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A Monte-Carlo Simulation of the Behaviour of Electron Swarms in Hydrogen using an Anisotropic Scattering Model

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Abstract

Hunter (1977) found that a Monte-Carlo simulation of electron swarms in hydrogen, based on an isotropic scattering model, produced discrepancies between the predicted and measured electron transport parameters. The present paper shows that, with an anisotropic scattering model, good agreement is obtained between the predicted and experimental data. The simulation code is used here to calculate various parameters which are not directly measurable.

1. Introduction

In a recent paper Hunter (1977; hereafter referred to as paper 1) described a Monte-Carlo simulation of the behaviour of electron swarms in a low pressure hydrogen gas under the influence of an electric field assuming only isotropic electron scattering. This work has now been extended to allow for the more realistic situation of nonisotropic electron scattering. We believe that only in this way can a consistent set of scattering cross sections be found for predicting the various transport and production coefficients with accuracy over a wide E/N range (E being the electric field and N the gas number density).

In the present paper, a more detailed set of inelastic cross sections is used in an attempt to confirm and expand upon the relationship between the photon and electron distributions described in paper 1. The photon distributions are obtained when electrons collide inelastically with the gas molecules, thereby producing radiation which may be observed as in the experiments of Blevin *et al.* (1976*a*, 1976*b*). An anisotropic scattering model should allow the relationship between the photon and electron distributions to be obtained with greater precision than was possible in the isotropic work of paper 1, thus allowing corrections to be made (where necessary) to the experimental results of Blevin *et al.* (1976*b*). This work will be reported elsewhere (Blevin *et al.* 1978).

The present simulations were performed over the E/N range $0{\cdot}5 \leqslant E/N \leqslant 200$ Td* using a consistent set of collision cross sections over the energy range from the threshold for each process to 50 eV. The gas number density used in the present simulations was $N = 3{\cdot}54 \times 10^{16}$ cm^{-3}, while the upper E/N limit was chosen such that excessive

* 1 Td (Townsend) $\equiv 10^{-21}$ V m^2.

amounts of computer time were not required to follow all the electrons produced in ionizing collisions. At E/N values below 0·5 Td, the electron scattering at collision becomes increasingly isotropic and elastic, allowing numerical Boltzmann solutions to accurately predict the transport parameters, as shown by Milloy and Watts (1977).

2. Collision Model Modifications

The collison model used in the present work has been described in paper 1 and is based upon the mean free time approximation. The largest assumption made in paper 1 was that the scattering of an electron after collision with a gas molecule was isotropic. This assumption led to a considerable discrepancy between the experimental and derived values for the transport and production coefficients. The anisotropic scattering data included in the present work are based on recent low energy angular scattering results.

(*a*) *Anisotropic Scattering*

The normalized weighting factor which determines the scattering angles θ and ϕ of the electron after collision with a gas molecule is given by

$$P(\theta, \phi) = \int_0^{\phi} \int_0^{\theta} I(\theta) \sin\theta \, \mathrm{d}\theta \, \mathrm{d}\phi \Big/ \int_0^{2\pi} \int_0^{\pi} I(\theta) \sin\theta \, \mathrm{d}\theta \, \mathrm{d}\phi ,$$

where $I(\theta)$ is the differential scattering cross section at a particular electron velocity V. The scattering angle ϕ may be found by calling a random number R_1, uniformly distributed on the interval [0, 1], and relating this number to ϕ by the expression $\phi = 2\pi R_1$.

The scattering angle θ is in turn related to a second random number R_2 by the relation

$$R_2(\theta) = \int_0^{\theta} I(\theta) \sin\theta \, \mathrm{d}\theta \Big/ \int_0^{\pi} I(\theta) \sin\theta \, \mathrm{d}\theta .$$

This expression is normalized by requiring that

$$\int_0^{\pi} I(\theta) \sin\theta \, \mathrm{d}\theta = 1 .$$

Defining the quantity $X = \frac{1}{2}(1 - \cos\theta)$, where $X = R_2$ for isotropic scattering, gives

$$R_2 = 2k \int_0^{X} Y(X) \, \mathrm{d}X , \tag{1}$$

with k a constant to be determined.

An examination of the angular scattering distribution data for electrons from molecular hydrogen (see e.g. Trajmar *et al.* 1970; Linder and Schmidt 1971; Teubner *et al.* 1974) indicates that a good approximation to the forward scattering differential cross section is given by $Y(X) = A\exp(-X/C)$, where A and C are energy-dependent constants that are to be determined. Similarly, in the case of backward scattering, $Y(X)$ may be approximated by $Y(X) = B\exp(X/D)$, where B and D are further

constants. Thus substituting into equation (1) and normalizing, such that $R_2 = 0$ and 1 when $X = 0$ and 1 respectively, gives

$$X = -C\ln(|1 - R_2\{1 - \exp(-1/C)\}|) \tag{2}$$

for forward scattering, and correspondingly

$$X = D\ln(|1 - R_2\{1 - \exp(1/D)\}|) \tag{3}$$

for backward scattering.

In the situation where the electron can be either forward or backward scattered, the differential scattering cross section may be approximated by

$$Y(X) = A' \exp(-X/C) + \exp(X/D),$$

with A' a constant between 0 and 1 which is dependent on the electron's energy at collision. To determine whether an electron is forward or backward scattered at collision, we call a random number R_1. Depending on whether $R_1 \gtrless A'$, we use either equation (2) or (3) respectively to determine the angular scattering of the electron by calling and substituting a second random number R_2. The scattering angle θ may then be found from the following relation

$$\theta = \arccos(1 - 2X).$$

(b) Collision Cross Sections

The collision cross sections used in this study differ considerably from those used in paper 1. In particular, separate singlet and triplet states have been included in an attempt to determine the dependence on E/N of the relative intensities of the radiation produced from the decay of these states. The importance of this knowledge is discussed elsewhere (Blevin *et al.* 1978).

The total scattering cross section Q_T is the same as that used in paper 1. The momentum transfer cross section Q_m was chosen to agree with that obtained by Crompton *et al.* (1969) up to 1·0 eV. Above 1·0 eV, the angular scattering distributions, and hence Q_m, were freely modified until reasonable agreement between the theoretical and experimental transport parameters was obtained over the entire E/N range used in this work.

Two rotational cross sections were used, namely the J_{0-2} cross section obtained by Crompton *et al.* (1969) and the J_{1-3} cross section obtained by Gibson (1970). The high energy tail of the cross section was extended to 100 eV by using the data of Srivastava *et al.* (1975*a*, 1975*b*). It was assumed that 12% of the molecules were in the $J = 0$ rotational ground state and that 65% occupied the $J = 1$ state. This approximately corresponds to the levels occupied by the gas molecules at room temperature. The remainder of the molecules were assumed to occupy the excited $J = 2$ and 3 states, and it was also assumed that super-elastic collisions and higher excitation transitions in these molecules have no effect on the swarm transport parameters. The electrons were taken to be isotropically scattered in rotational excitation collisions, as this has been shown by Linder and Schmidt (1971) and Srivastava *et al.* to be a good approximation for impact energies up to 10 eV.

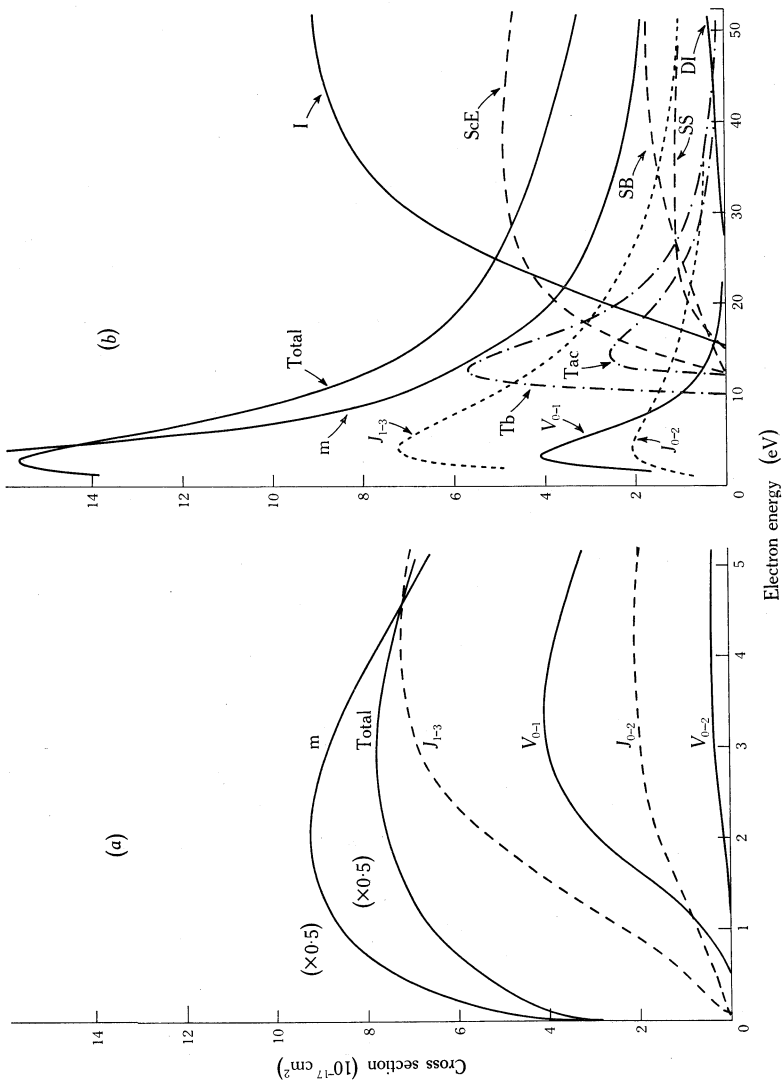

Fig. 1. Scattering cross sections for molecular hydrogen used in the Monte-Carlo simulations: (*a*) low energy cross sections and (*b*) high energy cross sections. Abbreviations used are; Total, total scattering; m, momentum transfer; *J*, rotational; *V*, vibrational; I, ionization; DI, dissociative ionization. The remaining abbreviations designate the singlet- and triplet-state excitation cross sections defined in Section 2*b*.

The chosen vibrational cross section for excitation to the $V = 1$ level was, up to 2 eV, that obtained by Crompton *et al.* (1969), with a smooth transition being made to that obtained by Linder and Schmidt (1971) at higher energies. The cross section for excitation to the $V = 2$ state was that obtained by Ehrhardt *et al.* (1968). The angular distribution of the scattered electrons in vibrational excitation is considerably anisotropic (Ehrhardt *et al.* 1968; Linder and Schmidt 1971), and consequently the scattering distribution of electrons from vibrational excitation was chosen to agree approximately with that observed experimentally at an impact energy of 3 eV.

There are a large number of singlet- and triplet-state excitation levels occupying the energy range between 11 and 15 eV (Sharp 1971), but it is impractical to derive cross sections for excitation to all these states, as no uniqueness can be claimed for any set that is thereby obtained. As a result, only three singlet- and three triplet-state cross sections were used to approximate these events. The three triplet-state cross sections used were: Q_{Tb} for excitation to the $\mathrm{b}\,^3\Sigma_u^+$ state; Q_{Tac} for excitation to the sum of the $\mathrm{a}\,^3\Sigma_g^+$ and $\mathrm{c}\,^3\pi_u$ states; Q_{TS} for excitation to the sum of the remaining triplet states. The thresholds for these cross sections were 8·9, 11·9 and 14·0 eV respectively. The three singlet-state excitation cross sections were: Q_{SB} for excitation to the $\mathrm{B}\,^1\Sigma_u^+$ state, with a threshold of 11·4 eV; Q_{ScE} for excitation to the sum of $\mathrm{c}\,^1\pi_u$ and $\mathrm{E}\,^1\Sigma_g^+$ states, with a threshold of 12·4 eV; Q_{SS} for excitation to the sum of the remaining singlet states with a threshold of 15·0 eV. The electron scattering in triplet-state excitation events was assumed to be isotropic while, for singlet-state excitation, the angular distributions measured by Hughes and McMillen (1932) were used. The justification for these cross sections has been discussed in paper 1.

The $^2\Sigma_u^+$ dissociative ionization cross section with a threshold energy of 23·0 eV measured by Crowe and McConkey (1973) and the ionization cross section of Cowling and Fletcher (1973) for excitation to the $\mathrm{X}\,^2\Sigma_g^+$ state, with a threshold of 15·5 eV, were used to simulate the ionization processes in molecular hydrogen. The angular distribution of the ejected electrons from both molecular and dissociative ionization events was assumed to be isotropic. The cross sections used in this work are shown in Figs 1*a* and 1*b*.

3. Results

To obtain accurate results for the transport and production coefficients at each E/N value, approximately 1000 electrons were sampled in each swarm, and the printout times were varied so that each electron experienced approximately 1000 collisions before the final printout time. The spatial and velocity coordinates for each electron were sampled after five constant time intervals, enabling the swarm drift velocity W to be obtained to within an accuracy of $\pm 1\%$, the lateral diffusion coefficient D_{T} to within $\pm 2\%$, the longitudinal diffusion coefficient to within $\pm 4\%$, and the mean swarm energy $\langle \varepsilon \rangle$ and production coefficients to within $\pm 2\%$.

The initial non-equilibrium time for the model was kept to a minimum by starting each electron with the energy possessed at the final printout time of the electron immediately preceding the one under study. This procedure automatically ensured that the electrons were started with the final energy distribution of the swarm, and since the electrons are randomly oriented at the start of the simulation, the time taken for the electron to reach equilibrium is negligible, allowing the whole of the electron's motion to be used to derive the various transport and production coefficients.

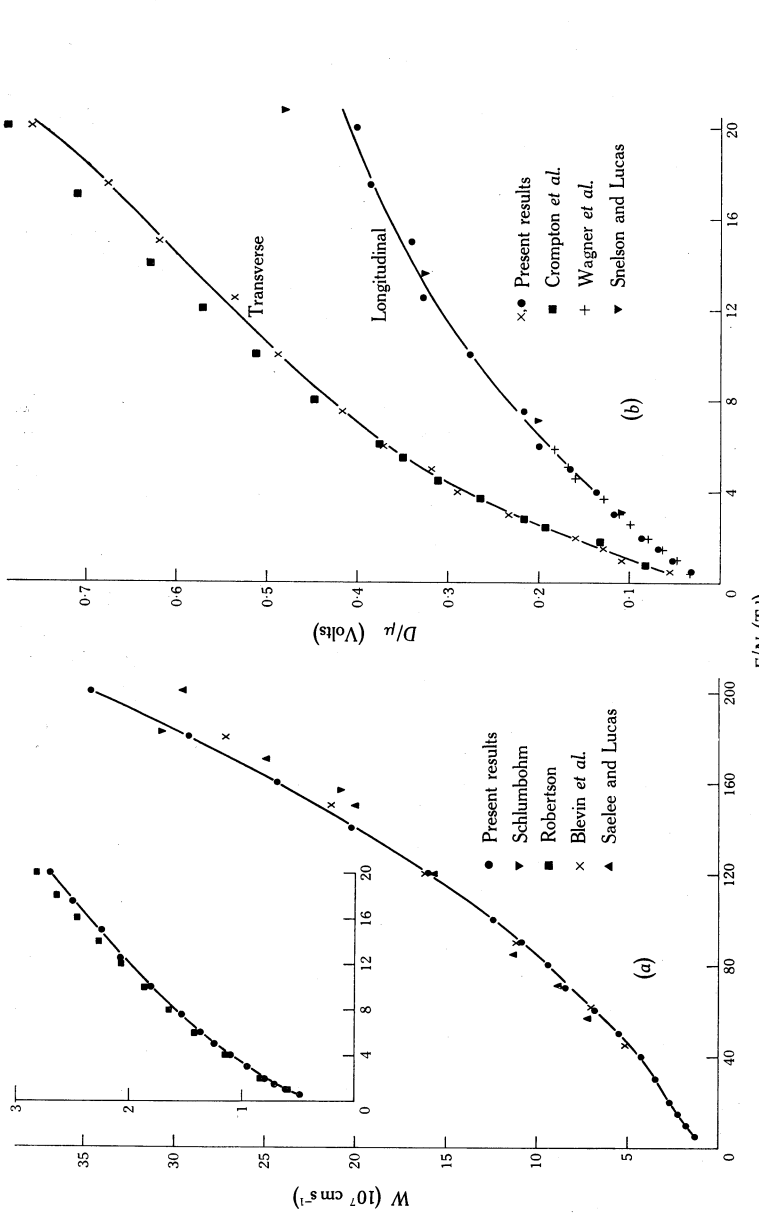

Fig. 2. Comparison between simulated and experimental results for (*a*) the electron drift velocity W and (*b*) the transverse and longitudinal diffusion coefficients at low E/N, shown as functions of E/N. Experimental data are from: Schlumbohm (1965*a*, 1965*b*); Crompton and McIntosh (1966), Crompton *et al.* (1968); Wagner *et al.* (1967); Robertson (1971); Snelson and Lucas (1975); Blevin *et al.* (1976*b*); Saelee and Lucas (1977).

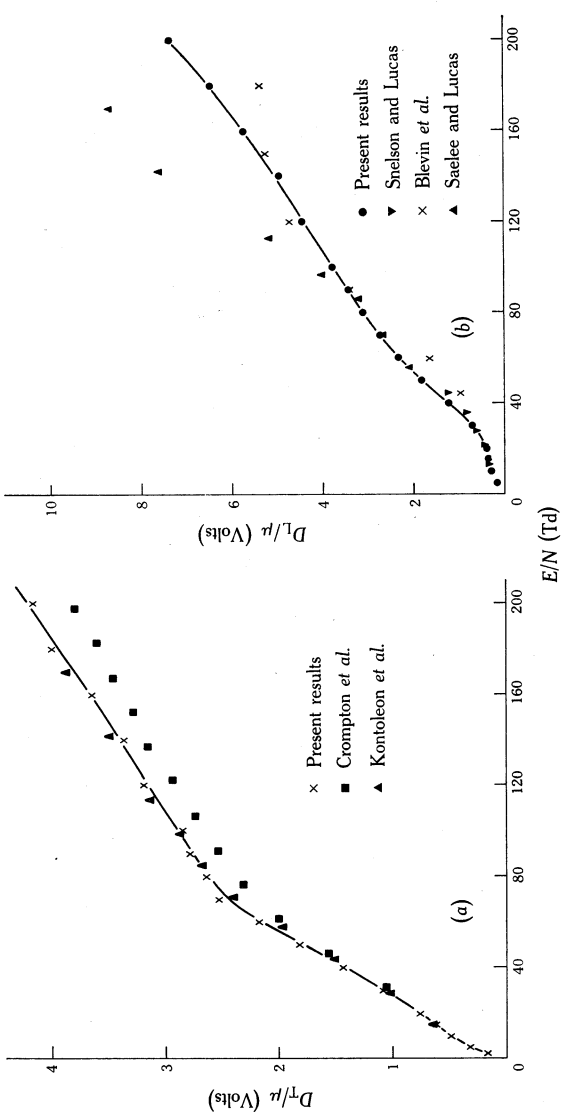

Fig. 3. Comparison between simulated and experimental results at high E/N for (*a*) the transverse diffusion coefficient D_T/μ and (*b*) the longitudinal diffusion coefficient D_L/μ. Experimental data are from: Crompton *et al.* (1965), Kontoleon *et al.* (1972), Snelson and Lucas (1975), Blevin *et al.* (1976*b*) and Saelee and Lucas (1977).

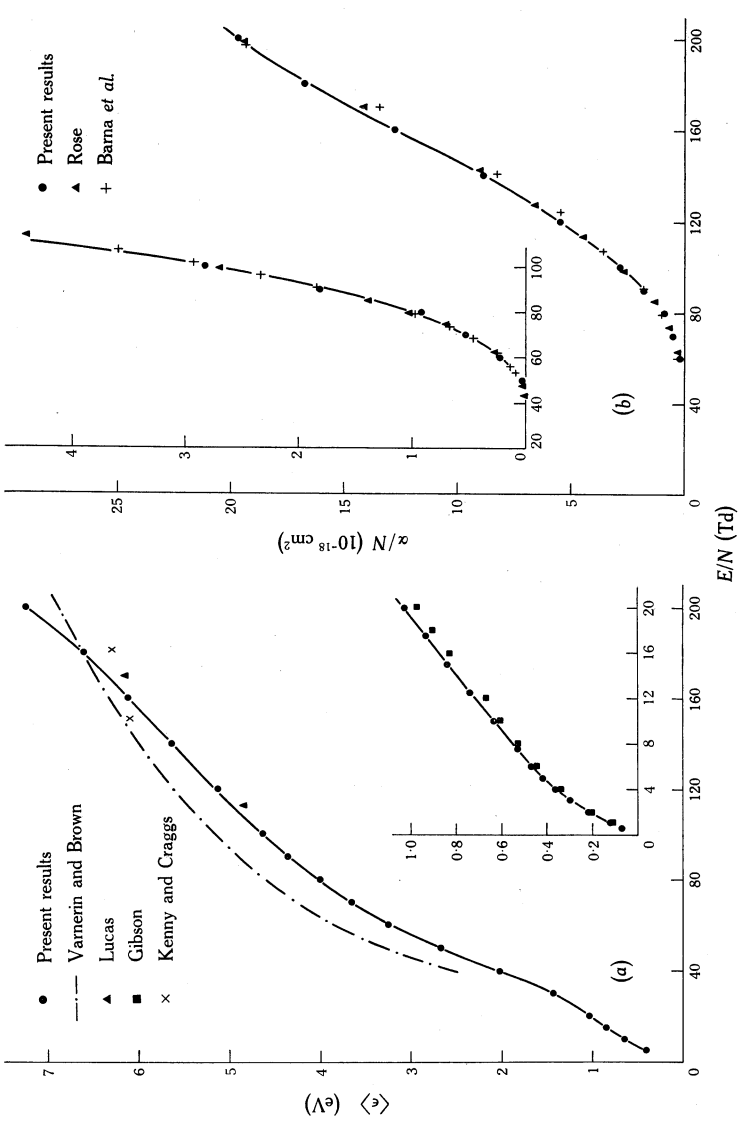

Fig. 4. Comparison between simulated and experimental results for (*a*) the electron swarm average energy $\langle \varepsilon \rangle$ and (*b*) the first ionization coefficient α/N. Experimental data are from: Varnerin and Brown (1950), Rose (1956), Barna *et al.* (1964), Lucas (1969), Gibson (1970) and Kenny and Craggs (1970).

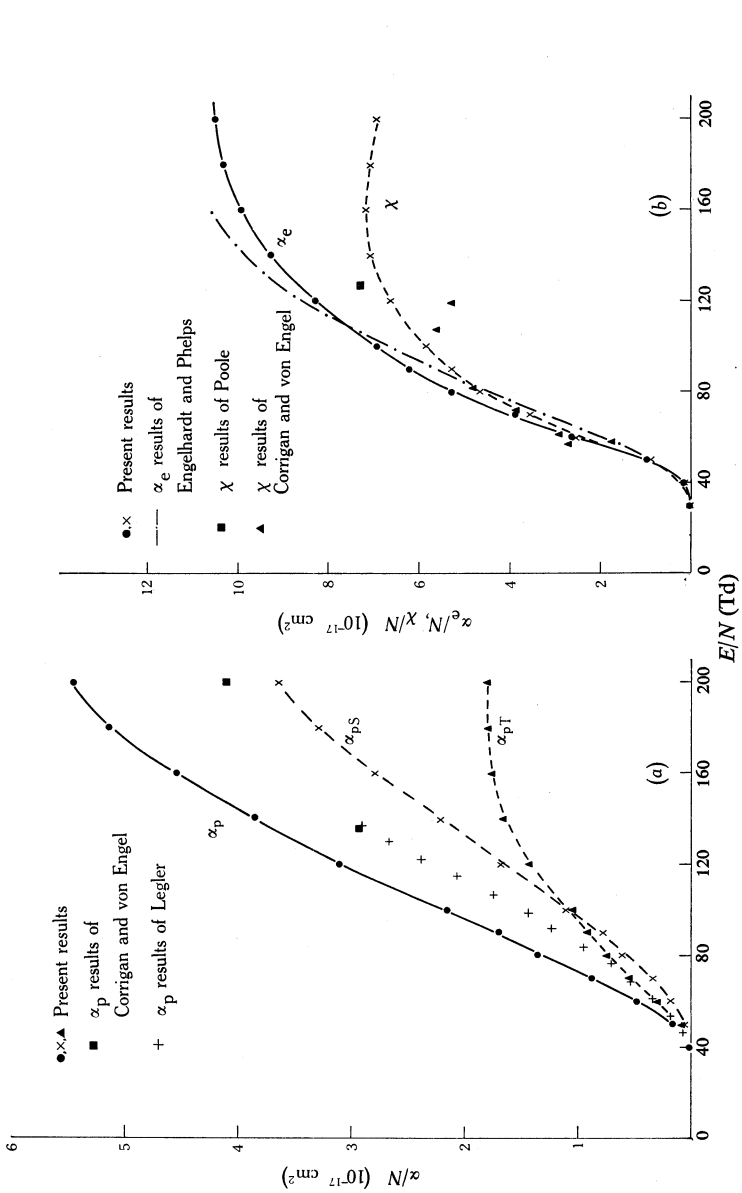

Fig. 5. Comparison between simulated and experimental results for (*a*) the total photon production coefficient α_p and its singlet and triplet coefficients α_{pS} and α_{pT} and (*b*) the total electronic excitation coefficient α_e/N and the dissociation coefficient χ/N. Experimental data are from: Poole (1937), Corrigan and von Engel (1958), Engelhardt and Phelps (1963) and Legler (1963).

In attempting to obtain a collision model that would enable the collision processes that occur in an electron swarm to be accurately simulated, the initial set of collision cross sections was varied until reasonable agreement was obtained between the experimental and simulation results for the three major transport parameters: W, ND_T and ND_L, and for the Townsend ionization coefficient α_T. The uniqueness and accuracy of the final set of cross sections given in Figs 1*a* and 1*b* are discussed in paper 1.

The drift velocity of the swarm is defined as $W = \Delta\langle z\rangle/\Delta t$ when the initial non-equilibrium region is negligibly small. The simulation results for the dependence of W on E/N are shown in Fig. 2*a*, where comparisons can be made for low E/N with the experimental results of Robertson (1971) and for high E/N with those of Schlumbohm (1965*a*, 1965*b*), Blevin *et al.* (1976*b*) and Saelee and Lucas (1977).

The ratio of the diffusion coefficient D to the electron mobility μ is given as

$$D/\mu = ND\,W^{-1}(E/N),$$

where D is either the lateral diffusion coefficient D_T, defined as

$$D_T = \langle\Delta R^2\rangle/4\Delta t = \langle\Delta R\rangle^2/\pi\Delta t,$$

with $R^2 = x^2+y^2$, or the longitudinal diffusion coefficient D_L, defined as

$$D_L = \Delta(\langle z^2\rangle-\langle z\rangle^2)/2\Delta t.$$

The low E/N values of D_L/μ and D_T/μ are given in Fig. 2*b* in comparison with the experimental results given by Wagner *et al.* (1967), Crompton *et al.* (1968), Huxley and Crompton (1975, results from Crompton and McIntosh 1966) and Snelson and Lucas (1975). The high E/N values of D_T/μ are given in Fig. 3*a* in comparison with results of Crompton *et al.* (1965) and Kontoleon *et al.* (1972), while the high E/N values of D_L/μ are shown in Fig. 3*b* in comparison with the results of Snelson and Lucas (1975), Blevin *et al.* (1976*b*) and Saelee and Lucas (1977).

The small disagreement between the simulation and experimental results for the above three transport parameters at the lower E/N values is expected to be primarily due to the assumed form of the low energy Q_m cross section, as the transport parameters are very sensitive to changes in this cross section. Similarly, at the upper E/N limit considered in this work, an increase in the Q_m cross section above approximately 20 eV would significantly reduce the simulation-derived transport parameters and improve the agreement with the experimental results.

The average energy of the electron swarm $\langle\varepsilon\rangle$ obtained from the simulations is shown in Fig. 4*a* in comparison with the values obtained by Gibson (1970) from a numerical solution of the Boltzmann equation at low E/N, while the high E/N results are compared with the experimental results of Varnerin and Brown (1950) and Kenny and Craggs (1970) and with the Boltzmann-derived values of Lucas (1969). The disagreement between the simulation and experimental results of Varnerin and Brown cannot be regarded as being significant, since the experimental results were obtained in a microwave discharge experiment, and consequently several assumptions must be made before values for $\langle\varepsilon\rangle$ can be obtained from the experimental results.

Production coefficients for all of the various inelastic processes occurring in the electron swarm may also be obtained from the simulations. The most important production mechanisms that can be studied in the present simulations are Townsend

ionization (coefficient α_T/N), photon production (coefficient α_p/N) and dissociation (coefficient χ/N). These coefficients are shown in Figs 4*b*, 5*a* and 5*b* respectively. It may be seen that there is good agreement between the simulation results for α_T/N and the experimental results of Rose (1956) and Barna *et al.* (1964) over the complete E/N range considered in this work.

Fig. 5*a* also displays the photon production coefficient obtained from excitation of the singlet states α_{pS}/N and of the higher triplet states α_{pT}/N, excluding direct excitation to the $b\,^3\Sigma_u^+$ state which decays via dissociation without the production of radiation. It may be noted that, at high E/N, the predominant production mechanism is via decay of the excited singlet states while, at lower E/N values, the decay of the higher triplet states produces the majority of the photons. This knowledge is important in determining the relative percentages of the radiation produced by the decay of singlet and triplet states, as observed in the experiments of Blevin *et al.* (1976*a*, 1976*b*). It is discussed by Blevin *et al.* (1978).

The dissociation coefficient χ/N is obtained by summing the production coefficient for excitation to all the triplet states and the dissociative ionization state. This coefficient is compared in Fig. 5*b* with the experimental results of Poole (1937) and Corrigan and von Engel (1958). Also shown in Fig. 5*b* is the production coefficient α_e/N for excitation to all the higher electronic states excluding ionization. A comparison is made with the results obtained by Engelhardt and Phelps (1963) from a numerical solution of the Boltzmann equation. It may be noted that, with few exceptions, there is good agreement between the simulation-derived production coefficients and the experimental. The differences that do exist cannot be adequately resolved at this stage, due to the lack of sufficiently accurate singlet- and triplet-state excitation cross sections and production coefficients for these processes.

The percentage power lost due to the elastic collisions and excitation to the low energy inelastic events at low E/N is shown in Fig. 6*a* in comparison with the Boltzmann results of Gibson (1970). The power lost due to all the processes at high E/N is shown in Fig. 6*b*. Gibson used a considerably larger J_{0-2} cross section than we have, and this is evidenced by the greater loss due to excitation to the J_{0-2} state in his work. Although the magnitudes of the power losses are considerably different in each case, there is very good agreement as to the relative shape of these curves as a function of E/N. It may be noted from Fig. 6*b* that, above $E/N \approx 120$ Td, the percentage power lost to the triplet excitation events gradually decreases, and this is confirmed by the decreasing trend in the dissociation production coefficient above $E/N \approx 140$ Td in Fig. 5*b*.

4. Conclusions

In conclusion, the present work has shown that it is now possible to reliably predict the electron swarm transport parameters over a wide range of E/N if accurate collision data are used in the simulation of the electron motion in the gas. Any discrepancies that exist between the experimental and simulation results have been considered above, and relatively small adjustments to the collision cross sections will correct any errors in the derived transport data. Although the set of cross sections that is thereby obtained is not unique, due to the large uncertainties that exist in a number of the cross sections (particularly in Q_m and the higher electronic excitation states), nevertheless it is thought that the simulation code is of sufficient sophistication to allow the collision processes in the electron swarm to be accurately simulated.

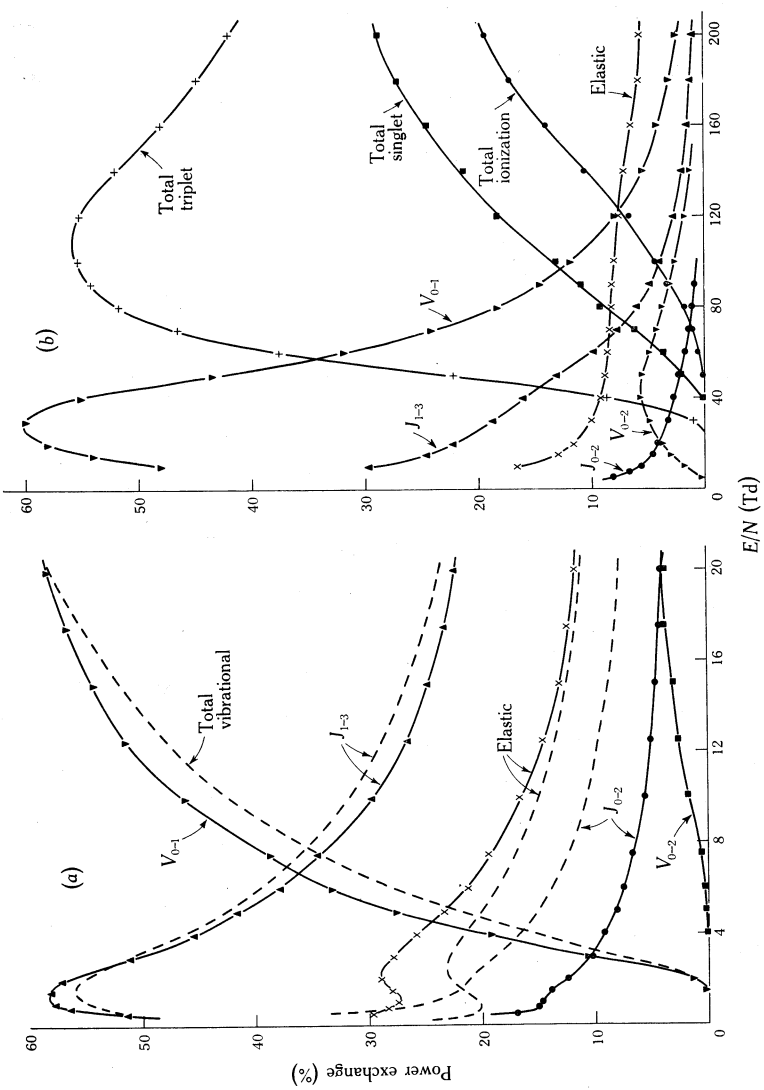

Fig. 6. Percentage power exchange as a function of E/N: (*a*) comparison between the present simulated results (full curves) and the Boltzmann calculations (dashed curves) of Gibson (1970) for the rotation and vibration states at low E/N and (*b*) simulated results at high E/N for all inelastic processes.

Finally, this study has shown the relative importance as a function of E/N of the singlet- and triplet-state excitations as photon production mechanisms. It has been observed that, at high E/N, the decay of the singlet states is the predominant source of photons while, at lower E/N values, the decay of the higher triplet states will produce more photons. This information is of importance in the correction of the photon distributions obtained in the experiments of Blevin *et al.* (1976*b*) as outlined by Blevin *et al.* (1978).

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