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Citation for published version (APA):

Hagelaar, G. J. M., Kroesen, G. M. W., & Klein, M. H. (2000). Energy distribution of ions and fast neutrals in microdischarges for display technology. *Journal of Applied Physics*, 88(5), 2240-2245. <https://doi.org/10.1063/1.1287758>

DOI:

[10.1063/1.1287758](https://doi.org/10.1063/1.1287758)

Document status and date:

Published: 01/01/2000

Document Version:

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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Energy distribution of ions and fast neutrals in microdischarges for display technology

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(Received 15 May 2000; accepted for publication 5 June 2000)

In this work we present simple theoretical predictions as well as full Monte Carlo calculations of the energy distribution of the ion and fast neutral fluxes impinging on the materials that surround the microdischarges in plasma display panels and plasma addressed liquid crystal displays. We consider various rare gas ion species in different microdischarge designs. Often simple theoretical distribution functions are in good agreement with the results of Monte Carlo calculations. Under the influence of symmetric charge transfer collisions the ion energy distribution is essentially different from a Maxwellian distribution, and the ion motion is strongly orientated along the electric field. The flux of the fast neutrals formed by symmetric charge transfer is usually even larger than the ion flux itself. © 2000 American Institute of Physics. [S0021-8979(00)10417-7]

I. INTRODUCTION

Recently, two alternative display technologies have emerged, that both make use of plasma. The first and best known is the plasma display panel (PDP) technology, using the ultraviolet emission from microdischarges to excite phosphors.¹ The other is the plasma addressed liquid crystal (PALC) technology, which uses very similar microdischarges as electrical switches for the addressing of a liquid crystal layer.^{2,3} In both technologies, an incident flux of energetic plasma particles causes damage to the materials surrounding the microdischarges, thus limiting the lifetime of the displays. Understanding and predicting this phenomenon requires knowledge of the energy distribution of the plasma particles impinging on the material surfaces. Of particular interest are ions and fast neutrals with an energy beyond the sputtering threshold, which varies from 10 to 100 eV, depending on surface material and incident particle species.⁴

Research on microdischarges leans heavily on discharge modeling tools, in view of experimental difficulties due to their small size. By the use of self-consistent fluid models, a good picture can be obtained of the electric fields and the particle densities and fluxes in microdischarges.⁵⁻⁹ However, these fluid models do not describe the energy distributions of the plasma particles. In this work, we attempt to predict—on the basis of the results of fluid models—the energy distribution of ions and fast neutrals impinging on the surface. We pursue both an elementary theoretical approach (Sec. II and III) and a more comprehensive Monte Carlo approach (Sec. IV).

Although many different designs exist for the PDP and PALC discharges, they all have very similar discharge characteristics. The inter-electrode gap is on the order of a few hundred microns, the gas pressure is a few hundred torrs. Typical discharge voltages are a few hundred volts, where

the reduced electric field can reach values of up to 500 V cm⁻¹ Torr⁻¹ locally in front of the surface. Figure 1 shows some typical profiles of the electric potential in the microdischarge sheaths. The sheaths are always highly collisional: typically the ion mean free path is in the submicron range, about two orders of magnitude smaller than the sheath thickness. This implies that the ion energy distribution is determined by the local electric field rather than the total sheath voltage, and that the ion mean energy is only a small fraction of this voltage. The discharge gas used in PDP and PALC displays is typically a rare gas or a mixture of rare gases. The main ion species are ionized discharge gas atoms. Typical ionization rate profiles are shown in Fig. 1. Molecular ions, such as He₂⁺ and Xe₂⁺, are also found, but tend to get dissociated in the sheaths: beyond a certain energy limit on the order of 1 eV, the dissociation cross sections become so large that survival is extremely unlikely.^{10,11}

We, thus, focus on atomic rare gas ions, moving toward the surface in a highly collisional sheath.

II. ION ENERGY DISTRIBUTION

On their path, the ions undergo collisions predominantly with neutral gas particles that are virtually at rest. Two different collision types are to be distinguished. First, there are elastic collisions, leading to the (more or less isotropic) scattering of the ions in the center of mass (CM) system of the colliding particles. Second, when moving in their parent gas, the ions are subject to symmetric charge transfer collisions. In such a collision the ion charge is transferred to a parent gas particle, while the velocities of both particles remain unaffected: after the collision the ion is virtually at rest. A symmetric charge transfer collision is equivalent to an elastic collision where the ion is scattered over 180° in the CM system.¹² As appears from Fig. 2, the cross sections of these collision processes are approximately constant over the en-

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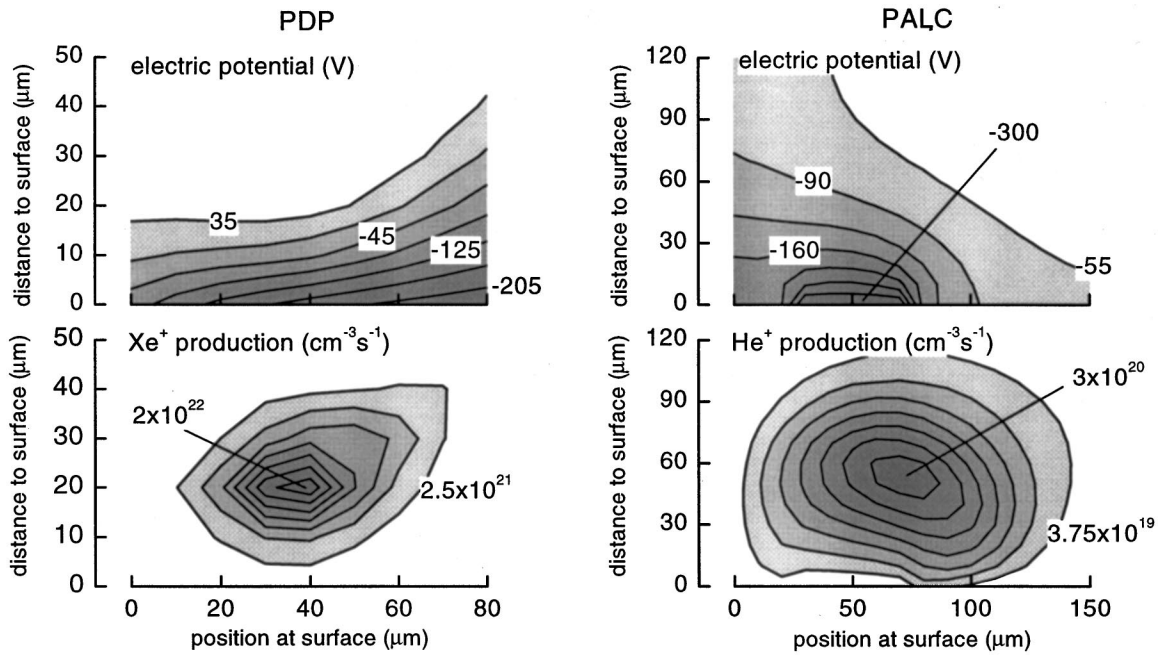


FIG. 1. Contour plots of the electric potential and the ion formation rate in front of the surface, at the position and the moment where the incident ion flux is the highest and the most energetic. A typical situation is shown for an alternating current PDP discharge in Ne–Xe 5% (left-hand side) and for a PALC discharge in He (right-hand side). These profiles were obtained with a fluid model.

ergy range of interest. The symmetric charge transfer cross section is usually considerably larger than the cross section for elastic collisions.

If symmetric charge transfer is the dominant collision process, the ion energy distribution can be predicted with an elementary theoretical approach. Consider an ion with mass m and charge q in a uniform electrostatic field E , being subject to a constant acceleration qE/m . Assume that the ion loses all its energy when it collides. The ion energy as a function of the time t after a collision is then

$$\epsilon = \frac{q^2 E^2}{2m} t^2. \quad (1)$$

When looking at an ensemble of such ions, the fraction of them with an energy in between ϵ and $\epsilon + d\epsilon$ is proportional to the average time \overline{dt} that one ion spends in this energy interval during a free path:

$$f(\epsilon) d\epsilon \propto \overline{dt}. \quad (2)$$

According to Eq. (1), the ion reaches the energy interval after

$$t = \frac{\sqrt{2m\epsilon}}{qE}, \quad (3)$$

that is, if it does not collide before. It then stays in the interval for a while dt :

$$dt = \left(\frac{dt}{d\epsilon} \right) d\epsilon = \frac{1}{qE} \sqrt{\frac{m}{2\epsilon}} d\epsilon. \quad (4)$$

On average, the time spent in the interval is

$$\overline{dt} = P(t) dt, \quad (5)$$

where P is the probability that no collision takes place before time t , which is related to the collision frequency ν according to

$$P(t) = \exp\left(-\int_0^t \nu(t') dt'\right). \quad (6)$$

Assuming the gas particles to be at rest, the collision frequency is a function of ϵ

$$\nu(\epsilon) = N\sigma(\epsilon) \sqrt{\frac{2\epsilon}{m}}, \quad (7)$$

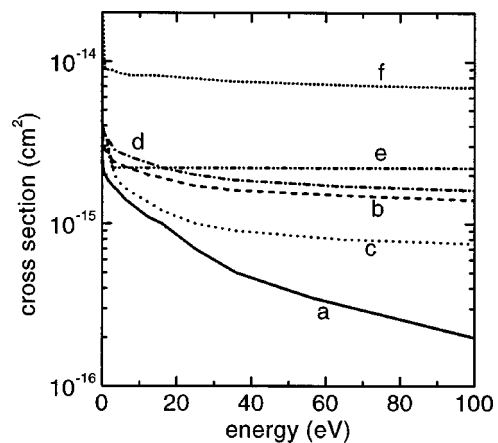


FIG. 2. Cross sections of some ion–neutral collisions relevant to PDP and PALC discharges. (a) elastic He⁺–He, (b) charge transfer He⁺–He, (c) elastic Ne⁺–Ne, (d) charge transfer Ne⁺–Ne, (e) elastic Xe⁺–Xe, (f) charge transfer Xe⁺–Xe. The data are taken from Refs. 13 and 14. A constant value derived from mobility data (Ref. 15) is assumed for cross-section (e), except at low energy, where it is assumed to be the same as (c). The energy on the horizontal axis is the ion impact energy.

where N is the gas particle density and σ the collision cross section. On combining Eqs. (2)–(7), one finds straightforwardly that

$$f(\epsilon)d\epsilon \propto \frac{1}{\sqrt{\epsilon}} \exp\left(-\frac{N}{qE} \int_0^\epsilon \sigma(\epsilon')d\epsilon'\right) d\epsilon. \quad (8)$$

The appropriate proportionality constant in Eq. (8) can be found by normalizing the distribution to unity. For a constant collision cross section this yields

$$f(\epsilon)d\epsilon = \frac{1}{\sqrt{\pi\omega\epsilon}} \exp\left(-\frac{\epsilon}{\omega}\right) d\epsilon, \quad (9)$$

where the parameter ω is given by

$$\omega = \frac{qE}{N\sigma}. \quad (10)$$

Averaging ϵ over the distribution (9) shows that ω is equal to twice the ion mean energy. Note that the distribution function (9) is essentially different from the Maxwellian distribution function: the Maxwellian function is zero at $\epsilon=0$, whereas Eq. (9) goes to infinity for $\epsilon \rightarrow 0$. Realize, furthermore, that all the ions move in the direction of the electric field: the ion motion is as anisotropic as can be. The ion drift velocity w is therefore found by simply averaging the velocity $(2\epsilon/m)^{1/2}$ over Eq. (9):

$$w = \int_0^\infty \sqrt{\frac{2\epsilon}{m}} f(\epsilon)d\epsilon = \sqrt{\frac{2\omega}{\pi m}}. \quad (11)$$

Rearranging Eq. (11) yields an alternative expression for ω :

$$\omega = \frac{\pi}{2} m w^2, \quad (12)$$

which is of great practical use, because it provides a way to estimate the ion energy distribution merely from the ion drift velocity, which is calculated in fluid models.

Expression (9) represents the energy distribution of the ion density. It is important to realize, that this is not the energy distribution of the ion flux hitting the surface. In view of the fact that all ions move in exactly the same direction, the latter distribution can be found by weighting Eq. (9) with the ion velocity $(2\epsilon/m)^{1/2}$. After normalization, we find

$$g(\epsilon)d\epsilon = \frac{1}{\omega} \exp\left(-\frac{\epsilon}{\omega}\right) d\epsilon \quad (13)$$

for the ion flux energy distribution. It follows from averaging ϵ over distribution (13) that the average energy of the incident ions is equal to ω , twice the ion mean energy.

III. FAST NEUTRALS

The ion–neutral collisions lead to the formation of energetic neutral particles. The ion flux is thus accompanied by a flux of fast neutrals. In this section we estimate the importance of this fast neutral flux. We consider the case that there is only one species of gas particles and that symmetric charge transfer is the predominant collision process for ions, as before. After a charge transfer collision, the neutral particle has the initial ion energy, which it loses again via elastic

collisions with gas particles that are virtually at rest. The energy transferred to a gas particle in an elastic collision is¹³

$$\Delta\epsilon = \frac{1}{2}(1 - \cos\theta)\epsilon, \quad (14)$$

where ϵ is the initial energy of the fast neutral and θ is the scattering angle in the CM system. In case the scattering is isotropic, the probability of $\cos\theta$ is uniformly distributed between -1 and 1 . Equation (14) then implies that the probability of $\Delta\epsilon$ is uniformly distributed between 0 and ϵ : the energy is redistributed completely randomly over the two colliding particles.

The density $N'(\epsilon)d\epsilon$ of the fast neutrals with an energy in between ϵ and $\epsilon+d\epsilon$ can be found by balancing the rates of their production and loss. Production of neutrals in the energy interval occurs by symmetric charge transfer and by elastic collisions of other fast neutrals with an energy larger than ϵ . The neutrals leave the energy interval through elastic collisions. Balancing the rates of these three processes, we write

$$\begin{aligned} N\sigma(\epsilon) \sqrt{\frac{2\epsilon}{m}} n_i f(\epsilon) d\epsilon \\ + \int_\epsilon^\infty \left[\frac{2d\epsilon}{\epsilon'} NQ(\epsilon') \sqrt{\frac{2\epsilon'}{m}} N'(\epsilon') \right] d\epsilon' \\ = NQ(\epsilon) \sqrt{\frac{2\epsilon}{m}} N'(\epsilon) d\epsilon, \end{aligned} \quad (15)$$

where n_i is the (total) ion density and Q is the cross section for elastic neutral–neutral collisions. The second term on the left—representing the elastic production rate—contains the factor $2d\epsilon/\epsilon'$. This is the probability that after an elastic collision between a neutral with energy $\epsilon' > \epsilon$ and a neutral at rest, one of the two particles ends up in the energy interval $d\epsilon$, assuming the energy redistribution to be completely random.

The density $N'(\epsilon)d\epsilon$ can be found from Eq. (15) by substituting $N'(\epsilon) = \epsilon^{1/2}h(\epsilon)/Q(\epsilon)$ or $N'(\epsilon) = \epsilon^{-5/2}h(\epsilon)/Q(\epsilon)$, differentiating the entire equation with respect to ϵ , and solving the resulting differential equation for $h(\epsilon)$. After backsubstitution of $h(\epsilon)$ we find

$$\begin{aligned} N'(\epsilon)d\epsilon = \frac{\sigma(\epsilon)}{Q(\epsilon)} n_i f(\epsilon) d\epsilon \\ + \frac{2}{Q(\epsilon)} n_i \epsilon^{-5/2} \int_\epsilon^\infty [\sigma(\epsilon') \epsilon'^{3/2} f(\epsilon')] d\epsilon' d\epsilon. \end{aligned} \quad (16)$$

The first term of this expression is the density one would find from Eq. (15) without the elastic production term: obviously it represents the neutrals that have been produced directly by charge transfer collisions. For constant cross sections σ and Q the ion energy distribution is given by Eq. (9), and Eq. (16) becomes

$$N'(\epsilon)d\epsilon = \frac{\sigma}{Q} \left(1 + \frac{2\omega}{\epsilon} + \frac{2\omega^2}{\epsilon^2} \right) n_i f(\epsilon) d\epsilon, \quad (17)$$

where $f(\epsilon)$ is once again given by Eq. (9).

The flux $\Gamma'(\epsilon)d\epsilon$ of the fast neutrals with an energy in between ϵ and $\epsilon+d\epsilon$ is the product of their density (17) and their average velocity. Since not all neutrals move in exactly the same direction, the average velocity is somewhat smaller than $(2\epsilon/m)^{1/2}$. An upper limit for the flux is thus found as

$$\begin{aligned} \Gamma'(\epsilon)d\epsilon &< \frac{\sigma}{Q} \left(1 + \frac{2\omega}{\epsilon} + \frac{2\omega^2}{\epsilon^2} \right) n_i \sqrt{\frac{2\epsilon}{m}} f(\epsilon)d\epsilon \\ &= \frac{\sigma}{Q} \left(1 + \frac{2\omega}{\epsilon} + \frac{2\omega^2}{\epsilon^2} \right) \Gamma_i g(\epsilon)d\epsilon, \end{aligned} \quad (18)$$

where Γ_i is the (total) ion flux and $g(\epsilon)$ is given by Eq. (13). Note, however, that the neutrals represented by the first term of Eq. (16) do all move in exactly the same direction, so that a lower limit for the flux is given by

$$\Gamma'(\epsilon)d\epsilon > \frac{\sigma}{Q} n_i \sqrt{\frac{2\epsilon}{m}} f(\epsilon)d\epsilon = \frac{\sigma}{Q} \Gamma_i g(\epsilon)d\epsilon. \quad (19)$$

Since σ is generally larger than Q , this means that the fast neutral flux influencing the surface exceeds the ion flux, for every energy.

IV. MONTE CARLO CALCULATIONS

Although the above theoretical treatment gives a good feel for the ion and neutral energy distributions, it is far from complete: the influence of (isotropic) elastic ion–neutral collisions is neglected, the electric field and cross sections are assumed to be constant, etc. A more complete picture of the energetic ions and neutrals can be obtained by Monte Carlo modeling. In this section we present the Monte Carlo calculation of the ion and fast neutral energy distributions in some typical microdischarge configurations.

The Monte Carlo model is basically the same as the model described in Ref. 16. It simulates the individual paths of a large number of ions and fast neutrals. The ions are sampled randomly from an ionization rate profile that has been calculated *a priori* with a fluid model, and then followed until they reach the surface. The electric field that accelerates the ions is also taken from the fluid calculation. The two-dimensional fluid model that we use for this purpose is described in Ref. 9. In addition to the ions, the neutrals with an energy higher than 1 eV are followed from all collision events occurring in the simulations. Both symmetric charge transfer collisions and isotropic elastic collisions are taken into account. The cross sections of the ion–neutral collisions are considered as functions of the impact energy, as represented in Fig. 2. For the cross-sections of neutral–neutral collisions, we assume constant values, derived from a hard-sphere model:¹⁷ $1.36 \times 10^{-15} \text{ cm}^2$ for He–He, $1.87 \times 10^{-15} \text{ cm}^2$ for Ne–Ne, $3.85 \times 10^{-15} \text{ cm}^2$ for Ne–Xe, and $6.53 \times 10^{-15} \text{ cm}^2$ for Xe–Xe.

The trajectory of an ion or fast neutral is simulated by the following procedure:

(1) A random value is chosen for the time until the next collision, taking into account the appropriate probability distribution given by the total collision frequency. For ions we use the null collision method.¹⁸

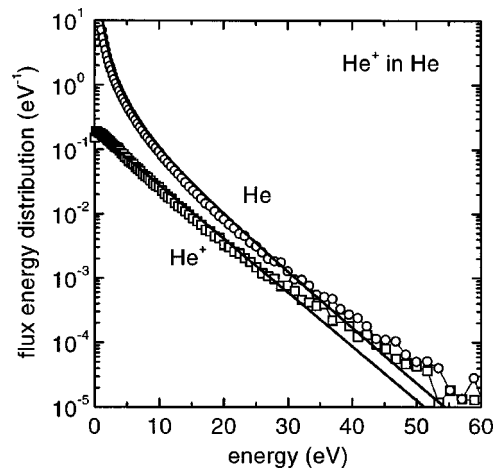


FIG. 3. Energy distribution of the He^+ flux and the accompanying fast neutral flux impinging on the cathode in a PALC discharge in pure He. The neutral flux energy distribution is not normalized to unity but related to the ion flux as $\Gamma'(\epsilon)/\Gamma_i$. The connected symbols represent the result of a Monte Carlo calculation, the lines are the theoretical functions (13) and (18).

(2) The free path covered during this time is calculated by integrating the equation of motion.

(3) The type of the collision is determined randomly, taking into account the relative probability of all possible collision types.

(4) The velocities of the colliding particles are transformed to the CM system.

(5) The effect of the collision is simulated. In the case of symmetric charge transfer, the ion and neutral velocities are simply swapped. In the case of an isotropic elastic collision, the velocities are turned to random (but mutually opposite) directions.

(6) The velocities are transformed back to the laboratory system.

(7) In the event that the laboratory energy of the original target particle now exceeds 1 eV, it is considered as a fast neutral and is followed, too.

The procedure, steps (1)–(7), is repeated until the ion or fast neutral reaches the surface. Neutrals are no longer followed once their energy has dropped below 1 eV. Reflection at the surface is not considered. For more details on the Monte Carlo model, see Ref. 16.

We applied the Monte Carlo model to a standard PALC discharge in pure helium. This case is relatively simple. The microdischarge operates in the direct current mode, where the main ion surface bombardment occurs at the cathode. The gas pressure is 150 Torr. The only ion species of importance is He^+ . The two-dimensional profiles of the electric potential and the ionization rate—input data of the Monte Carlo simulations—are shown in Fig. 1. We simulated the trajectories of ten million ions and all the neutrals beyond 1 eV formed in collisions. Figure 3 shows the calculated energy distribution $g(\epsilon)$ of the incident ion flux, as well as the energy distribution $\Gamma'(\epsilon)/\Gamma_i$ of the incident fast neutral flux. Note that the neutral energy distribution is not normalized to unity, but scaled along with the ion energy distribution. Besides the Monte Carlo results, the theoretical distribution

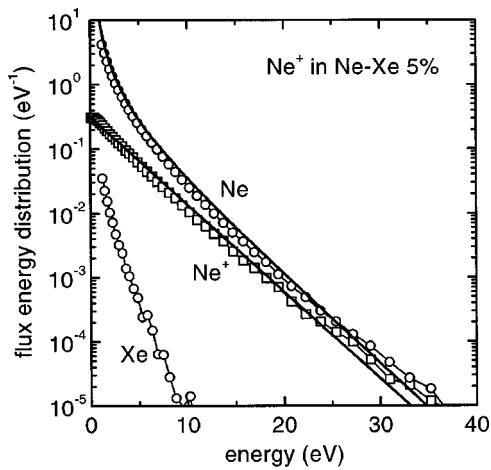


FIG. 4. Energy distribution of the Ne^+ flux and the accompanying fast neutral fluxes impinging on the surface in an alternating current DPD discharge in Ne–Xe 5%, at the moment and the position where they reach their maximum value. The neutral flux energy distribution is not normalized to unity but related to the ion flux as $\Gamma'(\epsilon)/\Gamma_i$. The connected symbols represent the result of a Monte Carlo calculation, the lines are the theoretical functions (13) and (18).

function (13) and the upper limit (18) are represented, where the parameter ω was found from the calculated drift velocity by Eq. (12). The fast neutral flux in the Monte Carlo simulation turns out to be close to the upper limit (18).

Then we considered a typical design¹⁹ for a surface-type alternating current PDP in a mixture of 95% Ne and 5% Xe. This case is more complicated. In contrast to the PALC discharge, the microdischarge in the PDP design has a transient character. However, the typical lifetime of individual ions is shorter than the typical time scale for the electric field variations. The gas pressure is 450 Torr. Two ion species are important: Ne^+ and Xe^+ . Figure 1 shows the electric potential and Xe^+ formation rate at the moment and position of maximum ion flux. The Ne^+ formation rate is not shown here but is similar to that of Xe^+ . The calculated energy

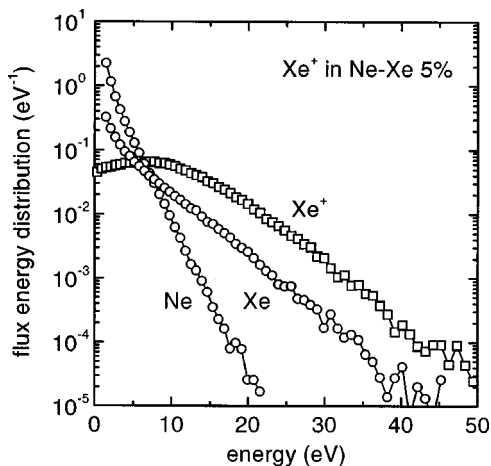


FIG. 5. Energy distribution of the Xe^+ flux and the accompanying fast neutral fluxes impinging on the surface in an alternating current PDP discharge in Ne–Xe 5%, at the moment and the position where they reach their maximum value. The neutral flux energy distribution is not normalized to unity but related to the ion flux as $\Gamma'(\epsilon)/\Gamma_i$. These data were calculated with a Monte Carlo model.

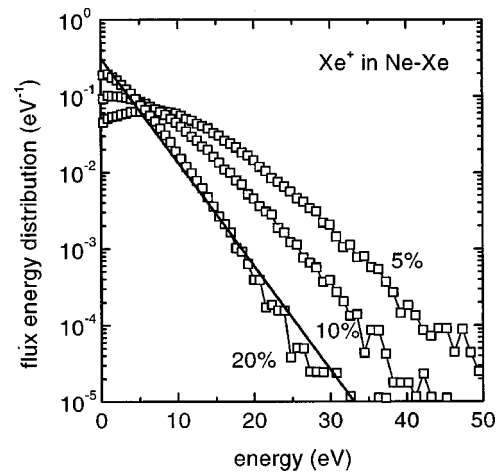


FIG. 6. Energy distribution of the Xe^+ flux impinging on the surface in an alternating current discharge in Ne–Xe, for different percentages of Xe. The connected squares represent the results of Monte Carlo calculations, the line gives the theoretical estimate (13) for 20% xenon.

distributions are shown in Figs. 4 and 5. For Ne^+ , Eqs. (13) and (18) are once again a good approximation of the Monte Carlo results.

For Xe^+ , however, the situation is entirely different. For these ions, the parent gas only constitutes a small percentage of the total gas mixture, so that the Xe^+ energy distribution is determined by elastic collisions with Ne rather than by symmetric charge transfer. The Eqs. (13), (18), and (19) do therefore not apply. At low energy ($\epsilon < 10$ eV), the Xe^+ distribution is underpopulated compared to the exponential function (13). In contrast to what we found for Ne^+ , the fast neutrals produced by Xe^+ are less important than the ions themselves. This is not surprising, since elastic collisions are far less efficient in forming fast neutrals than charge transfer collisions. Figure 6 demonstrates that if we increase the Xe percentage, the influence of symmetric charge transfer collisions rapidly grows. The Xe^+ energies decrease and the energy distribution approaches form (13). For 20% of xenon, Eq. (13) already gives a reasonable description.

V. CONCLUSIONS

For ions that mainly undergo charge transfer collisions, the energy distribution can be found from the drift velocity as a simple theoretical function, which is essentially different from the Maxwellian distribution function. Also, the energy distribution of the fast neutrals formed in symmetric charge transfer collisions is well described by a theoretical function. The motion of both ions and fast neutrals is strongly orientated along the electric field. In general, the fast neutral flux is larger than the ion flux itself. The simple functions fail to describe the energy distribution of ions that mainly undergo elastic collisions. Full Monte Carlo calculations are required in this case.

ACKNOWLEDGMENT

This work was supported by the Philips Research Laboratories in Eindhoven, The Netherlands.

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