Charge transfer between helium ions and metastable neon

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Absolute and relative cross sections were obtained for the charge-transfer reaction $He^+(1S) + Ne^* \rightarrow He^\ddagger + Ne^+$. The Ne* represents a composite of $Ne(3s\,^3P_2)$ and $Ne(3s\,^3P_0)$, and the $He^\ddagger$ represents various excited states of He. The studies were made by a merging-beams technique for a relative kinetic energy W of the reactants from 0.1 to 500 eV. The data indicate that the reaction is directed with most of the product Ne^+ scattered in the direction of the reactant Ne*. The cross section monotonically increases with W. The threshold for the reaction is near 0.1 eV. A modified Demkov approach is used to calculate cross sections, which agree very roughly with the experimental values above $W \approx 3$ eV. At lower W the agreement is poor.

I. INTRODUCTION

Experimental charge-transfer studies of ground-state reactants in the energy range from thermal to 10 eV are difficult and, as a result, not very numerous. Investigations involving excited reactants are even more scant.

In this paper we discuss an experiment and calculations involving charge transfer of ground-state ions with metastables covering this energy range and beyond. In particular, the reaction we have investigated is

$$He^+(1S) + Ne^* \rightarrow He^\ddagger + Ne^+ . \quad (1)$$

The Ne* represents a composite of $Ne(3s\,^3P_2)$ and $Ne(3s\,^3P_0)$. No state selection was made. The $He^\ddagger$ represents excited He, and Ne^+ is either $Ne^+(2p^5\,^2P_{3/2})$ or $Ne^+(2p^5\,^2P_{1/2})$.

Studies of the reaction were made in the range $0.1 \leq W \leq 500$ eV (where W is the interaction energy, or relative kinetic energy of the reactants) by measuring the product Ne^+ current and lab-energy distributions of Ne^+. In this range of W there are a number of exit channels, all of which result in endothermic processes. The contributing channels which we used in developing theoretical cross sections for reaction (1) will be listed later. Laboratory energies of different species will be designated by E with an appropriate subscript. For example, the lab energy of Ne* will be E_{Ne^*}.

II. EXPERIMENTAL

A schematic of the merging-beams apparatus has been shown previously.[1(a)] Only minor modifications of this schematic apply to the present experiment, and these have been described in two other papers.[1(b),1(c)] The general technique for obtaining data and the procedure for extracting cross sections from lab-energy distributions have been discussed.[1(a),1(d)]

With the aid of the schematic mentioned above a brief description of the experiment will be given. Helium ions were generated in the electron-impact source 1 and, after mass analysis in the merging magnet, passed into the interaction region. Neon ions were generated in the electron-impact source 2. After mass analysis in the analyzing magnet, the Ne^+ passed into a charge-transfer cell containing Na vapor at a pressure of about 0.8 mTorr. Some of the Ne^+ was converted to Ne* in this cell by near-resonant charge transfer. The remainder of the Ne^+ was either converted to ground-state Ne or did not react and was collected on the condenser plates following the cell. The neon-neutral beam from the cell then merged with the He^+ beam from source 1 in the merging magnet, and finally both beams entered the interaction region where Ne^+ was generated by reaction (1). The Ne^+ was detected in the detector assembly, which consisted, among other components, of a 180° spherical electrostatic condenser (that acted as an energy analyzer) and a Johnston MM-1 electron multiplier. Beam chopping resulted in a modulated output of the multiplier, which was fed into a lock-in amplifier followed by a recording system.

A potential P was applied to the interaction region so that Ne^+ formed inside this region would, upon leaving it, have a different energy than Ne^+ formed outside. The detector assembly could then be set to accept only Ne^+ formed inside.

For $W \leq 30$ eV, E_{He^+} inside the interaction region was fixed at either 1000 or 1100 eV, whereas for $W > 30$ eV, it was fixed at 1650 eV. The lab energy of Ne^+ from source 2 was adjusted to give the desired W.

The pressure in the interaction region, which was surrounded by a stainless-steel can pumped by Ti sublimation, was approximately 4×10^{-9} Torr.

III. BEAM COMPOSITION

Some studies of reaction (1) were made with pure beams of $He^+(1S)$ by maintaining the energy V_e of the ionizing electrons below the threshold of $He^+(2S)$ production, viz., 65.4 eV. However, many of the data were taken with $V_e \approx 150$ eV because more intense He^+ beams could be achieved.

To observe the effect of the higher V_e, a composite cross section for the reaction was measured with $V_e \approx 150$ eV and compared with the cross section for $V_e < 65.4$ eV. This was done at several W's. The cross sections were the same within experimental error. This indicates that either the cross section for $He^+(1S)$ and $He^+(2S)$ are the same at these W's or only a small percentage of $He^+(2S)$ is emitted from the source for $V_e \approx 150$ eV. Since the former possibility is extremely unlikely, we conclude that a virtually pure beam of $He^+(1S)$ is achieved with $V_e \approx 150$ eV.

The Ne beam consisted of Ne* and Ne ground-state [i.e., $Ne(2p^6\,{}^1S_0)$] particles. For our purposes, then, its composition can be given by the fraction f_g of $Ne(2p^6\,{}^1S_0)$. The fraction of $Ne({}^3P_{2,0})$ is $(1-f_g)$. As determined by an experimental technique described previously, $f_g = 0.57$.[2]

The ratios $Ne({}^3P_2):Ne({}^3P_0):Ne({}^1S_0)$ in the Ne beam have been previously calculated by a statistical-weight method.[2] These ratios are 5:1:6. Thus the calculated $f_g = 0.5$ is in good agreement with the experimentally determined value.

To observe the effect in our experiments of the ground-state component of the Ne beam, we merged $He^+(1S)$ and pure $Ne(2p^6\,{}^1S_0)$ beams at various W and searched for Ne^+ signals. These were negligible, and we conclude that charge transfer between $He^+(1S)$ and ground-state Ne [as well as collisional ionization of ground-state Ne by $He^+(1S)$] is small compared with charge transfer between $He^+(1S)$ and Ne*. The ground-state Ne beam was obtained by substituting Ne gas for Na vapor in the charge-transfer cell.

IV. THEORY

An appropriate theory for charge transfer in a system like that in reaction (1) has been developed by Demkov.[3] In such systems the energy separation of the potential curves at large internuclear distances is relatively small and large reaction cross sections exist.

To calculate cross sections that can be compared with our experimental values, we have used a modified Demkov approach developed by Olson[4] and subsequently employed by Olson and Smith[5] and Ice and Olson.[6] The method has been applied to the following reactions in which He in the 2S state is a product, the reactants and products can form molecular electronic states of the same species [under the Λ, S coupling scheme, Hund's case (a)],[7] and the angular momentum of the $2p^5$ core of $Ne(3s\,{}^3P_2)$ and of $Ne(3s\,{}^3P_0)$ is conserved. It is reasonable to assume that the angular momentum of this core is not perturbed since charge transfer occurs at large internuclear separations. A similar conservation rule has been applied, for example, by Ice and Olson.[6] The endothermicity ΔE of each reaction is shown.

$$He^+(1\,{}^2S_{1/2}) + Ne(3s\,{}^3P_2) \rightarrow \begin{cases} He(2\,{}^3S_1) + Ne^+(2p^5\,{}^2P_{3/2}) - 0.18 \text{ eV} & \text{(1a)} \\ He(2\,{}^1S_0) + Ne^+(2p^5\,{}^2P_{3/2}) - 0.97 \text{ eV}; & \text{(1b)} \end{cases}$$

$$He^+(1\,{}^2S_{1/2}) + Ne(3s\,{}^3P_0) \rightarrow \begin{cases} He(2\,{}^3S_1) + Ne^+(2p^5\,{}^2P_{1/2}) - 0.18 \text{ eV} & \text{(1c)} \\ He(2\,{}^1S_0) + Ne^+(2p^5\,{}^2P_{1/2}) - 0.98 \text{ eV}. & \text{(1d)} \end{cases}$$

A cross section was calculated for each of the reactions (1a)–(1d) using the modified Demkov approach mentioned above. The required dipole polarizabilities of excited Ne and He were obtained from papers by Olson and Smith[5] and by Robinson *et al.*[8] To obtain a total cross section Q for He(2S) production the individual cross sections for (1a) through (1d), i.e., Q_a through Q_d, were added with appropriate weighting factors C. Thus

$$Q = \tfrac{5}{6}(C_a Q_a + C_b Q_b) + \tfrac{1}{6}(C_c Q_c + C_d Q_d).$$

Reactions (1a) and (1b) were each weighted five times as much as (1c) and (1d) because the ratio of the beam intensity of $Ne(3s\,{}^3P_2)$ to that of $Ne(3s\,{}^3P_0)$ was assumed to be 5 (i.e., the ratio of the statistical weights). This assumption is based on the previously mentioned statistical-weight calculation, which provided an f_g in good agreement with the measured value.

The C's are associated with statistical weights of the exit channels, i.e., $2J+1$. Thus

$$Q = \tfrac{5}{6}(\tfrac{3}{4}Q_a + \tfrac{1}{4}Q_b) + \tfrac{1}{6}(\tfrac{3}{4}Q_c + \tfrac{1}{4}Q_d),$$

and finally

$$Q = \tfrac{1}{24}(15Q_a + 5Q_b + 3Q_c + Q_d).$$

Our rather unsophisticated, statistical approach can only be expected to result in a crude solution to this complex problem. Nevertheless, a rough theoretical Q could be useful, and our method does

simplify the problem. As W increases, statistical weights should reflect probabilities more accurately.[5]

Reactions leading to He in allowed $2P$ states are more endothermic than those resulting in He($2S$). Cross sections for such reactions were calculated and added in a fashion similar to that above to give a total cross section for He($2P$). This cross section was only 20% of the Q for He($2S$) at $W = 500$ eV and less at lower W. The inclusion of the He($2P$) cross section in our final calculation was not warranted because the error caused by its omission is probably not as large as errors introduced from our statistical approach.

Further discussion of our calculations and comparison with experimental results will be given later.

V. RESULTS AND DISCUSSION

A. Energy distributions

Laboratory-energy distributions of Ne^+ production from $He^+(1S) + Ne^*$ are shown in Fig. 1. As mentioned previously, the potential P was applied to the interaction region so that the detector assembly could be adjusted to accept Ne^+ formed inside the region and reject Ne^+ formed outside. The relation between $E^{in}_{Ne^+}$, the energy of Ne^+ generated inside the interaction region, and E_{Ne^+}, the energy of the Ne^+ after it leaves the region, is $E^{in}_{Ne^+} = E_{Ne^+} - P$.

The energy resolution of the detector is 0.95%. Thus, if the product Ne^+ is monoenergetic at energy E_{Ne^+}, the full width at half maximum (FWHM) of the distribution will be $(9.5\times10^{-3})E_{Ne^+}$.

In Fig. 1 the E_{Ne^+} at the center of the distribution for $W = 400$ eV is approximately 2448 eV, and the FWHM $\approx$ 23 eV. Since $(9.5\times10^{-3})2448$ eV $= 23$ eV, we conclude that this distribution is monoenergetic. It is also noted that at the center $E_{Ne^+} \approx E_{Ne^*} + P$ and, hence, $E^{in}_{Ne^+} \approx E_{Ne^*}$. Other distributions in the figure that appear to be monoenergetic except for a rather low-intensity, high-energy tail are those for $W = 1$, 10, and 30 eV. At the center of the monoenergetic part of these distributions $E_{Ne^+} \approx E_{Ne^*} + P$.

For a better understanding of such distributions Newton diagrams for reaction (1) are shown in Fig. 2. These diagrams are drawn for the case when the lab velocity of He^+, $|\vec{v}_{He^+}|$, is greater than the lab velocity of Ne*, $|\vec{v}_{Ne^*}|$. Shown is an assumed scattering sphere for Ne^+ in the center of mass (c.m.) system.

As will be shown later, reaction (1a) dominates the charge-transfer process. This reaction will be used to explain the distribution. We will des-

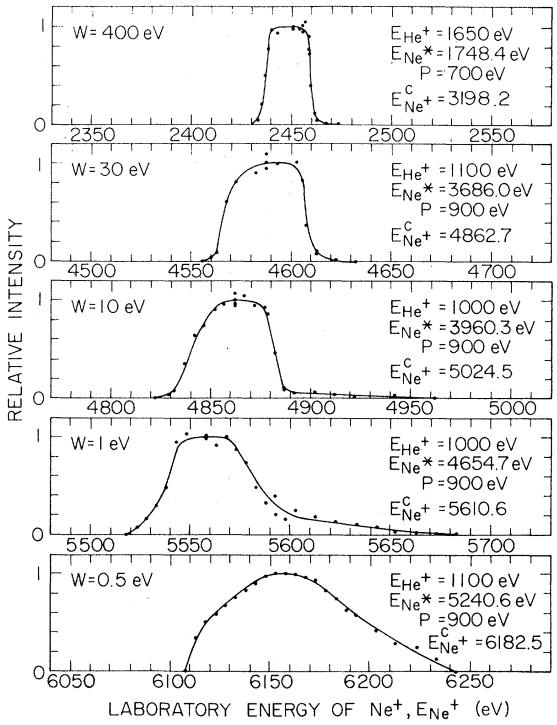

FIG. 1. Laboratory-energy distributions of Ne^+ production from $He^+(1S) + Ne^*$ for $He^+(1S)$ faster than Ne*. The energy of Ne^+ inside the interaction region is $E_{Ne^+} - P$, where E_{Ne^+} is the energy of the Ne^+ after it leaves the region, and P is the potential applied to the interaction region. When Ne^+ leaves the interaction region, it gains an energy equal to P. The energy of the reactant $He^+(1S)$ inside the interaction region is E_{He^+}; outside the region it is $E_{He^+} + P$. The energy of Ne^+ (inside the interaction region) if it had the velocity of the c.m. is $E^c_{Ne^+} - P$.

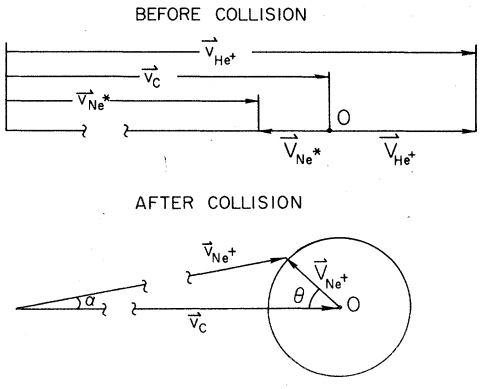

FIG. 2. Newton diagrams for $He^+(1S) + Ne^* \rightarrow He^\ddagger + Ne^+$. Subscript c refers to the c.m., $\vec{v}$ is for laboratory velocity, and $\vec{V}$ is for velocity in the c.m. system. The scattering angle in the lab system is α; in the c.m. system it is θ. When Ne^+ is in the same direction as Ne*, $\alpha = \theta = 0°$. The drawing is not to scale.

ignate W' as the relative kinetic energy of the products of this reaction. The scattering sphere shown in Fig. 2 is associated with this W'. The relation between W and W' is that $W' - W = \Delta E$, where ΔE is the endothermicity of the reaction. Each distribution for $W \geq 1$ eV can be explained by assuming that Ne^+ is scattered on the surface of the sphere and that $\theta = 0°$ for the monoenergetic part of the distribution and $0° < \theta \leq 180°$ for the tail. The E_{Ne^+} at the center of the monoenergetic part can easily be calculated and for each W is very close to (but slightly larger than) $E_{Ne^*} + P$. As noted above, this is the case in Fig. 1 for $W \geq 1$ eV.

The distribution for $W = 0.5$ eV can be explained in the same way even though it does not indicate a monoenergetic part and tail as clearly as do the distributions for the higher W. There could be two reasons for this. First, at $W = 0.5$ eV the diameter of the scattering sphere is comparable to the velocity resolution of the detector, and the monoenergetic part and tail tend to merge into a single, wide distribution. At the higher W's this is not the case since the diameter of the scattering sphere is large compared to the resolution. Second, at $W = 0.5$ eV a larger portion of the total scattering is associated with the tail.

From Fig. 1 it is noted that most of the Ne^+ is at $E_{Ne^+} < E^c_{Ne^+}$, where $E^c_{Ne^+}$ is the energy that Ne^+ would have outside the interaction region if it had the velocity of the c.m. inside. From Fig. 2 this indicates that most of the Ne^+ is scattered in the direction of the incident Ne*. Thus reaction (1) is directed, a conclusion that has been verified for charge transfer in other systems.

As mentioned above, the high-energy tail is associated with angular scattering. In Fig. 2 it is observed that lab energies in the tail that are less than $E^c_{Ne^+}$ are associated with $\theta < 90°$. In Fig. 1 it is noted that for $W \geq 10$ eV, $\theta < 90°$ for all scattering. For $W = 1$ and 0.5 eV there is scattering on either side of 90°.

Another conclusion concerning the kinetics of reaction (1) arises from the fact the $E_{Ne^+} \approx E_{Ne^*} + P$ at the center of the monoenergetic part of the distribution. This conclusion is that there is little momentum transfer in the reaction, which is not unexpected for charge transfer. In the lab system the momentum of most of the Ne^+ (i.e., the Ne^+ associated with the monoenergetic part) is only slightly larger than that of Ne* because of the small endothermicity of the reaction. Momentum differences between Ne^+ in the low-intensity tail and Ne* are somewhat larger.

For W greater than the ionization energy of ground-state Ne (i.e., 21.6 eV) and Ne* (ie., $\approx$ 5 eV) the possibility must be considered that some of the detected Ne^+ was due to collisional ionization of these species. The E_{Ne^+} for such ionization can be calculated and for a given W is greater than $E_{Ne^*} + P$ when the reactant He^+ is faster than Ne*. If this ionization were significant, evidence of it would appear in Fig. 1. Since there is no such evidence, we conclude that collisional ionization is negligible. Further proof that collisional ionization of ground-state Ne is negligible was discussed previously.

B. Cross sections

Experimental cross sections for reaction (1) were obtained for $0.1 \leq W \leq 500$ eV. To put these cross sections on an absolute scale we made an absolute measurement at $W = 150$ eV with the result that $Q(150) = 3.2 \times 10^{-15}$ cm^2.

Our measured and theoretical cross sections are shown in Fig. 3. The experimental data show the threshold to be near 0.1 eV. This is expected from the endothermicities of the reactions in the system. It is estimated that random errors in our experimental relative Q are ±10% whereas errors in our absolute Q are ±27%. We estimate that transverse velocities[9] increase our nominal, or quoted, W's in this experiment by an energy W_T no greater than 0.02 eV. A $W_T = 0.02$ eV could result in reductions of our Q of 9%, 2%, and 1% for nominal W's of 0.1, 0.5, and 1 eV, respectively.

In the W range of interest each of the theoretical cross sections Q_a through Q_d has about the same shape, rising from zero to a maximum of from $(50 \sim 70) \times 10^{-16}$ cm^2 and then decreasing very slowly. The zeros and maxima of Q_a and Q_c are at lower W than those of Q_b and Q_d because they have smaller $|\Delta E|$'s. In light of these remarks and the coefficients of the Q's used to obtain the total

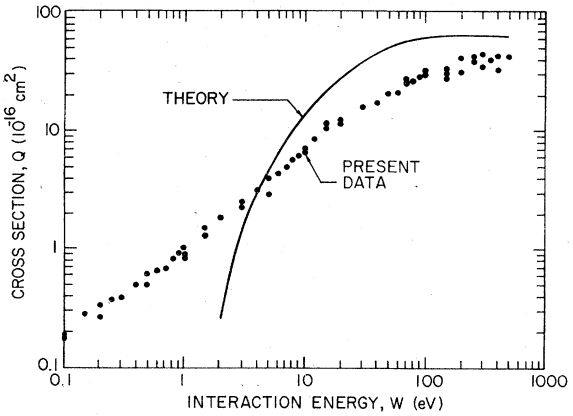

FIG. 3. Measured and theoretical cross sections for $He^+(1S) + Ne^* \rightarrow He^{\ddagger} + Ne^+$. For the theoretical curve, $He^{\ddagger}$ represents He in the 2S state.

charge-transfer cross section, it can be seen that reaction (1a) dominates the process over the entire range of W. For example, at $W = 2$, 50, and 500 eV reaction (1a) accounts for 88%, 86%, and 71% respectively, of the total theoretical Q.

The poor agreement of theory with experiment below $W \approx 3$ eV could be due to the failure of the Demkov approach to represent Q_a adequately at low W. On the other hand, if rotational coupling between molecular states had been considered in the theory, there may have been better agreement. The discrepancy cannot be significantly improved through the use of weighting different from statistical.

For $3 \lesssim W \lesssim 400$ eV the agreement between theory and experiment (the largest discrepancy is a factor of about 2.5) is not bad but can be improved with different weighting. Thus, it is difficult to assess the accuracy of the Demkov theory for such W. Even though the theoretical curve only crudely agrees with the data in this range, we feel it is a useful representation of Q.

Statistical weights should reflect probabilities of the various channels with fairly reasonable accuracy near 400 or 500 eV in Fig. 3. Thus, the fair agreement at such W (the calculation is about 30% greater than the data) suggests that the Demkov theory adequately represents the Q for reaction (1).

VI. SUMMARY

Reaction (1) is directed with most of the Ne^+ scattered in the direction of the reactant Ne*. The momentum transfer in reaction (1) is generally small and equal to that required to overcome the endothermicity. These observations on the kinetics of the reaction are consistent with other data on near-resonant charge-transfer processes. The threshold of reaction (1) appears to be near 0.1 eV, which is close to the smallest endothermicity of the process. The data indicate that the cross section monotonically increases with W, and at $W = 150$ eV is $(3.2 \pm 27\%) \times 10^{-15}$ cm^2. Theoretical calculations using a modified Demkov approach agree very roughly with the data above $W \approx 3$ eV. Agreement is poor at lower W.

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