>12</sup> calculated the correct (experimental) voltage-current behavior at voltages lower than 200 V, assuming that the electron energy distribution is Maxwellian. This assumption, however, does not agree with experimentally measured electron energy distributions;<sup>38</sup> the latter clearly resemble the distribution calculated in the present model, and it is generally accepted that the electron energy is rather high.

The fact that the current-voltage relations are satisfactorily described at low voltages but not at high voltages leads us to the conclusion that at low voltages the processes incorporated in the model are indeed the dominant ones, but that at higher voltages other processes come into play. First, at higher voltages and higher electron energies, the creation of multiply charged Ar ions can become more important. Ar<sup>2+</sup> ions have a much higher secondary electron emission coefficient<sup>39</sup> which results in an increase of the total current. However, Carman<sup>15</sup> found that the creation rate of  $Ar^{2+}$  is still about 20 times lower than that of  $Ar^+$  at 1000 V, so that incorporating this process will probably not have a large effect. Second, at higher voltages the Ar ions and fast Ar atoms created by charge transfer reach higher energies. Ionization of Ar atoms by impact of fast Ar ions and fast Ar atoms can then come into play. In Ref. 40 it was shown that this process can become dominant at high electrical fields ( $\mathcal{E}/n > 15$  kTd; where 1 Td= $10^{-21}$  V m<sup>2</sup>). Preliminary results have indeed already shown that these processes can have significant effect. Third, the more energetic Ar ions and fast atoms at higher voltages also cause more sputtering during cathode bombardment. The sputtered atoms can be ionized and these ions also contribute to the total electrical current. This effect was indeed found to be important in Ref. 41 where aluminum sputtering discharges in argon were investigated. Another effect that can come into play at high voltages is gas heating as a result of the increased power input. The gas heating effect at constant pressure causes a dilution of the gas according to the ideal gas law. This results in a longer mean free path of the electrons which yields less ionization and hence a lower electrical current. This effect therefore seems to be opposite to the three effects mentioned above and cannot explain the discrepancy between calculated and experimental currents. Finally it was suggested that stepwise ionization from the metastable levels can possibly play a role in explaining the present discrepancy. This effect can indeed be important at low voltages,<sup>42</sup> where a lot of electrons have energies too low for direct ionization from the ground state. However, at the voltages considered in this paper, the effect was found to be negligible,<sup>43</sup> since the electron energies are

![](_page_7_Figure_0.jpeg)

FIG. 12. Relative contribution of the primary, secondary, and thermalized electron group at the anode as a function of voltage at three different pressures.

much higher and an intermediate level for ionization is therefore not needed. Incorporation of the other four processes will however be necessary in future work in order to investigate their influence on the I-V results.

The influence of discharge voltage on other quantities related to the total current like particle densities and fluxes will not be correctly predicted either at voltages above 600 V and will hence not further be discussed here. Only those quantities of which it is believed that the pressure and voltage effects are correctly predicted are presented. From the Monte Carlo simulations it is possible to acquire information about the splitting up of electrons in

- a primary group of electrons that have traversed the whole discharge without collisions,
- a secondary group of electrons that have lost energy by collisions but are still energetic enough for inelastic collisions, and
- (iii) a group of thermalized electrons that are unable to cause inelastic collisions.

Figure 12 shows the splitting up of these three electron groups at the anode backplate as a function of discharge voltage at three different pressures. In contrast to the behavior of these three groups at the interface between CDS and NG.<sup>13,15</sup> the thermalized group is dominant at the end of the discharge plasma at all voltages and pressures (of the order of 80%) whereas the primary group is always smaller than 1.5%. The primary and secondary group increase with increasing voltage and decreasing pressure while the thermalized group shows the opposite behavior. This was to be expected since at lower pressures and higher voltages inelastic collisions are not so frequent and less energy is lost by electrons. The plasma potential seems rather independent of discharge conditions, although the model predicts a slight increase at increasing pressures and voltages. The position of electrical field reversal increases with increasing voltage and decreasing pressure.

# **IV. CONCLUSION**

A self-consistent hybrid Monte Carlo-fluid model is presented that describes a dc glow discharge in Ar. The Monte Carlo part simulates the fast electrons while the fluid part describes the gas ions and slow electrons. The calculated quantities are collision rates and energy distributions of the fast electrons, fluxes and densities of the plasma species, and the electric field and potential distributions. Comparison was made between Monte Carlo calculations carried out in the CDS only and similar calculations in the entire discharge. It was found that the influence of electrons from the NG that reenter the CDS was rather high. The calculations were carried out in a three-dimensional geometry corresponding to a typical discharge cell of the Fisons VG 9000 glow discharge mass spectrometer. The influence of three-dimensional scattering and the incorporation of side walls in the Monte Carlo simulations on the calculated quantities was investigated. The three-dimensional scattering enhanced the ionization rates and hence the particle densities and fluxes and yielded a more realistic total electrical current compared to simple forward scattering. Moreover the rate profiles showed a behavior that agreed better with experimental optical emission data. The incorporation of side walls in the discharge was found to be extremely important since the calculated quantities without side walls were far too high (i.e., the obtained electrical current was 230 instead of about 5 mA/cm<sup>2</sup>). The influence of pressure and voltage on the calculated quantities was studied. The pressure dependence seems somewhat too high. The voltage dependence is satisfactorily predicted at voltages lower than 600 V, but not yet at higher voltages, although the calculated values are close to the experimental ones. It is suggested that at higher voltages other processes can come into play, like

- (i) creation of  $Ar^{2+}$  ions,
- (ii)  $Ar^+$  ion and fast Ar atom impact ionization,
- (iii) contribution of sputtered atoms and ions and
- (iv) the effect of gas heating which is opposite to the three above processes. In future work these processes will be incorporated.

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# APPENDIX: THE EXPONENTIAL FINITE DIFFERENCE SCHEME

Consider the one-dimensional continuity equation [i.e., Eqs. (5) and (6)]:

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$$\frac{\delta n}{\delta t} + \frac{\delta j}{\delta x} = r \tag{A1}$$

and the flux equation of diffusion and migration [i.e., Eq. (8) and (9)]:

$$j=bn-a \frac{\delta n}{\delta x}$$
 ( $b=\pm \mu \mathcal{E}, a=D$ ). (A2)

Discretization of Eq. (A1) yields

$$\frac{n_{i,k+1} - n_{i,k}}{\Delta t} + \frac{j_{i+1/2,k+1} - j_{i-1/2,k+1}}{\Delta x} = r.$$
 (A3)

According to the Scharfetter–Gummel exponential scheme, j is assumed constant between mesh points. Integration of Eq. (A2) with  $j=\text{constant}=j_{i+1/2,k+1}$ , yields for  $j_{i+1/2,k+1}$  (time level k+1 is omitted):

$$j_{i+1/2} = \frac{b_{i+1/2}[n_{i+1} - n_i \exp(-\rho_{i+1/2})]}{1 - \exp(-\rho_{i+1/2})},$$
 (A4)

where

$$\rho_{i+1/2} = -\frac{b_{i+1/2}}{a_{i+1/2}} \,\Delta x$$

Inserting Eq. (A4) into Eq. (A3) leads finally to

$$\frac{n_{i,k+1}-n_{i,k}}{\Delta t} + \frac{X_i^{+}n_{i+1,k+1}+X_i^{-}n_{i-1,k+1}+X_i^{c}n_{i,k+1}}{\Delta x} = r,$$

where

$$X_{i}^{+} = \frac{b_{i+1/2}}{1 - \exp(-\rho_{i+1/2})} = f_{+}(V_{i}, V_{i+1}),$$

$$X_{i}^{-} = \frac{b_{i-1/2} \exp(-\rho_{i-1/2})}{1 - \exp(-\rho_{i-1/2})} = f_{-}(V_{i}, V_{i-1}),$$

$$X_{i}^{c} = -X_{i}^{+} \exp(-\rho_{i+1/2}) - X_{i}^{-} \exp(+\rho_{i-1/2})$$

$$= f_{c}(V_{i-1}, V_{i}, V_{i+1}).$$

Therefore, Eq. (A5) and hence also Eq. (A1) can be seen as function of the following unknowns:  $f(n_{i-1,k+1}, n_{i,k+1}, n_{i+1,k+1}, V_{i-1,k+1}, V_{i+1,k+1})$ .

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