Energy distributions of particles striking the cathode in a glow discharge

I. Abril, A. Gras-Marti, J. A. Vallés-Abarca
Departamento de Fisica, L.C.F.C.A., Facultad de Ciencias, Universidad de Alicante, Apartado 99, Alicante, Spain
(Received 27 June 1983)

Charge-exchange collision in the cathode fall region of an abnormal glow discharge is assumed to be the mechanism which limits the energy of the ions and generates the energetic neutrals bombarding the cathode. The model used by W. D. Davis and T. A. Vanderslice [Phys. Rev. 131, 219 (1963)] for the calculation of the ion distribution was not applied to calculate the distribution of neutrals. A transport formulation is proposed that allows the calculation of both the distributions of energetic ions and fast neutrals.

In a pioneering work, Davis and Vanderslice[1] measured the energy distributions of ions bombarding the cathode in abnormal glow discharges. They also modeled these distributions and remarked on the role of charge-exchange collisions between energetic ions and the atoms of the discharge gas in reducing the effective length over which the ions accelerate. These collisions take place in the dark space or cathode-fall region. The basic model proposed in Ref. 1, and the good agreement between the measured and calculated ion distributions, has been of interest to many researchers.[2] The energy distribution of fast energetic neutrals was not calculated in Ref. 1.

Interest in gas discharges has been recently renewed because of applications in dry etching and thin-film deposition.[3,4] Some of the stages in the deposition process have already been modeled by us, including the energy distributions of sputtered fluxes at given distances from the target,[5] the spatial profiles of sputtered particles diffusing in the gas phase,[6] and the calculation of thickness profiles of deposited films.[7] Full characterization of a plasma etching or sputtering system requires the knowledge of the energy distributions of energetic particles bombarding the cathode in the discharge, since they determine the sputtering rate of the target thereby located.[4,8-11]

Several papers[9-13] attempt to tackle the calculation of the energy spectra of neutrals and/or ions, occasionally modifying some of the assumptions proposed in Ref. 1. These calculations are not always transparent in the application of the statistics of the collision processes. It turns out that a simple formulation, to be presented here, allows calculation of both the energy distribution of energetic ions and neutrals. Our formulation reproduces the ion distributions derived in Refs. 1, 9, 11, and 13, whereas the distributions of neutrals are at variance with the results derived in Refs. 10 and 12 and are more general than the calculations of Ref. 9.

In abnormal glow discharges, the major part of the primary ionization processes take place in the negative-glow region.[14,15] Ions leaving this region with thermal energies feel the electric field and accelerate towards the cathode. The mean free path for symmetric (resonant) charge-exchange collisions λ is roughly constant over the energy range of interest.[16] Take *one* singly charged ion (a primary or "source" ion) at rest at the interface between the negative-glow and the cathode fall regions of the discharge, the plane $x=0$. Let $A(x,E)\,dE$ be the probability of finding an ion at x with kinetic energy in the interval (E,dE). This ion may either be the accelerated source ion, or an ion generated in a sequence of collisions initiated by the source ion. If J_0 (ions $cm^{-2}s^{-1}$) is the ion flux at $x=0$, then the ion current density at x, with energy in the interval (E,dE), is $J_0A(x,E)\,dE$. The spatial evolution of $A=A(x,E)$ is given by

$$\frac{\partial A}{\partial x}=e\frac{d\phi}{dx}\frac{\partial A}{\partial E}-\frac{A}{\lambda}+\frac{\delta(E)}{\lambda} . \tag{1}$$

The first term on the right-hand side is due to the accelerating field. $e=1.6\times10^{-19}$ C is the electron charge and $\phi(x)$ is the electric potential in the dark space. The effect of charge-exchange collisions is to reduce the number of energetic ions (the term $-A/\lambda$) and to increase the number of ions generated at rest [the source term $\delta(E)/\lambda$, where δ is Dirac's delta].

With the initial condition $A(0,E)=\delta(E)$ corresponding to the source ion initially at rest, the solution of Eq. (1) is

$$A(x,E)=\exp(-x/\lambda)\delta(E+e\phi(x))+\int_0^x(dx'/\lambda)\exp[-(x-x')/\lambda]\delta(E+e(\phi(x)-\phi(x'))) . \tag{2}$$

The first term in Eq. (2) represents the contribution of the source ion that has not undergone any collision from $x=0$ up to x, while the second term is the contribution due to generated ions. Since there is only one ion at any x, per source ion starting at $x=0$, the distribution is normalized,

$$\int_0^{E_m}dE\,A(x,E)=1 , \tag{3}$$

where

$$E_m=-e\phi(x) \tag{3'}$$

is the kinetic energy of a singly charged ion that travels from $x=0$ to x without colliding.

More explicitly, integrating Eq. (2) the distributions are given by

$$A(x,E)=\exp(-x/\lambda)\delta(E-E_m)+[\lambda e\phi'(x_0')]^{-1}\exp[-(x-x_0')/\lambda] , \tag{2'}$$

where $E\le E_m$, $\phi'=|d\phi/dx|$, and x_0' is the root of $e\phi(x')=E-E_m$. The complicated spatial dependence of the electric potential in the dark space is generally approxi-

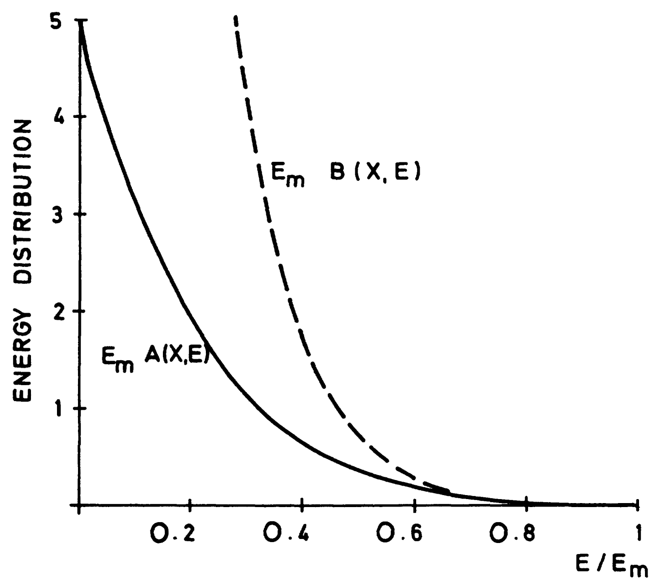

FIG. 1. Energy distributions of ions and neutrals, normalized to one ion entering the dark space at $x=0$. Solid line: ions, from Eq. (2′) (only the second term has been represented). Dashed line: neutrals, from Eq. (5). Dimensionless units in both axes are based on the maximum energy E_m, Eq. (3′). For linear electric field variation in the dark space and a ratio $x/\lambda=10$.

mated by a linear[9,13] or quadratic[1,10–13,17] variation with distance. The distributions in Eq. (2′) coincide with those given in Refs. 1, 9, 11, and 13. In general, they are monotonically decreasing functions of energy for $x/\lambda \gtrsim 1$ (cf. Fig. 1), and reverse trend for $x/\lambda \lesssim 1$ (cf. Fig. 4 in Ref. 1 and Fig. 3 in Ref. 11).

Hosokawa[10] proposes different models for the cases $\lambda \gtrsim x$ and $\lambda \lesssim x$. Owing to the use of erroneous attenuation factors in both cases, to treat the collision statistics, his results do not coincide with ours. It should be noted in this connection that Eqs. (1)–(2′) are valid irrespective of the value of the ratio x/λ.

Every charge-exchange collision in the dark space originates an energetic neutral with the energy of the parent ion. This neutral is assumed to proceed without colliding. The energy distribution of the neutrals $B(x,E)$, per source ion, is given by the evolution equation

$$\frac{\partial B(x,E)}{\partial x}=\frac{A(x,E)}{\lambda} . \quad (4)$$

With the initial condition $B(0,E)=0$, one gets

$$B(x,E)=\int_0^x (dx'/\lambda)A(x',E) , \quad (5)$$

where A is given by Eq. (2′). The total number of neutrals at x, per source ion, follows from Eqs. (3) and (5),

$$\int_0^{E_m} dE\, B(x,E)=x/\lambda . \quad (6)$$

This is an obvious result since λ^{-1} represents the average number of collisions per unit distance in a Poisson process[18] and an energetic neutral is generated in every collision.

The distributions calculated from Eq. (5) are monotonically decreasing functions of energy in the range of parameters of interest ($x/\lambda \gtrsim 1$). One particular case is plotted in Fig. 1. Some papers[9,10,12] have attempted to derive the distribution B, but the results in Refs. 10 and 12 differ from the ones given above. Only the uniform field case was treated in Ref. 9.

The physical origin of the distributions in Eqs. (2) and (5), as multiple-collision processes, can be made explicit by means of a discretized model where those results are obtained as a superposition of all possible sequences of events initiated by a given ion, comprising from zero up to an infinite number of collisions.[8,19,20]

When the distributions of fast neutrals are calculated from Eq. (5), it turns out that, under pressure conditions such that $x/\lambda \gg 1$, the sputtering rate in discharge systems may be dominated by energetic neutrals which are much more abundant than the secondary ions striking the target located at the cathode. In the particular example of Fig. 1, this result is clear when both energy distributions of ions and neutrals are compared. Details of these results, which are of interest in applications, will be given elsewhere.[8]

ACKNOWLEDGMENTS

This work is part of a research project under the auspices of the Spanish-CAICYT (Proyecto No. 599), in collaboration with the University of Salford (financially supported by The British Council and the Spanish MEC). The interest of Professor R. H. Ritchie and the constructive criticism expressed by Dr. O. H. Crawford, both at Oak Ridge National Laboratory, are warmly appreciated. H. Mase kindly provided a copy of the material quoted in Ref. 13.

[1]W. D. Davis and T. A. Vanderslice, Phys. Rev. 131, 219 (1963).
[2]A survey of the Science Citation Index (ISI) yields at least 70 quotations of Ref. 1 during the period 1964–1981.
[3]*Thin Film Processes*, edited by J. L. Vossen and W. Kern (Academic, New York, 1978).
[4]B. Chapman, *Glow Discharge Processes* (Wiley, New York, 1980).
[5]A. Gras-Marti and J. A. Vallés-Abarca, Le Vide-les Couches Minces 212, 397 (1982); J. Appl. Phys. 54, 1071 (1983).
[6]J. A. Vallés-Abarca and A. Gras-Marti, J. Appl. Phys. (in press).
[7]G. Gonzalez-Diaz, I. Martil, F. Sanchez-Quesada, M. Rodriguez-Vidal, A. Gras-Marti, and J. A. Vallés-Abarca, J. Vac. Sci. Technol. (in press).
[8]I. Abril, A. Gras-Marti, and J. A. Vallés-Abarca (unpublished).
[9]J. S. Logan, J. H. Keller, and R. G. Simmons, J. Vac. Sci. Technol. 14, 92 (1977); J. H. Keller and R. G. Simmons, IBM J. Res. Dev. 23, 24 (1979).
[10]N. Hosokawa, Jpn. J. Appl. Phys. 18, 889 (1979).
[11]Y. Chouan and D. Collobert, J. Phys. (Paris) 43, 279 (1982).
[12]D. G. Teer, J. Adhes. 8, 289 (1977).
[13]H. Mase and T. Tanabe, in Proceedings of the International Ion Engineering Congress ISIAT'83 and IPAT'83, Kyoto, Japan (unpublished).
[14]M. J. Druyvesteyn and F. M. Penning, Rev. Mod. Phys. 12, 87 (1940).
[15]A. V. Bondarenko, Zh. Tekh. Phys. 46, 2535 (1976) [Sov. Phys. Tech. Phys. 21, 1497 (1976)].
[16]R. S. Robinson, J. Vac. Sci. Technol. 16, 185 (1979).
[17]R. T. C. Tsui, Phys. Rev. 168, 107 (1968).
[18]J. M. H. Peters, Eur. J. Phys. 3, 19 (1982).
[19]R. N. Varney, Am. J. Phys. 39, 534 (1971).
[20]J. C. Ashley and R. H. Ritchie, Phys. Status Solidi 38, 425 (1970).