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Some Recent Studies of Electron Swarms in Gases*

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Abstract

Some recent studies of electron swarms in gases under the action of an electric field are introduced. The studies include a new type of continuity equation for electrons having a form in which the partial derivative of the electron density with respect to position and to time are interchanged, a method to deduce the time-of-flight and arrival-time-spectrum swarm parameters based on a Fourier-transformed Boltzmann equation, an examination of the correspondence between experimental and theoretical electron drift velocities, and an automatic technique to deduce the electron–gas molecule collision cross section from electron drift velocity data. We also briefly introduce a method for the deduction of electron collision cross sections with gas molecules having vibrational excitation cross sections greater than the elastic momentum transfer cross section by using a gas mixture technique, an integral type of method for solution of the Boltzmann equation with salient numerical stability, a quantitative analysis of the effect of Penning ionisation, and the behaviour of electron swarms under radio frequency electric fields.

1. Introduction

The significance for physics of studying electron swarms is threefold. The first is that the study deepens our understanding of the mechanism by which a group of charged particles drifts in an array of randomly dispersed point scatterers under an electric field. This also stimulates the development of techniques by which the properties of electron swarms are analysed. The second is that the parameters characterising the properties of swarms, often referred to as electron swarm parameters, are determined by the study. These parameters are important not only for characterising the properties of electron swarms in a variety of gases, but also for analysing, predicting and simulating the properties of gas discharges and gas discharge plasmas. We live in an era in which gas discharges and discharge plasmas play an important role in dealing with environmental problems, in manufacturing electronic devices such as VLSIs and solar cells, and in the design of electrical insulation which supports the present day electricity transportation systems. The third is that the study enables us to determine the electron–gas molecule collision cross sections. The technique by which the cross section is worked out by electron swarm studies is independent of other methods

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of determining the cross sections and has its own characteristic advantages in certain circumstances.

In what follows, some recent developments in the study of electron swarms and their applications are described. One thing that must be borne in mind is that the topics treated here are by no means comprehensive, and other authors would treat different features of the properties of electron swarms.

2. Arrival Time Spectrum Representation—A New Form of Continuity Equation

The equation of continuity of electrons in gases is important for representing the behaviour of the species which plays the main role in gas discharges and gas discharge plasmas. Until recently, a type of continuity equation represented by equation (1) was regarded as unique:

$$\frac{\partial n}{\partial t} = \nu_{\mathrm{i}} n - W_{\mathrm{r}} \frac{\partial n}{\partial z} + D_{\mathrm{L}} \frac{\partial^2 n}{\partial z^2} + D_{\mathrm{T}} \left(\frac{\partial^2 n}{\partial x^2} + \frac{\partial^2 n}{\partial y^2} \right) - D_3 \frac{\partial^3 n}{\partial z^3} + \ldots . \tag{1}$$

Here $n \equiv n(x, y, z, t)$ is the electron density, ν_{i} is the ionisation frequency, W_{r} is the centre-of-mass, or centroid, drift velocity, D_{L} and D_{T} are respectively the longitudinal and transverse diffusion coefficients and D_3 is a higher order coefficient. The electric field is assumed to be uniform and in the inverse z direction. Equation (1) has been used practically in all treatments in which continuity of electrons in electron swarms and in weakly ionised gases is considered quantitatively (e.g. Huxley and Crompton 1974). It is to be noted that, when integrated with respect to x, y and z, equation (1) reduces to the form

$$\frac{\mathrm{d}N}{\mathrm{d}t} = \nu_{\mathrm{i}} N , \tag{2}$$

where

$$N \equiv \int \int \int n(x, y, z, t) \, \mathrm{d}x \, \mathrm{d}y \, \mathrm{d}z$$

is the number of electrons in the electron swarm at time t.

In a previous paper (Tagashira *et al.* 1977) we pointed out that the drift velocity W_{r}, defined as the centre-of-mass velocity, and the drift velocity W_{m}, defined as the ratio of the distance between two positions in the field direction to the difference between the mean arrival times of the electrons at the positions, generally assume different values if electron impact ionisation of gas molecules and/or electron attachment to the molecules exist.

It is interesting to note that the definition of these two drift velocities are symmetric with respect to time and position:

$$W_{\mathrm{r}} = \frac{\mathrm{d} \langle z \rangle}{\mathrm{d}t} , \tag{3}$$

$$W_{\mathrm{m}} = \frac{\mathrm{d}z}{\mathrm{d}\bar{t}} . \tag{4}$$

Here, $\langle z \rangle$ is the average position of electrons in a swarm at a time in the field direction and $\bar{t}$ is the mean arrival time of electrons at a position in the field direction.

Another example of such a symmetry is the well-known formula for electron current growth in the steady-state Townsend experiment:

$$\frac{\mathrm{d}n(z)}{\mathrm{d}z} = \alpha_{\mathrm{T}}\, n(z)\,, \tag{5}$$

where

$$n(z) \equiv \int\int\int n(x,y,z,t)\,\mathrm{d}x\,\mathrm{d}y\,\mathrm{d}t$$

is the number density of electrons at z in the field direction.

The symmetry between equations (2) and (5) is obvious; t and ν_1 in equaton (2) are respectively replaced by z and α_{r} in equation (5). The symmetries between equations (3) and (4) and also between (2) and (5) strongly suggest that there might be another method of representing the development of electron swarms, in which the position z and the time t are interchanged.

Recently, Kondo and Tagashira (1990) worked out a representation along this line. The analysis starts with a Boltzmann equation, however, the continuity equation will be introduced here instead since it is of greater interest to investigators working on electron swarms in gases, gas discharges and weakly ionised plasmas. The new continuity equation is written as

$$\frac{\partial n}{\partial z} = \alpha_0\, n - \alpha_1 \frac{\partial n}{\partial t} + \alpha_2 \frac{\partial^2 n}{\partial t^2} - \alpha_3 \frac{\partial^3 n}{\partial t^3} + \ldots\,, \tag{6}$$

where

$$n \equiv n(z,t) = \int\int n(x,y,z,t)\,\mathrm{d}x\,\mathrm{d}y$$

is the number density of electrons in the field direction and the α are the electron swarm parameters as follows: $\alpha_0 \equiv \alpha_{\mathrm{T}}$ is the Townsend first ionisation coefficient, $\alpha_1 \equiv 1/W_{\mathrm{m}}$ is the reciprocal of the mean arrival time drift velocity as given by equation (4), α_2 is a parameter representing longitudinal diffusion and therefore closely related to the longitudinal diffusion coefficient, and α_3 is a higher order coefficient.

In the deduction of equation (6), it is assumed that $\partial n/\partial t$ is the small expansion parameter, in contrast to previous works where ∇n was the relevant expansion parameter. Interchanging the roles of z and t is equivalent to exchanging the roles of ω and k in the dispersion relation given subsequently.

It is interesting to note that equation (5) is deduced if both sides of equation (6) are integrated with respect to time t. This is similar to the way in which equation (2) is deduced from (1).

A few comments can be made on the new continuity equation (6). It has been shown that the 'α' parameters in (6) are related to the parameters which may

be referred to as 'ω' parameters (Tagashira *et al.* 1977; Kondo and Tagashira 1990) in (1) as follows:

$$\omega_0 - \alpha_0\,\omega_1 + (\alpha_0)^2\omega_2 - (\alpha_0)^3\omega_3 + \ldots = 0\,, \tag{7a}$$

$$\alpha_0 - \omega_0\,\alpha_1 + (\omega_0)^2\alpha_2 - (\omega_0)^3\alpha_3 + \ldots = 0\,, \tag{7b}$$

$$\omega_1^{-1} = \alpha_1 - 2\omega_0\,\alpha_2 + 3(\omega_0)^2\alpha_3 - \ldots\,, \tag{8a}$$

$$\alpha_1^{-1} = \omega_1 - 2\alpha_0\,\omega_2 + 3(\alpha_0)^2\omega_3 - \ldots\,, \qquad \text{etc.}\,, \tag{8b}$$

where $\omega_0 = \nu_\mathrm{i}$, $\omega_1 = W_\mathrm{r}$, $\omega_2 = D_\mathrm{L}$, $\omega_3 = D_3\,,\ldots$, and the α have already been given above.

A salient feature of the new treatment is that W_m is calculated directly from the Boltzmann equation, while the conventional treatment must calculate in principle an infinite number of parameters as equation (8b) shows. The new treatment is essentially based on the arrival time distribution of electrons and its moments at positions in the field direction (Kondo and Tagashira 1990), and therefore this type of treatment may be called the arrival time spectrum (ATS) analysis. In contrast, the conventional analysis based on equation (1) relies on the flight distance distribution of electrons and its moments, and may be called the flight distance spectrum (FDS) analysis. Kondo and Tagashira (1990) also treated eigenvalue problems associated with the ATS and FDS analyses.

In closing this introduction to the ATS analysis, it should be mentioned that only the continuity equation (6) in the field direction has been deduced so far. The continuity equation in the perpendicular direction to the field has not been obtained.

3. Swarm Parameters deduced from a Fourier-transformed Form of the Boltzmann Equation

It has been shown that the Boltzmann equation for electron swarms under the action of a uniform electric field in gases has a solution in terms of a Fourier integral (Parker and Lowke 1969; Tagashira *et al.* 1977; Kumar *et al.* 1980):

$$f(z,\boldsymbol{v},t) = \frac{1}{2\pi}\int \mathrm{d}k\; f(k,\boldsymbol{v})\exp\{\mathrm{i}\,kz - \omega(k)t\}\,, \tag{9}$$

where

$$f(z,\boldsymbol{v},t) = \int\int f(x,y,z,\boldsymbol{v},t)\;\mathrm{d}x\,\mathrm{d}y$$

is the electron velocity distribution, k is the Fourier parameter, $\boldsymbol{v}$ is the electron velocity and

$$\omega(k) = -\sum \omega_r(-\mathrm{i}\,k)^r\,, \tag{10a}$$

$$f(k,\boldsymbol{v}) = \sum f_r(\boldsymbol{v})(-\mathrm{i}\,k)^r\,. \tag{10b}$$

The physical interpretation of ω_r $(r = 0,1,2,\ldots)$ was given in the previous section, while $f(k,\boldsymbol{v})$ and $\omega(k)$ are respectively the electron velocity distribution of the k component and its eigenvalue.

Putting equation (9) into the usual Boltzmann equation for electron swarms, the following equation for $f(k, \boldsymbol{v})$ and $\omega(k)$ is obtained (Date *et al.* 1992):

$$\left(-\omega(k) + \mathrm{i} v_z k + \frac{eE}{m} \frac{\partial}{\partial v_z} + J \right) f(k, \boldsymbol{v}) = 0 . \tag{11}$$

Here J is the collision operator for electron–gas molecule scattering.

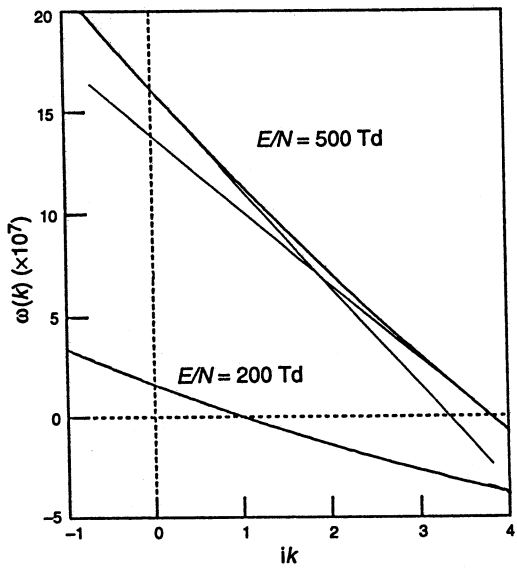

Fig. 1. Plot of $\omega(k)$ versus ik for krypton.

By solving equation (11), $\omega(k)$ is obtained, and by a curve-fitting with ik as the parameter, the ω_r $(r = 0, 1, 2, ...)$ are obtained. A plot of $\omega(k)$ for an actual krypton gas is shown in Fig. 1. As equation (10a) shows, the intersection of $\omega(k)$ with the vertical axis gives $\omega_0 = \nu_i$, the ionisation frequency, and the slope at the intersection equals $\omega_2 = W_r$, the centre-of-mass drift velocity. In contrast, it can be shown (Kondo and Tagashira 1990) that the intersection of $\omega(k)$ with the horizontal axis gives $\alpha_0 \equiv \alpha_T$, the ionisation coefficient, and the slope there represents $\alpha_1^{-1} \equiv W_m$, the mean arrival time drift velocity. Robson (1990, 1991) has shown the $\omega(k)$ versus ik relationship for a Fokker–Planck model collision operator and shown that the same conclusion as above is obtained. The plot of $\omega(k)$ as a function of ik in Fig. 1 for krypton clearly shows that $\omega(k)$ is downward convex and does not go through the origin. This fact demonstrates that the centre-of-mass drift velocity W_r is, in general, not equal to the mean arrival time drift velocity W_m when ionisation is present, in accordance with the prediction of a previous paper concerning the effects of ionisation on the electron swarm parameters (Tagashira *et al.* 1977). At the lower E/N value (=200 Td) shown in Fig. 1, the ionisation is less prominent and the $\omega(k)$ versus ik curve runs closer to the origin, giving a smaller ionisation frequency and ionisation coefficient. If ionisation is negligible, the curve should go through the origin, giving $\nu_i = \alpha_T = 0$ and $W_r = W_m$, the latter being in agreement with the result

predicted by Tagashira *et al.* (1977) and confirmed by Kondo and Tagashira (1990). The works by Robson (1990, 1991) are also in qualitative agreement with these results. This seems to support the prediction of Tagashira *et al.* (1977) on the effect of the presence of ionisation and/or attachment on the electron swarm parameters, at least qualitatively.

A numerical subtlety for deduction of the time-of-flight and arrival-time-spectrum swarm parameters from equation (11), as touched earlier, is explained here. Although the ω parameters $\omega_0, \omega_1, \omega_2, \ldots$ and the α parameters $\alpha_0, \alpha_1, \alpha_2, \ldots$ can in principle be deduced from the intersections and the derivatives at the horizontal and vertical axes of the $\omega(k)$ versus ik curve, as mentioned earlier, it would become increasingly difficult to deduce the parameters as the degree of the differentiation increases. A curve-fitting technique instead may be used for the purpose: the value of $\omega(k)$ is calculated from equation (11) for various given values of ik and then the $\omega(k)$ versus ik curve may be drawn for an actual gas as shown in Fig. 1, from which the ω and α parameters are deduced by a curve-fitting technique. A salient feature of the technique is that only equation (11) needs to be solved and the higher order equations, which are more difficult to handle because of their numerical instability, are now unnecessary for deduction of the higher order parameters ω_n and α_n, for $n \geq 1$. This method for deducing the parameters ω and α has been demonstrated to work for actual gases in Date *et al.* (1992).

The eigenvalue problem (11) and dispersion relation and the swarm parameter deduction and definition have been studied by many other authors; notably by Standish (1987, 1989), Phelps and Pitchford (1985), Yousfi *et al.* (1985) and Blevin and Fletcher (1984).

4. Correspondence between Experimental and Theoretical Electron Drift Velocities

Theoretical electron drift velocities may be defined either by the velocity W_r of the centre-of-mass motion, the reciprocal W_m of the derivative of the mean arrival time with respect to the observational position in the field direction, the simple mean W_v of the velocities of electrons in an isolated swarm, or the simple mean V_d of the velocities of electrons in a steady-state Townsend experiment (Tagashira *et al.* 1977). We note that in actual experiments, the electron drift velocity is sometimes measured in rather intuitive arrangements and analytical representation of the measurement is difficult. In this section, simple and well-known arrangements for the measurement of electron drift velocities are checked by a Monte Carlo simulation study.

The experimental arrangements studied here are those by Schlumbohm (1965) and Frommhold (1959). In Schlumbohm's experiment the current flowing in a parallel plane gap due to the drift of an isolated electron swarm is measured and the transit time of an electron swarm is defined as the interval T_SC between the generation of the electron swarm at the cathode by a brief ultraviolet flash and the time at which the current reaches the peak. The Schlumbohm drift velocity is defined by

$$W_\mathrm{SC} = d/T_\mathrm{SC}\,, \tag{12}$$

where d is the gap length.

Frommhold (1959) defined T_{FD} as the interval between the generation of electrons at the cathode by a brief flash in a parallel plane gap and the eventual intersection of the logarithmic rise of the time-integrated gap current and the saturation value of the time-integrated current. The Frommhold drift velocity is then defined as

$$W_{FD} = d/T_{FD}\,, \tag{13}$$

where d is the electrode separation.

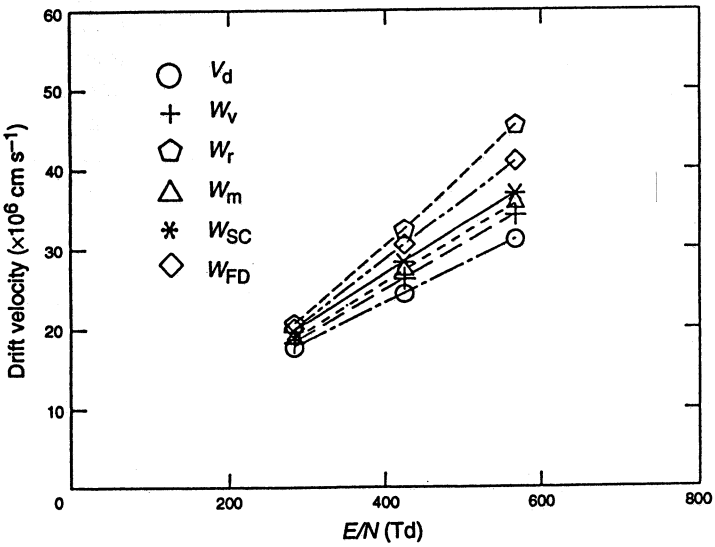

Fig. 2. Electron drift velocity as a function of E/N.

A calculation has been performed for CH_4 (Satoh *et al.* 1991) and the results are shown in Fig. 2. In deducing Schlummbohm's W_{SC} and Frommhold's W_{FD}, a differential form $\Delta d/\Delta T_{SC}$ or $\Delta d/\Delta T_{FD}$ is used, rather than the original definition given in (12) and (13), to eliminate the gap length dependence of W_{SC} and W_{FD} even at constant values of E/N, E and N being the electric field and the gas number density.

Fig. 2 shows that none of the theoretical drift velocities agree with W_{SC} or W_{FD}, and that W_{SC} and W_{FD} are essentially different drift velocities. Though not shown in the figure, a comparison was made between W_{FD} and $W = (W_r + W_m)/2$ which found that W_{FD} closely agrees with W. The nature of W was discussed in Satoh *et al.* (1991) and Tagashira *et al.* (1977); see also Phelps and Pitchford (1985; their Reference 16). Fig. 2 also shows that W_{SC} is close to but slightly larger than W_m, the mean arrival time drift velocity. Similar calculations have been performed for a model gas with constant collision frequency, and for argon and SF_6 to suggest that more or less similar results are obtained.

In the calculation, a few simplifying assumptions are made: the effect of the duration of the initial ultraviolet flash is considered to be negligibly small, and the motion of ions generated by ionisation and/or attachment is neglected. The effect of secondary phenomena at the cathode is also neglected. The model has to be improved to evaluate these effects in the near future.

With all these assumptions, however, the present results strongly suggest that the correspondence between experimental and theoretical drift velocities can be complicated and comparison between them must be done with care.

5. Deduction of the Momentum Transfer Cross Section from Electron Drift Velocity Data

The electron–gas molecule collision cross section may be determined either by quantum mechanical calculations, by single beam experiments or by the swarm technique. The swarm technique relies on the use of high precision measurement data of swarm parameters, such as electron drift velocities, diffusion coefficients and the ionisation coefficient, and on a high precision technique for solving the Boltzmann equation. Here, an example of the swarm method in which the momentum transfer cross section in argon is deduced (Suzuki *et al.* 1990) is introduced. The experimental data on the electron drift velocity in argon as a function of E/N obtained by Robertson (1977) are used.

An outline of the technique is as follows. A general property of the electron energy distribution is that its tail extends towards high energies as the reduced field E/N increases. The extended portion of the distribution may be used as 'searcher' of the cross section at around the electron energies of the extended portion. An algorithm is set up so that this 'search' is performed automatically by consulting the drift velocity data through evaluation of the magnitude of the cross section at the 'searched' energy region by a Boltzmann equation technique. The deduction of the cross section then proceeds from low E/N to high E/N values, or vice versa, and is performed automatically.

A technique of this kind has been developed by Suzuki *et al.* (1990) and applied to the momentum transfer cross section of argon. The result is shown in Fig. 3. We note that the result is obtained in principle without any prior knowledge of the momentum transfer cross section; that is to say, for the initial value of the cross section a constant value was assumed and the algorithm modified this value to deduce the cross section as in Fig. 3. Any shape for the initial cross section is found to give the same result.

The deduced momentum transfer cross section in argon shows good agreement with the quantum mechanical calculation by Dasgupta and Bhatia (1985). Note that the two techniques are totally independent. Agreement with the results of other authors is also good, except for Yau *et al.* (1980). This result also presents a valuable and comprehensive check of previous cross sections from the swarm method point of view, since the electron swarm parameters calculated from the deduced cross section agree almost perfectly with the experimental values.

6. Other Studies

Four topics are briefly mentioned. One is the application of the gas mixture technique for the determination of electron–gas molecule collision cross sections in monosilane (Kurachi and Nakamura 1991). Since monosilane has a greater cross section for vibrational excitation than for momentum transfer, an analysis should be carried out with a multi-term Boltzmann equation technique rather than the so-called two-term expansion technique, if a pure gas is to be analysed. However, if monosilane is diluted with argon, say, the cross section of which is far less controversial, then the analysis becomes much simpler since the momentum

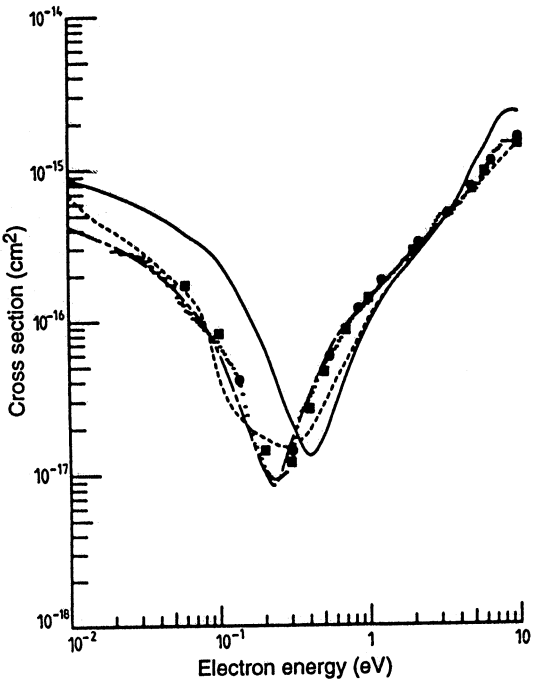

Fig. 3. Momentum transfer cross section of argon at low electron energies: step curve, present results; solid circles, Dasgupta and Bhatia (1985); solid squares, Bell *et al.* (1984); solid curve, Yau *et al.* (1980); dotted curve, Frost and Phelps (1964); and dashed curve, Haddad and O'Malley (1982).

transfer cross section for elastic collisions of the mixture is much greater than the vibrational cross sections of the mixture and the two-term technique may be used to far higher accuracy. Also, the mixing ratio may be changed to make more precise analyses. For the determination of comprehensive sets of collision cross sections of, for example, etching gases, which in most cases have large vibrational excitation cross sections in comparison with elastic momentum transfer, the technique developed by Kurachi and Nakamura (1991) for the gas mixture becomes very important.

An integral type of computational method for solution of the Boltzmann equation has been proposed by Ikuta and co-workers (Ikuta *et al.* 1991). Essential differences between this method and the path integral method (e.g. Skullerud and Kuhn 1983) have been discussed at past scientific meetings. Nevertheless, it is true that the method presents a useful and powerful means of deducing the electron energy distribution and swarm parameters under both dc and ac electric fields, in particular with excellent numerical stability.

Penning ionisation is important in such applications of weakly ionised gases as discharge lamps and plasma displays. Sakai *et al.* (1989) have recently studied the effects of Penning ionisation on the properties of electron swarms in a variety of gases.

Another topic is electron swarms in radio frequency (rf) fields. This is particularly inportant since rf fields are used in plasma processing of semiconductor devices and materials. Readers are referred to a recent review by Makabe (1991).

7. Concluding Remarks

Some recent investigations into the properties of electron swarms and their applications in gases have been introduced. The study of electron swarms has a long history, yet it has been producing very interesting and useful results which enchant many scientific workers. The author believes that this well of enchantment will not run dry in the near future, despite vigorous investigations by keen researchers.

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