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Theory of the density dependence of electron drift velocity in gases

G. L. Braglia and V. Dallacasa
Istituto di Fisica dell'Università, 43100 Parma, Italy
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We present a quantum-mechanical theory of the electron drift speed W in a gas in a field at low E/N and relatively high pressures. It is able to give a quantitative interpretation of the experimental results, as regards the dependence of W on both the gas number density and the gas temperature. As an application, we make a comparison between theory and experiment in He, H_2, N_2, and Ar.

I. INTRODUCTION

According to the conventional theory, the electron drift speed W in a gas in a dc electric field is a function only of the ratio E/N between the field strength E and the number density N.[1] But this theory is unable to account for the experimental results at low E/N and high pressures,[1-5] where the drift velocity has been found to be a function of N at constant E/N.[1,6-8] The first attempt to explain this density dependence was made by Frommhold,[2] who suggested that the phenomenon was owing to the electrons spending part of their transit time as temporary negative ions. However, the existence of the required states for resonance capture has never been confirmed either by theory or by experiments.[1] Moreover, Frommhold's theory is unable to explain the results relevant to monoatomic gases.[6] An alternative theoretical approach, proposed by Legler[3] (who treats the background gas as a continuum) explains some results at extremely high pressures, but does not appear to account for the majority of the results,[6] particularly at low densities.[1] In this paper we present a theory which is able to explain quantitatively the experimental data. It is quantum mechanical in character and particularly appropriate to the range of pressures commonly explored in practice. Contrary to Legler's theory, which is also quantum mechanical, our theory is (i) more appropriate at low (rather than high) densities, when the pressure effect begins to appear and (ii) able to explain the dependence of the drift velocity on both the gas number density N and the gas temperature T. As an application, we make a comparison between theory and experiments in He, H_2, N_2, and Ar.

II. THEORY

At sufficiently low values of E/N, the drift velocity is a linear function of E/N at any given N.[1] This happens when the electrons (mass m, charge e) have an energy distribution practically independent of E, i.e., when they are in almost thermal equilibrium with the background gas (of atoms or molecules of mass M). In this paper we are interested mainly in these values of E/N since it is in this region that the density dependence of the drift velocity is more evident. Since the highest pressures we are concerned with are around 3×10^4 Torr at $T = 293$ K, in gases with cross sections of the order of $(1-5)\times 10^{-20}$ m^2, one sees that the lower mean free paths are of an order around 2×10^{-8} m. The electron energy which corresponds to a wavelength $\lambda(\epsilon)$ of this order is about 4×10^{-3} eV. But the mean electron energies of interest are around 10^{-2} eV, so that only a small fraction of electrons have a wavelength λ of the order of the mean free path $l(\epsilon)$. It is in this respect that our theory differs from that of Legler,[3] who develops a theory where $\lambda \approx l$ for almost all the electrons.

Since E/N is so small that W is a linear function of E/N, we start from the following equation for the drift velocity (deduced from the Kubo formula[9]):

$$W = \lim_{\eta \to 0} \frac{eE}{3} \int_0^\infty dt \int_0^\beta d\lambda \langle \vec{v}(-i\hbar\lambda)\cdot \vec{v}(t)\rangle e^{-\eta t} \qquad (1)$$

where $\vec{v}(t)$ indicates electron velocity at time t, $\beta = 1/k_B T$, and the angular brackets are used to indicate the velocity autocorrelation function. Equation (1) presumes that the system is translationally invariant, which is true since our system is spatially homogeneous. When using this property and assuming that the atom number density is not too high (in order that, on the average, the electron mean free path is long with respect to the thermal wavelength) one can give Eq. (1) the standard approximate form[10]

$$W = \frac{e\hbar^3 E}{12\pi^4 m^2} \int_{-\infty}^{+\infty} d\epsilon \left(-\frac{df(\epsilon)}{d\epsilon}\right) \times \int_{\vec{k}} d\vec{k}\, k^2 [\mathrm{Im}\, G(\vec{k},\epsilon)]^2 , \qquad (2)$$

where

$$G(\vec{k},\epsilon) = 1/(\epsilon - \hbar^2 k^2/2m - \Sigma)$$

is the Green function [$\Sigma=\Sigma(\vec{k},\epsilon)$ is the self-energy which, in general, will depend on the energy ϵ and the wave vector $\vec{k}$ of the electron]. In Eq. (2), $f(\epsilon)$ is the distribution of the electron energy. Since we are assuming that the electrons are in almost thermal equilibrium, we are allowed to put $f(\epsilon)=Ce^{-\beta\epsilon}$, C being the normalization constant. But we may also assume that the scattering processes are isotropic,[11,12] from which it follows that Σ is $\vec{k}$ independent. Then, since we neglect atom-atom correlations, we have $\Sigma(\epsilon)=Nt(\epsilon)$, where $t(\epsilon)$ is the t matrix on the energy shell relevant to a single scattering center (atom or molecule). The consequence of this simplification is that, after some manipulations, Eq. (2) can be given the form

$$W=\frac{1}{3}\frac{eE}{m}\frac{\beta}{N}C\hbar \times\int_{\epsilon_0}^{\infty}d\epsilon\, e^{-\beta\epsilon}\frac{1}{\operatorname{Im}t(\epsilon)}[\epsilon-N\operatorname{Re}t(\epsilon)]^{3/2}\,, \qquad (3)$$

where ϵ_0 is the solution of the equation $\epsilon_0-N\operatorname{Re}t(\epsilon_0)=0$ if the solution is positive, otherwise $\epsilon_0=0$. Thus, by virtue of the optical theorem

$$\operatorname{Im}t(\epsilon)=\frac{\hbar}{\sqrt{2m}}[\epsilon-N\operatorname{Re}t(\epsilon)]^{1/2}Q(\epsilon)$$

[$Q(\epsilon)$ is the electron cross section], we can write

$$W=\frac{\sqrt{2}}{3}\frac{eE}{\sqrt{m}}\frac{\beta}{N}C\int_{\epsilon_0}^{\infty}d\epsilon\,\frac{e^{-\beta\epsilon}}{Q(\epsilon)}[\epsilon-N\operatorname{Re}t(\epsilon)]\,. \qquad (4)$$

As one can see, if we indicate by W_0 the conventional drift velocity, Eq. (4) says that $W\rightarrow W_0$ in the limit of low values of N, at fixed E/N.

In order to obtain W from Eq. (4) it is necessary to give an explicit expression of $\operatorname{Re}t(\epsilon)$. To this end, we make use of the fact that[13] $dN\operatorname{Re}t(\epsilon)/d\epsilon\rightarrow 1$ when $\bar{\lambda}(\epsilon)/l(\epsilon)\rightarrow 1$, i.e., as $\epsilon\rightarrow\epsilon_c$ where ϵ_c is defined by the equation $\bar{\lambda}(\epsilon_c)/l(\epsilon_c)=1$. From this it follows that for energies sufficiently close to ϵ_c one can assume that

$$\frac{dN\operatorname{Re}t(\epsilon)}{d\epsilon}\simeq\frac{\bar{\lambda}(\epsilon)}{l(\epsilon)}\,. \qquad (5)$$

In the following we shall assume that $dN\operatorname{Re}t(\epsilon)/d\epsilon=\bar{\lambda}(\epsilon)/l(\epsilon)$ for all the (small) electron energies where the conventional theory of the drift velocity fails.

Let us suppose first that, in the limit of low electron energies, $Q(\epsilon)$ is weakly dependent on ϵ. From Eq. (5) it follows immediately that

$$\operatorname{Re}t(\epsilon)\simeq\operatorname{Re}t(0)+\hbar\left(\frac{2\epsilon}{m}\right)^{1/2}Q(\epsilon)\,. \qquad (6)$$

According to this equation, since we consider only first-order corrections (with respect to N) to the drift velocity W, ϵ_0 must be approximated by $N\operatorname{Re}t(0)$, so if $\operatorname{Re}t(0)>0$ introduction of Eq. (6) into Eq. (4) leads one to write, after some algebra,[14]

$$W=\frac{2\sqrt{2}}{3}\frac{eE}{N}\left(\frac{\beta^5}{\pi m}\right)^{1/2} \times\int_0^{\infty}d\epsilon\, e^{-\beta\epsilon}\frac{\epsilon}{Q(\epsilon)}\left(1-2\frac{\bar{\lambda}(\epsilon)}{l(\epsilon)}\right)\,, \qquad (7)$$

where $\bar{\lambda}(\epsilon)=\hbar/\sqrt{2m\epsilon}$ and $l(\epsilon)=1/NQ(\epsilon)$. As one can see, the corrective term to the conventional equation for the drift speed involves the ratio $\bar{\lambda}(\epsilon)/l(\epsilon)$ so that the correction is no longer negligible when the condition for the applicability of classical mechanics begins to fail. It is essential to observe that this result is substantially different from that of Legler[3] who is led to assume that $\operatorname{Re}t(\epsilon)\simeq\operatorname{Re}t(0)$ while our corrective term in Eq. (7) comes from the second term of the right-hand side of Eq. (6). In other words, contrary to Legler, we find a result which is appropriate in the semiclassical limit, in accord with our original assumption that $\bar{\lambda}\ll l$ for a large fraction of the electrons.

If we perform the integration in Eq. (7), we find that

$$W=W_0(1-\alpha b)\,, \qquad (8)$$

where

$$\alpha=\sqrt{2\pi}\,\frac{\sqrt{mk_BT}}{\hbar}a \quad\text{and}\quad b=2\pi\frac{\hbar^2}{m}\frac{Na}{k_BT}\,.$$

As regards the parameter a it must be noted that, in order to write the last member of Eq. (8), we take advantage of the fact that $Q(\epsilon)$ is slowly varying with ϵ, to put $Q(\epsilon)=\overline{Q}=4\pi a^2$ (constant). Therefore, a is the scattering length if $\overline{Q}=Q(0)$, in which case b is the parameter introduced by Legler.[3]

The above results are expected to represent correctly the density dependence of the drift velocity in gases such as He, H_2, N_2 where the cross section is almost constant for small energies and the scattering length $a>0$, since this implies that

$$\epsilon_0\propto\operatorname{Re}t(0)=2\pi\hbar^2a/m>0\,.$$

But Eqs. (7) and (8) are expected to fail in a gas such as Ar, where the cross section is strongly dependent on ϵ for small electron energies and the scattering length a is negative, which means that $\operatorname{Re}t(0)<0$, i.e., $\epsilon_0=0$. In this case Eq. (4) yields

$$W=\frac{\sqrt{2}}{3}\,\frac{eE}{\sqrt{m}}\,\frac{C}{N}\int_0^\infty d\epsilon\,\frac{e^{-\beta\epsilon}}{Q(\epsilon)}\,[\epsilon-N\mathrm{Re}t(0)-NA(\epsilon)]\,, \tag{9}$$

where $A(\epsilon)\equiv\mathrm{Re}t(\epsilon)-\mathrm{Re}t(0)$. On the other hand, the quantity $\mathrm{Re}t(0)$ is the dominant part of $\mathrm{Re}t(\epsilon)$ for the small energies of our interest, so that $A(\epsilon)\ll\mathrm{Re}t(0)$. We have made a numerical analysis to test the relative importance of $\mathrm{Re}t(0)$ and $A(\epsilon)$ in Ar and have found that $A(\epsilon)$ is about a power of 10 lower than $\mathrm{Re}t(0)$ so that we are allowed to assume $\mathrm{Re}t(\epsilon)\simeq\mathrm{Re}t(0)$. As a consequence in a gas such as Ar we expect that the drift velocity is well represented by the equation

$$W=\frac{\sqrt{2}}{3}\,\frac{eE}{\sqrt{m}}\,\frac{C}{N}\int_0^\infty d\epsilon\,\frac{e^{-\beta\epsilon}}{Q(\epsilon)}\,[\epsilon-N\mathrm{Re}t(0)]\,. \tag{10}$$

Thus, in this case, we find a corrective term which differs from that of Legler only by a factor of 2 and the presence of $Q(\epsilon)$ instead of $Q(0)$.

All the above results are expected to be correct only in the limit of very low E/N. In fact it is necessary that the electrons are in almost thermal equilibrium with the background gas, in accord with the fact that the electron energy distribution in Eqs. (7) and (10) is the Maxwellian distribution at the gas temperature T. In this limit the drift velocity is a linear function of E/N. But if we increase the value of E/N the energy distribution ceases to be a Maxwellian distribution. If the inelastic processes can be neglected, it becomes a Davydov distribution[1]

$$F(\epsilon)=c\exp\left(-\int_0^\epsilon\frac{\alpha(\epsilon)d\epsilon}{1+\alpha(\epsilon)k_BT}\right)\,, \tag{11}$$

where c is the normalization constant and

$$\alpha(\epsilon)=3\,\frac{m}{M}\left(\frac{m}{e}\right)^2\left(\frac{N}{E}\,Q(\epsilon)\right)^2\epsilon\,. \tag{12}$$

As mentioned,[12] $Q(\epsilon)$ is the momentum-transfer cross section. Simultaneously, the drift velocity ceases to be a linear function of E/N and, in general, this happens at values of E/N so low that the density dependence of the drift velocity at fixed E/N is still of the same order of magnitude as of that which we have in the limit $E/N\to 0$.[6] Therefore it is interesting to see, at this point, how one can represent the density dependence of the drift velocity also in the transition region from very low E/N, where this density dependence is more evident, to high E/N, where it disappears.

The extension of Eqs. (7) and (10) is rather natural when observing that at high E/N the drift velocity must be[1]

$$W=W_0=-\left(\frac{2}{m}\right)^{1/2}\frac{eE}{3N}\int_0^\infty\frac{\epsilon}{Q(\epsilon)}\,\frac{dF(\epsilon)}{d\epsilon}\,d\epsilon\,, \tag{13}$$

while at low E/N, from Eqs. (7) and (10), W is of the form

$$W=-\left(\frac{2}{m}\right)^{1/2}\frac{eE}{3N}\,C\int_0^\infty\frac{\epsilon}{Q(\epsilon)}\,\frac{de^{-\beta\epsilon}}{d\epsilon}\,[1-B(\epsilon)]\,d\epsilon \tag{14}$$

with the meaning of $B(\epsilon)$ being obvious. Then, one is naturally led to assume that the general equation for the drift velocity is

$$W=-\left(\frac{2}{m}\right)^{1/2}\frac{eE}{3N}\int_0^\infty\frac{\epsilon}{Q^*(\epsilon)}\,\frac{dF(\epsilon)}{d\epsilon}\,d\epsilon\,, \tag{15}$$

$$Q^*(\epsilon)=Q(\epsilon)/[1-B(\epsilon)]\,. \tag{16}$$

Using $Q^*(\epsilon)$ in place of $Q(\epsilon)$ in the expression (12) of $\alpha(\epsilon)$ is not expected to have an important effect as the weight of $B(\epsilon)$ is non-negligible only at small electron energies. In fact, we have had a numerical confirmation of this. However, if we want to be logically consistent, we must also replace $Q(\epsilon)$ by $Q^*(\epsilon)$ in Eq. (12). This substitution is easily seen to be required by the balancing momentum equation. Therefore, when the effect of elastic collisions dominate, as a general formula for the drift velocity, one is allowed to use the result of the conventional theory, i.e., Eqs. (11)–(13), with only the substitution of $Q^*(\epsilon)$ for $Q(\epsilon)$. Of course, we must expect that this extension will probably be applicable only to monoatomic gases. In molecular gases, rotational and vibrational excitations can make the use of Eq. (11) invalid for any E/N. In these cases, our theory is expected to account correctly for experimental results only in the limit $E/N\to 0$, i.e., for E/N so small that $F(\epsilon)$ is a Maxwellian distribution with practically the same temperature of the background gas. Under this condition the inelastic collisions are involved by the equation for W in a negligible way through the cross section Q of Eq. (13) (which is intended to be the total momentum-transfer cross section). In this limit, our theory does not indicate any difference between the results for $W-W_0$ in gases such as H_2 and N_2, if E and T are fixed. But, if E/N is not sufficiently small, if the thresholds for rotational and vibrational excitations are not very small, and, finally, if also T is not very small (for instance when $T=293$ K) we must expect that the corrective term in a molecular gas will be soon influenced by inelastic processes in a non-negligible way. Differences between experimental results relevant, for instance, to n-H_2, p-H_2, or D_2 may be still consistent, under these conditions, with our theory. However, a theoretical analysis of the effect of the inelastic collisions goes beyond the purpose of this paper. The situation is unsatisfactory even from the experimental point of view. So, we shall limit ourselves to point out that in the limit $E/N \to 0$ (and for not too high pressures) a large number of the experimental results presently available, can be explained simply by a quantum theory such as that developed above. Of course, our treatment does not exclude that for certain gases other effects, such as resonance trapping in molecular gases or polarization phenomena in highly polarizable gases, may also be important. In these cases the simple approach given here is clearly insufficient. However, it is worth noting that all these effects do not seem to be dominant for the gases, E/N, and pressures we have taken into consideration.

III. RESULTS

The density dependence of the drift velocity has been studied in a number of gases.[1,6] We have made a comparison of our theory with the experimental data relevant to the most common and most studied gases. In particular, we have considered gases such as He, H_2, N_2 for which the scattering length is positive and the cross section is almost constant in the limit of small electron energies (cf. Fig. 1). Moreover, we have considered Ar as an example of a gas where the scattering length is negative and the cross section is strongly dependent on the electron energy (cf. Fig. 2).

In Fig. 3 we present some of the results obtained

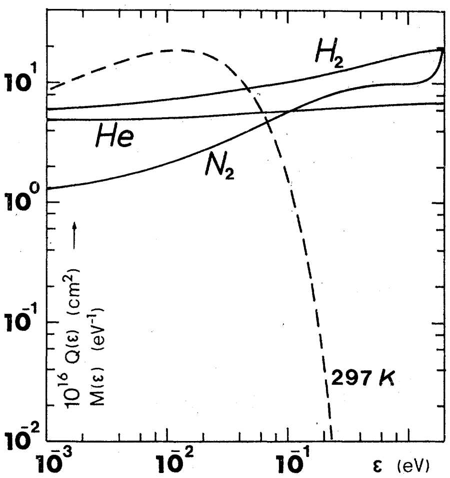

FIG. 1. Momentum-transfer cross sections of electrons in He, H_2, and N_2 (Refs. 1 and 15). As one can see, the cross sections are weakly dependent on ϵ over the entire energy interval of our interest, where the Maxwellian distribution $M(\epsilon)$ (broken curve) assumes non-negligible values.

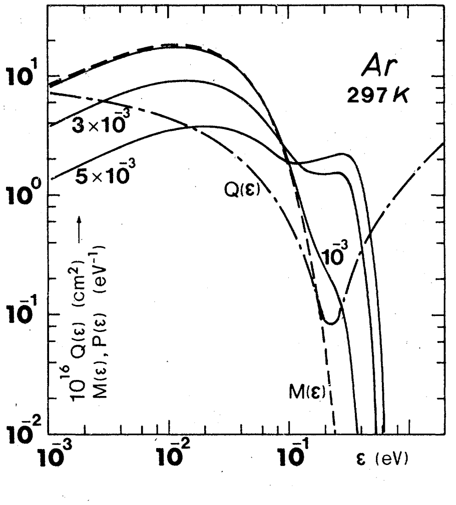

FIG. 2. Momentum-transfer cross section $Q(\epsilon)$ for electrons in Ar (chain curve) (Ref. 17). As one can see, the cross section is strongly dependent on ϵ in the energy interval of our interest. The electron energy distribution is no longer Maxwellian (broken curve) for $E/N = 10^{-20}$ V cm^2. For comparison, in this figure, we give also some other distributions of importance in this paper, which we mark with the corresponding value of E/N (1 Td $= 10^{-17}$ V cm^2).

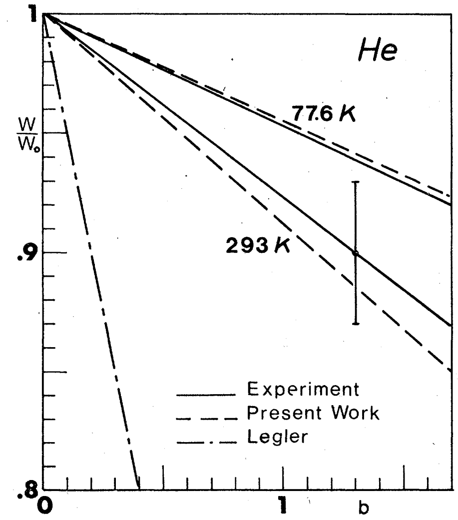

FIG. 3. Theoretical and experimental behavior of W/W_0 as a function of the parameter b in He, at low E/N. The data of Bartels (Ref. 6) at $T = 77.6$ K agree almost perfectly with our theory. The slight discrepancy between theory and Grunberg's (Ref. 7) data at 293 K is within the experimental error.

for He, using Eq. (8). In this case, there is no trouble as regards the value of the scattering length, since the cross section is practically constant over a sufficiently wide energy interval [so that $\bar{Q} \approx Q(0)$] and the scattering length is well known to be equal to 0.63×10^{-10} m. As one can see, at $T = 77.6$ K our theory agrees perfectly with the experimental results of Bartels.[6] We only note a certain discrepancy with the experimental results of Grunberg[7] at 293 K although the slight disagreement is fully consistent with the experimental error. On the contrary the theory of Legler[3] indicates a universal behavior of W/W_0 as a function of b, which is extremely different from the expected behavior at $T = 77.6$ and 293 K.

The comparison between our theory, i.e., Eq. (8), and experiments for H_2 is given in Fig. 4. Here, the agreement is very good at both $T = 77.6$ and 293 K. It is worth noting also that the experimental errors are now very small. The results of Fig. 4 have been obtained by using the value $a = 0.85 \times 10^{-10}$ m that we have found from drift-velocity data. This value agrees with that suggested by Legler[3] and also used by Bartels.[6]

As regards N_2, the results are given in Fig. 5. We have a reasonable amount of experimental data only at $T = 293$ K and also in this case the experimental error appears to be large. In spite of this, the agreement between theory and experiments is good enough. But we must note that the value $a = 0.73 \times 10^{-10}$ m instead of 0.61×10^{-10} m, as suggested by Legler, has been used for N_2.

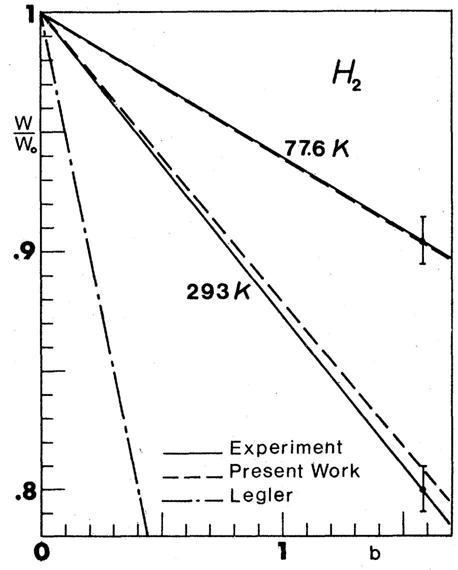

FIG. 4. Theoretical and experimental behavior of W/W_0 as a function of b in H_2, at low E/N. The data of Bartels (Ref. 6) at $T = 77.6$ K and Grunberg (Ref. 7) at 293 K agree well with our theory.

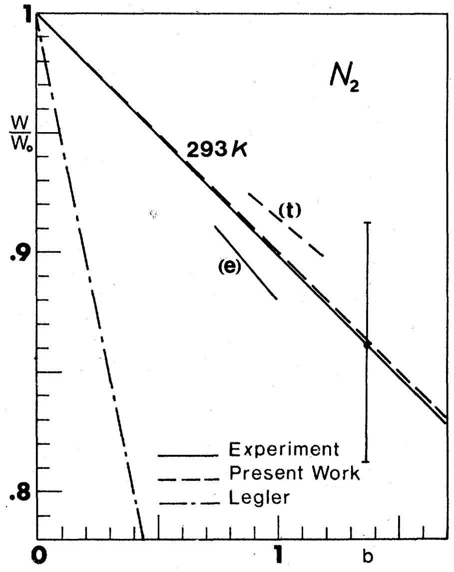

FIG. 5. Theoretical and experimental behavior of W/W_0 as a function of b in N_2 at low E/N. The data obtained by Grunberg (Ref. 7) at $T = 293$ K agree well with our theory if we assume that $a = 0.73 \times 10^{-10}$ m. But, even for $a = 0.61 \times 10^{-10}$ m, the difference between theory (line t) and experiment (line e) is compatible with the experimental error.

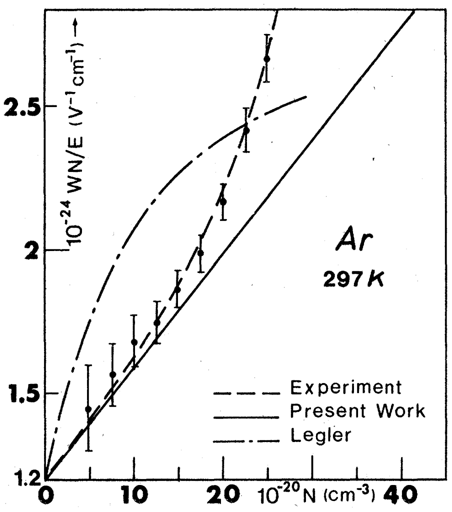

FIG. 6. Theoretical and experimental behavior of $W/(E/N)$ as a function of N in Ar at $T = 297$ K for very low E/N. The experimental results (points) are from Bartels (Ref. 6). As one can see, Eqs. (10) and (17) agree perfectly with the experimental results for sufficiently low values of N. In practice we are within the experimental errors for $N < 1.5 \times 10^{21}$ cm^{-3} corresponding to a gas pressure of $\sim 4.5 \times 10^4$ Torr at 297 K.

Our value has been obtained from an inference of the cross section at $E/N = 1.214\times10^{-1}$ Td where[15] $W_0 = 2.55\times10^3$ m/s. In fact, the full line of Fig. 5 represents the experimental result just at this value of E/N. However, the reader can also see in Fig. 5 that, even when using the value $a = 0.61 \times10^{-10}$ m the difference between our theory and experiment is consistent with the experimental errors. Note that the theory of Legler provides an incorrect behavior also in the two molecular gases H_2 and N_2.

Only the results for Ar remain now to be discussed. In this case, owing to the particular behavior of the cross section, we have been obliged to use numerical means to obtain W from Eq. (10) even at very low E/N. As regards the quantity $\mathrm{Re}\,t(0)$ we have assumed that

$$N\,\mathrm{Re}\,t(0) = -2\pi N\frac{\hbar^2}{m}\left(\frac{Q(\frac{3}{2}k_BT)}{4\pi}\right)^{1/2} \qquad (17)$$

which is equivalent to the introduction of an effective scattering length corresponding to the cross section at the thermal energy.[16] At $T = 297$ K, in particular, $Q(\frac{3}{2}k_BT) \simeq 2\times10^{-20}$ m^2 (cf. Fig. 2) so that $N\,\mathrm{Re}\,t(0) \simeq (6\times10^{-7})p$ eV. The result of our analysis is given in Fig. 6. As one can see, if N is not too high, in practice for N lower than 1.5×10^{21} cm^{-3} corresponding to a pressure of 45 500 Torr, the agreement between theory and experiment is very good and we always remain within the experimental error. But one must note that the disagreement increases at higher values of N, which is expected, since our theory presumes that the gas pressure is so low that we are allowed to neglect corrective terms involving powers of N higher than the first. When studying Ar we have also examined in detail the validity of Eqs. (15) and (16) for representing the drift velocity in the transition region from very low to relatively high values of E/N. This case lends itself particularly to this study since the energy distribution in Ar ceases to be Maxwellian at very low values of E/N (as shown in Fig. 2), when the density dependence of the drift velocity, at fixed E/N, is still large. In Fig. 7 we summarize our results. As one can see, Eq. (15) is able to represent correctly the drift velocity over the entire range of E/N and for all values of gas pressures at which our theory is expected to be correct.

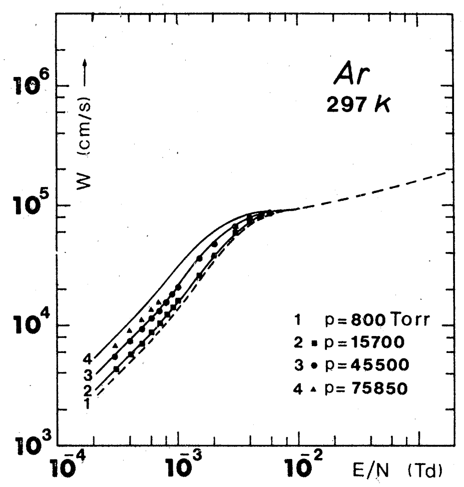

FIG. 7. Comparison between theoretical curves and experimental points for drift velocities of electrons in Ar at $T = 297$ K at three different gas pressures. As one can see, Eq. (15) agrees with the experimental results for any value of E/N and $p \lesssim 4.5\times10^4$ Torr. For comparison we also give the drift velocity at $p = 800$ Torr where the density dependence has been found completely negligible (see also Ref. 17).

ACKNOWLEDGMENT

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in the brackets of Eq. (4) has the meaning of a potential energy. In effect the t matrix $t(\epsilon)$ is linked to the scattering amplitude $f(\theta)$ (Ref. 11), which is well known to be defined in terms of the electron-atom interaction potential. It is also interesting to observe that, as regards Eq. (7), the correction to the conventional drift velocity appears to be independent of the cross section, i.e., a function of E, m, and k_BT. Therefore, it appears to be the same for any gas for which the scattering length is positive and the cross section is a slowly varying function of ϵ for small electron energies. Really, in the comparison of our results with those of other authors, we shall have recourse to representations where this feature is not so evident.

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