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Mobilities and Reaction Rates of Neon Ions in Neon*

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The mobilities of Ne^+ and Ne_2^+ ions in neon have been measured at 300°K. The mobilities of Ne^+ and Ne_2^+ at low E/p_0 are given by the equations $K(Ne^+)=4.07[1+8.0\times10^{3}(E/p_0)^2+2.0\times10^{-6}(E/p_0)^4]^{-1/8}$ and $K(Ne_2^+)=6.14\{[1+8.3\times10^{-3}(E/p_0)^2]/[1+2.0\times10^{-3}(E/p_0)^2+8.4\times10^{-6}(E/p_0)^4]\}^{1/4}$, with E/p_0 in V/cm Torr. The Ne^+ was observed to convert to Ne_2^+ by a three-body reaction with a rate coefficient ranging from about 7×10^{-32} cm^6/sec at $E/p_0=2.0$ V/cm Torr to about 5×10^{-32} cm^6/sec at $E/p_0=6.0$ V/cm Torr.

I. INTRODUCTION

THE set of measurements reported here is an extension to neon of a previous set of measurements made in helium.[1] The quantities measured were the mobilities and diffusion coefficients of the ions produced by a discharge in pure neon and the rate of reaction of Ne^+ with the neon to produce Ne_2^+. In several respects, the results of this experiment represent a significant improvement over previously reported measurements in neon; among other things, these measurements indicate that the interpretation of reported neon mobilities is not as simple as has been previously assumed.

The mobility referred to in this paper is reduced to a standard gas density of 2.69×10^{19} atoms/cm^3. It is defined by

$$K=(v/E)(p_0/760), \tag{1}$$

where v is the ionic drift velocity in cm/sec, E is the magnitude of the applied electric field in V/cm, and $p_0=p(273/T)$ (p is the gas pressure in Torr and T the gas temperature in °K). The measurements reported here were made at $T=300\pm1$°K.

In the case of ions reacting with the gas, the rate of loss of the reactant ions can be expressed as

$$\partial\rho/\partial t=-\alpha\rho, \tag{2}$$

where ρ is the ion density and α is the reaction frequency in sec^{-1}. For the conversion of Ne^+ to Ne_2^+, the quantity α will be referred to as the attachment frequency.

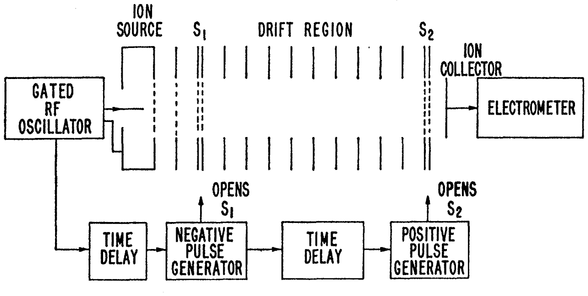

FIG. 1. Schematic illustration of the experimental apparatus. S_1 and S_2 are electrical shutters.

II. APPARATUS AND DATA ANALYSIS

The apparatus and data analysis routine were the same as used in the analogous measurements in helium and have been described in detail in a previous article[1] so that only a brief review is included here. Figure 1 shows a schematic diagram of the mobility tube and some of the associated electronics. The mobility tube consists of an ion source, a 10-cm-long drift region where a uniform electric field is maintained, and an ion collector. Two electrical shutters S_1 and S_2 are located at either end of the drift region. These shutters are biased to prevent the passage of positive ions. At appropriate times each shutter can be opened by reversing the field using a square voltage pulse applied to one of the grids in the shutter pair. A cycle of operation begins by striking a discharge in the source region to produce some ions. Positive ions in the afterglow diffuse out of the source region and drift toward shutter S_1. At some variable time after the discharge, S_1 is opened and a well-defined pulse of ions is let into the drift region. These ions drift to the vicinity of shutter S_2, where they can pass through to the collector only if this shutter is opened at the right time. The data consist of tabulations of the average current to the collector as a function of the time delay between the opening of shutters S_1 and S_2.

The situation is rather well described by assuming that the spatial distribution of ions after passing S_1 is a δ function, and the data are the distribution of arrival times of these ions at the second shutter. Figure 2 shows a sample set of data with the experimental values represented by dots. The continuous curves are the results of calculations and their sum provides a fit to the data. For ions which do not react with the gas, the distribution of arrival times at the second shutter is approximately a Gaussian. The mean of this distribution provides a value for the average time of flight of the ions, and from this value the drift velocity and mobility can be calculated. The width of the distribution provides a value for the diffusion coefficient of the ions. In Fig. 2, two Gaussian peaks are indicated. These correspond to two different ion species having different times of flight through the drift

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[1] E. C. Beaty and P. L. Patterson, Phys. Rev. 137, A346 (1965).

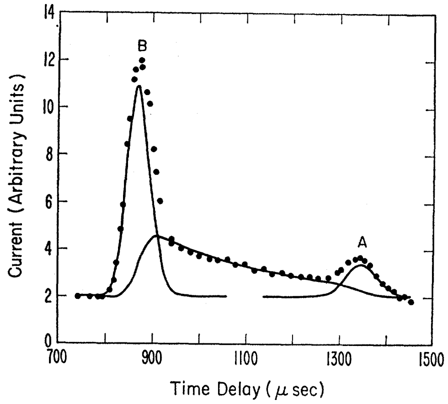

FIG. 2. Typical ion current measured at the collector versus the time delay between the openings of the shutters. Dots are data points; the solid curves are the results of a numerical decomposition of the data into the currents due to the different kinds of ions. Ion A is identified as Ne^+ and ion B as Ne_2^+. The current at intermediate time delay is due to Ne_2^+ produced by a reaction of Ne^+ with the gas. For this data $p_0=4.12$ Torr, $E/p_0=2.42$ V/cm Torr. The results of the numerical analysis for this data were: $K(Ne^+)=4.07$; $K(Ne_2^+)=6.30$; $\beta=6.5$; the measured diffusion coefficients were larger than the thermal values [Eq. (5)] by 7% (Ne^+) and 44% (Ne_2^+).

region. There is also some secondary ion current present, occurring at time delays between the times of flight of ions A and B. This current is the result of ion A reacting in flight to form ion B. The distribution of this secondary ion current is approximately a segment of an exponential, and in the limit of negligible diffusion the decay constant is simply related to the reaction rate constant. Hence, for a set of data such as that shown in Fig. 2, a relatively simple analysis suffices to extract approximate values for the mobilities and the reaction rate. However, the data were actually subjected to more complex numerical analysis to obtain values for diffusion coefficients as well and in order to study extraneous effects such as space charge. As described in a previous paper,[1] the continuity equation for the ions has been solved, including diffusion, drift, and a unidirectional reaction. The experimental data were numerically fitted to the solutions, with the mobilities, diffusion coefficients, and the reaction rate treated as independent variables. The method accounted completely for diffusion in the direction of the field and included a correction for the times of flight across the shutters. Radial diffusion was only partially accounted for, and space charge was not taken into account at all. Qualitatively, space charge has little effect on the mobility determinations, but it causes the diffusion-coefficient determinations to be too large and has an unpredictable effect on the reaction rate. The increase of the diffusion coefficients over the expected values provided some measure of the space-charge effects.

Since contaminants having a low ionization potential are preferentially ionized, it was necessary to take precautions to secure and maintain purity of the neon gas. The electrode assembly was sealed in a Pyrex envelope connected to a bakeable ultrahigh vacuum system. The source of neon was a commercial Pyrex flask, and the gas was further purified by a cataphoresis discharge (see Ref. 1). During operation, the system was disconnected from the pumping station by a mercury cutoff. Gas pressures were measured by either a mercury manometer or a mercury McLeod gauge; and a liquid-nitrogen trap served to prevent mercury vapor from contaminating the gas. When used with extreme care, the mercury manometer readings were reproducible to a precision of ±0.02 Torr. The McLeod gauge could be used for pressures as high as 7.0 Torr and was used to extend the range of measurement down to about 1.0 Torr.

III. DATA

The measured mobilities of Ne^+ and Ne_2^+ are shown in Fig. 3 as a function of E/p_0. The slower ion could be identified with certainty as Ne^+ because of its unique mobility. At room temperature, the Ne^+ mobility is largely controlled by resonant charge transfer with the neutral gas atoms. Thus the Ne^+ ion has a mobility that is considerably lower than any other ion in neon.

To assist in an extrapolation of the Ne^+ mobilities to lower E/p_0 and to otherwise characterize the data in a convenient way, a numerical fit was made to the

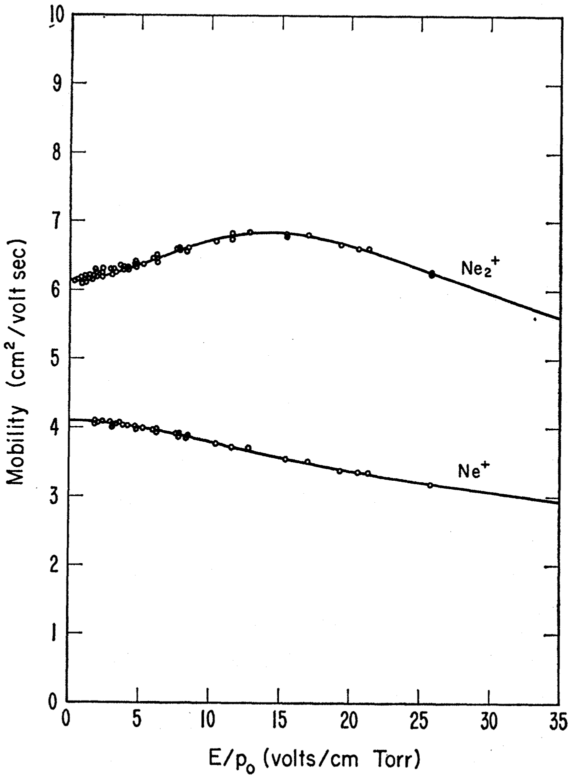

FIG. 3. Results of mobility determination. The solid lines are analytical representations of the data. The equations are given in the text.

function

$$K=K_0/[1+a_1(E/p_0)^2+a_2(E/p_0)^4]^{-1/8}. \qquad (3)$$

The solid line in Fig. 3 represents this function with $K_0=4.07$ cm²/(V sec), $a_1=8.0\times10^{-3}$ (cm² Torr²/V²), and $a_2=2.0\times10^{-6}$ (cm⁴ Torr⁴/V⁴). Since $v(E)=-v(-E)$, it can be seen that the mobility is an even function of electric field. The scattering of a Ne^+ ion by a Ne atom can be reasonably well represented by the hard-sphere model (isotropic scattering with a cross section independent of energy), especially at high energies. Wannier[2] has shown that for this model, the mobility at high E/p_0 goes as $(E/p_0)^{-1/2}$. Equation (3) is an even function of E with the desired asymptotic form. The data reported here do not extend to sufficiently high E/p_0 to determine the asymptotic limit. The values of constants K_0, a_1, and a_2, given above, were chosen so that Eq. (3) provides a good representation of our data and also fits well the data reported by Hornbeck[3] which extends to quite high E/p_0. In addition, this equation is within the experimental error of the other recognized determinations of the mobility of Ne^+ in Ne.[3-7] The zero-field mobility K_0 is also consistent with theoretical estimates.[8-10]

At pressures less than about 6 Torr, the Ne^+ was observed to react with the gas while in flight through the drift tube. The product of this reaction was the ion species we have called Ne_2^+. (The conclusion of this paper is that the ion is indeed the ground electronic state of Ne_2^+. However, it is impossible to be certain and much of the other data is valid even if this conclusion should be incorrect.) At pressures greater than 6 Torr, the reaction proceeded so fast that only the production was observed in the mobility spectrum. The rate of this reaction was determined from the data, and the results are presented in a later paragraph. It was concluded from a study of the data presented below and the analogy with helium that the observed reaction was the three-body attachment

$$Ne^+ + 2Ne \rightarrow Ne_2^+ + Ne. \qquad (4)$$

This conversion of Ne^+ into Ne_2^+ is qualitatively analogous to that previously reported for ions in helium.[1] Madson, Oskam, and Chanin[11] have recently confirmed the helium-ion identifications, using a direct mass analysis.

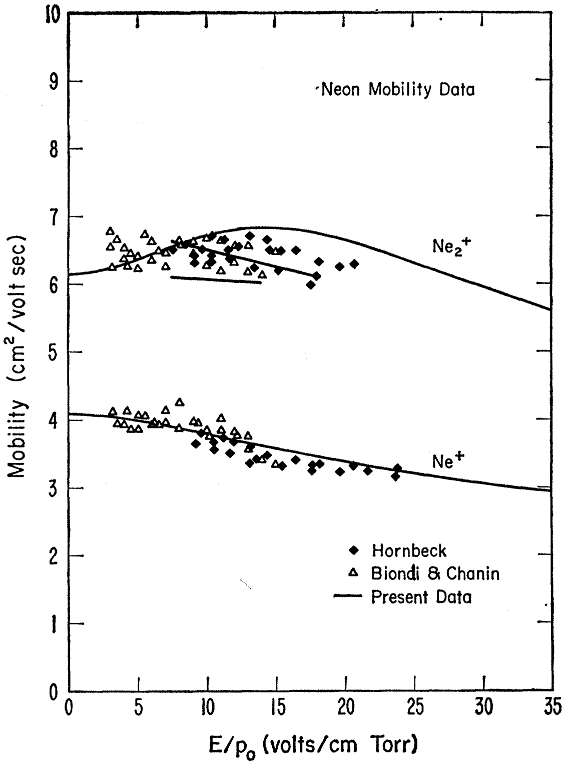

FIG. 4. Comparison of present measurements with some previous data (Refs. 3 and 4). The data curves for Ne^+ and Ne_2^+ mobilities are labeled. The ions represented by the two short lines are unidentified.

To achieve an analytic representation of the Ne_2^+ data, it was necessary to use an equation more complicated than Eq. (3). The curve through the data of Fig. 3 is a graph of

$$K=K_0\{[1+b_1(E/p_0)^2]/[1+b_2(E/p_0)^2+b_3(E/p_0)^4]\}^{1/4},$$

with

$$K_0=6.14 \text{ cm}^2/(\text{V sec}),\ b_1=8.3\times10^{-3} \text{ cm}^2\text{Torr}^2/\text{V}^2,$$
$$b_2=2.0\times10^{-3} \text{ cm}^2 \text{ Torr}^2/\text{V}^2,$$

and

$$b_3=8.4\times10^{-6} \text{ cm}^4 \text{ Torr}^4/\text{V}^4.$$

The mobility determinations at low E/p_0 are the most accurate, making the extrapolation to $E/p_0=0$ relatively reliable.

Data on the reaction of Ne^+ with the neon gas are displayed in Table I. The attachment frequency α was determined by the use of the analysis program as the value providing the best fit to the data. For the three-body reaction (4), α is expected to be proportional to the square of the gas density. Hence, the rate coefficient $\beta=\alpha/n^2$ was also computed and is displayed in the fourth column of Table I. (The authors have pre-

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[2] G. H. Wannier, Bell System Tech. J. **170**, 84 (1952).

[3] J. A. Hornbeck, Phys. Rev. **84**, 615 (1951).

[4] M. A. Biondi and L. M. Chanin, Phys. Rev. **94**, 910 (1954).

[5] L. M. Chanin and M. A. Biondi, Phys. Rev. **106**, 473 (1957).

[6] H. J. Oskam and V. R. Mittelstadt, Phys. Rev. **132**, 1435 (1963).

[7] R. Hackham, Brit. J. Appl. Phys. **2**, 191 (1966).

[8] T. Holstein, J. Phys. Chem. **56**, 832 (1952).

[9] Yu. P. Mordvinov and B. M. Smirnov, Zh. Eksperim. i Teor. Fiz. **48**, 133 (1965) [English transl.: Soviet Phys.—JETP **21**, 91 (1965)].

[10] A. Dalgarno, Phil. Trans. Roy. Soc. London **A250**, 428 (1958).

[11] J. M. Madson, H. J. Oskam, and L. M. Chanin, Phys. Rev. Letters **15**, 1018 (1965).

viously published[12] a preliminary value for the rate coefficient based upon an analysis of only a portion of the data. The rate coefficients listed in Table I supercede this earlier value. The previous data is consistent with that reported here, except that the explicit dependence on E/p was not recognized.)

The rate coefficients listed in Table I are plotted versus E/p_0 in Fig. 5. There is systematic dependence of β on E/p_0. This indicates that the applied electric field causes some preferential "heating" of the ions even at these relatively low values of E/p_0. This heating also caused increased values for the observed diffusion coefficients for both Ne^+ and Ne_2^+.

In addition to ion mobilities and the reaction rate, the data analysis yielded values for the diffusion coefficients of the ions. For very small values of E/p_0, the distribution of ion velocities is only slightly different from a Maxwellian. In this limit the diffusion coefficient D is a scalar related to the mobility by

$$K=15.27Dp_0/T, \tag{5}$$

where D is in cm²/sec, p_0 in Torr, and T in °K. It is in this limit that the equations used in the data analysis are appropriate. At higher values of the electric field the concept of diffusion is still useful, but the diffusion coefficient must be a tensor rather than a scalar. In the present application, however, the concern is almost entirely with diffusion in the direction of the electric field. As a result, the analysis equations are still applicable, in the absence of a reaction, even at higher E/p_0 with a value of the diffusion coefficient somewhat larger than that given by (5). The increase in apparent diffusion coefficients can be interpreted as an increase in ion "temperature." If the two ions have appreciably different "temperatures" radial diffusion may cause errors in the reaction rate determinations.

No explicit effort was made to get accurate data on the diffusion coefficients; the study was conducted to check consistency. The most serious source of systematic error in the diffusion data was space-charge effects. In the limit of low E/p_0 and small ion densities, the diffusion coefficients satisfied Eq. (5) within a tolerance of ±20%. At $E/p_0=20$ V/cm Torr, the diffusion coefficients for Ne^+ and Ne_2^+ were, respectively, 1.6 and 3.0 times the values given by Eq. (5). The estimated uncertainties were about 20%. Within this tolerance the diffusion coefficients were linear functions of E/p_0 in the range 0–20. (Since the diffusion coefficient is an even function of E, this linear dependence cannot be accurate for small values of E/p_0.)

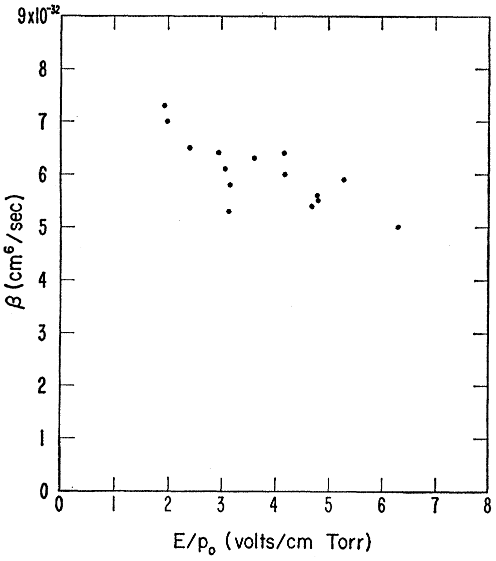

FIG. 5. Data for the three-body rate constant for the reaction of Ne^+ with Ne to make Ne_2^+. The dependence on E/p_0 is presumed to be a manifestation of a temperature dependence of the rate constant.

TABLE I. Reaction rate data for the reaction $Ne^+ + 2Ne \rightarrow Ne_2^+ + Ne$.

p_0 (Torr)	E/p_0 (V/cm Torr)	α (1/sec)	β (10^{-32} cm⁶/sec)
2.58	1.95	602	7.3
3.16	3.15	664	5.3
3.16	6.30	622	5.0
3.17	4.71	680	5.4
3.35	2.02	979	7.0
3.35	2.96	901	6.4
4.12	2.42	1377	6.5
4.12	3.63	1332	6.3
4.70	3.18	1614	5.8
4.70	4.23	1664	6.0
4.70	5.30	1628	5.9
5.13	3.07	2010	6.1
5.17	4.81	1828	5.5
5.17	4.81	1864	5.6
5.93	4.19	2805	6.4

IV. DISCUSSION

A first-order approximation to the solution of the Boltzmann equation[13] provides a relation between the diffusion cross section Q_d and the mobility:

$$K=\frac{2.71\times10^{-8}}{Q}\left(\frac{1}{\mu T}\right)^{1/2}. \tag{6}$$

The quantity μ is the reduced mass of ions and gas molecules (in grams), and Q is given by the expression

$$Q=\frac{1}{2}\int_0^\infty x^2Q_d(x)e^{-x}dx,$$

where $x=\mu v^2/2kT$ and v denotes the relative velocity. For the Ne^+ collisions with Ne, the diffusion cross

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[12] E. C. Beaty and P. Patterson, in *Proceedings of the Sixth International Conference on Ionization Phenomena in Gases*, edited by P. Hubert and E. Cremieu-Alcan (S.E.R.M.A., Paris, 1964), Vol. III, p. 289.

[13] S. Chapman and T. G. Cowling, *The Mathematical Theory of Nonuniform Gases* (Cambridge University Press, New York, New York, 1960), p. 165.

section (Q_d) is approximately twice the charge transfer cross section.[10] Assuming that the charge transfer cross section is independent of energy, Eq. (6) and the mobility data require that it be $47\pi a_0^2$.

No very accurate theory is available for the mobility of Ne_2^+ ions in neon. Treating the Ne_2^+ as spherical and taking into account only a polarization interaction, a mobility of 5.99 cm^2/V sec is obtained. Including additional terms in the interaction causes the mobility to have a dependence on gas temperature and E/p_0. At 300°K, the low-field mobility (K_0) reported here is somewhat higher than the calculated polarization limit, and the observed increase of mobility with E/p_0 indicates that a positive slope with temperature is to be expected.

Many previous measurements of the mobilities of neon ions in neon have been reported.[3–7] However, in the case of the Ne_2^+ ion, it is not clear that all experiments were actually dealing with the same ion species. The standard technique has been to get "pure" neon, ionize it, and sort the ions for mobility. Generally two mobilities have been found. One could be identified with certainty as Ne^+, and the other was presumed to be ground-state Ne_2^+. In analogous measurements in helium, however, this procedure has been shown conclusively to be inadequate for identifying the molecular ion.[11,12,14–16] The problem is that it is possible to preferentially produce other ion species as well. In addition to the possibility of producing impurity ions, there is the possibility of producing the molecular ions in an excited state or even doubly charged ions.

The ion we have identified as Ne_2^+ is that ion which is the product of a reaction of the Ne^+ with the gas. Ions of this same mobility are also produced in the discharge and are presumed to be the same as those formed from the Ne^+ reaction. However, by a judicious choice of discharge conditions, two other ions with mobilities very close to that of Ne_2^+ could be produced in the purest neon available. Our data on the mobilities of these two ions are shown in Fig. 4 along with the Ne^+ and Ne_2^+ mobilities. The identities of these two ions are unknown. Both were produced only in a spark discharge and disappeared shortly after the breakdown. Two possible sources of these ions cannot be ruled out. The discharge conditions permitted the production of ions in excited states or doubly charged ions which may have survived long enough. Also impurities sputtered from the electrodes may have become ionized in sufficient quantities to have been observed. Failure to identify these two ions has no bearing on the validity of the identification of Ne^+ and Ne_2^+; hence their mobilities have no bearing on the main part of this paper. The only reason for including data on these unidentified ions is the possibility that they may be helpful in interpreting other experimental results.

For comparison with our data, there are included in Fig. 4 some of the measurements that have been reported previously for Ne_2^+.[3,4] In view of the existence of several ion types with approximately the same mobility, it is impossible to tell from the mobility data alone which of the ions in our experiment correspond to the ions studied in the previous experiments. Only in the case of Hornbeck's measurements was a more controlled method of producing the ions employed. Hornbeck obtained his mobilities from an analysis of transient currents in the gas following a photoelectrically induced avalanche. As a result, Hornbeck was at least able to rule out the possibility that he was dealing with doubly charged ions.

The discharge conditions could be chosen so that the unidentified ions were not produced. In addition, the rather complicated data-analysis procedure used for the data reported here permits distinguishing two ions of different mobilities even when the mobilities are very close together. Thus, there was no difficulty in separating the ion here identified as Ne_2^+ from other ions.

The measured rates of reaction of (4) that are listed in Table I are consistent with theoretical estimates. Thomson[17] considered the general problem of complex ion formation using the assumption that the binding forces are caused by the charge interacting with an induced dipole. Using his formulas, a value of $\beta=10^{-31}$ cm^6/sec is obtained for reaction (4). In the previous article on helium,[1] the present authors have tried to compute the attachment frequency of ions where the dominant scattering mechanism is charge transfer. The technique involved approximating the rate at which two atoms and an ion get close enough together to permit rapid charge transfer among the three particles. The reaction rate was assumed to be this collision rate multiplied by a parameter s which denoted the fraction of these collisions resulting in a bound molecular ion. Setting $s=0.4$ in formula (18) of Ref. 1, the calculated rate for reaction (4) is 6.4×10^{-32} cm^6/sec.[18] Mahan[19] has attempted to improve on the Thomson formulation of the problem by including some of the effects of charge transfer. His result for reaction (4) is 6.2×10^{-32} cm^6/sec.

While all three of the above theoretical treatments contain some serious deficiencies, they can nevertheless be expected to give correct order-of-magnitude estimates of the reaction rate. The same cannot be said for the theoretical treatment put forth by Niles and

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[14] Kenneth B. McAfee, Jr., D. Sipler, and D. Edelson, Phys. Rev. **160**, 130 (1967).

[15] E. C. Beaty, J. C. Browne, and A. Dalgarno, Phys. Rev. Letters **16**, 723 (1966).

[16] E. C. Beaty, in *Proceedings of the Fifth International Conference on Ionization Phenomena in Gases* (North-Holland Publishing Co., Amsterdam, 1961), Vol. I, p. 183.

[17] J. J. Thomson, Phil. Mag. **47**, 337 (1924).

[18] In applying this formula to the formation of He_2^+, a numerical error was made. To duplicate the experimental He_2^+ results, the value of s required is 0.4 instead of 4.0 as reported in Ref. 1.

[19] B. H. Mahan, J. Chem. Phys. **43**, 3080 (1965).

Robertson,[20] who attempted to make an *a priori* calculation of the rate of the inverse of reaction (4) under thermal equilibrium conditions. This calculation requires some assumption about the cross section for the dissociation of molecular ions upon impact by atoms. Niles and Robertson implicitly assumed that this cross section was a constant above threshold. Furthermore, all the dissociation was assumed to result directly from collisions of ground-state ions. In particular, there was no allowance for any vibrational excitation of the molecular ion. Until the dependences of the cross section on energy and vibrational states are known, it is doubtful that this method can be relied upon as even an order-of-magnitude estimate of the reaction rate.

It is necessary to consider the possibility that reaction (4) was not the only reaction occurring under the experimental conditions. The most serious possibility is a charge-exchange reaction of the Ne^+ with an impurity. The rate of such a reaction would depend linearly on the concentration of the impurity. This concentration could be expected to vary a great deal with gas sample and with time. No such variation was observed.

Measurements of the rate of reaction (4) have been reported on two previous occasions. Hackham[7] reported a rate coefficient of 1.6×10^{-32} cm^6/sec at $T=294$°K, while Sauter, Gerber, and Oskam[21] reported a value of 4.2×10^{-32} cm^6/sec at $T=335$°K. Both of these values were deduced from studies of decaying plasmas in late afterglows of neon. Our experimental technique was quite different, and our rate coefficients listed in Table I are all larger than the values reported from the afterglow studies.

V. UNCERTAINTIES

A principal source of uncertainty in the mobilities at high E/p_0 values was in the determination of pressures. Fields greater than 25 V/cm were not used because of problems of breakdowns occurring inside the tube. The lowest pressure used was 0.96 Torr, and the uncertainty in the pressure measurements was estimated to be ±0.02 Torr. At very low E/p_0 values, pressures as high as 20.00 Torr were used, and the pressure measurements were quite reliable. Fields less than 5 V/cm were not used because of the uncontrolled effects of stray fields and space charge. The uncertainty in the Ne_2^+ mobilities at low E/p_0, due mainly to a lack of definition of the drift distance, is estimated to be about 1%. For Ne^+, mobility data could not be obtained at E/p_0 values less than about 2.0 because of the high rate of conversion of the Ne^+ to Ne_2^+.

The fraction of the atomic ions which get through the drift space is $\exp(-\alpha d/v_1)$, where v_1 is the Ne^+ drift velocity and d is the drift distance. The argument of this exponential must be of the order of unity for the reaction rate measuring technique to work well. For small values of $\alpha d/v_1$, noise is a significant contributor to the uncertainty. The noise originated in the ion source and was coupled to the ion collector by a photoelectric effect. Most of the photoelectric effect was eliminated from the data by subtracting a collector-current measurement made with the first shutter biased past cutoff. Noise in the photoelectric current was not removed, and for some of the data there was a small spurious component of collector current which was changed by the bias change. This incomplete correction for the background effect caused the reaction rate to be systematically low for low $\alpha d/v_1$. For large values of $\alpha d/v_1$, the data were rather insensitive to the reaction rate. Space charge and inadequate accounting for radial diffusion contribute other possible systematic errors in the rate coefficient measurements. An uncertainty of about 20% is estimated for the rate coefficients listed in Table I.

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[20] F. E. Niles and W. W. Robertson, J. Chem. Phys. **40**, 3568 (1964).

[21] G. F. Sauter, R. A. Gerber, and H. J. Oskam, Phys. Norvegica. **32**, 1921 (1966).