

calculated and await comparison to future neutron scattering experiments. Unless the substrate were to have a very strong disordering effect upon the monolayer, neutron scattering should give the monolayer lattice structure and the degree of localization of the helium atoms.

Generalizing these results, it would seem that the Gauss-Hartree approach to the ground state

of the adsorbed film is quite reasonable. However, for certain substrates it might be necessary to incorporate a more detailed treatment of motion perpendicular to the surface.

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¹F. D. Manchester, *Rev. Mod. Phys.* **39**, 383 (1967).

²M. Bretz, G. B. Huff, and J. G. Dash, *Phys. Rev. Lett.* **28**, 729 (1972).

³C. E. Campbell, F. J. Milford, A. D. Novaco, and M. Schick *Phys. Rev. A* **6**, 1648 (1972).

⁴L. Passell (private communication).

⁵A. Sjölander, in *Thermal Neutron Scattering*, edited by P. A. Egelstaff (Academic, New York, 1965).

⁶R. A. Guyer, in *Solid State Physics*, edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1969), Vol. 23.

⁷C. E. Campbell and M. Schick, *Phys. Rev. A* **3**, 691 (1971).

⁸E. L. Pollock, T. A. Bruce, G. V. Chester, and J. A. Krumhansl, *Phys. Rev. B* **5**, 4180 (1972).

⁹The Gauss-Hartree approximation is used here to mean use of a variational wave function constructed as a product of Gaussians having negligible overlap.

¹⁰L. W. Bruch and I. J. McGee, *J. Chem. Phys.* **46**, 2959 (1967).

¹¹D. E. Hagen, A. D. Novaco, and F. J. Milford, in *Proceedings of the Second International Symposium on Adsorption-Desorption Phenomena, Florence* (Academic, London, to be published).

¹²H. F. Schaefer, D. R. McLaughlin, F. E. Harris, and B. J. Alder, *Phys. Rev. Lett.* **25**, 988 (1970); P. Bertonicini and A. C. Wahl, *Phys. Rev. Lett.* **25**, 991 (1970).

¹³D. C. Hickernell, E. O. McLean, and O. E. Vilches, *Phys. Rev. Lett.* **28**, 789 (1972).

¹⁴M. Bretz and J. G. Dash, *Phys. Rev. Lett.* **27**, 647 (1971).

Mobilities and Longitudinal-Diffusion Coefficients of Mass-Identified Positive Ions in Carbon Monoxide Gas*

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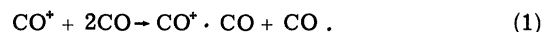
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We have measured with a drift-tube mass spectrometer the mobilities of CO^+ , $\text{CO}^+ \cdot \text{CO}$, and C^+ ions in carbon monoxide gas at 300 °K. The measurements were made over a substantial range of E/N , where E is the drift-field intensity and N is the gas number density. Zero-field reduced mobilities were determined for $\text{CO}^+ \cdot \text{CO}$ and C^+ ions. Their values are 1.90 ± 0.03 and $2.7 \pm 0.1 \text{ cm}^2/\text{V sec}$, respectively. Measurements were also made of the longitudinal-diffusion coefficient of CO^+ ions in CO at 300 °K as a function of E/N .

I. INTRODUCTION

This paper presents the results of an investigation of the drift and longitudinal diffusion of low-energy positive carbon monoxide ions in carbon monoxide gas at room temperature. The data were obtained with a drift-tube mass spectrometer of ultrahigh-vacuum construction. The mobility of CO^+ , $\text{CO}^+ \cdot \text{CO}$, and C^+ ions in CO and the longitudinal-diffusion coefficient of CO^+ in CO were determined as a function of E/N . The results are an extension of similar studies on H_2 , $^{1,2} \text{D}_2$, $^2 \text{N}_2$, $^{3,4} \text{O}_2$, 5 and NO^6 performed in this laboratory. The only previous data⁷ on the mobility of CO^+ and $\text{CO}^+ \cdot \text{CO}$ ions in CO obtained with a drift-tube

mass spectrometer apparently were seriously affected by the ion-molecule reaction



No previous measurements on diffusion of ions in CO have been reported. In the present study, the effect of reaction (1), as well as other reactions, has been carefully considered and accounted for. In Paper II, immediately following this paper, the results of a study of reaction (1) will be presented.

II. GENERAL

As a slow ion moves through a gas under the influence of a static uniform electric field, on the average it gains energy from the field between

collisions with molecules and loses energy during collisions. Since the ionic mass is usually comparable to the molecular mass, only a few collisions are normally required for ions to attain a steady-state condition after they are produced in the gas. The ratio of the electric field intensity to the gas number density, E/N , is the parameter that determines the average ionic energy acquired from the field in steady-state drift, above the energy associated with the thermal motion. In this paper, the quantity E/N will be expressed in units of 10^{-17} V cm², called the Townsend (abbreviated Td).

If the value of E/N is small and constant, and if steady-state conditions have been achieved, the motion of an ensemble, or swarm, of ions of a given kind consists of a slow uniform drift in the field direction superimposed on the much faster random motion which produces diffusion. Under such conditions, the average energy the ions have acquired from the field is small with respect to their thermal energy, and their mean drift velocity v_d in the field direction is proportional to the field strength E . Thus, $v_d = KE$, where the constant K is called the *mobility* of the ions and is expressed in units of cm²/V sec.

At higher values of E/N , where the ions are no longer in thermal equilibrium with the gas, the drift velocity may no longer be proportional to the electric field strength. However, the mobility, defined as the ratio of drift velocity to electric field strength, still serves as a convenient quantity to describe the transport of ions through the gas.

The mobility K is inversely proportional to the gas number density N , and in order to facilitate comparisons between data taken at various temperatures and pressures, experimental results are usually presented as the mobility which would result if N were 2.69×10^{19} cm⁻³, which corresponds to 0 °C and 760 Torr. This practice leads to the definition of a *reduced mobility* $K_0 = K(p/760) \times (273.16/T)$, where p and T are the pressure in Torr and temperature in degrees kelvin at which the measurement is performed. The *zero-field reduced mobility* is defined as the limiting (constant) value that K_0 approaches as E/N approaches zero.

Whenever a number-density gradient of ions exist in a gas of uniform total pressure, there will be a motion of the ions to eliminate the gradient, the process known as diffusion. If the gradient is small, and if the value of E/N is very low, the flux density will be proportional to the magnitude of the gradient and opposite in direction. The constant of proportionality, known as the *diffusion coefficient* D , can be related to the ionic mobility by a classical relation known as the Einstein equation⁸:

$$D/K = kT/e, \quad (2)$$

where k is Boltzmann's constant and e is the ionic charge. Equation (2) is exact in the limit of small ionic concentrations and vanishing electric field strength, and approximately correct so long as the ions are close to being in thermal equilibrium with the gas molecules.

At values of E/N where the ions acquire an average energy appreciably above the thermal energy of the gas molecules, the ionic flow due to diffusion becomes anisotropic and the diffusion flux density is given by $\vec{j} = -\vec{D} \cdot \vec{\nabla} n(\vec{r}, t)$, where $n(\vec{r}, t)$ is the ionic number density, and \vec{D} is the diffusion tensor. \vec{D} has the form⁹

$$\vec{D} = \begin{vmatrix} D_T & 0 & 0 \\ 0 & D_T & 0 \\ 0 & 0 & D_L \end{vmatrix}, \quad (3)$$

where D_T is the *transverse-diffusion coefficient* which describes the rate of diffusion in directions perpendicular to the electric field, and D_L is the *longitudinal-diffusion coefficient* characterizing diffusion in the field direction.

III. APPARATUS

The apparatus that was used to perform the present research has been described in detail in several reports,¹⁰⁻¹⁴ so a detailed description will not be given here. Briefly, the apparatus consists of a large ultrahigh-vacuum chamber containing a drift tube, ion source, and sampling and analysis chamber. A very pure sample of carbon monoxide is admitted into the drift tube through a servo-controlled leak valve, and the gas continuously flows from the drift tube through a small aperture and into the main kettle, where it is pumped away by a 6-in. oil-diffusion pump. An MKS "Baratron" capacitance manometer continuously monitors the pressure in the drift tube, and, through a Granville-Phillips pressure controller, maintains any selected pressure in the range of 0.026–0.795 Torr.

An ion swarm can be followed from creation to detection in Fig. 1. The ion source, which is movable over almost the entire length of the 44-cm drift tube, creates short bursts (typically 1–5 μ sec in length) of ions in the drift tube. The ions are created by electron impact with the carbon monoxide gas and are gated out of the source into the drift region by a double-grid shutter located at the ion entrance aperture.

Each ion swarm migrates down the drift region under the influence of a uniform electric field maintained by the drift-field guard rings. During this migration the swarm spreads owing to diffusion, and may undergo ion-molecule reactions with the neutral gas. When the ions reach the bottom of the drift

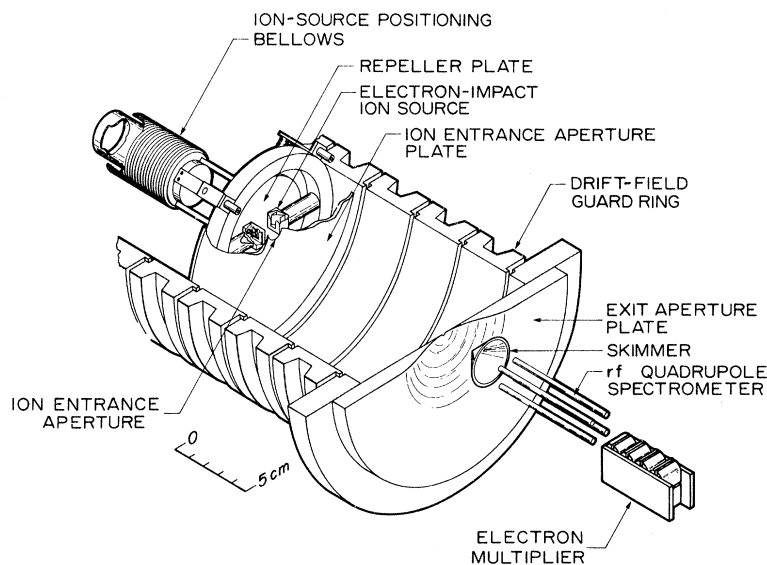


FIG. 1. Interior components of the drift tube and ion-sampling apparatus. The drift tube is vertical, with the ion source toward the top. The exit aperture plate separates the "high-pressure" drift region from the evacuated regions below it.

tube, those close to the axis are swept out through the exit aperture, and the core of the emerging jet of ions and gas molecules is cut out by a conical skimmer and directed into a radio-frequency quadrupole mass spectrometer. Ions of only a selected charge-to-mass ratio traverse the length of the spectrometer; all other ions are rejected in the mass-selection process. The selected ions are then detected individually by an electron multiplier operated as a pulse counter, and the resulting pulses are electronically sorted as to their arrival time by a 256-channel time-of-flight analyzer. A spectrum of arrival times is built up by accumulating data from about 10^6 ion bursts for a given source position, to provide a time profile of the swarm of ions of the given species at a particular distance from the ion-entrance aperture.

IV. MOBILITIES

The differential equation describing the number density of an ion species created only in the source, gated into the drift tube as an axially thin disk of constant areal number density s and radius r_0 , and diffusing and reacting as it drifts down the drift tube, is

$$\frac{\partial n(x, y, z, t)}{\partial t} = D_T \left(\frac{\partial^2 n}{\partial x^2} + \frac{\partial^2 n}{\partial y^2} \right) + D_L \frac{\partial^2 n}{\partial z^2} - v_d \frac{\partial n}{\partial z} - \alpha n + sS(x^2 + y^2 - r_0^2) \delta(z) \delta(t), \quad (4)$$

where α is the depleting reaction frequency and $S(\varphi) = 0$ if $\varphi > 0$, $S(\varphi) = 1$ otherwise. From the solution of this equation one can obtain an equation for the flux of ions leaving an exit aperture of area a located on the axis z cm from the source^{3,12}:

$$\Phi(z, t) = [ase^{-\alpha t} / 4(\pi D_L t)^{1/2}] (v_d + z/t) \times (1 - e^{-r_0^2/4D_T t}) e^{-(z-v_d t)^2/4D_L t} \quad (5)$$

Figure 2 compares the experimental results (shown as the histogram) and the predictions of this equation (shown as the solid curve).

If the mean arrival time, defined by

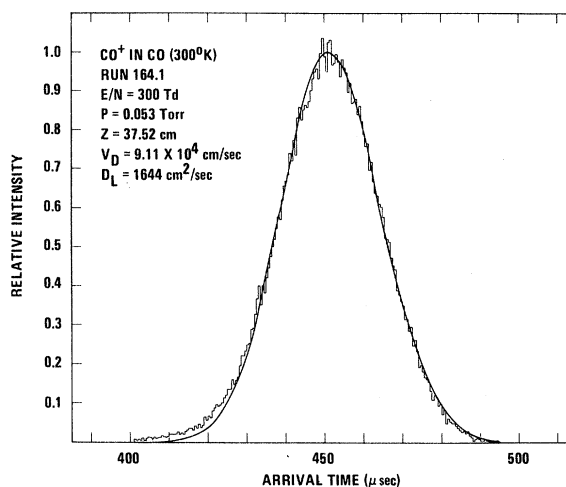


FIG. 2. Sample experimental arrival-time spectrum (histogram) compared with the corresponding best-fit analytical profile (smooth curve). The curve-fitting procedure used to obtain the latter is described in Sec. V. The slight disagreement between the shapes of the experimental and analytical spectra may be attributable to the asymmetry of the ionic velocity distribution at the high E/N employed for the measurement [J. H. Whealton (unpublished)].

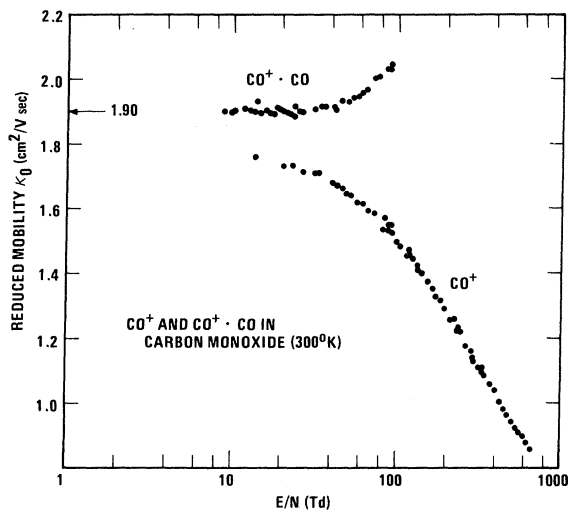


FIG. 3. Experimental mobility data for CO^+ and $\text{CO}^+ \cdot \text{CO}$ ions in carbon monoxide gas at 300°K. The number 1.90 by the arrow at the left-hand side of the drawing indicates the zero-field value of K_0 for $\text{CO}^+ \cdot \text{CO}$ ions in CO obtained by extrapolating the experimental data. The data for CO^+ do not extend to E/N low enough for a determination of the zero-field value of K_0 .

$$\bar{t} = \int_0^\infty t \Phi(t) dt / \int_0^\infty \Phi(t) dt,$$

is used to calculate the drift velocity $v_d = z/\bar{t}$, then two corrections to the drift-velocity calculation must be considered. These corrections arise from the fact that we are making time averages at a fixed distance instead of space averages at a fixed time. One of the corrections is due to longitudinal and transverse diffusion, and one is due to longitudinal diffusion and a depleting reaction rate.¹² Both of these corrections can be estimated analytically. The pure diffusion term is virtually eliminated by the use of a differencing technique in the drift-velocity calculation, where the drift velocity is obtained from the difference in mean arrival times for two or more drift distances, $v_d = (z_i - z_j)/(\bar{t}_i - \bar{t}_j)$. The combination diffusion-reaction correction cannot be so eliminated, and the appropriate correction must be applied to the drift-velocity results.¹⁵ It should be noted that for most of the data presented here, the reaction correction was within the scatter of the data and could be ignored.

Figures 3 and 4 show the reduced-mobility values for CO^+ , $\text{CO}^+ \cdot \text{CO}$, and C^+ ions in CO at 300°K as a function of E/N . In the case of CO^+ , the ion-molecule reaction converting CO^+ into $\text{CO}^+ \cdot \text{CO}$ prevented us from obtaining data at values of E/N low enough to determine the zero-field mobility. The zero-field reduced mobilities of $\text{CO}^+ \cdot \text{CO}$ and C^+ are 1.90 ± 0.03 and $2.7 \pm 0.1 \text{ cm}^2/\text{V sec}$, respectively.

CO^+ and C^+ are true primary ions, created by electron-impact within the ion source. By keeping the gas pressure relatively low within the drift tube, the effects of depleting reactions can be made small or insignificant. $\text{CO}^+ \cdot \text{CO}$, however, is a reaction-produced ion. At relatively high pressures, the ion-molecule reaction (1) can be driven to completion early in the drift, and meaningful results can be obtained for the reaction-produced ions also.

The principal source of error in measurements of the mobility of CO^+ and $\text{CO}^+ \cdot \text{CO}$ was in the pressure measurement. The accuracy of the MKS "Baratron" was checked by remeasuring the mobility of potassium ions in nitrogen gas at low values of E/N , a measurement made by several experimenters¹⁶ and considered well known. The accuracy of the pressure measurements is estimated to be 1%. For C^+ ions, the principal source of error is due to the random scatter in the mean-arrival-time measurements. This random scatter was larger in the case of C^+ ions than for the other ions due to the low abundance of the C^+ ions. C^+ is formed by electron impact much less frequently than is CO^+ . If the total ion production had been increased in order to increase the number of C^+ ions formed, the space-charge effects due to the other, more numerous, ions could have distorted the results obtained for C^+ . This effect was noticed in hydrogen by Albritton *et al.*¹⁷ while studying the mobility of H^+ in hydrogen. In that case H_3^+ was

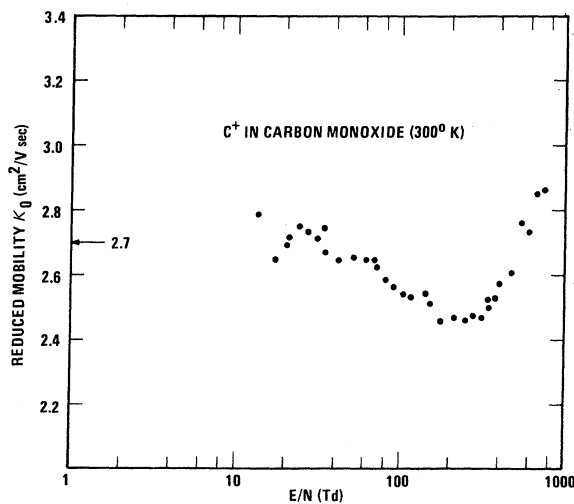


FIG. 4. Experimental mobility data for C^+ ions in carbon monoxide gas at 300°K. The scatter in the data at low E/N is due mainly to the very low abundance of the C^+ ions under low-field conditions. The number 2.7 by the arrow at the left-hand side of the drawing indicates the zero-field value of K_0 obtained by extrapolating the experimental data. The rise in the mobility at very high E/N is unusual, but experimental checks lead us to believe that it is not an experimental artifact.

the more abundant ionic species. The random error for the C^+ arrival-time measurements was about $\pm 3.0\%$, significantly decreasing the accuracy of this measurement.

Two other minor reactions were detected, each contributing slightly to the possible error bars, but neither believed to contribute significantly to the results. The charge-exchange reaction $C^+ + CO \rightarrow C + CO^+$ was indicated by a small number of CO^+ ions arriving at the detector with abnormally short drift times. The fastest ion observed was C^+ , and the leading edge of the CO^+ arrival-time spectrum corresponded to the drift velocity of C^+ . As with the clustering reaction (1), ions apparently drifted part of the drift distance as C^+ , reacted with the CO gas molecules, and drifted the rest of the way as CO^+ .

The other minor reaction observed was the break-up of the $CO^+ \cdot CO$ cluster: $CO^+ \cdot CO + CO \rightarrow CO^+ + 2CO$. The evidence of this reaction was the arrival of CO^+ ions with the drift times corresponding to the dimer ions and not characteristic of CO^+ . These fast CO^+ ions did not have drift times as short as ions traveling partially as C^+ , but they formed a small peak about the drift time of $CO^+ \cdot CO$.

Impurities with masses in the range of 41–47 amu were seen on occasion, but these impurities were removable by exposing the input gas to a refrigerating-vapor-bath cold trap for a period of several days.¹⁸ No errors were believed to have been caused by the impurities when they were present.

V. LONGITUDINAL-DIFFUSION COEFFICIENTS

No statistical characteristic of the arrival-time spectrum has been found that measures the longitudinal-diffusion coefficient as accurately as the mean arrival time measures the drift velocity. Qualitatively, the width of the spectrum is a function of the longitudinal-diffusion coefficient, but the only way we have found to determine D_L accurately is by a computerized curve-fitting procedure.

Basically, the curve-fitting procedure involves the matching of an experimental arrival-time spectrum with the analytical function described by Eq. (5), starting with an assumed value of D_L which can be varied until the best fit is achieved. This matching procedure might be assumed to require a many-parameter fit, since (5) contains several parameters that have not been determined. However, it was shown by Moseley *et al.*¹⁹ that D_T and α strongly affect only the over-all magnitude of the spectrum; their effect on the shape of the spectrum is very weak. In the analysis used on the data presented in this paper, D_T was set equal to D_L and α was set equal to zero. Checks on the propriety of our assignment of the indicated values to D_T were made by varying these values between

wide limits and examining the resulting changes in our final values of D_L —these changes were very small. Since the ion-source density s is not a measured quantity and several experimental transmission factors are not known, a normalization between the experimental arrival-time spectra and the theoretical profiles must be made. In order to reduce the effects of random scatter in the normalization of the experimental spectrum, the data are smoothed before the determination of the peak height is made. The unsmoothed data are used for the actual comparison with the theory, however.

As noted in the paper by Snuggs *et al.*,⁵ in addition to the smoothing normalization, the experimental spectrum must be shifted in time with respect to the theoretical profile, since the theoretical profile contains no allowance for end effects (such as the time required for the ions to travel from the drift-tube exit aperture to the detector). This shift is made initially by matching the peaks of the profiles in time, and then the detailed curve fitting is begun. First, a reasonable match is obtained by varying the value of D_L . Step (1): This procedure involves the examination of the sum of cubed differences on both sides of the peak of the experimental profile and the adjustment of D_L in order to make these differences a minimum. The cubes of the differences are used instead of the squares in order to retain the sign of the differences. Step (2): Then a secondary over-all time ad-

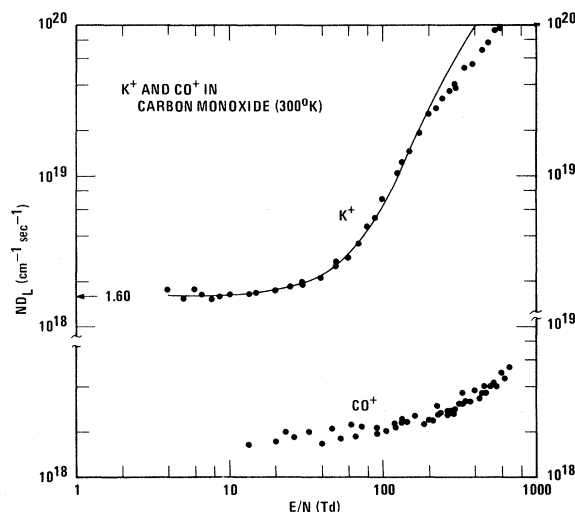


FIG. 5. Experimental results (symbols) on longitudinal diffusion of CO^+ and K^+ ions in CO at 300°K. N is the gas number density and D_L is the longitudinal-diffusion coefficient. The solid curve is a plot of a theoretical expression for ND_L of K^+ ions in CO, described in Ref. 20. The accuracy of the K^+ experimental data is believed to be high at E/N below 225 Td, but gradually decreases as E/N is raised above this value, as explained in Ref. 20.

justment is made by translating the experimental spectrum in time until the best fit is obtained. Subsequently, steps (1) and (2) are repeated as necessary to determine the best final value of D_L .

Of the ionic species discussed thus far in this paper, CO^+ is the only one for which diffusion coefficients are reported. Except for the very lowest values of E/N and, therefore, the highest values of N , the arrival-time spectra of $\text{CO}^+ \cdot \text{CO}$ were influenced by the production of dimer ions in the drift tube and, therefore, could not be adequately represented by Eq. (5). Our experimental values of D_L for $\text{CO}^+ \cdot \text{CO}$ at low values of E/N merely confirmed the prediction of the Einstein equation (2). In the case of C^+ the inadequate statistics that introduced the large random error in the measurement of K made D_L virtually impossible to measure. The measurement of the drift velocity requires only a knowledge of the first moment of the arrival-time distribution, while the measurement

of the longitudinal-diffusion coefficient requires matching the entire arrival-time distribution. Errors due to poor statistics do not affect the determination of the first moment as greatly as they affect the matching of the entire distribution.

Figure 5 shows the diffusion coefficients for CO^+ ions in CO. Since D_L varies as $1/N$, the results are expressed as the product ND_L vs E/N . The data cover the range of E/N from 14 to 660 Td. The values of ND_L for K^+ ions in CO are presented in another paper,²⁰ and are shown here only for comparison with the results for CO^+ . The striking difference between the two sets of data is the very small increase in value of ND_L for CO^+ with increasing E/N when compared with ND_L for K^+ . The slower increase in ND_L for CO^+ may be attributed to the role that resonant charge exchange plays in the total scattering interaction for these ions. Similar behavior has been seen⁴⁻⁶ for N_2^+ in N_2 , O_2^+ and O_2^- in O_2 , and NO^+ in NO.

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¹D. L. Albritton, T. M. Miller, D. W. Martin, and E. W. McDaniel, *Phys. Rev.* **171**, 94 (1968).

²T. M. Miller, J. T. Moseley, D. W. Martin, and E. W. McDaniel, *Phys. Rev.* **173**, 115 (1968).

³J. T. Moseley, I. R. Gatland, D. W. Martin, and E. W. McDaniel, *Phys. Rev.* **178**, 234 (1969).

⁴J. T. Moseley, R. M. Snuggs, D. W. Martin, and E. W. McDaniel, *Phys. Rev.* **178**, 240 (1969).

⁵R. M. Snuggs, D. J. Volz, J. H. Schummers, D. W. Martin, and E. W. McDaniel, *Phys. Rev. A* **3**, 477 (1971).

⁶D. J. Volz, J. H. Schummers, R. D. Laser, D. W. Martin, and E. W. McDaniel, *Phys. Rev. A* **4**, 1106 (1971).

⁷M. Saporoschenko, *J. Chem. Phys.* **49**, 768 (1968).

⁸E. W. McDaniel, *Collision Phenomena in Ionized Gases* (Wiley, New York, 1964), p. 491.

⁹G. H. Wannier, *Bell System Tech. J.* **32**, 170 (1953).

¹⁰D. L. Albritton, D. W. Martin, E. W. McDaniel, T. M. Miller, and J. T. Moseley, Technical Report,

Georgia Institute of Technology, Atlanta, Ga., 1967 (unpublished). Copies available from authors.

¹¹T. M. Miller, D. W. Martin, E. W. McDaniel, J. T. Moseley, and R. M. Snuggs, Technical Report, Georgia Institute of Technology, Atlanta, Ga., 1968 (unpublished). Copies available from authors.

¹²J. T. Moseley, D. W. Martin, E. W. McDaniel, R. M. Snuggs, and T. M. Miller, Technical Report, Georgia Institute of Technology, Atlanta, Ga., 1968 (unpublished). Copies available from authors.

¹³R. M. Snuggs, D. J. Volz, J. H. Schummers, R. D. Laser, I. R. Gatland, D. W. Martin, and E. W. McDaniel, Technical Report, Georgia Institute of Technology, Atlanta, Ga., 1970 (unpublished). Copies available from authors.

¹⁴J. H. Schummers, G. M. Thomson, D. R. James, E. Graham, IV, I. R. Gatland, D. W. Martin, and E. W. McDaniel, Technical Report, Georgia Institute of Technology, Atlanta, Ga., 1972 (unpublished). Copies available from authors.

¹⁵This small correction is given by Eq. (147) on p. 89 of Ref. 12. It results in a downward adjustment of the measured drift velocity by approximately the amount $2\alpha D_L/v_d$ and is too small to have a noticeable effect except at our lowest values of E/N .

¹⁶E. W. McDaniel and D. W. Martin, *Rev. Sci. Instr.* **42**, 157 (1971).

¹⁷D. L. Albritton *et al.*, Ref. 10, p. 129.

¹⁸L. J. Puckett, M. W. Teague, and D. G. McCoy, *Rev. Sci. Instr.* **42**, 580 (1971).

¹⁹J. T. Moseley *et al.*, Ref. 12, pp. 97-99.

²⁰G. M. Thomson, J. H. Schummers, D. R. James, E. Graham, I. R. Gatland, M. R. Flannery, and E. W. McDaniel, *J. Chem. Phys.* (to be published).