

Temperature Dependence of Dissociative Recombination*

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Calculations of the dissociative-recombination rate for diatomic molecular ions and electrons are reported for various arrangements of the potential-energy curves. The temperature variation is shown to be different for the two possible heating mechanisms: selective electron heating or total gas heating. An attempt is made to correlate the afterglow and shock-tube measurements of the recombination rate for nitric oxide and neon. This attempt fails, and this suggests the need for further studies.

I. INTRODUCTION

For molecular ions in plasmas, the dominant electron-ion recombination mechanism is often dissociative recombination. Considerable progress has been made in recent years in the measurement of dissociative-recombination rates.^{1,2} Particular attention has been given to the temperature dependence of the rate. A knowledge of this dependence is required for the analysis of ionization in the upper atmosphere. Theoretical studies have so far shown little success in *ab initio* calculation of the dissociative-recombination coefficient but have contributed to the discussion of the temperature dependence.

The simplest theory of dissociative recombination^{1,3-5} suggests that the recombination rate for ions in the ground state should vary with the electron temperature according to the law

$$\alpha_0(T_e) = C T_e^{-0.5} . \quad (1)$$

This prediction can be tested in afterglow experiments by applying a microwave field for selective heating of the electrons. Most recent experiments of this type give results close to this simple law.⁶⁻⁹

In other situations, the whole gas is heated and a different dependence of the recombination rate on temperature might be expected. The results of shock-tube experiments¹⁰⁻¹² and the analysis of atmospheric recombination¹³ suggest that for some ions, the rate may decrease more rapidly with increasing temperature than is predicted by Eq. (1). The shock-tube data on neon and argon are of particular interest. At low temperatures, the rate varies as $T^{-0.49}$ and $T^{-0.67}$, respectively, in agreement with the afterglow results. At higher temperatures, the behavior in both gases is as $T^{-1.5}$. The transition between these two variations is near 900°K for neon and near 700°K for argon. The experimental results for neon are shown in Fig. 1.

The difference in the results of afterglow and shock-tube measurements has been attributed to a strong dependence of the recombination rate on the

vibrational state of the ion. O'Malley^{14,15} has suggested that the results on neon and argon are consistent with a simple model in which the recombination rate is taken to be zero for all excited vibrational states. We will try to determine whether this model can be justified.

II. THEORY OF DISSOCIATIVE RECOMBINATION

In this section we will outline the results of the theory which has been described elsewhere.^{1,5} We will consider the recombination of a diatomic ion AB^+ in its ground electronic state. The recombination reaction can be considered as the formation and dissociation of an intermediate state AB'' :



The initial capture of the electron can be caused by the transfer of energy either to the nuclear or electronic motion of the target. The transfer of energy directly to nuclear motion leads to the formation of a Rydberg state in an excited vibrational or rotational level. This capture mechanism will be discussed briefly in Sec. V. We assume here that in the capture step a target electron is excited and the incident electron drops into an unoccupied orbital. Thus, it is most likely that the state AB'' will have two electrons in excited orbitals.

The strength of the configuration interaction responsible for the formation of AB'' is expressed in terms of the capture width Γ_c .^{1,5,16} In accord with the Born-Oppenheimer separation of electronic and nuclear motion, the capture is most likely to occur near the nuclear separation R_c at which the difference in the electronic energies for AB'' and AB^+ is equal to the kinetic energy ϵ of the incident electron. The capture cross section, then, must be proportional to the probability of the nuclear separation being close to R_c in the initial ionic state. The cross section can be expressed as

$$\sigma_{\text{cap}}(\epsilon) = \frac{\pi^2}{m\epsilon} \frac{r}{2} \Gamma_c \frac{|\xi_\alpha(R_c)|^2}{|U'(R_c)|} , \quad (3)$$

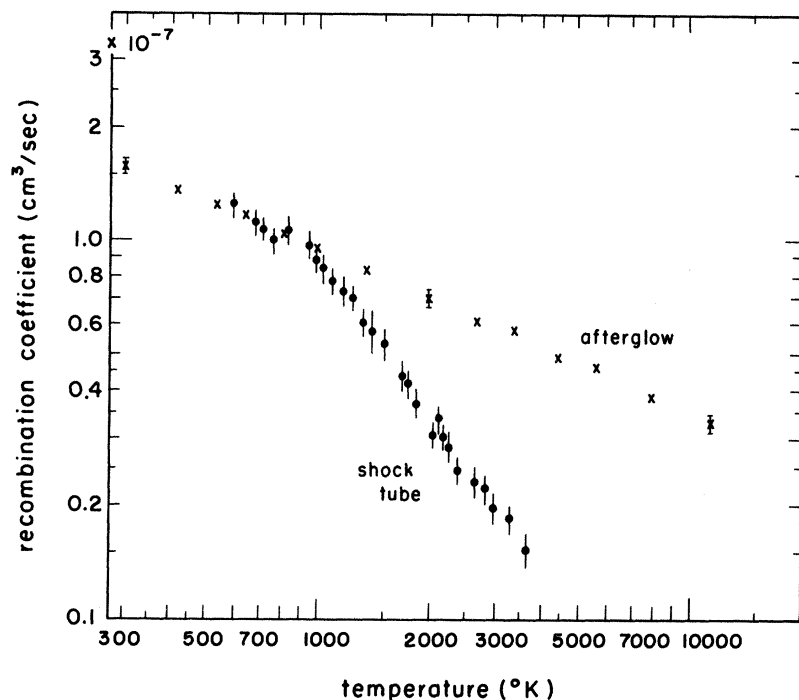


FIG. 1. Measured recombination coefficients for neon as a function of temperature; xxx denote afterglow data of Frommhold, Biondi, and Mehr using selective electrons heating; ●●● denote shock-tube data by Cunningham and Hobson with shock heating of the ions and electrons.

where m is the electron mass, r is the ratio of the multiplicities of the states AB'' and AB^+ , $\zeta_\alpha(R)$ is the nuclear wave function for the initial ion, and $U'(R)$ is the slope of the potential curve for the state AB'' .

For recombination to be completed, the state AB'' must decay by dissociation rather than by autoionization. The potential curve for AB'' is almost always repulsive, and the nuclei will be forced apart after the formation of the intermediate state. As the nuclei move apart, autoionization can take place but the autoionization rate decreases rapidly once the separation is greater than the range of the vibrations of the initial ion AB^+ . The probability of dissociation without electron emission is called the survival factor and is denoted by $S_f(\epsilon)$. If we assume a Maxwellian distribution of electron velocities at a temperature T_e , the recombination rate can then be expressed as

$$\alpha_v(T_e) = 4\pi^3 \hbar^2 r / (2\pi m k T_e)^{1.5} \int d\epsilon \Gamma_c \frac{|\zeta_\alpha(R_c)|^2}{|U'(R_c)|} \times S_f(\epsilon) \exp\left(-\frac{\epsilon}{kT}\right) \quad (4)$$

The subscript v on the recombination rate denotes the vibrational state of the initial ion. For a gas of molecules with a Boltzmann distribution of vibrational states appropriate to a temperature T_v , the effective rate is

$$\alpha(T_e, T_v) = \left[\sum_v \alpha_v(T_e) \exp\left(-\frac{E_v}{kT_v}\right) \right] \times \left[\sum_v \exp\left(-\frac{E_v}{kT_v}\right) \right]^{-1} \quad (5)$$

The vibrational energy levels of the ion are here denoted by E_v .

For most molecules, Eq. (4) can be simplified by making the following assumptions. (i) The survival factor $S_f(\epsilon)$ is close to unity. For atmospheric gases, the stabilization time is typically 10^{-15} sec, whereas, the autoionization rate is only of the order of 10^{13} sec $^{-1}$. (ii) The incident electron energy is small, say less than 1 eV. For such energies, the capture width varies little with energy. This condition implies temperatures below 10 000 °K. (iii) The slope of the potential curve for AB'' is sufficiently large so that the variation of the capture point R_c with incident energy can be neglected. We will replace R_c by R_s , its value at zero electron energy (R_s is the separation at which the potential curves for AB^+ and AB'' cross). For atmospheric ions, typical slopes are 25 eV/Å and this implies a difference between R_c and R_s of order 10^{-3} Å for electrons of energy 0.025 eV. With these approximations the energy integration in Eq. (4) can be carried out and gives

$$\alpha_v(T_e) = \frac{2\pi^3 \hbar^2 r}{(2\pi m^3 k T)^{0.5}} \Gamma_c \frac{|\zeta_\alpha(R_s)|^2}{|U'(R_s)|} \quad (6)$$

This expression gives the contribution of one state AB'' to the recombination rate. It can be seen that the most important states are those whose potential curves cross that of AB^* within the range of the initial nuclear vibrations. For most molecules, there are probably several suitable states and their contributions should be summed. However, for H_2 and He_2 , there seem to be no states with optimal potential curves. Thus, the following discussion does not apply to these two molecules.

This qualitative theory predicts the simple dependence of the recombination rate on electron temperatures that was discussed in Sec. I. In Sec. III, we report numerical calculations of the recombination rate in which we relax conditions (i) and (iii) and assess the deviations from Eq. (6). In addition, we discuss the dependence of the effective recombination rate $\alpha(T_e, T_v)$ on the ionic vibrational temperature T_v .

III. NUMERICAL SOLUTION

The computation of recombination rates can be divided into two parts. First, the important intermediate states AB'' must be identified and their properties evaluated. This aspect of the problem will not be discussed here. Instead, we will examine how the recombination cross section depends on the properties of the intermediate states. When the electronic structure of the intermediate state is known, the wave equation for the nuclear function $\xi(\vec{R})$ is set up in the form⁵

$$[-(\hbar^2/2M)\nabla^2 + U(R) - E]\xi(\vec{R}) = J_i(\vec{R}) + J_f(\vec{R}). \quad (7)$$

The left-hand side of Eq. (7) has the usual form for a nuclear wave equation with a potential energy $U(R)$. The two terms on the right-hand side of Eq. (7) represent the formation of the state AB'' and its decay by autoionization. The first term is proportional to the initial ionic nuclear function $\xi_{\alpha'}(\vec{R})$ and to the configuration interaction matrix element $V_{\alpha E}$ which is responsible for the capture of the electron

$$J_i(\vec{R}) = V_{\alpha E} \xi_{\alpha'}(\vec{R}). \quad (8)$$

The decay term contains a contribution from all the possible final states of the ion $\xi_{\alpha'}(\vec{R})$. It has the form

$$J_f(\vec{R}) = \sum_{\alpha'} \beta_{\alpha' E} \xi_{\alpha'}(\vec{R}) \int d\vec{R}' \xi_{\alpha'}(\vec{R}') \xi(\vec{R}'), \quad (9)$$

with

$$\beta_{\alpha' E} = \int dE' \frac{|V_{\alpha' E'}|^2}{E - E' + i\delta}, \quad (10)$$

where δ is an infinitesimal positive number.

In order to evaluate this decay term, we need to know the matrix element $V_{\alpha' E'}$ for all α' and E' , and so we will replace $\beta_{\alpha' E}$ by $\beta_{\alpha E}$ and take only the

contribution from the pole at $E' = E - i\delta$. However, in making this approximation, we lose the property that $J_f(\vec{R})$ describes only the decay and not the generation of the intermediate state. To recover this property, we modify the argument of $J_f(\vec{R})$ leaving its magnitude unchanged. The result of these two operations is

$$J_f(\vec{R}) = i\pi |V_{\alpha E}|^2 \frac{\xi(\vec{R})}{|\xi(\vec{R})|} \left| \sum_{\alpha'} \xi_{\alpha'}(\vec{R}) \right| \times \int d\vec{R}' \xi_{\alpha'}(\vec{R}') \xi(\vec{R}'). \quad (11)$$

The capture width Γ_c discussed in Sec. I is given by

$$\Gamma_c = 2\pi |V_{\alpha E}|^2. \quad (12)$$

Thus, the autoionization rate is of the order of Γ_c/\hbar , but varies with the internuclear separation. Typical widths are of the order of 0.1 eV. With such values, the survival factor is close to unity, and the approximation made above introduces errors of the order of 1%.

The nuclear equation (7) can be solved by the standard integration techniques. The results in Sec. IV were obtained by using the Numerov method for solving second-order differential equations. The integral in the decay term was treated by iteration. The first solution was obtained by neglecting autoionization and three iterations were usually sufficient for convergence to four significant figures.

IV. RESULTS

The important features of the potential curve for the intermediate state AB'' are the position of the stabilization point R_s and the slope $U'(R_s)$. These quantities have been treated as variable parameters and $U(R)$ was taken to be linear close to R_s . The other parameters in the calculation were the capture width Γ_c and the ionic vibrational spacing $\hbar\omega$. These parameters were chosen to represent the atmospheric ion NO^+ and the rare-gas ion Ne_2^+ .

A. NO^+

It has been suggested⁵ that there are four states of NO that may contribute significantly to the recombination of NO^+ . There is sufficient spectroscopic data on two of these states $B^2\Pi$ and $B'^2\Delta$ to determine their potential curves. The stabilization points are characterized by the dimensionless variable ρ defined by

$$\rho = (R_s - R_0)/a.$$

Here R_0 is the equilibrium separation and a is the classical amplitude of the ground-state vibration for the ion NO^+ . The value of the parameters suggested by the spectroscopic data is given in the

TABLE I. Parameters for the calculations for NO.

Case	ρ	$U' (R_s)$ (eV/Å)	Γ_c (eV)
A	+0.5	30	0.075
B	+0.5	30	1.2
C	-1.0	30	0.075
D	+1.0	30	0.075

first row of Table I. The recombination rate appropriate to these values is shown on a logarithmic scale in Fig. 2A. At each temperature T , the rate $\alpha_0(T)$ for the ground-state ions is shown together with the effective rate $\alpha(T_e, T_v)$, with $T_e = T_v = T$. Since our purpose is to examine the dependence on temperature, the absolute magnitude of the rate is not given. To aid analysis of the results, straight lines have been drawn to indicate simple power-law variations $T^{-\gamma}$. It can be seen that over the range of temperatures studied $\alpha_0(T_e)$ behaves approximately as $T_e^{-0.5}$, as predicted by the simple theory. However, the presence of excited vibrational ions reduces the effective rate $\alpha(T_e, T_v)$ below $\alpha_0(T_e)$.

By analysis of the iteration procedure, it was found that the survival factor was always greater than 0.95. In order to see the effects of increased autoionization, the capture and decay widths were multiplied by a factor of 16, and the results are shown in Fig. 2B. Both $\alpha_0(T_e)$ and $\alpha(T_e, T_v)$ de-

crease more rapidly with increasing temperature.

The dependence of the recombination rate on the position of the stabilization point was studied by varying the parameter ρ . In Figs. 2C and 2D, the results are shown for $\rho = -1$ and $\rho = +1$, corresponding to intersections at the left and right extremes of the Franck-Condon region for the ground state of the initial ion. In Fig. 2C, it can be seen that the excited vibrational states lead to an increased recombination rate $\alpha(T_e, T_v)$. This behavior is to be expected for most negative values of ρ and also for large positive values.

None of the results in Fig. 2 show the rapid decrease of the rate with temperature as $T^{-1.5}$ that has been suggested by some authors. Furthermore, it is not possible to correlate the low-temperature afterglow measurements^{17,18} with the high-temperature shock-tube data^{19,20} by any of our temperature variations. In Fig. 3, the experimental results are compared with the theoretical estimates of the contributions of the two states $B^2\Pi$ and $B'^2\Delta$ to the recombination rate. Since the nature of afterglows is understood to a greater extent than that of shock-tube gases, we conclude that the shock-tube measurements give recombination rates which are too low.

B. Ne_2^+

For the rare gases other than helium, very little is known about the potential curves for either the ions or neutral molecules. In particular, the

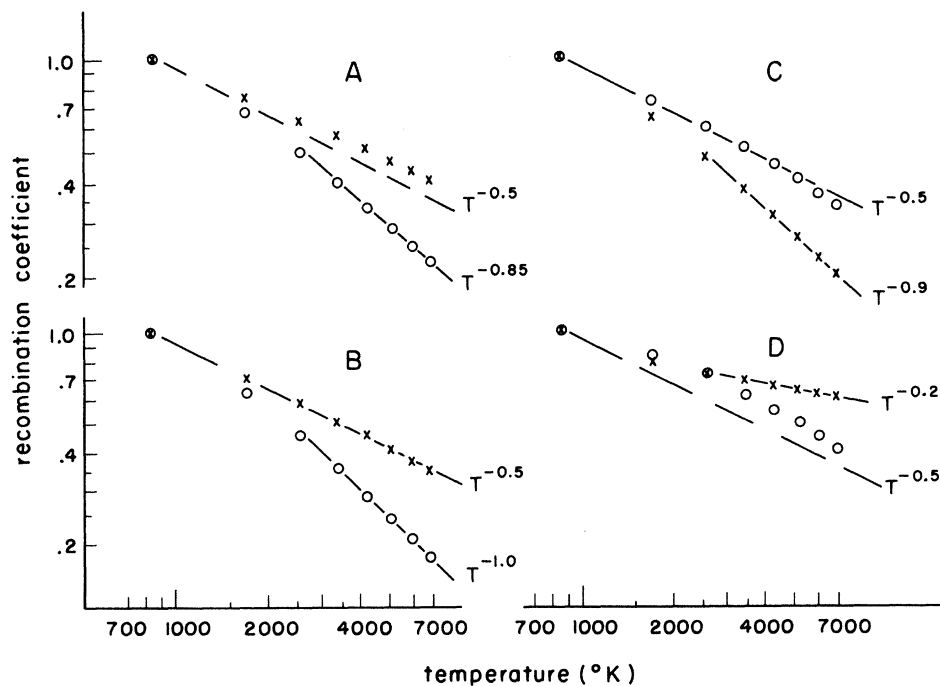


FIG. 2. Calculated variation of the recombination coefficient for nitric oxide for four sets of parameters described in Table I; x denotes electron heating and O denotes total heating.

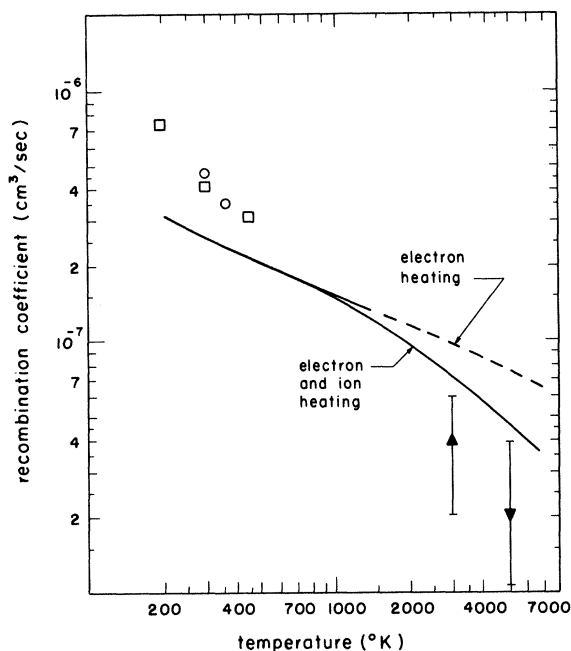


FIG. 3. Theoretical and experimental recombination coefficients for nitric oxide. O, Gunton and Shaw (afterglow); □, Weller and Biondi (afterglow); ▲, Stein *et al.* (shock tube); ▼, Lin and Teare (shock tube). The curves represent the calculated contribution of two intermediate states of NO to the coefficient. The broken line refers to selective electron heating and the unbroken line to total gas heating.

TABLE II. Parameters for the calculations for Ne.

Case	ρ	$U' (R_s)$ (eV/Å)	Γ_c (eV)
A	0.0	5	0.075
B	0.0	2.5	0.075
C	1.0	2.5	0.075

spacing of the ionic vibrational levels is not well established. Quantum-mechanical calculations quoted by Wahl *et al.*²¹ suggest a spacing of 0.08 eV. This value was supported by Cunningham and Hobson^{11,12} since it is close to this energy that the shock-tube measurement of the recombination rate changes from a $T^{-0.5}$ to a $T^{-1.5}$ dependence. Our primary purpose, therefore, was to see if we could reproduce the afterglow and shock-tube results with this value of the spacing.

O'Malley^{14,15} has suggested that the experimental results can be explained by assuming that the recombination rate for excited ions is negligible. The most rapid decrease of the recombination rate with increasing vibrational energy is found when the potential crossing is near the equilibrium nuclear separation ($\rho = 0$). Such a crossing was assumed in our first two calculations with the results shown in Figs. 4A and 4B. The parameters appropriate to these results are given in Table II. With a potential slope of 5 eV/Å, the variation of $\alpha_0(T_e)$ remains close to $T^{-0.5}$, up to around 1500 °K. The effective rate $\alpha(T_e, T_v)$ decreases more rapidly,

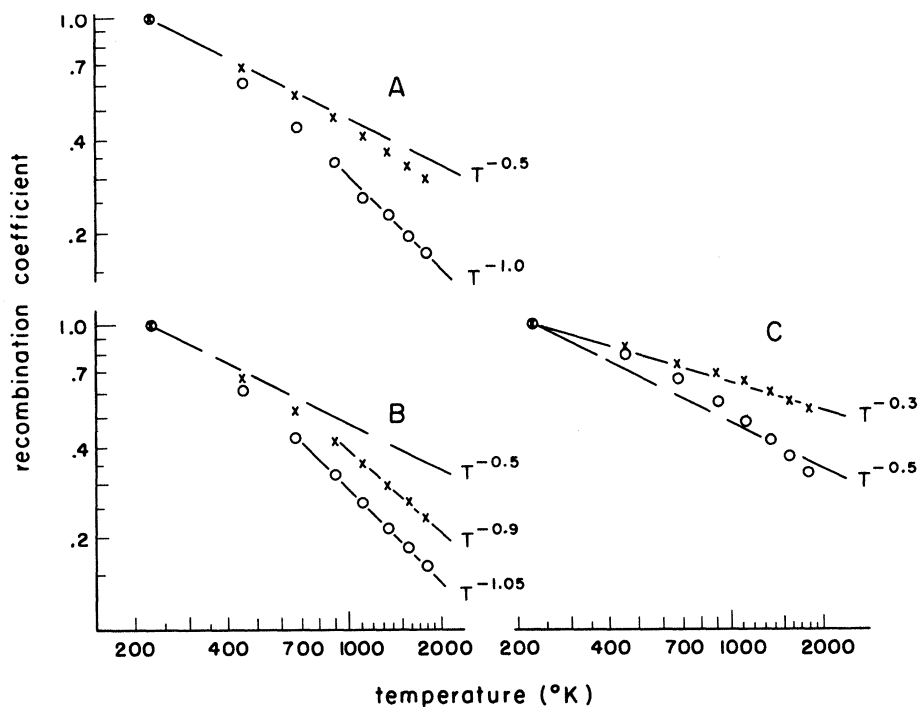


FIG. 4. Calculated variation of the recombination coefficient for neon for three sets of parameters given in Table II; x denotes electron heating and O denotes total heating.

but at 1800 °K the decrease is only as $T^{-1.0}$. In order to obtain a faster rate of decrease, the slope was reduced to 2.5 eV/Å. Although this produces the desired effect in $\alpha(T_e, T_v)$, it is accompanied by a significant deviation of $\alpha_0(T_e)$ from the $T^{-0.5}$ law. A similar effect is found if the autoionization rate is increased.

Again, we find that we cannot correlate the afterglow and shock-tube results, even with a potential crossing at the center of the Franck-Condon region. Furthermore, since there are very many states of the molecules Ne_2 and Ar_2 , it would seem surprising that the significant states for recombination all cross the ionic curve near its minimum. The rate appropriate to a crossing with $\rho = 1$ is shown in Fig. 4C. If there are significant states with crossings outside the Franck-Condon region of the ground state but within the range of excited vibrations, these should contribute more to the effective rate $\alpha(T_e, T_v)$ than to $\alpha_0(T_e)$.

There seem to be two possible conclusions from these results. First, the vibrational spacing for Ne_2^+ could be much smaller than 0.08 eV. For values below 0.05 eV, the rate at around 2000 °K could conceivably behave as $T_e^{-0.5}$ under electron heating and $T^{-1.5}$ under total heating. Alternatively, one must conclude that in the shock tube the vibrational temperature was not as stated and that the vibrational and translational degrees of motion were not in equilibrium when the recombination measurements were made.

V. CONCLUSIONS

We have confirmed earlier predictions that a significant difference can exist between the effects of electron heating and total heating on the recombination rate. For the atmospheric ions this difference should be small below 1000 °K, since at such temperatures the ions are predominantly in

the ground vibrational state. At higher temperatures, also, the difference does not seem to be large enough to explain the measurements of recombination coefficients in shock-tube experiments. A similar situation is found in neon, although the difference between the results for the two heating methods becomes apparent at lower temperatures.

This incompatibility of theory and experiment suggests that further study should be given to the distribution of vibrational states in the ions undergoing recombination. This is of particular importance for shock-tube experiments but is probably also necessary for afterglow and atmospheric conditions.

Some experiments have indicated the possibility of significant deviations of the recombination rate from the $T^{-0.5}$ dependence, even at low temperatures. One explanation for such deviations would be the occurrence of indirect recombination⁵ in which a vibrationally excited Rydberg state is formed and subsequently decays by predissociation. However, there is so far no firm evidence, either from experiments or theory, for the importance of this alternative recombination mechanism.

Further theoretical work is needed to test the validity of the approximations made in this treatment. Of particular interest is the effect of the Rydberg states on the direct-recombination process.²² O'Malley¹⁵ has suggested that the presence of these states could lead to a significant reduction in the survival factor. This point should be investigated since it is not yet clear whether this is an important effect or whether a smaller survival factor could lead to better agreement between theory and experiment. In the calculations reported here, the survival factor was kept above 0.4, since smaller values led to deviations from the results of the experiments using selective electron heating.

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PHYSICAL REVIEW A

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Delayed-Coincidence Study of $O^+ + Ne$ Collisions at 50–200 keV*

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Delayed-coincidence techniques have provided measurements of the distributions in values of Q , the energy transferred to inelastic processes, during $O^+ + Ne$ collisions at 50–200 keV. The average values \bar{Q} of the Q distributions remain almost constant as a function of r_0 , the distance of closest approach of the colliding particles. For fixed r_0 , the \bar{Q} values vary slowly with bombarding energy and lie, for example, approximately 40 eV higher at 100 keV than at 50 keV. The flatness of the $\bar{Q}(r_0)$ functions is reflected in a similar independence of r_0 found in measurements of the average charges of the particles after collision. These results indicate that the collisions produce excitations only among L -shell electrons. However, some data describing the widths of certain Q distributions indicate that K -shell excitations may occur infrequently among collisions that produce O^{+4} ions. Supplementing the data for inelastic energy transfers are determinations of average ionic charge states after collision, measurements of angular distributions of scattering cross sections, and calculations of inelastic energy-loss cross sections deduced from the Q data.

I. INTRODUCTION

A number of recent papers^{1–3} have presented data describing features of close-encounter atomic collisions. During these collisions, an interesting variety of phenomena occur as the electron shells interpenetrate and readjust to their rapidly changing environment. The complexity of the interpenetrating-shell phenomena has made them difficult to explain in detail, and further understanding requires systematic experimental studies. Several such studies, aimed at various aspects of close-encounter phenomenology, are continuing at a number of laboratories. Two papers have already presented some studies of O^+ reactions: an investigation² of $O^+ + Ar$ and a recent report³ by Knystautas, Kessel, Del Boca, and Hayden describing $O^+ + Kr$. In an effort to describe more clearly the systematics of O^+ collisions with noble gases, this study examines $O^+ + Ne$ reactions at 50–200 keV.

Section II is an exposition of the four types of data obtained in these measurements: inelastic energy transfers, scattering angular distributions, ionization probabilities, and energy-loss cross sections derived from the energy-transfer data. Section III contains a discussion of interrelationships among these data and a comparison of the

data with the earlier studies of O^+ collisions with noble gases.

II. EXPERIMENTAL RESULTS

A. Methods and Notation

The experimental technique used in accumulating most of the data reported here is the method of delayed coincidences, described in detail in several earlier publications.^{1,2} The particular apparatus and nearly all the procedures used in this study are described in Ref. 2. This section contains merely a review of notation for the reader's convenience and a description of one new procedure.

In this report (as well as in Ref. 2) a " $10/mn$ " process is a scattering reaction in which a beam particle of charge +1 strikes a neutral target particle and scatters, with charge + m , into a detector while the target particle recoils, with charge + n , into another detector. The angles through which the beam and target particles scatter are, respectively, θ and φ , both measured from the beam direction. The symbol Q_{mn} denotes the amount of energy transferred to inelastic processes during the $10/mn$ collision. A simple kinematic expression, which neglects thermal motion of the