Aust. J. Phys., 1984, **37**, 23–34

Model Calculations of Negative Differential Conductivity in Gases

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Abstract

Negative differential conductivity in gases has been studied using simple models of elastic and inelastic collision cross sections for electron scattering. The use of such models has demonstrated features of the cross sections that lead to the phenomenon, and shown that it can occur without a Ramsauer–Townsend minimum (and even without a sharply rising momentum-transfer cross section) or a special combination of inelastic cross sections.

1. Introduction

Negative differential conductivity (NDC), that is, decreasing electron drift velocity with increasing electric field strength, occurs in semiconductors and gases. It has received considerable attention because, on the one hand, a number of applications are dependent on it, while on the other it can cause undesirable instabilities. The role of NDC in gas discharge physics has been stressed by Lopantseva *et al.* (1979) (see also Petrushevich and Starostin 1981), who made both experimental and theoretical studies of instabilities in externally sustained discharges in Ar–N_2 and Ar–CO mixtures and in pure Ar. Investigations of the phenomenon are particularly important in relation to the operation of Ar–N_2 lasers (Searles 1974; Ault *et al.* 1974; Ault 1975; Bychkov *et al.* 1980), CO lasers (Willett 1974; Garscadden 1981) and diffuse discharge switches (Christophorou *et al.* 1982; Schoenbach *et al.* 1982), and to the detection of nuclear radiation (Mathieson and El-Hakeem 1979; Al-Dargazelli *et al.* 1981).

In the gas phase, NDC has been most commonly observed in very dilute mixtures of molecular gases with argon. There were some early reports of NDC in *pure* argon but it is now known that these were the result of the presence of molecular impurities (Long *et al.* 1976, and references therein) and it is well established that NDC is not present in the pure gas (Robertson 1977). The phenomenon has also been observed experimentally and/or predicted 'theoretically' in a number of other gas mixtures and even in pure gases; Table 1 lists some examples.

Almost all investigations of NDC relate it to the presence of a Ramsauer–Townsend minimum in the electron momentum-transfer cross section σ_m, which is exhibited by

0004-9506/84/010023$02.00

some atomic and molecular gases, and one or more inelastic energy loss processes in the region of that minimum. This situation may be present in a single molecular gas (e.g. methane) or it may be produced by adding a small quantity of a molecular gas to a heavy monatomic gas.

A more general situation that arises when small quantities of molecular gas are added to an atomic gas is that of enhanced electron conductivity, that is the value of the drift velocity for the mixture exceeds the values for both constituent gases at the same E/N (Long *et al.* 1976; Garscadden *et al.* 1981; Foreman *et al.* 1981).

Table 1. Occurrence of NDC in various pure gases and gas mixtures

Reference	Theory	Experiment
Pack and Phelps (1961)		N_2 (77 K)
Pack *et al.* (1962)		CO (77 K)
Lowke (1963)		N_2 (77 K)
Hurst *et al.* (1963)		H_2O–CH_4, H_2O–N_2
Klots and Reinhardt (1970)		Various hydrocarbons
Christophorou (1971)		Data and references for various gases
Long *et al.* (1976)	Ar–N_2, Ar–CO	
Kleban and Davis (1977)	CH_4	
Kleban and Davis (1978)	CH_4, CD_4	
El-Hakeem and Mathieson (1978)		Ar–CH_4
Mathieson and El-Hakeem (1979)	CH_4, Ne–CH_4, Ar–CH_4, Ar–CO_2	
Lopantseva *et al.* (1979)		Ar–N_2, Ar–CO
Lin *et al.* (1979)	CH_4	
Elford (1980)		Hg (due to presence of mercury dimers)
Foreman *et al.* (1981)		Ar–CH_4, He–CH_4
Kleban *et al.* (1981)	He–CH_4, Ar–CH_4	
Garscadden (1981)	CO–Ar–He	
Christophorou *et al.* (1982)		Ar–C_3F_8, Ar–CF_4
Haddad (1983*a*)		Ar–N_2
Haddad and Milloy (1983)		Ar–CO
Haddad (1984)		CH_4

Explanations of two examples of NDC in gases have recently been given, one by Long *et al.* (1976) and the other by Kleban and Davis (1977, 1978). Long *et al.* based their argument on the variation with E/N of the mean collision frequency for momentum transfer $\langle \nu_{\rm m} \rangle$, while Kleban and Davis were concerned with the effect of the degree of anisotropy of the electron velocity distribution function. Both groups of authors dealt with the situation when a Ramsauer–Townsend minimum is present.

The aim of the present paper is to point out the importance of certain features of the elastic and inelastic collision cross sections in inducing NDC. Model calculations have been used to show that the presence of a Ramsauer–Townsend minimum is not a necessary condition for NDC to occur, and to clarify further the proposed explanations. We believe that one can get a much clearer physical picture of the primary cause of NDC when one uses simple model cross sections rather than the cross sections for real gases.

2. Theoretical Description

The physical situation that leads to NDC for low electric fields can be understood in general terms in the following way. The well-known formula for the drift velocity in terms of the electron speed c (based on the so-called 'two-term approximation'),

$$v_{\mathrm{dr}} = -\frac{4\pi}{3}\frac{eE}{m}\int_0^\infty \frac{c^3}{\nu_{\mathrm{m}}(c)}\frac{\mathrm{d}f_0}{\mathrm{d}c}\,\mathrm{d}c\,,$$

can be transformed by partial integration to

$$v_{\mathrm{dr}} = \frac{eE}{3mN}\langle c^{-2}\frac{\mathrm{d}}{\mathrm{d}c}\frac{c^2}{\sigma_{\mathrm{m}}(c)}\rangle\,,$$

provided that $c^3 f_0(c)/\nu_{\mathrm{m}}(c) = 0$ in the limit $c \to 0$ or $c \to \infty$ (Huxley and Crompton 1974). In these formulae $\sigma_{\mathrm{m}}(c)$ and $\nu_{\mathrm{m}}(c) = N\sigma_{\mathrm{m}}(c)c$ are the energy-dependent momentum-transfer cross section and momentum-transfer collision frequency respectively. It follows that provided the cross section does not vary too rapidly the formula for v_{dr} reduces to

$$v_{\mathrm{dr}} = FeE/m\langle\nu_{\mathrm{m}}\rangle\,, \tag{1}$$

where F is a factor near unity (e.g. $F = 0\cdot85$ if σ_{m} is constant), which is constant or varies slowly with E/N (Huxley and Crompton 1974; see also Long *et al.* 1976; Lin *et al.* 1979).

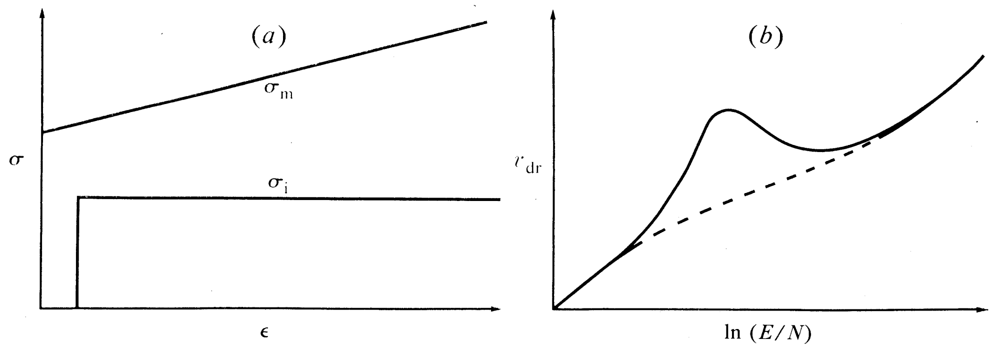

Fig. 1. Simple model cross sections (*a*) and the corresponding drift velocity (*b*) used to illustrate the primary cause of negative differential conductivity. The dashed curve in (*b*) shows the drift velocity that would be expected if there was no inelastic scattering.

We use equation (1) to predict qualitatively the variation of the drift velocity with E/N for the simple situation illustrated in Fig. 1. For low values of E/N essentially all the electrons have energies below the inelastic threshold. Therefore, because elastic scattering is the only energy loss process, the mean electron energy and $\langle\nu_{\mathrm{m}}\rangle$ rise relatively rapidly with increasing E/N, and v_{dr} increases only slowly. At a sufficiently large value of E/N a significant fraction of the electrons in the swarm have energies above the threshold. There is now a much larger average energy loss per collision, the average energy and $\langle\nu_{\mathrm{m}}\rangle$ increase less rapidly with E/N, and v_{dr} therefore increases more rapidly than it would if there was no inelastic channel (see equation 1). This is illustrated in Fig. 1*b*. At very large values of E/N, elastic scattering again becomes the dominant energy loss process, since the average energy loss per elastic collision increases linearly with electron energy, whereas the energy loss per inelastic collision remains constant, so that the drift velocity must approach asymptotically the dashed curve which corresponds to the case of no inelastic scattering. At intermediate values of E/N there is at least the possibility that v_{dr} will decrease with increasing E/N, as shown in Fig. 1b. Whether or not this actually occurs depends on the combination of elastic and inelastic cross sections and the threshold energy of the inelastic process.

Explanations of NDC as observed in specific cases have been given by several authors. Kleban and Davis (1977, 1978) discussed the phenomenon in terms of the degree of anisotropy of the velocity distribution. They considered gases such as methane where the threshold of vibrational excitation coincides with a Ramsauer–Townsend minimum in σ_m. In the range of values of E/N where the maximum in the distribution of electron speeds is somewhat above that corresponding to the excitation threshold, the average electron energy is kept relatively low by the inelastic collisions even though E/N is relatively large. Moreover, because σ_m is small the frequency of elastic collisions, whose effect is to randomize the velocity vectors without significantly reducing their magnitude, is small. Under these conditions the distribution function may become markedly anisotropic, a condition which Kleban and Davis described as 'streaming anisotropy'. At higher values of E/N, where the distribution of electron speeds has its maximum in the region where σ_m is large, there is enhanced elastic scattering, especially of those electrons whose motion is predominantly in the direction of the electric force and whose velocities are therefore largest. The consequence is enhanced randomization of the velocities and reduced anisotropy. Thus, although the average electron *speed* is increased, the average *velocity* (the drift velocity) may be reduced, i.e. NDC may occur.

While affording a new insight into the nature of the phenomenon, the Kleban and Davis description does not provide the basis for a criterion for its occurrence, whereas the preceding argument and equation (1) can provide at least an approximate criterion (see Section 3).

A somewhat different approach was taken by Long *et al.* (1976) who based their argument on equation (1) but implied that NDC occurs in a transition region where the mean energy of the swarm lies between two inelastic processes with widely separated thresholds.

Our argument presented at the beginning of this section suggests that NDC has a simpler explanation than that given by Long *et al.*, and the model calculations described in Section 3 support this view.

Lopantseva *et al.* (1979) also based their discussion on equation (1) and used a relatively simple argument to develop from it a criterion for NDC. This equation predicts that NDC will occur when $\langle v_m \rangle$ increases more rapidly than E (see Section 3), but does not say anything about the characteristics of the elastic and inelastic cross section(s) that will lead to this situation. Following Lopantseva *et al.*, we write an approximate energy balance equation in the form*

$$\langle v_i \rangle \varepsilon_i = v_{dr} eE, \tag{2}$$

* Lopantseva *et al.* (1979) have taken the energy loss per collision to be the characteristic energy ε_k. Consequently their NDC criterion differs from equation (3).

where $\langle \nu_i \rangle$ is the mean inelastic collision frequency, ε_i is the energy loss per inelastic collision and, for the sake of simplicity in this discussion, the energy loss in elastic scattering is neglected. Combining equations (1) and (2) we get*

$$\frac{\partial v_{\mathrm{dr}}}{\partial(E/N)} = \frac{F\varepsilon_i}{2mv_{\mathrm{dr}}} \frac{\langle \nu_i \rangle}{\langle \nu_m \rangle} \left(\frac{1}{\langle \nu_i \rangle} \frac{\partial \langle \nu_i \rangle}{\partial(E/N)} - \frac{1}{\langle \nu_m \rangle} \frac{\partial \langle \nu_m \rangle}{\partial(E/N)} \right), \tag{3}$$

where it is assumed that $F(E/N)$ is constant.

While the many approximations contained in the derivation of equation (3) make it far from adequate in a realistic situation, nevertheless it is useful as a guide to the relationship between elastic and inelastic processes that leads to NDC. It is clear, for example, that a rapid increase of $\langle \nu_m \rangle$ or decrease of $\langle \nu_i \rangle$ with E/N will induce a negative slope of v_{dr} versus E/N, i.e. an NDC.

Finally, we note that a more exact criterion is derived by Robson (1984; present issue p. 35) using momentum-transfer theory. His approach is more general than those of Long *et al.* (1976) and Lopantseva *et al.* (1979).

3. Model Calculations

We have performed a number of calculations using model cross sections to explore the conclusions of the previous section. One conclusion was that a Ramsauer–Townsend minimum in σ_m was not a necessary condition for NDC, although it has almost always been taken to be so in the literature. Our models were constructed with the intention of verifying this conclusion as well as investigating more generally the features of the elastic and inelastic cross sections that lead to the phenomenon. Our choice of the models was guided by the general arguments given in the previous section.

Model 1. We chose for the first model a momentum-transfer cross section that linearly increases with energy and is given by

$$\sigma_m = 5 + 1{\cdot}95\,\varepsilon \tag{4a}$$

(all cross sections are in 10^{-16} cm^2 when the energy ε is given in eV), and an inelastic cross section that is constant above a threshold energy ε_T, i.e.

$$\sigma_i = 0 \qquad (\varepsilon < \varepsilon_T) \tag{4b}$$

$$= 0{\cdot}03 \;\; (\varepsilon \geqslant \varepsilon_T). \tag{4c}$$

This model corresponds to that on which the discussion at the beginning of Section 2 is based. The threshold energy is the parameter in this model and was given the values 0·05, 0·1 and 0·2 eV. The inelastic energy loss was taken to be equal to ε_T. Room

* Inclusion in equation (2) of energy losses in elastic collisions would result in an additional term always greater than zero (Robson 1984) on the right-hand side of equation (3). This term would compete with terms that can be negative, thus reducing the range of situations where NDC occurs. Equation (3) predicts that NDC occurs whenever $d(\nu_i/\nu_m)/d(E/N) < 0$, which is not true (see e.g. the results for Model 5).

temperature and a molecular mass of 28 a.m.u. were assumed. Drift velocities were calculated using the usual two-term spherical harmonics representation of the velocity distribution function.

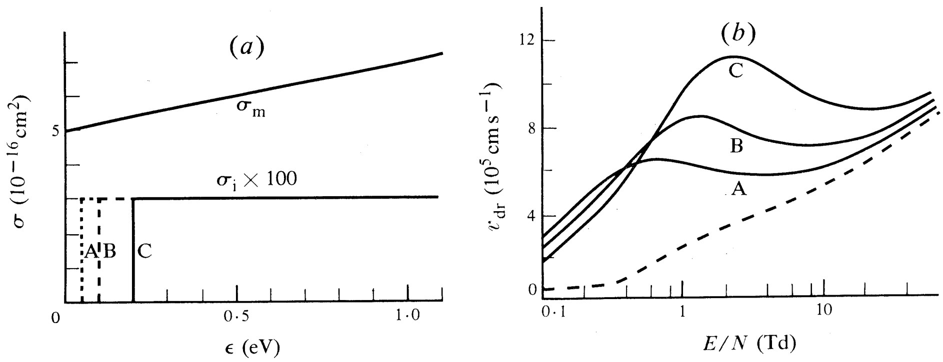

Fig. 2. Model 1: (*a*) Cross sections, where the inelastic thresholds are A, $\varepsilon_T = 0\cdot05$ eV; B, $\varepsilon_T = 0\cdot1$ eV; C, $\varepsilon_T = 0\cdot2$ eV. (*b*) Calculated drift velocities, where the dashed curve shows results for the case when only the elastic process is present. (1 Td $\equiv 10^{-21}$ V m^2.)

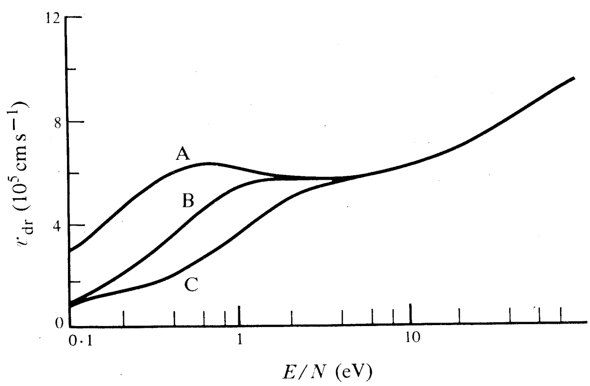

Fig. 3. Calculated drift velocities for Model 2 with the threshold energy values: A, $\varepsilon_T = 0\cdot05$ eV; B, $\varepsilon_T = 0\cdot3$ eV; C, $\varepsilon_T = 1$ eV. In each case the inelastic energy loss is $\varepsilon_i = 0\cdot05$ eV.

The results are shown in Fig. 2. As predicted, NDC occurs even with cross sections as unspectacular as those for this model and where there is no Ramsauer–Townsend minimum. Note that as ε_T increases NDC is postponed to higher E/N, but the effect is much more pronounced. This is because the larger inelastic losses in the region of maximum loss further suppress the mean electron energy, leading to a reduction in $\langle v_m \rangle$ and increase in v_{dr}. Since the drift velocities converge at high E/N, such an increase must lead to enhanced NDC.

Model 2. Here we chose σ_m and σ_i to have the same form as in Model 1, but kept the energy loss per collision ε_i the same in all cases ($0\cdot05$ eV per collision). The threshold energies for the inelastic process were $0\cdot05$, $0\cdot3$ and $1\cdot0$ eV. The results are shown in Fig. 3. As the difference between ε_i and ε_T increases, NDC becomes less pronounced until, in case C, the dependence of v_{dr} on E/N becomes monotonic.

This is because the delay in the onset of inelastic scattering allows the mean electron energy to rise to a point where the elastic energy losses become comparable with the inelastic losses, so that the situation is little different from that for pure elastic scattering where NDC cannot occur.

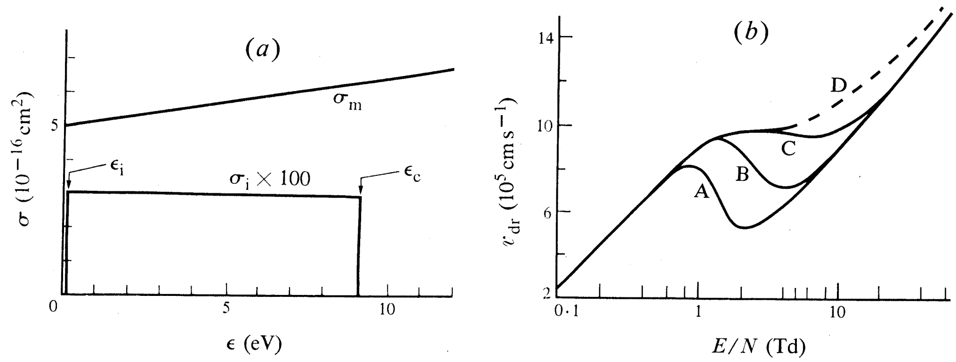

Fig. 4. Model 3: (*a*) Cross sections, where the inelastic cross section cutoffs have the values A, $\varepsilon_c = 1$ eV; B, $\varepsilon_c = 3$ eV; C, $\varepsilon_c = 9$ eV (illustrated); D, $\varepsilon_c = \infty$. In each case $\varepsilon_i = \varepsilon_T = 0{\cdot}1$ eV. (*b*) Calculated drift velocities, where the dashed curve for D shows the case of no cutoff.

Model 3. With the third model we set out to show how a sudden decrease of σ_i induces NDC (see equation 3). Initially the inelastic cross secuion of Model 1, case B, was used and the slope of σ_m reduced (to $0{\cdot}15$) until no NDC was observed (case D in Fig. 4*b*). Then σ_i was set to zero above different 'cutoff' energies ε_c. The set of cross sections (in 10^{-16} cm^2) is

$$\sigma_m = 5 + 0{\cdot}15\,\varepsilon, \tag{5a}$$

$$\sigma_i = 0 \qquad (\varepsilon < 0{\cdot}1\text{ eV};\quad \varepsilon > \varepsilon_c) \tag{5b}$$

$$= 0{\cdot}03\ (0{\cdot}1\text{ eV} \leqslant \varepsilon \leqslant \varepsilon_c), \tag{5c}$$

where the parameter ε_c has the values $1{\cdot}0$, $3{\cdot}0$, $9{\cdot}0$ eV and ∞, and $\varepsilon_i = 0{\cdot}1$ eV. The results shown in Fig. 4 demonstrate that NDC can be induced by reducing the cutoff energy, and that it becomes more pronounced as ε_c is further reduced. This is evidently due to the fact that above ε_c a situation is rapidly reached, as E/N increases, when inelastic scattering is unimportant; that is, one reaches the asymptotic region discussed under Model 1 much more rapidly. Correspondingly, NDC can occur with a momentum-transfer cross section that increases much less rapidly with energy when the inelastic cross section has this form.

Model 4. It was evident from earlier work that NDC is enhanced when σ_m increases rapidly with energy. We illustrate this by using the following model cross sections:

$$\sigma_m = 5 + A\varepsilon, \tag{6a}$$

$$\sigma_i = 0 \qquad (\varepsilon < 0{\cdot}1\text{ eV}) \tag{6b}$$

$$= 0{\cdot}03\ (\varepsilon \geqslant 0{\cdot}1\text{ eV}), \tag{6c}$$

where A is a parameter taking the values $9{\cdot}95$, $0{\cdot}45$ and 0. The results presented

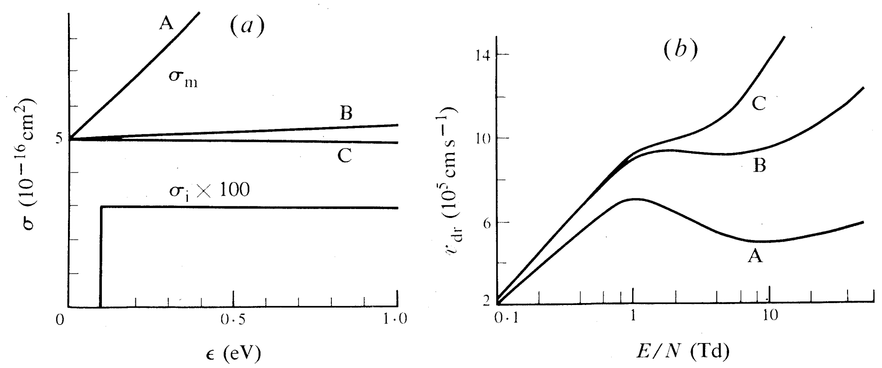

Fig. 5. Model 4: (*a*) Cross sections, where the slopes of σ_m have the values A, $A = 9{\cdot}95$; B, $A = 0{\cdot}45$; C, $A = 0$. (*b*) Calculated drift velocities.

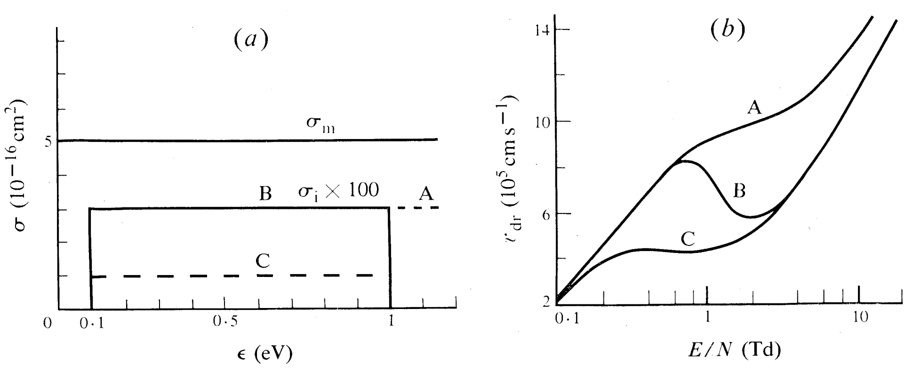

Fig. 6. Model 5 (see text): (*a*) Cross sections. (*b*) Calculated drift velocities.

in Fig. 5 not only show that NDC is promoted by a steeply rising σ_m, but also that the phenomenon can occur with an energy dependence that is much weaker than that usually found on the high energy side of a Ramsauer–Townsend minimum. The model also shows that if σ_m increases too slowly with energy the reduced anisotropy of the velocity distribution function as E/N increases will be more than offset by the increased average speed of the electrons, thus eliminating NDC.

Model 5. Finally, we demonstrate a perhaps unexpected result that NDC can occur even when σ_m is not increasing. Our model has the following characteristics

$$\sigma_m = 5, \qquad \sigma_i = 0 \quad (\varepsilon < 0{\cdot}1)\,; \tag{7a, b}$$

$$\text{case A:} \quad \sigma_i = 0{\cdot}03 \quad (\varepsilon \geqslant 0{\cdot}1)\,; \tag{7c}$$

$$\text{case B:} \quad = 0{\cdot}03 \quad (0{\cdot}1 \leqslant \varepsilon \leqslant 1{\cdot}0); \quad \sigma_i = 0 \quad (\varepsilon > 1{\cdot}0)\,; \tag{7d}$$

$$\text{case C:} \quad = 0{\cdot}01 \quad (0{\cdot}1 \leqslant \varepsilon \leqslant 1{\cdot}0); \quad = 0 \quad (\varepsilon > 1{\cdot}0)\,. \tag{7e}$$

As shown in Fig. 6, case B shows that NDC can in fact occur in this situation. Note that a cutoff in σ_i is essential for NDC to occur (compare curves A and B in Fig. 6*b*), and that this cross section must be sufficiently large (compare curves B and C). Both conditions are necessary to ensure a sufficiently rapid decrease in anisotropy with increasing E/N.

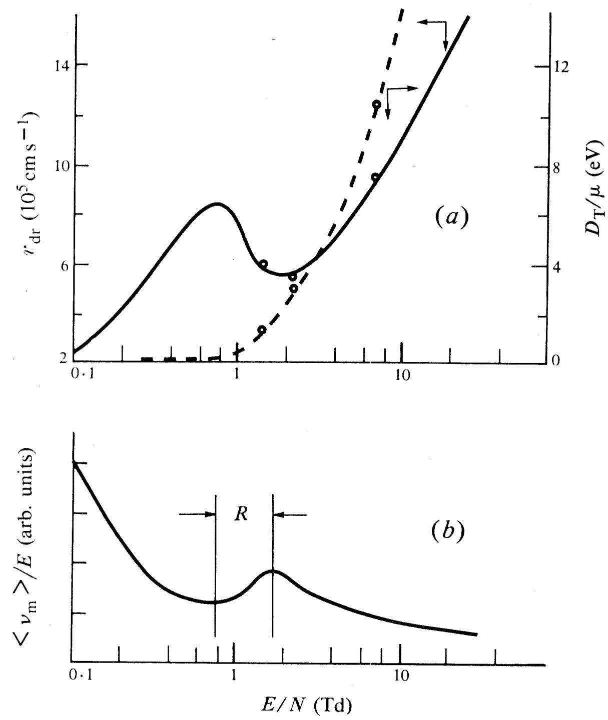

Fig. 7. Model 5, case B: (*a*) Calculated drift velocities (solid curve) and the ratio D_T/μ (dashed curve). The multiterm calculations for both v_{dr} and D_T/μ are represented by circles on the corresponding curves. (*b*) The E/N dependence of $\langle \nu_m \rangle/E$. The region where $\langle \nu_m \rangle$ rises more rapidly than E, and therefore NDC occurs, is indicated by R.

4. General Comments about Model Calculations

The results presented in the previous section were obtained using the Boltzmann code developed by Gibson (1970). In order to determine the significance of errors arising from the so-called two-term approximation inherent in the solution of Boltzmann's equation, upon which Gibson's code is based (Holstein 1946), we have recalculated v_{dr} using a multiterm code (Lin *et al.* 1979; see also Haddad *et al.* 1981) for the typical case of Model 5, case B. The results are compared with the two-term results in Fig. 7*a*. Fig. 7*a* also shows results for D_T/μ, the ratio of transverse diffusion coefficient to mobility, which is more subject to error from this approximation. The errors arising from the approximation are negligible in each case, a result which is not surprising given the ratio of the elastic and inelastic cross sections (Reid 1979). The validity of the two-term approximation in this instance also shows that the degree of anisotropy required for NDC to occur need not be large (Kleban and Davis 1977).

We have also used Gibson's (1970) code to calculate $\langle \nu_m \rangle$ and plotted the ratio $\langle \nu_m \rangle/E$ in Fig. 7*b*. The range of E/N where $\langle \nu_m \rangle$ increases more rapidly than E (denoted R) corresponds to the region of NDC, in accordance with equation (1), and illustrates the adequacy of the explanations based on this formula despite the approximations inherent in its derivation.

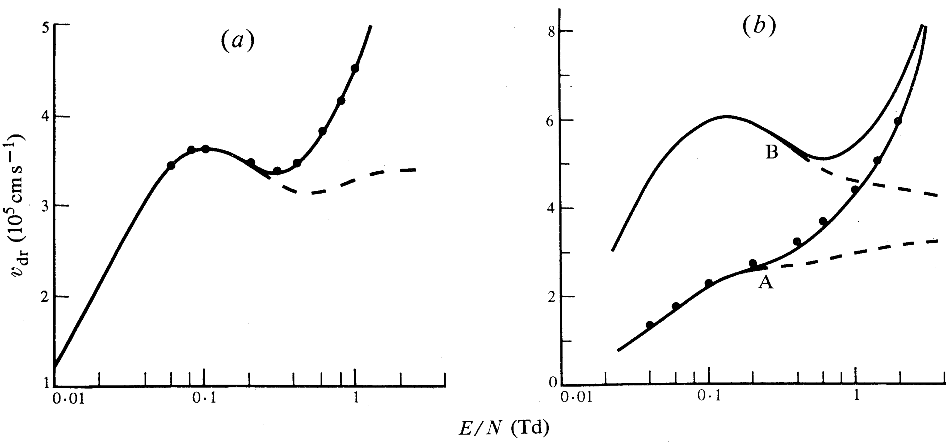

Fig. 8. Drift velocities in nitrogen at low values of E/N for (*a*) $T = 77$ K and (*b*) $T = 293$ K. The solid curves correspond to the inclusion of all the relevant processes, and dashed curves to the omission of vibrational excitation. The points are the experimental results of Lowke (1963). In (*b*) curves A and B correspond respectively to when superelastic processes are included and omitted.

5. NDC in Real Gases

Most studies of NDC have concentrated on gases or gas mixtures in which the threshold or thresholds for vibrational excitation lie near or above a Ramsauer–Townsend minimum. Consequently such conditions are generally taken to be necessary for the phenomenon to occur. However, its occurrence in nitrogen and carbon monoxide at 77 K for small values of E/N (Pack and Phelps 1961; Pack *et al.* 1962; Lowke 1963) shows that a Ramsauer–Townsend minimum is not necessary, and also suggests that rotational rather than vibrational excitation may be the relevant inelastic process in these instances. Moreover, in the case of CO, where σ_m *decreases* with increasing energy in the relevant range of swarm energies, the fact that NDC occurs confirms one of the conclusions reached with the models, namely that σ_m need not increase with energy for the effect to occur.

From an examination of experimental data for v_{dr} and D_T/μ in N_2, it seems to us unlikely that vibrational excitation is responsible for NDC in the 77 K data for this gas. We investigated this point more fully using the two-term Boltzmann code referred to in the previous section, together with rotational excitation cross sections calculated using the Gerjuoy and Stein (1955) formula and the momentum-transfer and vibrational excitation cross sections of Pitchford and Phelps (1982). By suppressing the vibrational cross sections we could show that the phenomenon is entirely due to rotational excitation (see Fig. 8*a*); in fact their removal considerably increases the range of E/N over which NDC occurs without affecting its onset. Thus, in this case the presence of a second inelastic process reduces NDC rather than promotes it (Long *et al.* 1976).

In real gases superelastic collisions with rotationally excited molecules also have a considerable influence on NDC. In N_2 at 293 K, NDC is not observed experimentally, and our calculations show that it does not occur even when the vibrational cross sections are suppressed (see Fig. 8*b*). Thus, its disappearance cannot be accounted for by the reduction in the energy gap between the threshold for rotational excitation of the most populated state and the lowest vibrational threshold that results from the higher temperature. On the other hand, as shown by curve B in Fig. 8*b*, the suppression of the superelastic cross sections restores the phenomenon at this temperature even when vibrational losses are included.

The energy dependence of σ_m in the relevant energy range is clearly less favourable for NDC in CO than it is in N_2. On the other hand, the decrease in the rotational excitation cross section at higher energies, which is characteristic of polar molecules (Takayanagi 1966), favours NDC so that our discussion of the phenomenon for N_2 is expected to be valid for CO also.

6. Conclusions

From the general arguments developed in this paper and the illustrations provided by the model calculations the following conclusions can be drawn:

(1) NDC cannot take place in the absence of an inelastic process (Robson 1984).

(2) The more rapid the increase of the momentum-transfer cross section with energy, the more likely is NDC to occur.

(3) An inelastic cross section that rapidly decreases with increasing energy enhances NDC or may even produce it under otherwise unfavourable conditions; for example, when the momentum-transfer cross section does not increase rapidly with energy. It has been shown, for example, that it is possible to induce NDC with a constant momentum-transfer cross section.

(4) The relative magnitude of the elastic and inelastic cross sections plays a key role, but it is not necessary to have such a low elastic cross section as is usually encountered at a Ramsauer–Townsend minimum. NDC can occur when the inelastic cross section is quite small compared with the elastic cross section, that is, under conditions for which the degree of anisotropy is never very large and a two-term spherical harmonics representation of the velocity distribution function is valid. On the other hand, NDC may fail to occur even if there is an apparently appropriate combination of characteristics of the elastic and inelastic cross sections (e.g. rapidly increasing momentum-transfer cross section, rapidly decreasing inelastic cross section etc.).

Finally we note that all the results we have presented can be explained using arguments developed by Kleban and Davis (1977), unless by their use of their term 'streaming anisotropy' they are implying that NDC is associated in this instance (CH_4) with the breakdown of the two-term approximation. Similarly, our results are consistent with the argument developed by Long *et al.* (1976). However, we note that the presence of a second inelastic process is not necessary for NDC to occur, although in certain 'realistic' situations the second process could play an important role if the thresholds of the two processes are close to each other. Its role would then be to suppress or reduce NDC produced by the first process rather than promote it. Most of all, it is interesting to note that all our results and conclusions are consistent with the criterion developed by Robson (1984) independently of our work. However, the essential part of our paper is the demonstration of the fact that NDC may occur in a much wider set of circumstances than has previously been discussed, and that its underlying causes may be simply conceptualized.

Acknowledgments

The authors wish to thank Drs R. E. Robson and M. T. Elford for some very useful comments on the work in general and on the manuscript. One of us (Z.Lj.P.) is grateful to the Australian National University for the provision of a postgraduate scholarship.

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Manuscript received 2 June, accepted 10 October 1983