

Anisotropic scattering of electrons by N_2 and its effect on electron transport

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As part of a systematic study of approximations commonly made in solutions of the Boltzmann equation for electrons in molecular gases, we have investigated the effects of anisotropic scattering on electron transport coefficients in N_2 and have extended our study of the multiterm expansion technique to higher E/n . A critical survey of published data yields a set of differential and integral cross sections for electron energies from 0.003 to 10^4 eV. For electric-field-to-gas-density ratios E/n between 10 and 500 Td (1 Td = 10^{-21} V m²), the changes in the commonly measured transport and reaction coefficients resulting from the introduction of anisotropic elastic and inelastic scattering, while keeping the elastic momentum-transfer cross section constant, are less than 1%. These calculations were made with use of the multiterm spherical-harmonic expansion solution of the Boltzmann equation. For $500 < E/n < 1500$ Td the changes in scattering anisotropy **cause changes in transport and reaction coefficients which increase with E/n to about 10%. The errors in drift velocity, mean energy, and the reaction coefficients resulting from the use of the two-term spherical-harmonic expansion rather than a six-term expansion are less than 3% at 1500 Td. However, the errors in the diffusion coefficients become large ($> 25\%$) at our highest E/n . The calculated transport coefficients are in generally good agreement with experiment for E/n less than 300 Td, but the differences increase at higher E/n . The importance of proper interpretation of ionization and excitation experiments at high E/n is illustrated by calculations which model either an exponential growth of density in time or an exponential growth with position. The calculated ionization coefficients are low compared to most experiments for E/n less than 200 Td. At $E/n > 600$ Td the agreement is good for the spatial growth experiments, but the calculated values are below experiment from the temporal growth experiments. The calculated excitation coefficients are generally higher than experiment at low and high E/n but in agreement with experiment at E/n near 150 Td.**

I. INTRODUCTION

The availability of differential scattering data for electrons in N_2 and the development of the multiterm spherical-harmonic expansion techniques for solution of the electron Boltzmann equation¹ make possible this investigation of the effects of anisotropic scattering on electron transport in N_2 . The present work is part of a systematic study¹⁻³ of the effects of various simplifying approximations on the solutions of the Boltzmann equation for electrons in molecular gases, such as N_2 . Our primary objective has been to develop and test techniques for improving the accuracy of the calculated values of measured transport and reaction-rate coefficients. Although this series of papers has included some changes in electron collision cross sections which improve the agreement between calculations and experiment, we are much more concerned with the effects of the computational techniques, secondary-electron distributions, and angular distribution on these calculated transport coefficients. The initial paper,¹ which we will refer to as I, describes techniques for adding higher-degree spherical harmonics (Legendre polynomials) to the two-term or Lorentz ap-

proximation when solving the electron Boltzmann equation. This work demonstrated the changes in calculated **transport and reaction, coefficients which occur when the inelastic cross section becomes comparable with the elastic momentum-transfer cross section, as in N_2 at energies near 2 eV.** In the second paper,² which will be referred to as II, we showed that at low E/n in N_2 the use of the two-term approximation causes large errors in the calculation of excitation rate coefficients, but that for low and moderate E/n the errors in transport coefficients are generally small, e.g., less than 8%. More recently³ in paper III, we examined the effects of various approximations to the distribution in energy of secondary electrons produced in the process of electron-impact ionization. We found that the primary consideration is to include the new electron in the Boltzmann equation. The results are sufficiently insensitive to the details of the assumed secondary-electron energy distribution so as to allow the use of delta function secondary-electron energy distributions adjusted to satisfy energy and momentum conservation.

The question of the effects of anisotropic scattering on solutions of the Boltzmann equation and on electron

transport calculations have been considered for moderate E/n and electron energies and for some model angular distributions.^{4,5} These calculations show that for realistic angular distributions the errors in the calculated transport coefficients are small, i.e., less than 5%. In a preliminary report on the present work⁶ we assembled a set of anisotropic cross sections for electrons in N₂ and applied it to the calculation of electron-energy distribution functions and transport coefficients at moderate E/n . In the present paper we discuss these cross sections and extend their application to higher electron energies and higher E/n . At moderate E/n and mean energies, we find that the calculated effects of anisotropic scattering are small enough so as to be observed only in precision swarm experiments. At higher E/n and higher electron energies where electron scattering is known to be highly anisotropic, we find increasingly important errors in the solutions of the electron Boltzmann equation obtained using the usual assumptions of momentum-transfer elastic scattering and isotropic inelastic scattering. We will limit E/n to 3000 Td and below, since experiments⁷ and recent theory⁸ for N₂ indicate the onset of departures from the

and Lucas⁹ and Kunhardt and Tzeng⁹ have investigated the effects of anisotropic scattering on electron motion in N₂ using Monte

$$+ \frac{h_{\rho\rho}(\mathbf{v})}{n^2} \frac{1}{\rho} \frac{\partial}{\partial \rho} \left[\rho \frac{\partial n_e(\mathbf{r}, t)}{\partial \rho} \right] \frac{\partial^2}{\partial z^2} \quad (2)$$

Here $f(\mathbf{v})$ is the velocity distribution in the absence of density gradients, $\rho = |\mathbf{r}| \sin \phi$ is the angle of the position vector \mathbf{r} with the direction of electron acceleration, $n_e(\mathbf{r}, t)$ is the electron density, $g_z(\mathbf{v})$ and $g_\rho(\mathbf{v})$ are the velocity distributions associated with the gradient of the electron density in the direction of acceleration and the direction perpendicular to the acceleration, and $h_{zz}(\mathbf{v})$ and $h_{\rho\rho}(\mathbf{v})$ are the velocity distributions associated with the second derivative of the electron density in the direction of \mathbf{a} and at right angles to \mathbf{a} . Substitution of Eq. (2) into Eq. (1) and the integration over all velocities yields the continuity equation for the electron density, i.e.,

$$\frac{\partial n_e(\mathbf{r}, t)}{\partial t} = \nu n_e(\mathbf{r}, t) - W \frac{\partial n_e(\mathbf{r}, t)}{\partial z} + D_L \frac{\partial^2 n_e(\mathbf{r}, t)}{\partial z^2} + \frac{D_T}{\rho} \frac{\partial}{\partial \rho} \left[\rho \frac{\partial n_e(\mathbf{r}, t)}{\partial \rho} \right], \quad (3)$$

where

$$\begin{aligned} \nu &= n \int C[f(\mathbf{v})] d^3v, \\ W &= \mu E = \int v_z f(\mathbf{v}) d^3v + \int C[g_z(\mathbf{v})] d^3v, \\ n D_L &= \int v_z g_z(\mathbf{v}) d^3v + \int C[h_{zz}(\mathbf{v})] d^3v \end{aligned}$$

itial energy distribution. Note that the integrals over the collision operator $C[f]$ in Eqs. (4)–(7) were not present in I and II because electron production and loss were not included in the Boltzmann equation.

Equations for $f(\mathbf{v})$, $g_z(\mathbf{v})$, $g_\rho(\mathbf{v})$, $h_z(\mathbf{v})$, and $h_{\rho\rho}(\mathbf{v})$ are obtained by substituting Eq. (2) into Eq. (1) and setting coefficients of $n_e(\mathbf{r}, t)$ and its derivatives equal to zero. The equation for $f(\mathbf{v})$ is

$$\mathbf{a} \cdot \nabla_{\mathbf{v}} f(\mathbf{v}) = nC[f(\mathbf{v})] - \nu f(\mathbf{v}). \quad (9)$$

The $\nu f(\mathbf{v})$ term accounts for the temporal growth and was added in III with the resultant requirement that Eq. (9) be solved as an eigenvalue problem. The differential equation for $g_z(\mathbf{v})$ is

$$\mathbf{a} \cdot \nabla_{\mathbf{v}} g_z(\mathbf{v}) = nC[g_z(\mathbf{v})] - \nu g_z(\mathbf{v}) - n(W - v_z)f(\mathbf{v}). \quad (10)$$

Integration of this equation over all velocities gives Eq. (5). The solution of Eq. (10) has been discussed by Tagashira *et al.*¹¹ Because of difficulties* encountered in the evaluation of $g_z(\mathbf{v})$ in the case of N_2 , but not other gases, we will not present values of D_L or of the second integral of Eq. (5). We will assume that this correction to W can be neglected in our evaluation of the effects of anisotropic scattering or of the effects of the truncation of the spherical-harmonic expansion. In this case the solution of Eq. (10) simplifies to the equations discussed in II and III. The differential equation for $g_\rho(\mathbf{v})$ is

$$\mathbf{a} \cdot \nabla_{\mathbf{v}} g_\rho(\mathbf{v}) = nC[g_\rho(\mathbf{v})] - \nu g_\rho(\mathbf{v}) + n\nu \sin\theta[f(\mathbf{v})]. \quad (11)$$

As for W , we neglect the contribution of the second integral in Eq. (7) to the value of D_T .

Similar but more complicated equations are obtained*, for $h_z(\mathbf{v})$ and $h_{\rho\rho}(\mathbf{v})$. Since these equations were not solved for this paper, they will not be given here.

The transport and ionization coefficients of Eq. (3) are sometimes termed time-of-flight (TOF) coefficients,*, since Eq. (3) is often applicable to the analysis of data obtained from time-of-flight experiments.^{13,14} We prefer to call these coefficients the density gradient expansion coefficients and reserve the time-of-flight terminology for the experiments and the transport coefficients derived by fitting solutions to Eq. (3) to these experiments. This procedure is consistent with ¹⁵

$$\frac{\nu}{n} - \frac{\alpha_i}{n} W + \left\{ \frac{\alpha_i^2}{n} (nD_L) \right\} = 0. \quad (13)$$

Equation (13), thus establishes the connection^{13,14} between the density, gradient expansion coefficients ν , W , and D_L .

Use of this form" instead of Eqs. (10) and (13) allows one to considerably simplify the Boltzmann calculation for steady-state experiments in which α_i is determined from the variation of $n_e(z)$ with position or with electrode spacing as in the steady-state Townsend experiment.," Thus, substitution of Eq. (14) into Eq. (1) yields

$$\mathbf{a} \cdot \nabla_{\mathbf{v}} f_{ss}(\mathbf{v}) + v_z \alpha_i f_{ss}(\mathbf{v}) = nC[f_{ss}(\mathbf{v})]. \quad (15)$$

The spatial ionization coefficient α_i is obtained by integrating Eq. (15) over all velocities, i.e.,

$$\frac{\alpha_i}{n} = \frac{\int v Q_0^i(v) f_{ss}(\mathbf{v}) d^3v}{\int v_z f_{ss}(\mathbf{v}) d^3v} \equiv \frac{k_{iss}}{V_d}, \quad (16)$$

where $Q_0^i(v)$ is the total ionization cross section. Equations (15) and (16) must be solved iteratively to obtain $f_{ss}(\mathbf{v})$, α_i/n , etc. Note that the α_i/n calculated by Eq. (16) is the same as that which would be obtained by extending the density gradient expansion and Eq. (13) to an infinite number of terms. Generally, the ionization rate coefficient k_{iss} defined by the numerator of Eq. (16) is smaller than $\nu/n \equiv k_i$ at the same E/n , i.e., the diffusion of electrons toward the cathode due to the density gra-

is the root of the equation

We will consider only cases in which either ν or α_i is zero, although one might well analyze experiments in which it is a good approximation to assume that both ν and α_i were nonzero. Note that in Eqs. (15) and (17) the magnitude of α_i is not restricted by the requirement of rapid convergence of the density gradient expansion as in Eq. (13).

We next make use of the spherical-harmonic expansion technique for solving for the electron velocity or energy distribution. In this discussion we will consider only Eq. (17), since the extension of the method to equations such as Eqs. (10) and (11) is straightforward. As in II, the

velocity distribution $\mathbf{f}(\mathbf{v})$ is written in the form

$$f(\mathbf{v}) = \sum_{j=0}^{m-1} f_j(\epsilon) P_j(\cos\theta), \quad (18)$$

where ϵ is the electron energy, m is the number of terms in the expansion, $P_j(\cos\theta)$ is the zero-order Legendre polynomial of degree j , and $f(\epsilon)$ is normalized by

$$\int_0^\infty \epsilon^{1/2} f_0(\epsilon) d\epsilon = 1. \quad (19)$$

With these assumptions Eq. (9) can be written as a series of coupled equations of the form

$$\begin{aligned} \frac{eE}{n} \frac{j}{(2j-1)} \left[\epsilon \frac{df_{j-1}}{d\epsilon} - \frac{(j-1)}{2} f_{j-1} \right] + \frac{eE}{n} \frac{(j+1)}{(2j+3)} \left[\epsilon \frac{df_{j+1}}{d\epsilon} + \frac{(j+2)}{2} f_{j+1} \right] \\ + \epsilon \frac{\alpha_i}{n} \left[\frac{j}{(2j-1)} f_{j-1} + \frac{(j+1)}{(2j+3)} f_{j+1} \right] = B[f_j], \end{aligned} \quad (20)$$

where $0 \leq j \leq m-1$ and the modified collision operator is given by

$$\begin{aligned} B[f_j(\epsilon)] = -\epsilon f_j(\epsilon) Q_T(\epsilon) + \epsilon f_j(\epsilon) Q_j^0(\epsilon) + \sum_{k=1}^p (\epsilon + \epsilon_k) f_j(\epsilon + \epsilon_k) Q_j^k(\epsilon + \epsilon_k) \\ + \frac{2m}{M} \delta_{j0} \frac{d}{d\epsilon} [\epsilon^2 Q_m^0(\epsilon) f_0(\epsilon)] + \int_{2\epsilon+u_i}^\infty u q_{\text{sec}}^i(j, u, \epsilon) f_j(u) du + \int_{\epsilon+u_i}^{2\epsilon+u_i} u q_{\text{sca}}^i(j, u, \epsilon) f_j(u) du - \frac{\epsilon}{v} \frac{\nu}{n} f_j(\epsilon). \end{aligned} \quad (21)$$

Here $\delta_{il} = 1$ for $j=l$ and $\delta_{il} = 0$

(22)

$$C[f_1(\epsilon)] = -\epsilon Q_m^0(\epsilon) f_1(\epsilon) - \sum_{k=1}^p \epsilon Q_0^k(\epsilon) f_1(\epsilon) + \sum_{k=1}^p (\epsilon + \epsilon_k) Q_1^k(\epsilon + \epsilon_k) f_1(\epsilon + \epsilon_k). \quad (28)$$

We see that in both of these equations the effects of elastic collisions are represented by the momentum-transfer cross section $Q_m^0(\epsilon)$. In particular, in the usual application of the conventional two-term approximation,²⁰ where the inelastic cross sections are assumed to be small compared to the elastic cross section, the only cross section entering Eq. (28) is $Q_m^0(\epsilon)$. For this reason we have chosen to make comparisons of the isotropic and anisotropic models of N_2 by keeping the elastic momentum-transfer cross section constant rather than by keeping the total elastic cross section $Q_0^0(\epsilon)$ constant as was done by Kunhardt and Tzeng.⁹ On the other hand, since we have shown in III that the term representing the reentry of anisotropic inelastic scattered electrons in Eq. (21) can often be neglected, we will keep the total inelastic cross sections $Q_0^k(\epsilon)$ constant. Note that the inelastic momentum-transfer cross section implied by Eq. (21) and analogous to Eq. (25), is not the same as that needed for use in fluid models of electron motion. See Appendix A.

In summary, we will present results in this paper for two models of electron scattering in N_2 . One model uses the full set of anisotropic scattering cross sections and will be referred to as the anisotropic model. In the second model we will use the anisotropic factor $Q_1^0(\epsilon)/Q_0^0(\epsilon)$ to obtain the momentum-transfer cross section for elastic scattering $Q_m^0(\epsilon)$. We will set all $Q_j^k(\epsilon)/Q_0^k(\epsilon)$ for $j \geq 1$ equal to zero. Thus, we use $Q_m^0(\epsilon)$ for the elastic cross section and the various $Q_0^k(\epsilon)$ or total cross sections for inelastic scattering. This model will be referred to as the isotropic inelastic model and is the model used in most previous solutions of the electron Boltzmann equation.^{13,20} Note that this model does not assume isotropic elastic scattering as is often stated.

III. CROSS SECTIONS

In this section we present the set of anisotropic collision cross sections for electrons in N_2 which we have assembled and used to investigate the effects of anisotropic scattering on the electron energy distributions and on the transport and reaction-rate coefficients. Because of the importance of collisions at high electron energies, we have examined much more thoroughly than previously the

TABLE I. Ratios of spherical-harmonic components of differential scattering cross sections for N_2 .^a

Process	Q_1^k/Q_0^k	Q_2^k/Q_0^k	Q_3^k/Q_0^k	Q_4^k/Q_0^k	Q_5^k/Q_0^k
Elastic scattering (10^{-2} – 10^4) ^b	$\frac{0.2\epsilon^{1/2}}{0.025 + \epsilon^{1/2}}$	$-\frac{0.046}{0.01 + \epsilon}$	$\frac{0.7\epsilon}{65 + \epsilon}$	$\frac{0.61\epsilon^2}{2000 + \epsilon^2}$	$\frac{0.55\epsilon^2}{4000 + \epsilon^2}$
	$+\frac{1.2[16\epsilon^{1/2} + \epsilon]}{[100 + 16\epsilon^{1/2} + \epsilon]}$	$+\frac{1.04\epsilon(2400 + \epsilon)}{[60\,000 + 3000\epsilon + \epsilon^2]}$	$+\frac{0.36}{3000 + \epsilon}$	$+\frac{0.39\epsilon}{5000 + \epsilon}$	$+\frac{0.45\epsilon}{5000 + \epsilon}$
Resonant vibrational excitation (1.7–3.6)	0	$\frac{5}{7}$	0	$\frac{8}{7}$	0
$A^3\Sigma_u^+, B^3\Pi, W^3\Delta$ (7–50)	$-\frac{\epsilon^2}{1500 + \epsilon^2}$	$\frac{\epsilon^2}{10^4 + \epsilon^2}$	0 ^c	0	0
$C^3\Pi$ (1 I–50)	–0.2				
$a^1\Pi_g$ (8.5–500)	$\frac{200\,000 + 160\epsilon^2 + \epsilon^4}{200\,000 + 2600\epsilon^2 + \epsilon^4}$	$\frac{\epsilon^2}{1600 + \epsilon^2}$	$\frac{\epsilon^2}{3500 + \epsilon^2}$	$\frac{\epsilon^2}{3500 + \epsilon^2}$	$\frac{\epsilon^2}{3500 + \epsilon^2}$
“Sum of singlets” (13–500)	$\frac{\epsilon^2}{2500 + \epsilon^2}$	$\frac{\epsilon^2}{2500 + \epsilon^2}$	$\frac{\epsilon^2}{3500 + \epsilon^2}$	$\frac{\epsilon^2}{3500 + \epsilon^2}$	$\frac{3}{3500 + \epsilon^2}$

^aNote that from Fig. 3 we would now choose somewhat lower values for D in some cases. e.g., 1000 and 300 for $A^3\Sigma_u^+$, $B^3\Pi$, and $W^3\Delta$ and for $j = 1$ and 2, respectively.

^bThe numbers in parentheses give the energy range in eV of the data used to obtain the formulas in this table. Only in the cases of the singlet states are the formulas of significance outside these energy ranges.

^cFrom Fig. 3 we would now take this entry to be $-\epsilon^2(5000 + \epsilon^2)^{-1}$.

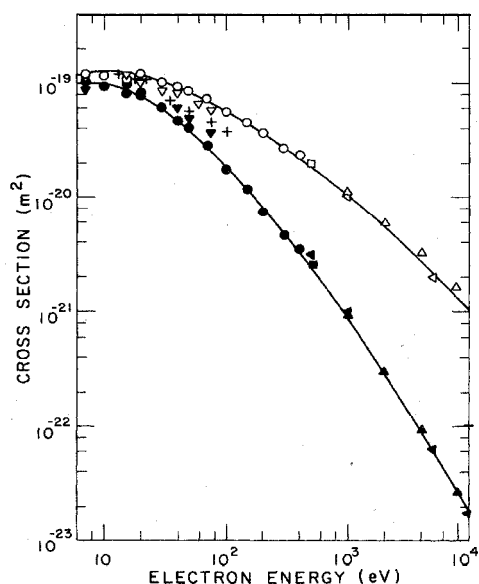
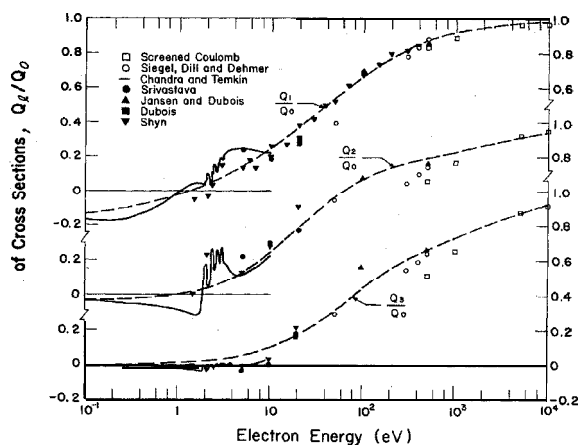


FIG. 1. Elastic scattering cross sections for N₂. The upper curve and the open points and crosses are the total cross sections, while the lower curve and the solid points are the momentum-transfer cross sections. Symbols and authors are the following: \square , \blacksquare , Bromberg (Ref. 23); +, Finn and Doering (Ref. 23); ∇ , \blacktriangledown , Srivastava et al. (Ref. 23); \circ , \bullet , Shyn and Carignan (Ref. 23); A, A, Riley et al. (Ref. 24); \triangleleft , \blacktriangleleft , Strickland et al. (Ref. 24).



higher-degree spherical-harmonic components of the cross sections Q_j^0 to the total elastic cross section Q_0^0 . In terms of these ratios and the tabulated values of Q_m^0 , the magnitude of the j th component of the elastic scattering cross section is given by

$$Q_j^0 = Q_m^0 \frac{Q_j^0}{Q_0^0} \left(1 - \frac{Q_1^0}{Q_0^0} \right)^{-1}. \quad (29)$$

Here we have suppressed the dependence on electron energy ϵ . The use of simple analytical approximations to the Q_j^k/Q_0^k values averages through the structure present, for example, near 2 eV and is justified by the relatively slowly varying electron energy distributions.

Figure 2 shows values of the ratios Q_1^0/Q_0^0 , Q_2^0/Q_0^0 , and Q_3^0/Q_0^0 determined from experiment^{23,28} and theory.^{24,29} In a few cases the theoretical results are available in a form equivalent to Q_j^0 , but in most cases it was necessary to resolve plotted differential scattering cross-section data into spherical-harmonic components with the associated uncertainties resulting from incomplete data at small and large scattering angles. The dashed curves of Fig. 2 show the values of the analytical expressions given in Table I and used to approximate the experimental and theoretical data. In view of the structure in the theoretical curves near 2 eV and the scatter in the higher-energy data shown in Fig. 2, it does not seem worthwhile to attempt better fits to the data at this time. As listed in Table I, we carried out the analysis to $j = 5$ in anticipation of six-term solutions of the Boltzmann equation. Of some concern to us is the tendency of the Q_j^0/Q_0^0 ratios to saturate at values well below unity at high-electron energies. This behavior may indicate a tendency of experiments and/or our analyses to inaccurately represent the forward scattering peak of the differential scattering cross section. Accordingly, we used a smooth interpolation between ratios determined from experimental data at energies below 300 eV and ratios determined from theory at energies above 5 keV.

B. Rotational and vibrational excitation

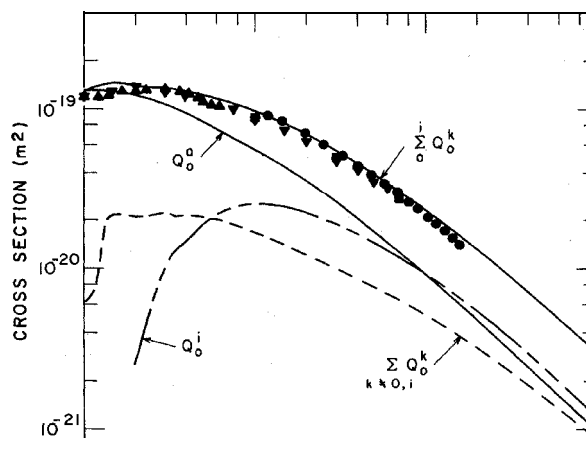
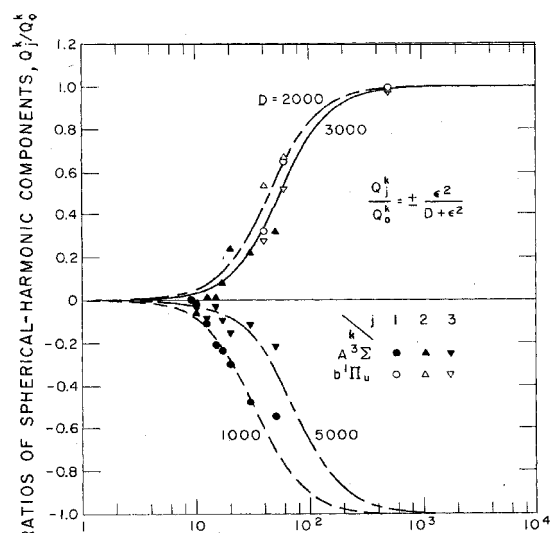
Electron-beam experiment³⁰ and theory^{29,31} have shown that rotational excitation cross sections are very large in the vicinity of 2 eV. Because of the shortage of cross-section data and in order to reduce the computer space required to store and use the very large number of energy-dependent rotational excitation cross sections in the resonance region, we have used the single-level approximation for the cross section for rotational excitation as derived from gas-heating data. See Appendix B. This cross section has a magnitude equal to the sum of the magnitudes of the vibrational excitation cross sections discussed next and is assigned an energy loss of 0.02 eV. Using this cross section we find that for the E/n considered in this paper the energy loss to resonance rotational excitation is as much as 11% of the input energy at mean electron energies near 0.6 eV. The rotational excitation was assumed to be isotropic, although now one could use the formulas of Wong and Dubé³⁰ for the resonance region.

The cross sections for electron excitation from the $v''=0$ vibrational level of $X^1\Sigma_g^+$ of N_2 to higher vibra-

tional levels ($v'=1$ to 8) of the $X^1\Sigma$ state come from a variety of sources. The cross section for excitation of the $v'=1$ level at electron energies below 1.7 eV is that found by EPR (Ref. 22) with a small modification near 1.5 eV to allow for resonance rotational excitation. For resonance excitation of the $v'=1$ to 8 levels at electron energies between 1.7 and 3.6 eV, we have used as a reference set the results of Schulz³² as normalized to Spence, Mauer, and Schulz³³ by Kieffer.⁴ Note that these resonance cross sec-

are based on electron beam data. Agreement between calculated and measured $C^3\Pi_u$ excitation coefficients¹⁸ and ionization coefficients¹ at $E/n < 300$ Td, was found¹⁸ by adjusting the magnitudes of these cross sections. We use factors of 2.0 for the $C^3\Pi_u$ state and 0.67 for all other triplet states.

The choice of cross sections for excitation of the important states at the higher-electron energies is based on the analyses of Zipf and McLaughlin⁴³ and of Jackman *et al.*⁴⁴ In particular, the Zipf and McLaughlin results have been used to construct the sum of cross sections for the excitations of the various $^1\Pi_u$ states, the $c'^1\Sigma_u^+$ and $b'^1\Sigma_u^+$ states, and unidentified states with energy losses of 15.8 and 17.3 eV.



sum of the excitation cross sections Q_0^k for all k not equal to 0 or i , and the total ionization cross section Q_0^i . The solid points show experimental values of the total cross section.⁵³ Note that the elastic, excitation, and ionization cross sections become comparable at the higher-electron energies and that the sum of these cross sections, as shown by the upper solid line, is equal to the total cross section to within the combined uncertainties. At electron energies above about 50 eV these curves agree reasonably well with the model of Jackman and Green.⁵² It is not clear which of the cross sections should be reduced in order to improve the agreement with the total cross-section measurements. The excitation cross sections would seem to be the most subject to error, although there is some disagreement among the measured ionization cross sections.⁴⁶

The principal improvements in the magnitudes and energy dependences of cross sections discussed in this paper from those of our previous work^{17,18,22} are (a) the significantly larger electronic excitation cross sections at high energies, (b) more accurate representations of electron-beam data for excitation of the $C^3\Pi_u$ and $E^3\Sigma_g^+$ states while retaining consistency with excitation coefficient data, and (c) the introduction of an effective resonance rotational excitation cross section. We have not attempted to optimize the cross sections by comparison with transport and excitation coefficient data, although we will show in Sec. V that the agreement of calculated and experimental swarm data is generally good.

IV. CALCULATED TRANSPORT COEFFICIENTS AND DISTRIBUTION FUNCTIONS

A. Effects of anisotropic scattering

The transport and reaction-rate coefficients calculated using the procedures of Sec. II and the cross sections of Sec. III were used to obtain the data of this section. As an example of these results, the calculated spherically symmetric components of the electron energy distributions $f_0(\epsilon)$ for $E/n = 1500$ Td are shown in Fig. 5. Starting from the uppermost curve at high energies the curves show the calculations for six-term anisotropic, six-term isotropic, two-term anisotropic, and two-term isotropic models. We see that at high energies, $\epsilon > 80$ eV, the anisotropic models yield about 20% more high-energy electrons than do the isotropic models. However, at these high energies the largest change is the result of including more terms in the spherical-harmonic expansion, as will be discussed in Sec. IV B. Note that for the two-term approximation the fact that the elastic momentum-transfer cross section was kept constant means that the changes observed in the distribution functions as the result of the change from isotropic to anisotropic scattering are due to the effects of the dipole ($j = 1$) components of the inelastic scattering. The somewhat larger change in $f_0(\epsilon)$ observed when changing the scattering model with the six-term approximation includes the effects of higher-degree components of the elastic and inelastic scattering. Although not readily seen in Fig. 5, the curves of $f_0(\epsilon)$ for the anisotropic and isotropic models cross such that at energies

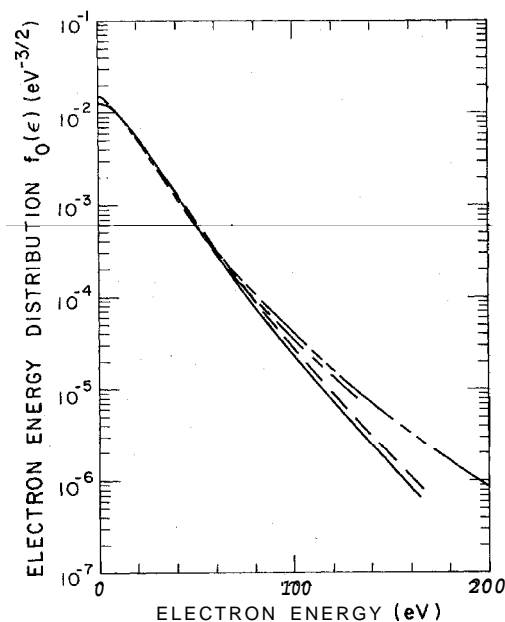


FIG. 5. Spherically symmetric component of electron energy distribution functions showing effects of anisotropic scattering

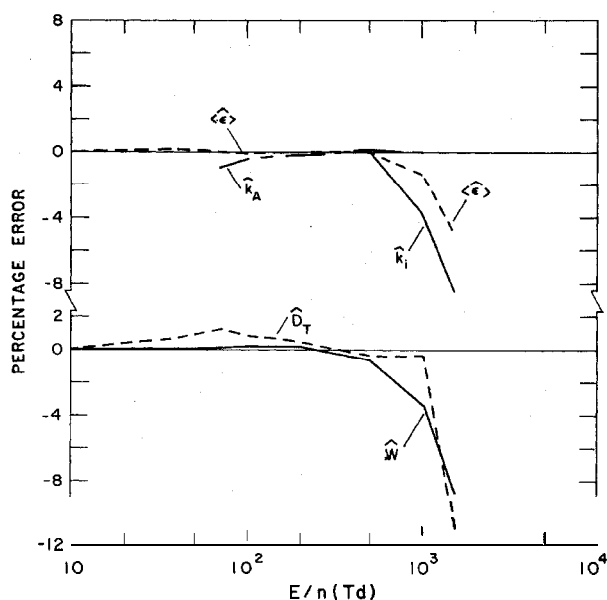


FIG. 6. Percentage change in transport and reaction coefficients caused by change from isotropic to anisotropic scattering models. Coefficients and symbols are the following: $\langle \epsilon \rangle$, average energy; \hat{k}_A , triplet excitation; \hat{k}_i , ionization; \hat{D}_T , transverse diffusion; \hat{W} , drift velocity. The lines simply connect calculated values.

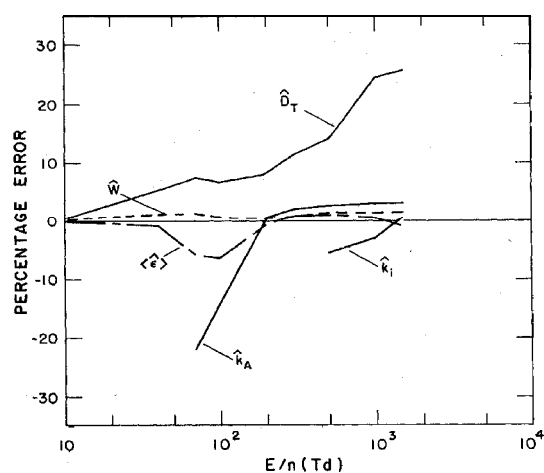
curves of Fig. 6 show that most of the errors increase rapidly with E/n . The large difference in the errors for the triplet excitation coefficient and the ionization coefficient at high E/n occurs because of the crossover of the electron energy distributions with and without anisotropic scattering shown in Fig. 5 and because the triplet excitation cross-sections peak at energies near the crossover while the ionization cross section peaks at energies above the crossover. The higher mean electron energy found with anisotropic scattering is consistent with the larger number of high-energy electrons shown in Fig. 5, while the higher-electron drift velocity is consistent with a lower contribution of inelastic scattering to the momentum-transfer cross section in this case. The much larger transverse diffusion coefficient D_T for the anisotropic model at 1500 Td is at first surprising in view of the decrease in D_T/ν found by Kiiciikarpaci and Lucas⁹ for N₂ and by Haddad *et al.*⁵ for H₂. However, it is accompanied by a much larger increase in D_L so that the relative increases are as expected. We have not shown the errors for the longitudinal diffusion coefficient since questions have been raised^{5,4} regarding the accuracy of the multiterm calculations of D_L for N₂ and because of erratic results obtained in the present calculations. Unfortunately, the issue is clouded by the question of runaway effects, i.e., Kunhardt and Tzeng⁹ find that their set of anisotropic cross sections results in the failure of the calculated diffusion coefficients to reach steady-state at $E/n = 1500$ Td as required for the validity of the hydrodynamic model. At lower E/n their results show changes with the scattering model which are similar to ours in sign but usually larger in magnitude. An important question here is the precision of their Monte Carlo calculations. The changes

in W and $\langle \epsilon \rangle$ shown in Fig. 6 are significantly smaller than those calculated by Kiiciikarpaci and Lucas⁹ using less accurate differential scattering data.

B. Effects of two-term approximation

In the course of the present calculations we have extended the comparisons of results obtained using the two-term and six-term spherical-harmonic expansions to higher E/n for electrons in N₂. Representative results for the electron energy distributions at high E/n (1500 Td) are shown in Fig. 5, where at high energies the lower pair of curves were calculated using the two-term approximation and the upper pair were calculated using the six-term approximation. The relatively large change in the numbers of high-energy electrons is ascribed to the failure of the two-term spherical-harmonic approximation when the ratio of the inelastic cross section to the elastic cross section is large. This tendency toward streaming in the direction of the acceleration is in part due to decreasing inelastic cross sections with increasing electron energy at high energies. We have verified the relation between large fractional excitation cross sections and the error in $f_0(\epsilon)$ for the two-term approximation using model cross sections and a simplified Boltzmann equation.⁶ Note that for the high E/n of Fig. 5 the energy gained per elastic collision by low-energy electrons is about 10 eV.

The results of the calculations of the errors in transport and rate coefficients for the two-term approximation relative to the six-term approximation are summarized in Fig. 7. These calculations are for the anisotropic model. At low and moderate E/n these results are similar to those of I for isotropic scattering. Particularly noticeable are the increasing errors in the triplet excitation coefficients and the ionization coefficients as E/n decreases. These errors are caused by the systematic deficiency in high-energy electrons for the two-term approximation in the presence of large inelastic cross sections.^{2,6} At high E/n the increase in the error in the calculated transverse dif-



E/n . The solid curves show the results of the calculations made using the anisotropic cross section set discussed in Sec. III

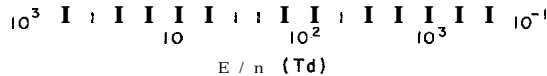


FIG. 8. Electron drift velocity W and characteristic energy K versus E/n for N_2 . The anisotropic cross section set from Sec. III and the six-term solution of the Boltzmann equation were used to calculate the solid curves. The isotropic inelastic set and the two-term approximation were used to obtain the dashed curves. The points show selected experimental data. Symbols and authors are the following: \bullet Lowke (Ref. 55); \triangle , Fletcher and Reid (Ref. 55); ∇ , Warren and Parker (Ref. 56); \blacksquare , Crompton and Sutton (Ref. 56); $+$, Schlumbohm (Ref. 55); \times , Kontoleon *et al.* (Ref. 56); ∇ , Roznerski and Leja (Ref. 56).

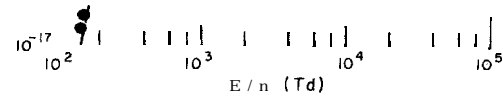


FIG. 9. Comparison of theoretical and experimental temporal growth constants v/n for the electron density in N_2 at high E/n . The calculated curves are designated as in Fig. 8. The points show experimental data with the following symbols and authors: \bullet , Frommhold (Ref. 57); $+$, Dibbem (Ref. 57); \triangle , Felsenthal and Proud (Ref. 57).

and the six-term spherical-harmonic expansion, while the dashed curves show the results of calculations made using the isotropic inelastic model and the two-term spherical-harmonic expansion form discussed in Sec. III. The points are from experimental data selected as the most accurate and/or recently available data in the various E/n ranges.

For E/n below 100 Td there is generally good agreement between our calculations and experimental⁵⁵ time-of-flight drift velocities shown in Fig. 8. It must be kept in mind that our drift velocities are calculated without the ionization corrections given by the second term in Eq. (5) and the resultant iteration implied by Eq. (10). From the calculations of Taniguchi *et al.*²⁶ we expect that our calculated drift velocities will be lower than the experimental time-of-flight velocities when ionization effects are important. Thus at E/n near 400 Td in Fig. 8, the two-term results of Taniguchi *et al.* and our approximate calculations show that the second term of Eq. (5) increases the drift velocity by about 10% and so improves agreement with experiment. However, the lower experimental values observed at higher E/n are not predicted²⁶ by the theory of Sec. II and may be the result of inapplicability of the hydrodynamic model or of the difficulties of measurement of such high drift velocities. Because of the common application of the two-term, isotropic inelastic model, we show the results of this model by the dashed curve. As is also indicated by Figs. 5 and 6 this model is

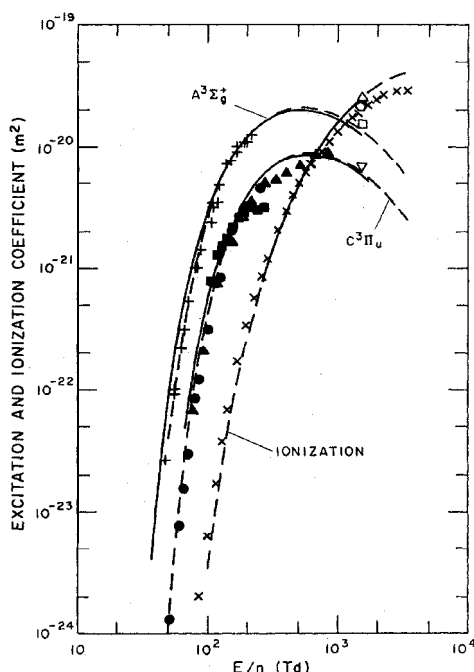


FIG. 10. Comparison of theoretical and experimental spatial excitation and ionization coefficients for electrons in N₂. The curves are calculated as in Fig. 8. The symbols and authors for the selected experimental data are the following: +, Levron and Phelps (Ref. 17); A, Legler (Ref. 18); ●, Tachibana and Phelps (Ref. 18); ■, [unclear]

to measurements of current ratios in a drift tube with a restricted area cathode.¹³ The comparison in Fig. 8 of calculated values of characteristic energy shows reasonable agreement with experiment⁵⁶ at E/n below 40 Td. For E/n between 40 and 200 Td the comparison is made difficult by the differences among experiments and the large uncertainties assigned to some of the data. This is unfortunate since it is this E/n range which is important in comparisons of measured excitation and ionization coefficients, such as shown in Figs. 9 and 10. Also noticeable are the differences in the ϵ_K values calculated using the isotropic cross-section set and the two-term expansion versus the anisotropic set with the six-term spherical-harmonic expansion. As shown in Sec. IV most of the differences arise from the use of the different spherical-harmonic approximations. The decrease in the measured ϵ_K values below our calculated values at E/n above 300 Td may be caused by our failure to include ionization effects in the calculations for the mobility and transverse diffusion coefficient or may be caused by departures from the hydrodynamic limit. The values of ϵ_K for high E/n calcu-

lated by Taniguchi *et al.*²⁶ using the two-term approximation including the ionization correction of Eq. (7) lie between the curves of Fig. 8.

The temporal growth constant ν found by the solution of the Eq. (9) describes the growth of electron density when the effects of electron-density gradients are negligible as when observing the spatially integrated growth of light output or of electron current in a pulsed Townsend experiment^{11,16,57} or the growth of ionization in magnetically confined discharge with a toroidal electric field.⁵⁸ In Fig. 9 we compare measured and calculated temporal growth constants. At the lower E/n the experimental values lie somewhat above the two-term, isotropic calculations as expected from the systematic errors in the two-term calculations.⁶ At high E/n the fact that the experimental results are too high may be the result of difficulties of interpretation of the measurements.⁵⁷ Runaway effects, if present, would be expected to reduce the temporal growth constant below the maximum value of the ionization rate coefficient for monoenergetic electrons indicated by the chain line.

Since most measurements of excitation and ionization coefficients are made using the steady-state drift-tube technique, Fig. 10 shows calculated and measured spatial growth coefficients as a function of E/n . The normalized spatial ionization coefficients α_i/n shown by the smooth curves of Fig. 10 were calculated from the values of ν and W neglecting the $\alpha_i^2 D_L$ term in Eq. (13). The α_i/n values calculated using Eq. (13) are higher by as much as a factor of 1.6 at $E/n = 1500$ Td. In order to test these simplified calculations of the spatial growth constant α_i/n , we have solved Eqs. (17)–(21) with $\nu/n = 0$, $m = 2$, and isotropic scattering for $E/n = 1500$ Td. This result is indicated by the open circle in Fig. 10 and is to be compared with the result obtained with $\alpha_i = \nu/W$ as indicated by the open triangle. The ratio of 1.2 is indicative of the expected difference between the solid curve of Fig. 10 and the exact calculation. These comparisons show that there is a cancellation of the effects of the increase in α_i/n caused by the $(\alpha_i/n)^2(nD_L)$ and higher terms in Eq. (13) and the decrease in α_i/n caused by the increase in W resulting from the contribution of ionization in Eq. (5). The agreement between calculated and measured⁵⁹ ionization coefficients for E/n less than 1000 Td is good since, as in our previous analysis,^{*} most of the electronic excitation cross sections were adjusted, i.e., reduced by a factor of $\frac{2}{3}$ from electron-beam results, in order to obtain the fit. The agreement at higher E/n is also good when one takes into account the correction indicated by the open points. Similar agreement has been found by Taniguchi *et al.*²⁶

Figure 10 also compares calculated and measured spatial excitation coefficients for the $A^3\Sigma_u^+$ and $C^3\Pi_u$ states. The measured excitation coefficients for the $A^3\Sigma_u^+$ state"

using the two-term isotropic model (dashed curve) and experiment, but that at low E/n the values calculated using the anisotropic six-term model (solid curve) model are too high. As shown in Sec. IV the difference is largely due to errors introduced by the two-term model rather than to errors from the isotropic scattering assumption. The better fit for the two-term model is the result of the adjustment of the magnitude of the vibrational excitation cross sections so as to fit the excitation coefficient data rather than to fit transport data. These trade-offs have been examined previously¹⁷ using the two-term approximation. We note that because of the higher excitation coefficients obtained with the six-term approximation, the scaling factor which multiplies the reference set of resonance vibrational cross sections could be raised from 1.5 to 1.9 so that the calculated $A^3\Sigma_u^+$ excitation coefficients fit the experimental values. Evidence for such large vibrational excitation cross sections has been obtained by Schulz⁶⁰ using electron-beam techniques. However, this change results in ϵ_K values which are about 3% below the values shown in Fig. 8. This seems outside the spread of the experimental results and the discrepancy found earlier¹⁷ is unresolved. (See Note added in *proof*.) The effect of the spatial gradient on the calculated spatial excitation coefficient at 1500 Td, as calculated using Eq. (16) with Q_0^i replaced by the sum of the triplet Q_0^k values, is indicated by the difference between the dashed curve and the open square. At this E/n the excitation rate coefficient k_{kss} is relatively insensitive to changes in the electron-density distribution caused by the density gradient²⁶ since the mean electron energy is about equal to the energy of the cross-section maximum. Therefore, the higher α_d/n value is caused principally by the lower value of the V_d used in the analog to Eq. (16) compared to the value of W used to calculate the smooth curves.

Figure 10 shows curves of the spatial excitation coefficients for the $C^3\Pi_u$ state as calculated using the same procedure as for the $A^3\Sigma$ state. Here we include the small contribution of cascading from the $E^3\Sigma_g^+$ state. The experimental data from several experiments are shown by the points. Although the calculated curves for $C^3\Pi_u$ excitation agrees in shape with the results¹⁸ of Tachibana and Phelps and the lower E/n data of Legler, it is higher by 1.3 to 1.9 for low E/n , depending on the technique for solving the Boltzmann equation. This difference between calculation and experiment could be removed by lowering the cross sections for $C^3\Pi_u$ and $E^3\Sigma_g^+$ excitation. On the other hand, the lower E/n data of Urošević *et al.*¹⁸ lies above the calculation at the lower E/n and below it at the higher E/n . The effect of the spatial gradient on the calculated excitation coefficient at 1500 Td is indicated by the difference between the dashed curve and the inverted open triangle. Judging from this calculation and from the calculated corrections to the excitation coefficients for spatial gradient and ionization effects of Taniguchi *et al.*,²⁶ we cannot explain the deviations from the E/n dependence of our calculations observed in the data of Legler and of Urošević *et al.* at high E/n . Note that the excitation coefficients calculated²⁶ for the $C^3\Pi_u$ state at high E/n by Taniguchi *et al.* and by Tagashira *et al.* are much lower than ours for $E/n \geq 200$ Td.

VI. DISCUSSION

The results of this paper show that when the electron Boltzmann equation is solved using realistic approximations to the differential cross sections for electron scattering by N_2 the transport and reaction coefficients for moderate and low E/n (< 500 Td) differ by less than 2% from those in which the inelastic scattering is assumed isotropic and the elastic momentum transfer or diffusion cross section is unchanged. At high E/n (> 500 Td) the importance of anisotropic scattering increases with E/n . We have shown that for most transport and reaction coefficients it is more important to allow for the anisotropy of the electron energy distribution beyond the dipole term than to include anisotropic scattering. For the analysis of precision measurements it is necessary to include anisotropic scattering and to use more than two-terms in the spherical-harmonic expansion of the electron energy distribution. Also of considerable importance at high E/n are the corrections to the transport and reaction coefficients arising from the combined effects of electron-density gradients and ionization. In the course of this work we have improved the set of collision cross sections for N_2 at high energies and, through the inclusion of resonance rotational excitation, at low energies.

In this paper we have followed the lead of Blevin¹⁵ by minimizing the number of transport and reaction coefficients calculated for comparison with experiment. We have also attempted to maintain a clear distinction between coefficients calculated using various approximations to the Boltzmann equation and coefficients derived from experimental data. Thus, application of the density gradient expansion to the Boltzmann equation in the hydrodynamic limit yields a rate equation for the electron density and a differential equation for each of what we have called the density gradient expansion transport and reaction coefficients. Appropriate solutions of the rate equation for the electron density can be used to analyze experiments performed using time-of-flight techniques or current growth experiments in time or space. The transport coefficients determined from analyses of these experiments are labeled by the experimental technique and can be compared with the calculated density gradient expansion coefficients. For the analysis of 'parallel-plane, steady-state, or time-integrated experiments the assumption of an exponential spatial dependence for the electron energy distribution and the electron density reduces significantly the number of velocity-dependent differential equations to be solved. The hydrodynamic spatial growth constant obtained from the solution of this equation can be compared directly with that obtained from experiment. The spatial growth constant can be also calculated from the density gradient expansion transport coefficients when the growth constant is small enough.

There is satisfactory to good agreement between calculated and measured transport coefficients for E/n less than about 100 Td. At higher E/n the difference between calculations and experiment become large, e.g., 30%. At the present time it is not clear whether the source of the discrepancy is in the approximations we used in solving the Boltzmann equation, in the cross section set, or in the

experimental data. From the theoretical side it is important that better techniques be developed for predicting the behavior of electrons at higher E/n where departures from the hydrodynamic model become important. Techniques discussed in the present paper are limited to those for solution of the Boltzmann equation when perturbations caused by spatial gradients are small or when the spatial or temporal gradients can be expressed as an exponential growth. It is important to define the range of validity of these techniques by solving the Boltzmann equation without restrictions as to the variation in space or time. At present such solutions are usually obtained by the Monte Carlo simulation method,^{8,9,54} although a few authors⁶¹ have discussed other approaches. Such techniques would also be useful in the treatment of problems where the initial conditions or boundary conditions exert a large influence on the solution, e.g., the backscattering or injected electrons at moderate and low E/n .

The extrapolation of the present results to other gases is difficult because of our lack of knowledge of the range of anisotropic scattering parameters. For example, for H₂ it appears¹⁴ that anisotropic scattering effects are small at low and moderate E/n , but that they may well be larger than for N₂ at high E/n because of indications^{25,62} of greater anisotropy and a more rapid decrease in $Q_m(\epsilon)$ with ϵ at high electron energies for H₂ than for N₂. Anisotropic scattering at low energies and low E/n may well be of considerable importance in gases, such as H₂O and other polar gases, where the highly anisotropic inelastic rotational excitation cross section is dominant.⁶³

$$Q_m^k(\epsilon) = Q_0^k(\epsilon) - (\epsilon + \epsilon_k) Q_1^k(\epsilon + \epsilon_k) f_1(\epsilon + \epsilon_k) / \epsilon f_1(\epsilon) .$$

$$\min \approx 3\pi a_0^2 \frac{\mathcal{R}\epsilon_i}{\epsilon^2} \left| \frac{z_{0n}}{a_0} \right|^2 ,$$

Note Added in Proof.

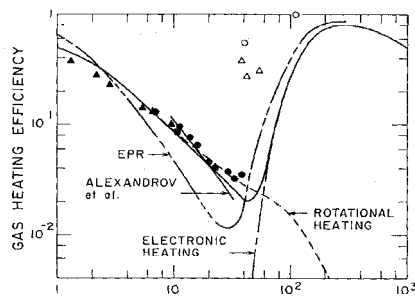
$$+ \frac{8\pi^2}{m} \int_0^\pi$$

where θ and θ_s are the polar angles measured from the direction of acceleration and the direction of the incident electron, respectively; $\theta' = \theta - \theta_s$; and

$$\mathcal{Q}_m^k(\epsilon) = Q_0^k(\epsilon) - \left[\frac{(\epsilon - \epsilon_k)}{\epsilon} \right]^{1/2} Q_1^k(\epsilon). \quad (\text{A4})$$

The square-root factor in Eq. (A4) is particularly important near the excitation threshold. Applying the simplified Born approximation used in Eq. (A2), \mathcal{Q}_m^k

$$\mathcal{Q}_m^k(\epsilon) \approx 2\pi a_0^2 \frac{\mathcal{R}\epsilon_i}{\epsilon^2} \left| \frac{z_{0n}}{a_0} \right|^2 \ln \left[\frac{4\epsilon}{\epsilon_k} \right].$$



this model and the experimental measurements. A number of attempts⁷⁵ have been made to explain positive-column temperature-rise data. Some of the factors considered

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