

INTERACTION OF THERMAL ELECTRONS WITH POLAR MOLECULES

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Summary

Drift velocities of thermal electron swarms have been measured in pure ethylene gas and in mixtures of acetone, ethanolamine, ethylene diamine, ethanol, n-propanol, isopropanol, chlorobenzene, diethyl ether, diethylamine, and propane with ethylene.

Cross sections for momentum transfer from electron to polar molecule deduced from these experiments and the literature are compared with the "point dipole" theories of Altshuler (1957) and Mittleman and Von Holdt (1965).

I. INTRODUCTION

Recently a considerable amount of experimental information has become available on the interaction of subexcitation electrons (energies below that required to excite atomic transitions) with molecules that possess permanent electric dipole moments (Pack, Voshall, and Phelps 1962; Hurst, Stockdale, and O'Kelly 1963; Lowke and Rees 1963; Cottrell and Walker 1965; Parks and Hurst 1965; Christophorou, Hadjiantoniou, and Hurst 1965). An exact theory that relates the cross section for momentum transfer (electron to molecule) to the dipole moment μ of a "point" dipole has now been published by Mittleman and Von Holdt (1965). As μ increases, the exact theory deviates strongly from the first Born approximation calculation of Altshuler (1957) (see Fig. 3), which has been used in previous comparisons of experiment with theory (Pack, Voshall, and Phelps 1962; Hurst, Stockdale, and O'Kelly 1963; Christophorou, Hadjiantoniou, and Hurst 1965).

We have recently measured drift velocities of thermal electron swarms in pure ethylene and in mixtures of a number of polar gases with ethylene. Momentum transfer cross sections deduced from these experiments and from the literature are presented here and are compared with the theoretical estimates.

II. EXPERIMENTAL METHOD

The method used in the present investigation for the determination of swarm transit times derives from the work of Stevenson (1952) and has been developed by Bortner, Hurst, and Stone (1957), Bortner and Hurst (1958), Hurst, Stockdale, and O'Kelly (1963), and Christophorou, Hadjiantoniou, and Hurst (1965). The apparatus has been described in some detail by Christophorou, Hadjiantoniou, and Hurst and is shown schematically in Figure 1. The experimental method need only be outlined here and may be understood by referring to the figure.

A small pulsed xenon discharge tube S (Edgerton Germeshausen and Grier type FX-6U), whose special glass envelope is largely transparent to wavelengths

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down to 2000 \AA , is used as an ultraviolet light source and is located outside the drift chamber behind a quartz window Q. Electron swarms are ejected by photo-emission from the gold plated photocathode C, which also acts as the (negative) high voltage electrode of a parallel plate ionization chamber. Each swarm drifts under the influence of the applied electric field E (V/cm) across a 9 cm gap to the earthed anode A, where some of the electrons may pass through a small centrally located aperture (0.1 by 0.4 cm) and trigger the proportional counter P. The ultraviolet light source is pulsed 500 times per second, each light pulse having a rise time $< 0.2 \mu\text{sec}$ and a half-width of $\sim 1 \mu\text{sec}$. Its output is monitored by a photodiode D (E.G. and G. type SD-100) whose spectral sensitivity closely

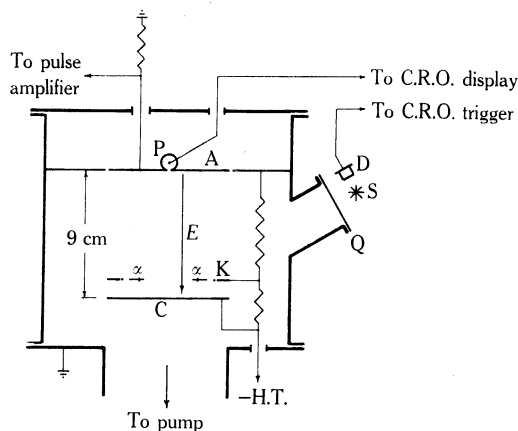


Fig. 1.—Schematic diagram of the apparatus; A, earthed anode; C, gold plated photocathode; D, photodiode; E , electric field; K, α -particle source; P, proportional counter; Q, quartz window; S, xenon discharge tube.

matches the output of the discharge tube. The photodiode output triggers a Dumont Fairchild model 766H oscilloscope and swarm transit times are estimated by displaying the counter output on the oscilloscope sweep. Transit times may be estimated with an accuracy of $\pm 0.1 \mu\text{sec}$, in the range $2\text{--}50 \mu\text{sec}$, in non-attaching gases.

The apparatus also contains a collimated ring α -particle source K (^{239}Pu thinly plated on platinum to provide negligible self-absorption of the emitted α -particles) located in a guard ring and resistance divider structure. The source emits α -particles centrally, parallel to the planes of the electrodes. The electron swarms of uniform size that are produced by the α -particle ionization of the chamber gas are normally used in studies of electron attachment when the apparatus is operated as a pulse ion chamber, but were used in this experiment to monitor gas purity and consistency as a function of time while the drift velocity measurements were in progress. (The magnitude of the pulse induced at the anode by an electron swarm as it drifts through the chamber gas is a function of both the drift velocity w (cm/ μsec) and the attachment coefficient (Bortner and Hurst 1958), so that changes in gas characteristics are reflected by changes in monitored pulse heights.)

The chamber, constructed of stainless steel, could be operated at temperatures up to 120°C and was heated, if necessary, to this temperature for several hours between fillings. It was evacuated by a 2 in. diameter silicone oil diffusion pump and rotary backing pumps to approximately 10^{-5} torr. The background rate of pressure rise before filling was always $< 10^{-3}$ torr/min, which was not sufficient to affect the accuracy of the transit time measurements.

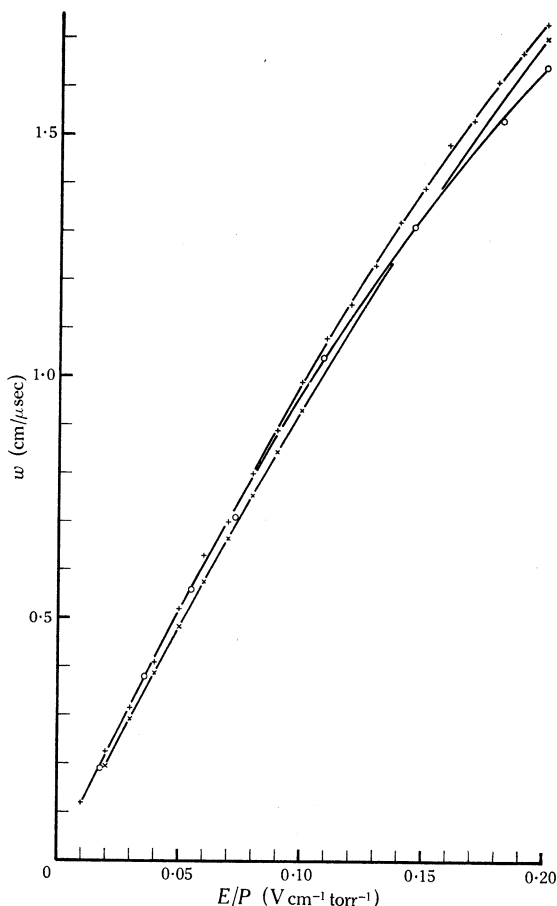


Fig. 2.—Electron swarm drift velocities in ethylene; ○, 150 torr, 353°K, and +, 150 torr, 298°K (present work); ×, 100 torr, 295°K (Parks and Hurst 1965).

Cylinder ethylene gas was obtained from commercial and industrial gas companies and was specified as 98% pure; the bulk of impurities being nitrogen and water vapour. It was passed through silica gel and 4 Å molecular sieve traps and was triply distilled through a system of liquid nitrogen traps filled with copper turnings before admission to the chamber. Further distillations did not improve its quality as judged from pulse height measurements using the α -particle generated

swarms. Cylinder propane used was Thermal Traders commercial grade, diethylamine was Fluka reagent grade ($\geq 98\%$ pure), diethyl ether and acetone were BDH A.R. grade, ethanol and isopropanol were BDH special grade for spectroscopy, n-propanol was Unilab reagent grade, ethylene diamine was May and Baker $> 98.5\%$ quality, chlorobenzene was BDH reagent grade, and ethanalamine was May and Baker reagent grade. Before admission to the chamber these substances were stored in a separate trap connected to the vacuum system and were periodically pumped over a period of 1 or 2 days to remove any dissolved vapours.

On introduction into the chamber the pressures of the polar vapours were monitored with an M.K.S. Baratron capacitance manometer (Head type 77H-10 with range $\sim 10^{-4}$ to 10 torr) until an equilibrium pressure was reached. Ethylene was then admitted to the chamber from the distillation chain to the required total pressure, which was measured with a Wallace and Tiernan diaphragm gauge. The concentration of polar vapour was changed by repeating this procedure to avoid errors likely to arise in a dilution procedure where a new unknown equilibrium may be set up between vapour adsorbed on the walls and vapour in the chamber.

III. RESULTS

Figure 2 shows the measured drift velocities in pure ethylene at 298 and 353°K in the pressure range 100–150 torr for $0.01 < E/P < 0.2 \text{ Vcm}^{-1}\text{torr}^{-1}$. Other measurements at 298°K at pressures up to 400 torr showed no pressure dependence of drift times. Our results at 298°K are in good agreement with the accurate time-of-flight measurements of Parks and Hurst (1965); there are no published measurements of w in ethylene at other temperatures.

For polar gas mixtures with ethylene, as in the work of Hurst, Stockdale, and O'Kelly (1963), plots of w^{-1} against $f_1 P/f_2 P$ ($f_1 P$ = polar gas pressure in torr, $f_2 P$ = ethylene pressure in torr; $f_1 P \ll f_2 P$) resulted in straight lines of the form

$$\{w(E/P)\}^{-1} = \{w_2(E/P)\}^{-1} + S(E/P) f_1 P/f_2 P, \quad (1)$$

where $w_2(E/P)$ is the drift velocity for electrons in pure ethylene.

Hurst, Stockdale, and O'Kelly have shown that an expression of the same form as (1) may be obtained from the well-known relation (Allis 1956)

$$w = \frac{4\pi}{3} \frac{e}{mnP} \frac{E}{P} \int_0^\infty f_0 \frac{d}{dv} \left(\frac{v^2}{\sigma} \right) dv \quad (2)$$

connecting drift velocity w and momentum transfer cross section σ , provided the cross sections for momentum transfer to both polar gas and ethylene are of the form $\sigma = A/v^2$ (e and m are the charge and mass of the electron, n is the number density of the gas molecules at 1 torr and 295°K , v is the velocity of the electron, and f_0 is the spherically symmetric term in the expansion of the electron velocity distribution; A is a constant equal to, say, A_1 for polar molecules and to A_2 for ethylene).

Christophorou, Hadjiantoniou, and Hurst (1965) have presented evidence (from electron drift velocities in pure ethylene) that, for ethylene, the v^{-2} form of the cross section is to be preferred to the v^{-1} dependence that might be expected

on theoretical grounds, since ethylene has no permanent dipole moment. The v^{-2} dependence for polar molecules follows from the nature of the long range r^{-2} dipole force.

Comparison of (1) and the analogous expression obtained from (2), namely,

$$w^{-1} = w_2^{-1} + w_2^{-1}(A_1/A_2)(f_1 P/f_2 P),$$

suggests that

$$S(E/P) \times w_2(E/P) = A_1/A_2. \quad (3)$$

Values of S and w_2^{-1} obtained from least squares fitting of the experimental drift velocities in polar gas-ethylene mixtures are shown in Table 1. In calculating momentum transfer cross-section ratios, average values of Sw_2 were used for the range $0.01 \leq E/P \leq 0.1 \text{ V cm}^{-1} \text{ torr}^{-1}$, for which the electron swarm contains a thermal (Maxwellian) distribution of electron velocities (Stockdale and Hurst 1964). Knowledge of the velocity distribution makes possible theoretical estimates of A_1/A_2 from the theories of Altshuler (1957) or Mittleman and Von Holdt (1965).

Figure 3 summarizes the experimental data, from the present work and from the literature, on momentum transfer cross sections for electrons to polar molecules. It also includes the theoretical estimates of Altshuler and of Mittleman and Von Holdt. Cottrell and Walker (1965) did not report momentum transfer cross sections but listed drift velocities in a number of pure vapours. The points shown as due to Cottrell and Walker were estimated using the low E/P linear portions of their drift velocity curves for pure polar gases. In this region the electron swarm energy distribution is thermal (Pack, Voshall, and Phelps 1962).

The error estimates shown in Figure 3, except in the case of ethanolamine, are standard deviations calculated from the A_1/A_2 estimates obtained at the various E/P values. For the case of ethanolamine, where only a single $f_1 P/f_2 P$ value was used in the measurements, a deviation of $\pm 20\%$ has been assigned by comparison with the range of deviations observed in the other listed substances when a single $f_1 P/f_2 P$ value is used to calculate A_1/A_2 . This procedure gives some indication of the quality of the least squares straight line fits and of the internal consistency of the data; however, it neglects the possible occurrence of systematic errors. Because of the precautions taken in measurement it is felt that systematic errors should be small.

IV. DISCUSSION

In general there is fairly good agreement between theory and experiment for the majority of compounds when it is considered that the theoretical estimates do not include the effects of short-range induced polarization. For dipole moments less than about 2 Debye units there seems little to choose between the exact quantum mechanical theory of Mittleman and Von Holdt and the first Born approximation calculation of Altshuler. At higher dipole moments, where the two theories begin to diverge rapidly, there are few experimental points. The data for acetone (taken as a whole) and ethanolamine favour the Altshuler theory, but there is obvious need for further measurements at high dipole strengths. Measurements using compounds with very high dipole moments are difficult because they tend to have high electron capture cross sections and high boiling points.

TABLE 1
VALUES OF w_2^{-1} AND S APPEARING IN EQUATION (1) FOR MIXTURES OF INDICATED GASES WITH ETHYLENE
Values are in units of 10^6 sec/cm

Gas	μ^* (10^{-18} e.s.u.)	Coefficient	E/P ($V\text{ cm}^{-1}\text{ torr}^{-1}$)				A_1/A_2^\dagger	$f_1P + f_2P^\ddagger$ (torr)	Range of f_1P/f_2P (torr)
Acetone	2.89	$\left\{w_2^{-1}\right\}_S$	—	1.56	1.12	0.97	0.69	150	0-0.04
Ethylenediamine	1.99	$\left\{w_2^{-1}\right\}_S$	—	378	251	182	149	150	0-0.04
			—	2.42	1.60	1.23	1.03	150	0-0.04
Ethanol, n-Propanol§	1.69	$\left\{w_2^{-1}\right\}_S$	—	225	154	115	92	150	0-0.14
			—	2.31	1.68	1.23	0.88	150	0-0.04
Isopropanol	1.69	$\left\{w_2^{-1}\right\}_S$	—	172	110	83	76	150	0-0.04
			4.09	2.12	1.39	1.06	0.91	150	0-0.04
Chlorobenzene	1.67	$\left\{w_2^{-1}\right\}_S$	287	147	102	76	58	100	0-0.01
			—	2.22	1.56	1.17	0.99	150	0-0.04
Diethyl ether	1.15	$\left\{w_2^{-1}\right\}_S$	—	142	113	84	60	150	0-0.04
			5.20	2.60	1.77	1.34	1.15	150	0-0.04
Diethylamine	0.92	$\left\{w_2^{-1}\right\}_S$	143	74	51	40	28.5	150	0-0.04
			123	5.24	2.56	1.64	1.23	150	0-0.04
Propane	0	$\left\{w_2^{-1}\right\}_S$	—	67	47	35	26	100, 150	0-0.04
			5.81	3.09	2.08	1.59	1.44	150	0-0.04
Ethanolanine	2.6	$\left\{E/P\right\}_S$	6.0	-4.0	-2.6	-1.4	-0.6	150	0-0.04
			0.074	0.112	0.149	0.186	—	150	0-0.04
		$\left\{E/P\right\}_S$	183	124	85	65	—	150	0-0.04
			0.093	0.130	0.168	0.204	—	150	0-0.04
			148	101	77	62	—	150	0-0.04

* Dipole moments from the compilation of McClellan (1963).

† Errors shown are standard deviations calculated from the listed w_2^{-1} and S values, except for ethanolanine, where an estimate of $\pm 20\%$ has been assigned based on deviations observed in the other listed substances when a single f_1P/f_2P value is used with w^{-1} for pure ethylene to calculate A_1/A_2 .

‡ Spot checks for pressure dependence of drift times in the range 50-200 torr were made.

§ n-Propanol measurements were made at $f_1P/f_2P = 0.03$ and 0.04 , and were then discontinued due to similarity to ethanol measurements.

|| Ethanolanine was measured at 80°C (measurements were made at 25°C for all other gases) and values of $f_1P/f_2P = 0.011$ and $\{w(E/P)\}^{-1}$ for pure ethylene at 80°C were used to calculate the S values.

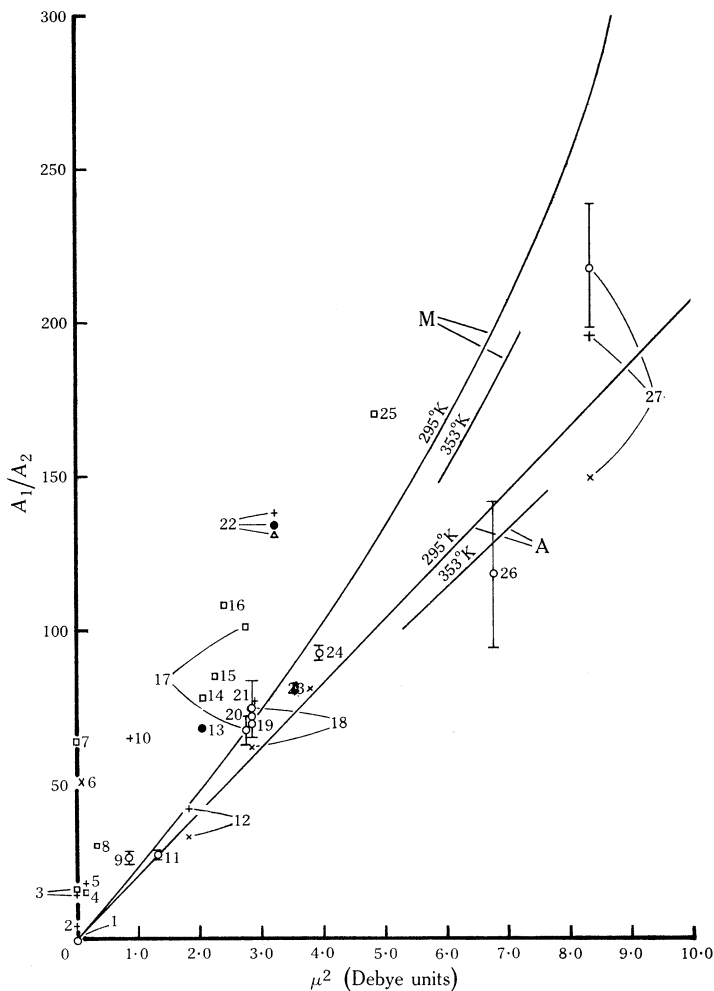


Fig. 3.—Ratio of momentum transfer cross section parameters A_1/A_2 related to the dipole moment μ for various polar gases added to ethylene. The continuous lines are theoretical estimates (A, Altshuler 1957; M, Mittleman and Von Holdt 1965). \circ , Present work; \triangle , Parks and Hurst (1965); \square , Christophorou, Hadjiantoniou, and Hurst (1965); \times , Cottrell and Walker (1965); $+$, Hurst, Stockdale, and O'Kelly (1963); \bullet , Pack, Voshall, and Phelps (1962) (the result of Lowke and Rees (1963) for water vapour is the same as that of Pack, Voshall, and Phelps). 1, Propane; 2, nitrous oxide; 3, benzene; 4, *m*-xylene; 5, toluene; 6, AsH_3 ; 7, *p*-dichlorobenzene; 8, *o*-xylene; 9, diethylamine; 10, H_2S ; 11, diethyl ether; 12, dimethyl ether; 13, NH_3 ; 14, *o*-bromotoluene; 15, *m*-dichlorobenzene; 16, bromobenzene; 17, chlorobenzene; 18, ethanol; 19, isopropanol; 20, *n*-propanol; 21, methanol; 22, H_2O (the Hurst, Stockdale, and O'Kelly point for H_2O includes their result for D_2O also); 23, methyl chloride; 24, ethylenediamine; 25, *o*-dichlorobenzene; 26, ethanolamine; 27, acetone. All experimental points in the present work are at 298°K except for 26 (ethanolamine), which was obtained at 353°K. The experimental points of the other authors shown were obtained at 295°K.

The good agreement, in the cases of water and dimethyl ether (data of Hurst, Stockdale, and O'Kelly 1963) and ethanol (present work), and fair agreement in the case of acetone (points from Hurst, Stockdale, and O'Kelly and present work) between momentum transfer cross sections estimated by the present method and those obtained from drift velocities in pure polar vapours (Pack, Voshall, and Phelps 1962; Lowke and Rees 1963; Cottrell and Walker 1965; Parks and Hurst 1965) support the method of analysis for mixtures used here and by Hurst, Stockdale, and O'Kelly and Christophorou, Hadjiantoniou, and Hurst.

It is notable that experimental momentum transfer cross sections for AsH_3 , NH_3 , H_2S , H_2O , D_2O , and a number of aromatic compounds are much higher than would be expected from either theory using the pure r^{-2} long-range dipole force. (The cross section we have obtained for chlorobenzene is considerably less than that reported by Christophorou, Hadjiantoniou, and Hurst and does not deviate strongly from theory. Our results for amines in general do not show the deviation observed by Pack, Voshall, and Phelps for ammonia, though diethylamine shows some discrepancy.)

These high deviations cannot be explained by induced polarization effects (Hurst, Stockdale, and O'Kelly 1963; Christophorou, Hadjiantoniou, and Hurst 1965). Enhanced rotational excitation due to interaction between the incident electron and π electron structure of the molecule may occur in aromatic compounds as suggested by Christophorou, Hadjiantoniou, and Hurst, but comparable effects in H_2O , D_2O , H_2S , and AsH_3 seem less likely.

Hydrogen bonding, which might increase the effective dipole moment, occurs in the above vapours and in ammonia. Victorova (1964) has calculated the percentage of dimers present in water vapour under conditions similar to those we have used. Making the overestimate of a doubling of the dipole moment in dimer formation, only a negligible fraction of the discrepancy in the case of water vapour is removed; dimer formation does, however, introduce the possibility of internal rotational excitation about the hydrogen bond.

Barua and Das Gupta (1963) and Barua, Chakrabati, and Saran (1965) have considered dimer formation in polar gases due to dipole-dipole interaction. Their results also do not afford an explanation of the momentum transfer cross section observations. Aggregation effects in general might be expected to result in pressure dependence of the momentum transfer cross section. Bradbury and Tatel (1934) and Kuffel (1959) have reported a pressure-dependent attachment process for thermal electron swarms in pure water vapour, but in more recent experiments by Pack, Voshall, and Phelps (1962), using pure water vapour, and by Hurst, Stockdale, and O'Kelly (1963) and Parks and Hurst (1965), using water vapour-ethylene mixtures, no measurable permanent electron attachment was observed.

It is possible that the large discrepancies from theory may be explained in terms of autoionizing negative ion states or of negative ions in which the electron is bound with an energy $\lesssim kT$ (Hurst, O'Kelly, and Stockdale 1962). Recent observations of resonance scattering of low energy electrons by the noble gases and mercury, e.g. Fleming and Higginson (1963), Schulz (1963), Simpson and Fano

(1963), Kuyatt, Simpson, and Mielczarek (1965), and of high vibrational excitation cross sections of various diatomic gases, e.g. Schulz (1962, 1964), Briglia and Rapp (1965), Cermak (1966), have been related to the formation of temporary negative ion states.

Theoretically, recent developments in the understanding of these resonances have been based to a large extent on analogous developments in nuclear resonance theory, particularly by Feshbach (1958, 1962). Application of the theory to the problem of resonant rotational excitation may help to provide a detailed explanation of the discrepancies mentioned above, but at present this is a difficult task for the more complex molecules considered here. A comprehensive review of resonant electron scattering and photon absorption has been given by Burke (1965).

In a very recent paper Turner (1966) has shown theoretically the possibility of temporary electron capture by a polar molecule with simultaneous rotational excitation. Using idealized spherically symmetric wave functions and assuming a point dipole interaction in both initial (free) and final (bound) states of the electron-molecule system, Turner shows that temporary negative ion lifetimes of the order of 10^{-13} sec (approximately one rotation time) are possible. The capture probability is proportional to the ratio $(\mu/I)^2$, where I is the moment of inertia of the molecule about a perpendicular axis passing through the centre of the dipole. Electron capture by rotational excitation is most probable therefore for polar molecules that have a large dipole moment and a small moment of inertia. For this process to make effective contributions to momentum transfer cross sections measured in swarm experiments, the spacing of pure rotational levels in the molecule must be of the order of thermal energies so that a relatively large number of electrons can be captured and released.

The two conditions, right spacing of rotational levels and a relatively large value of μ/I , are met only by a few light molecules, among them H_2O , D_2O , H_2S , NH_3 , HF , HCl , and H_2O_2 . The theory therefore suggests an explanation for the discrepancies that have previously been observed with the first four of these molecules and also, possibly, with AsH_3 .

Moments of inertia of deuterated "light" molecules, such as these, deduced from spectroscopic observations of pure rotational transitions are about twice the values obtained for the non-deuterated molecules (e.g. Landolt and Bornstein 1951). This is so for D_2O compared with H_2O and holds through the series NH_3 , PH_3 , AsH_3 , SbH_3 (Blevins, Jache, and Gordy 1955). While the rotational levels of the deuterated compounds are shifted to lower energies, it remains unexpected that H_2O and D_2O should provide identical momentum transfer cross sections (Fig. 3). A further observation of some interest is the discrepancy apparently shown by AsH_3 . We have

$$(\mu/I)_{\text{NH}_3}^2 \div (\mu/I)_{\text{AsH}_3}^2 \simeq 400.$$

However, NH_3 shows a smaller apparent deviation than AsH_3 .

Further electron swarm drift velocity measurements at low E/P , on a range of the simple hydrides—both normal and deuterated—would be of great interest in

connection with the model proposed by Turner. These measurements should be carried out over as wide a range of temperatures as possible.

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