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Evaluation of Electron Swarm Parameters in SF_6 Using Monte Carlo Method

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تقدير عوامل النقل فى SF_6 باستعمال محاكاة مونت كارلو

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ازداد الاهتمام بالتفريغ كثيرا في الأعوام الأخيرة وذلك لأهميته في تقنيات الإلكترونيات الصغيرة الدقيقة، وخاصة الاهتمام بخصوصية التفريغ في غاز سالب التكهرب الذي يكثر استعماله في التطبيقات التكنولوجية. في هذا البحث نقدم بعض الخصوصيات المفصلة لعوامل النقل مثل متوسط الطاقة، سرعة الإنسياق ومعامل التأيين الموجودة عن طريق محاكاة مونت كارلو في غاز سالب التكهرب.

Keywords: *Monte Carlo simulation, Plasmas, SF6, Swarm parameters.*

ABSTRACT

The motion of electrons in sulfur hexafluoride (SF_6) in uniform electric fields is simulated using a Monte Carlo method. The swarm parameters are evaluated and compared with experimental results of drift velocity, electron mean energy, ratio of ionization coefficient and attachment coefficient. The electron molecule collision cross sections adopted in the simulation result in a good agreement with the experimental values over the range of E/N investigated (E is the electric field and N is the gas number density of background gas molecules).

Introduction

Renewal of interest in the study of glow discharge has recently become quite active, largely due to the many fields of application. Glow discharges serve extensively as plasma processing devices in microelectronics, e. g., for ion etching, thin film deposition, and plasma treating of surfaces, and they also find application as atomization, excitation (laser excitation), ionization sources in analytical chemistry [1-6]. Remote plasmas are also used to generate oxygen atoms for in situ growth of high temperature super conducting thin films and for substrate cleaning prior to deposition [7,8]. To attain better results in these application fields; a quantitative understanding of the glow discharge is required. Especially interesting are the properties of discharges in electronegative gases, which are most frequently used for technological applications. In particular, studies of SF_6 have been motivated by the importance of this gas for plasma etching of metals and silicon, for negative ion sources, and in development of gaseous dielectrics. Fast and selective etching of different materials has been demonstrated in pure SF_6 and its mixtures with inert gases and with O_2 [9-13].

Due to the importance of glow discharge process, there has been an increased effort to understand and model them. Electron swarm parameters are important in the design and modeling of gas lasers, plasma switches, and gas-filled detectors. Typically, electron swarm parameters are used in fluid simulations of plasmas to determine the time-dependent evolution of densities of radicals and charged particles; in particular, they are needed to solve the continuity equation for electrons, which includes drift, diffusion, and electron multiplication processes [14]. Proper understanding of the electron dynamics in the plasma is essential since it is electron collisional processes that produce the radicals that act as etching or deposition precursors [15]. A detailed knowledge of electron swarm parameters in gases is necessary for accurate simulations of plasma processes. Theoretical work to predict the transport and ionizing properties of electrons using the measured collision cross sections is based on a numerical analysis of the Boltzmann equation in which the various mechanisms, by which the electrons lose energy, are included. The solution of Boltzmann equation is usually found by utilizing the Lorenz approximation in which the first two terms of the spherical harmonic expansion are considered. Though the effects of including higher order terms have been investigated in other gases such an analysis is not available for SF_6. The numerical solution of the Boltzmann equation yields the electron energy distribution with the electric field E and gas number density N as parameters. Appropriate integration of the energy distribution function yields the transport and ionizing properties of the electron swarm. Monte Carlo simulation of electron drift in a uniform electric field has the advantage that the motion of the electron at all stages during its passage in the discharge is traced.

In this paper, we have studied the behavior of electrons in uniform electric fields by a Monte Carlo method. Swarm parameters are determined as a function of E/N for different rates of increase of the electric field. The calculation has been performed in sulfur hexafluoride. The motivation for the development of the computer simulation described in this paper has its basis in the need to quantitatively understand microelectronic plasma processing.

Simulation Method

The electron transport in a gas under the influence of an electric field E can be simulated with the help of a Monte Carlo method [15-20]. Every electron, during its transit in the gas, performs a succession of free flights punctuated by elastic or inelastic collisions with molecules of gas defined by collision cross sections. During the successive collisions for every electron, certain information

(velocity, position, etc.) is stored in order to calculate, from appropriate sampling methods, transport coefficients and macroscopic coefficients.

In a spherical coordinate system, a background gas of SF_6 molecules with a number density of $N=3.29.10^{22}$ m^{-3}, which corresponds to a gas pressure of 1 Torr at 20°C is considered. To avoid large negative powers of 10, a unit of $1Td = 10^{-21}$ V. m^2 is used. The applied electric field E is antiparallel to the z-axis. n_o electrons with a constant energy ε_0 are injected from the origin of the coordinate system assuming a cosine distribution for the angle of entry with respect to the z-axis. At t = 0 an electron observes a free flight time with a randomly selected angle of entry depending on the distribution. Interactions with the electrodes are not considered. The mean collision time T_m of an electron is inversely dependent upon the total collision cross section Q_T, the gas number density N and the electron velocity $|V|$ and accordingly

$$T_m = \left(N.Q_T.|V|\right)^{-1} \tag{1}$$

The probability of collision in the time step ΔT is

$$P = 1 - \exp\left(-\Delta T/T_m\right) \tag{2}$$

The new position and energy of the electron are calculated according to the equation of motion. The collision is simulated by comparing P with R_1 at the end of each step, where R_1 is a random number uniformly distributed between 0 and 1. When the electron undergoes a collision

$$P = \left[1 - \exp\left(-\Delta T/T_m\right)\right] \geq R_1 \tag{3}$$

The nature of the collision is determined in the following way: $P_{2,j}$ is the probability that collision process j takes place, j = 1, 2, 3,... n, including momentum transfer, attachment, vibration, excitation, and ionization collisions. This leads to the system

$$P_{2j} = Q_j / Q_T$$

$$\sum P_{2,j} = 1$$

$$P_{2,1} \leq P_{2,2} \leq P_{2,j} \leq P_{2,N}$$

$$P_{2,1} + P_{2,2} + P_{2,j-1} \leq R_2 \leq P_{2,1} + P_{2,2} + ... P_{2,j} \tag{4}$$

which determines the *j-th* collision process.

The total collision cross section is defined as

$$Q_T = Q_{el} + Q_{At} + Q_v + Q_{ex} + Q_{ion} \tag{5}$$

where Q_{el} is the elastic differential cross section, which is replaced by the momentum transfer collision cross section in the simulation; Q_{At} is the attachment cross section, Q_{ex} the total electronic excitation cross section, Q_v the vibrational cross section, and Q_{ion} is the total ionization cross section.

The sum of the fractional probabilities is equal to unity, and the interval [0,1] is divided into segments with lengths corresponding to these fractional probabilities. A new random number R_2 between 0 and 1 is generated, and the interval into which this random number falls, determines the type of collision that occurs. The new energy and direction after the collision depends upon the type of collision: for excitation, the new energy ε is given by: $\varepsilon = \varepsilon_0 - \varepsilon_{exc}$, where ε_{exc} is the excitation threshold energy, and ε_0 is the electron energy before collision. For ionization, the total energy before collision is divided between the primary (original) electron and the secondary electron created in the ionization collision. For elastic collisions, the new kinetic energy of the electron is calculated by

$$\varepsilon = \varepsilon_0\left[1 - 2(m/M)(1 - \cos\chi)\right], \tag{6}$$

which is deduced from the hard sphere model. χ is the scattering angle of the electron after collision, where m and M are the masses of electron and molecule of SF_6, respectively. After a collision the angles are determined by isotropic distribution. Hence, after the event of a collision if the probabilities of inelastic collisions fail the collision is deemed to be elastic. If the electron is attached it is lost in the swarm and its subsequent fate is ignored. All the electrons in the swarm moving forward and backwards, including the electrons formed during the ionization process, are traced until the termination time or loss due to attachment.

Results and Discussion

In the Monte Carlo technique, the electron trajectories are calculated and collisions of electrons with molecules in the gas are simulated. The swarm parameters are obtained after following seed electrons from initial conditions for a long period of distance or time. One of the difficulties in using the Monte Carlo technique in a highly electron attaching gas such as SF_6 is that the initial electrons released at the cathode get lost due to the high electron attachment cross section of SF_6. The cross sections set of SF_6 employed is that of Hayes *et al* [21], Peach [22,23], and Itoh *et al* [24], and is shown in Fig. 1. The computing time for the Monte Carlo technique depends upon the number of test electrons released from the cathode and the number of collisions occurring while each electron travels the distance from the cathode to the anode. In order to evaluate the swarm parameters for better simulation, at least 10000 test electrons are required, for zero field and low E/N. At low E/N values in gases such SF_6 having high attachment cross sections at low energy, most seed electrons, after a few free flights, can be attached. In this case, the Monte Carlo simulation is not able to calculate electron swarm parameters with enough precision, if the seeded electrons are less than 10000. The initial electrons are injected as a point source at t=0 and r=0 with a cosine distribution with a mean energy of 2 eV.

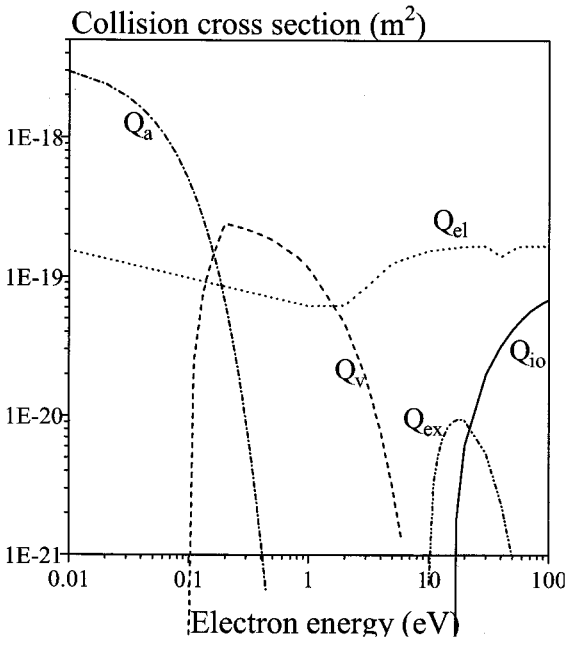

Fig. 1. Summary of collisional cross sections of SF_6, Q_a: attachment, Q_v: vibrations, Q_{ex}: excitations, Q_{io}: ionization, Q_{el}: elastic momentum transfer.

Under zero field conditions, Fig. 2.1 and Fig. 2.2 show electron mean energy, drift velocity, and also longitudinal and transverse diffusion coefficients, and the reduction electron number density. Probably one of the most convincing validity tests of the treatment of low energy electron molecule collisions with the Monte Carlo method is to determine distribution functions and transport coefficients under zero field conditions. Indeed, for an electron swarm or beam released (with known initial energetic and angular distributions) through a gas under zero electric field conditions, it is well established that this electron swarm relaxes after a greater or lesser period of time (depending on initial conditions and background gas) towards an equilibrium distribution, whatever the initial distribution or the nature of the background gas. Such an equilibrium is obviously characterized by a classical behavior: The electron distribution function becomes Maxwellian at the background gas temperature, there is no more electron drift, and diffusion becomes completely isotropic (i.e. longitudinal and transverse diffusion coefficients are identical). Fig. 2.1 shows drift velocity and electron mean energy, versus time, which relaxes towards gas energy. Electrons emitted along the forward direction, after a relatively few collisions, lose their initial anisotropic angular distribution so that the initial directed velocity becomes rapidly negligible. Fig. 2.2 shows the longitudinal (ND_L) and transverse (ND_T) diffusion coefficients versus time. In this short time scale, the longitudinal diffusion coefficient, after an overshoot effect due to the anisotropy of the initial distribution, tends towards transverse diffusion.

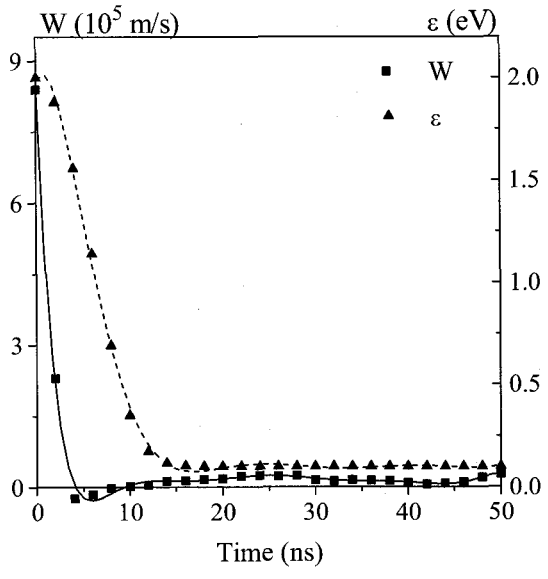

Fig. 2.1. Zero field electron mean energy (ε) (ND_L), transverse and drift velocity (W) as a function of time.

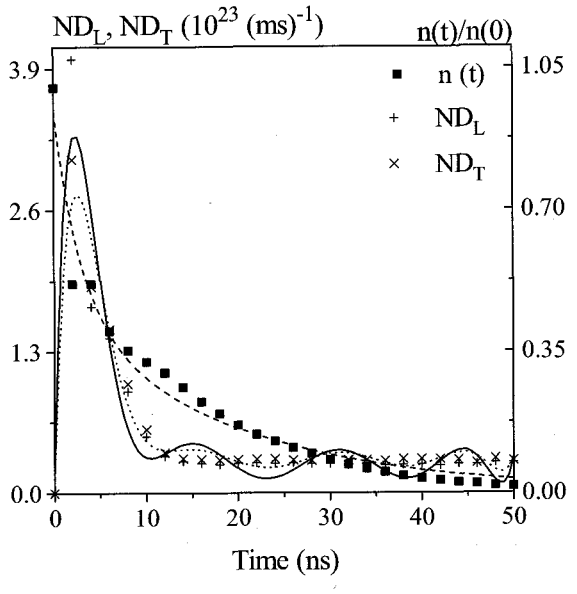

Fig. 2.2. Zero field longitudinal (ND_T) diffusion coefficients, and reduction electron number density (n(t)/n(0)) as a function of time

For zero field and a lower E/N value, the decrease of the number density n(t) is more pronounced due to the higher attachment efficiency in SF_6 (Fig. 3.1 and Fig. 3.2). One notices that for E/N = 10 Td the transport parameters are slightly higher than those for a zero field. The fluctuation of the transport coefficients in the first is attributed to the non-equilibrium of the electron energy distribution. The fluctuation in the latter is attributed to the statistical scatter since the number of electrons decreases rapidly due to attachment. To reduce the scatter it was thought that a considerable number of electrons needed to be injected into the drift space in order to reduce the

fluctuation. The Monte Carlo method is the preferred method at high values of E/N because it directly simulates the experimental method and also provides a check on whether the electrons have attained equilibrium. The computing time for the Monte Carlo technique depends upon the number of test electrons released from the cathode and the number of collisions occurring while each electron travels the distance from the cathode to the anode.

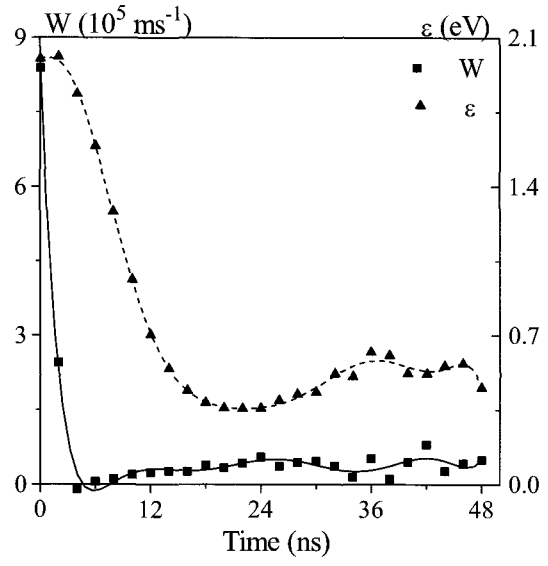

Fig. 3.1. Electron mean energy (ε) and drift velocity (W) as functions of time, E/N=10Td

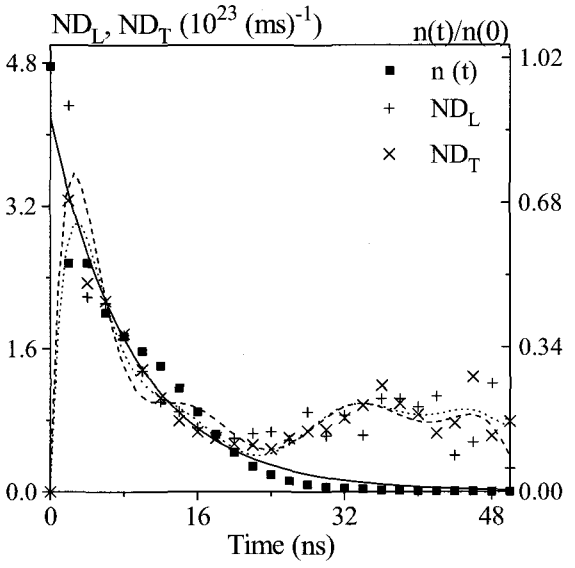

Fig. 3.2. Longitudinal (ND_L), transverse (ND_T) diffusion coefficients, and reduction electron number density (n(t)) as functions of time, E/N=10Td

Fig. 4 shows the variation of the ionization coefficient (α) and the average electron energy (ε) with time. The initial electrons are injected with a mean energy of 0.1 eV. This energy is low enough to not influence the behavior of the swarm at later times. t0 is the time required for the average electron energy to reach its steady state value. There is a time lag between the onset of steady state for the average energy and the onset of steady state for the ionization coefficient. The transient in the ionization coefficient (α) occurs because initially, t ~ t0, the number of accumulated ionizing collisions is small, hence the ionization coefficient has not reached steady state. Fig. 4 also shows that the mean electron energy fluctuates, with diminishing amplitude of fluctuation, because the number of electrons in the avalanche is small enough to reduce computational costs. The drift velocity, has the same qualitative time behavior as α, but reaches steady state in a shorter time.

Figs. 5.1 and 5.2 show the variation of the drift velocity and the mean electron energy, versus E/N. There is good agreement among the data for the electron drift velocity [25,26], between various experiments, and the values calculated from the Monte Carlo method. A fairly good agreement is found for, between our values for the electron mean energy and those of the various experiments [27-31]. The calculated drift velocities with Monte Carlo method compared with the calculated values, on the basis of the Boltzmann equation in the E/N range 30 to 550 Td [32]. The present results are about 20% higher than those referred in above at high reduced electric field. The electronic component constitutes only a small fraction of the total current collected at the anode, the attachment processes being dominant; most of the electrons emitted at the cathode are readily attached. Indeed, the dominance of an equilibrium region in the gap is not evident and may not

warrant the hypotheses inherent in the approximation of the hydrodynamic regime in the Boltzmann analysis.

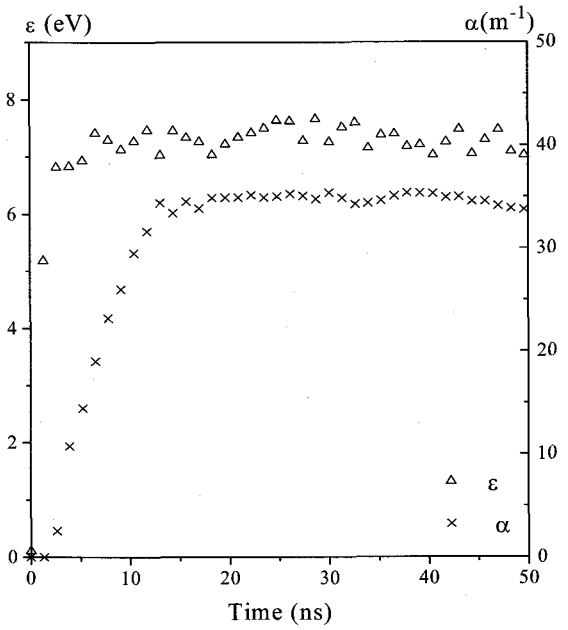

Fig.4. Mean electron energy (ε) and ionization coefficient (α) as a function of time, E/N=300 Td.

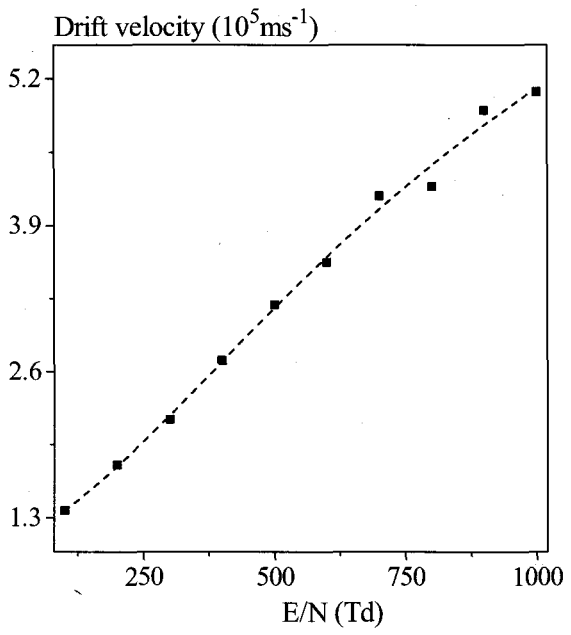

Fig. 5.1. Electron drift velocity as a function of E/N, fitted curve is the dashed line.

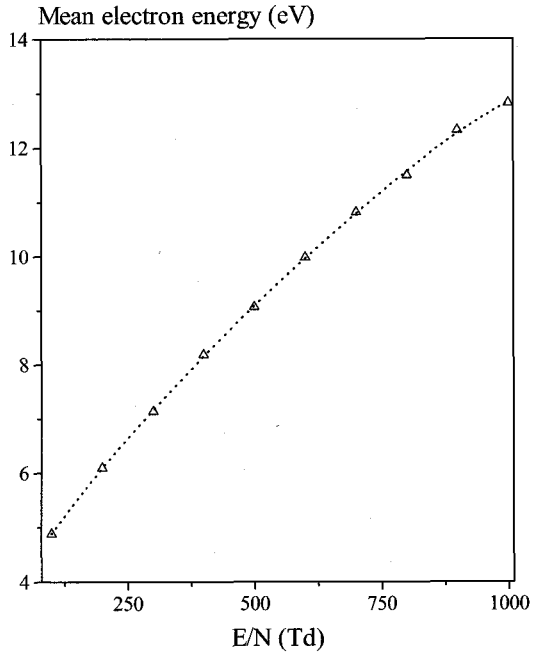

Fig. 5.2. Mean electron energy as a function of E/N, fitted curve is the dashed line.

The experimentally electron drift velocity determined using the voltage transient method [33] within the range 360 Td < E/N < 720 Td agrees quite well with the present results at the middle of the referred range, but rather well in the low and high reduced electric field of this interval. The present data agree well with the values of the electron drift velocity measured with the pulsed

Townsend technique in [34] over the range of the reduced electric field strength E/N, from 50 to 360 Td. The electron drift velocity plotted as a function of E/N, in the range 200 to 1000 Td, there is good agreement [35] among the data for the electron drift velocity from experiments and the our values calculated from the Monte Carlo method.

The very good agreement between experimental data and the simulation results shown in Figs. 5.1 and 5.2 indicates that the collision calculations accurately predict the growth of electron pulses into electron avalanches.

In view of practical importance to engineers, the swarm parameters α/N (reduced ionization coefficient) and η/N (electron attachment coefficient divided by gas number density) generated by the simulation technique in this gas are shown in Figs. 6.1 and 6.2. The attachment coefficient is a measure of the probability that an electron will attach to a gas molecule in traveling a unit distance in the electric field direction and is, ideally, only a function of E/N for a given gas. The reduced ionization coefficient α/N is related to E/N according to the semi-empirical equation:

$$\alpha/N = A.\exp\left(-B/(E/N)\right) \qquad (7)$$

where A and B are constants characteristic of the gas.

There have been a number of measurements of the ionization coefficient; the measured and calculated data from the Monte Carlo method are in reasonable agreement [25, 27, 29-31, 36]. In general, the calculated data of the attachment coefficients with the Monte Carlo Technique agree well with the experimental results [25, 27, 29-31, 36]. Here, we confirm that the Monte Carlo method is valid for deduction of the swarm parameters at low, intermediate and high-reduced electric field E/N values despite the fact that SF_6 is a strongly electronegative gas.

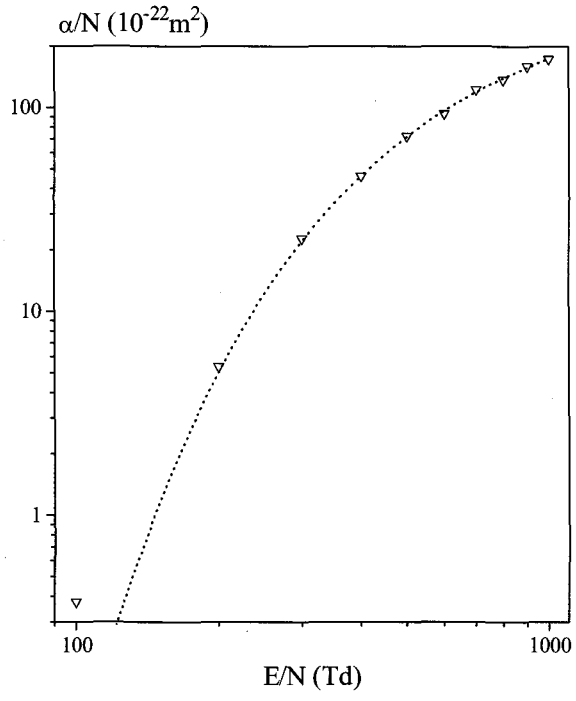

Fig. 6.1. Reduced ionization coefficient α/N as a function of E/N, curve from equation 3 is the dashed line.

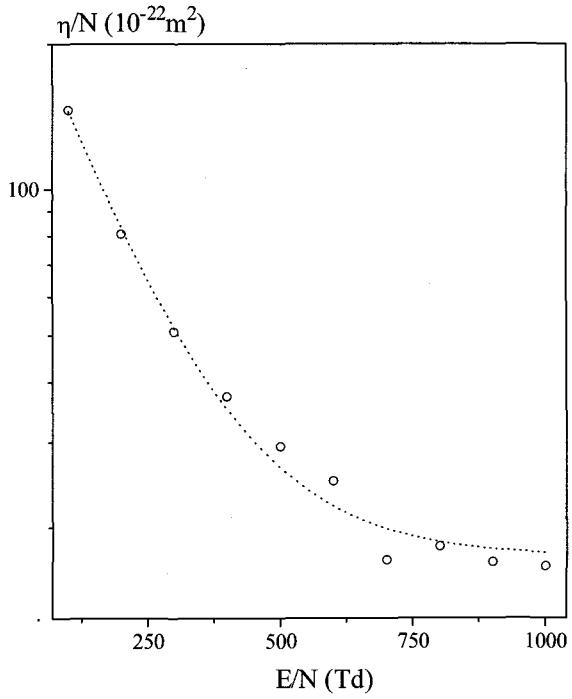

Fig. 6.2. Reduced electron attachment coefficient η/N as a function of E/N, fitted curve is the dashed line.

For a set of n particles and a given simulation time, the electron energy distribution function can be determined. To obtain good statistics, a large number of particles are required. The electron energy distribution at E/N=200 Td and 400 Td conditions are shown in Fig. 7.1 and Fig. 7.2. The anisotropic parts of the distribution are quite small in the case of E/N=200 Td as shown in Fig. 7.1 by contrast, the distribution at E/N=400 Td with the anisotropic parts is much greater than that with isotropic part alone. This implies that the low E/N fluctuation results from the acceleration of each electron during the interval between collisions in the low energy region where the direction of the electrons is easily changeable by the electric field. The dot and full lines show the Maxwellian distribution at the same energy. There is a good agreement between the present calculated distribution function and those results obtained by Yoshizawa *et al* [37].

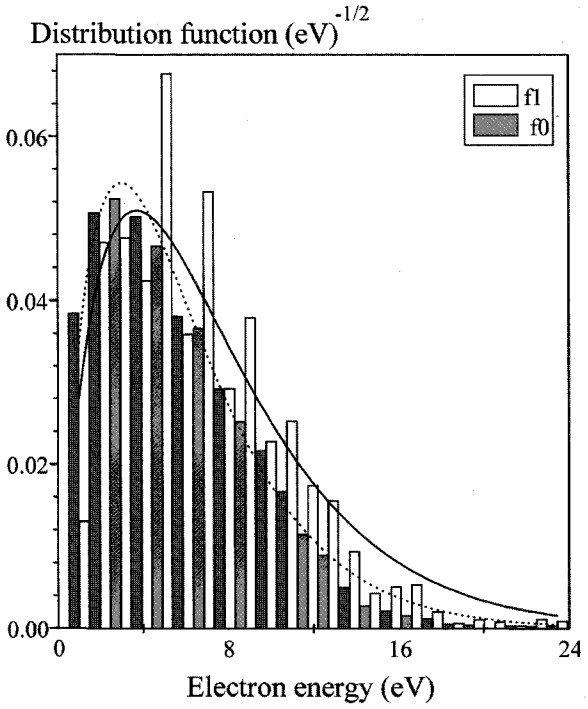

Fig. 7.1. Electron energy distribution at E/N=200Td,
f0: isotropic part, f1: with anisotropic parts.

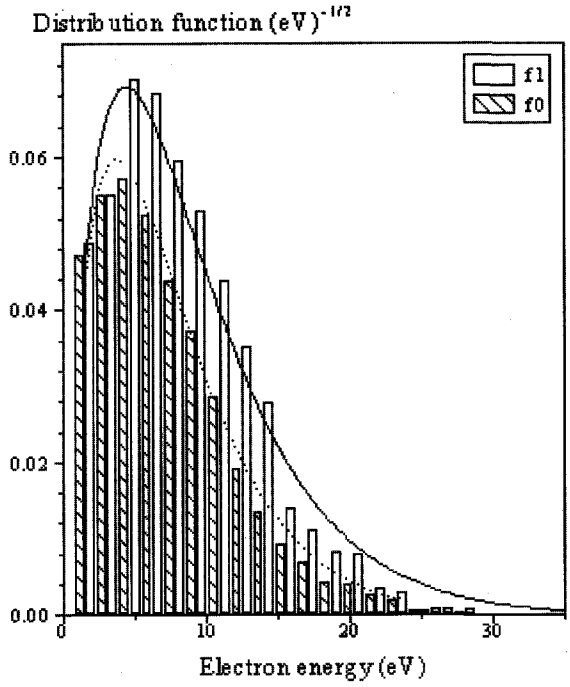

Fig. 7.2. Electron energy distribution at E/N=400Td,
f0: isotropic part, f1: with anisotropic parts.

Monte Carlo methods as applied to gas discharge problems involve evaluating the percentage of a given species of particles emanating from a given source, after experiencing energy loss and gain, terminate in defined categories. Its technique is realistic because a large number of particles are followed from the source through their life history. The fundamental physical concept is the mean free path or the mean free flight time, with the collision equations being formulated to the conditions such as the number density, and electric field intensity. From the history of each species the average properties are obtained by an efficient tracing method and compared with the respective measured quantity. In problems connected with collision physics (corona, breakdown, neutron scattering, photon diffusion etc.) it is necessary to have a source of random numbers distributed normally in the interval [0, 1].

Conclusion

In this study, we have examined the behavior of electrons in uniform electric fields using a Monte Carlo simulation. Electron swarm parameters were calculated as a function of reduced electric fields E/N. Binary electron neutral gas molecule collisions are the essential mechanism in electron avalanche growth. The simulation results give values for electron drift velocity, electron mean energy, ionization and attachment coefficients, longitudinal and transverse diffusion coefficient, and electron energy distribution as functions of time and reduced electric fields. In this work, we confirm that the Monte Carlo method is valid for deduction of the swarm parameters at low, intermediate and high reduced electric field E/N values despite the fact that SF_6 is a strongly electronegative gas. The reduced attachment coefficient becomes small for the high values of the reduced electric field due to the large ionization cross section of SF_6. We consider that inelastic collision reduce electron energy and improve dielectric strength. The good agreement between calculated and measured swarm parameters demonstrates the validity of the binary collision simulation techniques, and the large number of electrons to be studied for obtaining stable values of the coefficients high resolution at low values of E/N. Energy distributions obtained by the simulation indicate Maxwellian tail behavior at corresponding mean energies.

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