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Dissociative-recombination product states and the dissociation energy D_0 of Ne_2^+

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Final product states of Ne_2^+ dissociative recombination were studied using time-of-flight spectroscopy to determine the kinetic energies released. The dissociative recombination occurred in a sustained discharge in the presence of a variable magnetic field and discharge voltage, at pressures of 5–15 mTorr. Under different conditions various excited states were observed ranging from the lowest 3s metastable states to higher Rydberg states within 0.000 54 eV of the dissociation limit. From their narrow widths, it is deduced that these higher states arose from Ne_2^+ ions with subthermal energies. From two of these narrow distributions, we obtain an improved value for the dissociation limit $D_0(\text{Ne}_2^+) = 1.26 \pm 0.02$ eV. [S1050-2947(97)01109-8]

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I. INTRODUCTION

Dissociative recombination (DR) of rare-gas molecular ions has been the subject of intense theoretical interest and many experimental studies for nearly 40 years. DR reactions are important in the study of low-temperature, low-pressure, ionized gases such as planetary ionospheres, laboratory electrical discharges, and interstellar clouds because DR is the only fast two-body process that can remove thermal free electrons and therefore is the dominant electron loss mechanism in these cases. The addition of ion storage rings with electron coolers to the experimental methods used to study DR has resulted in renewed interest in the reaction. Three international conferences [1–3] in the past several years have been devoted to experimental and theoretical discussions of the DR reaction.

The DR of Ne_2^+ and other rare-gas molecular ions was extensively studied by Biondi and co-workers [4–8], who used optical spectroscopy to identify the final product states of the DR reaction. They produced the molecular ions by microwave excitation in an afterglow apparatus. A more recent study of DR of Ne_2^+ by Malinovsky *et al.* [9], reporting the production of the 3p and 3d states, used similar optical techniques.

The traditional view of the DR reaction of rare-gas molecular ions heavier than He_2^+ , for example,

was that the reaction yielded one atom in the np^6 ground-state configuration and the other predominantly in the $np^5(n+1)p$ excited state [10]. The reason for this belief was twofold. First, it was thought that the repulsive potential energy curves leading to the $(n+1)s$ excited product states passed inside (at smaller internuclear separations) the turning point of the ground-state ion potential curve and therefore

did not intercept it as is required for the direct DR process [11]. Second, in studies of Biondi and co-workers and Malinovsky *et al.* [5–9], the instrumentation used was not sensitive to the vacuum ultraviolet (vuv) radiation from the $(n+1)s$ states, and the $(n+1)p \rightarrow (n+1)s$ transitions were the lowest that were observable, so the available experimental evidence supported the first belief.

We have used time-of-flight (TOF) spectroscopy to study the final electronic states in the product atoms from the DR reaction of Ar_2^+ [12] and Kr_2^+ [13] and have reported the production of the unexpected ground-state atoms in the DR of Ne_2^+ and Ar_2^+ [14]. It will be seen that TOF spectroscopy reveals information not available from traditional optical methods. Here we report our TOF spectroscopy studies of the production of excited product states in the DR of Ne_2^+ .

II. TIME-OF-FLIGHT METHOD

The kinetic energy released to the product atoms by the DR reaction can be calculated from

$$E_{\text{KE}} = E_i + E_e - D_0 - E_{\text{FS}}, \quad (2)$$

where E_{KE} is the total kinetic energy released in the reaction, E_i is the ionization energy of the atom, D_0 is the dissociation energy of the molecular ion, E_{FS} is the energy of the final state in the product atom, and E_e , the electron energy, is assumed to be zero. E_e is assumed to be zero because DR reaction rates decrease rapidly with increasing E_e , and, at the pressures used in our discharge the slow electrons are thermalized [8]. E_i for Ne is 21.559 eV [15], and D_0 for Ne_2^+ is 1.30 eV [16]. E_{KE} and hence E_{FS} can be determined from

$$E_{\text{KE}} = M V_{\text{DR}}^2 \quad (3)$$

since the molecular ion is homonuclear. E_{KE} is the kinetic energy for the two product atoms, each of mass M . Here, we ignore the small contribution from the isotope effect since neon is 90.8% Ne_{20} and 8.9% Ne_{22} . V_{DR} is measured directly by the TOF method.

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TABLE I. Final product states observed (states used to fit the data).

State	J	E_{FS} (cm^{-1})	Energy (eV)	Velocity (m/sec)	Channel	Width (m/sec)
$2p6\ 1s$	0	0	0	9823	3	
$3s\ [\frac{3}{2}]$	2	134 043.8	16.62	4173 ^a	7	650
$3s'\ [\frac{1}{2}]$	0	134 820.6	16.72			
$3p\ [\frac{1}{2}]$	1	148 259.8	18.38	3020	10	525
$3p\ [\frac{5}{2}]$	3	149 659	18.55	2880	10.6	500
$3p\ [\frac{5}{2}]$	2	149 826.2	18.58	2855 ^a	11	700
$3p\ [\frac{3}{2}]$	1	150 123.6	18.61			
$3p\ [\frac{3}{2}]$	2	150 317.8	18.64			
$3p'\ [\frac{3}{2}]$	1	150 774	18.69	2763	11.1	300
$3p'\ [\frac{3}{2}]$	2	150 860.5	18.70	2754	11.1	300
$3p'\ [\frac{1}{2}]$	1	151 040.4	18.73	2734	11.2	350
$3p'\ [\frac{1}{2}]$	0	152 972.7	18.97	2518	12.2	200
$4s\ [\frac{3}{2}]$	2	158 603.1	19.66	1738	17.6	100
$3d'\ [\frac{5}{2}]$	3	162 412.1	20.14	880 ^a	34.8	90
$3d'\ [\frac{3}{2}]$	2	162 421.9	20.14			
$3d'\ [\frac{3}{2}]$	1	162 437.6	20.14			
$4p\ [\frac{5}{2}]$	3	162 832.7	20.19	727	42.1	70
$4p\ [\frac{3}{2}]$	1	163 014.6	20.21	649	47.2	30
$4p\ [\frac{3}{2}]$	2	163 040.3	20.21	638	48.1	10
$4p\ [\frac{1}{2}]$	0	163 403.3	20.26	439	69.8	28

^aAverage of the states listed.

We have shown previously [14] that the DR atom flux dj is given in time space by

$$dj(t) = (c/t^4) \exp(-\{[(L/T) - V_{DR}]/V_p\}^2) dt. \quad (4)$$

FIG. 1. A typical time spectra with source conditions that inhibit the DR reaction. The time channel width is 40 μsec , and t_0 is at channel zero. The peak of this distribution occurs in channel 33 which corresponds to a most probable velocity V_p of 930 m/sec.

FIG. 2. A time spectrum exhibiting some DR final product states. (b) is an expansion of the region around the DR peak. The solid line is a fit to the data. The data were fit to two DR product states with velocities of 4173 m/sec (a weighted average of the velocities of the $3s[\frac{3}{2}] J=2$ and $3s'[\frac{1}{2}] J=0$ product states) and 2855 m/sec (a weighted average of the velocities of the $3p[\frac{5}{2}] J=3$, the $3p[\frac{5}{2}] J=2$, and $3p[\frac{3}{2}] J=1$ states).

In this expression c is a constant related to the flux of atoms from a particular transition, L is the flight path length, t is the time of flight, V_{DR} is the velocity of the product atom, and V_p is the most probable velocity of the velocity distribution of the parent Ne_2^+ ions, which is assumed to be a Maxwellian velocity distribution.

III. APPARATUS

The molecular beam apparatus used here is the same as the one used in our previous TOF studies of DR [13]. The DR reaction occurs in a continuous discharge, effusive source maintained by electrons produced by a directly heated thoriated tungsten ribbon (2.5×0.025 mm) cathode. The source is surrounded by a solenoid that produces a variable

FIG. 3. A time spectrum illustrating DR to some $3p$ states. The data were fitted to a line with a velocity of 2855 m/sec (a weighted average of the velocities of the $3p[\frac{5}{2}] J=3$, the $3p[\frac{3}{2}] J=2$, and the $3p[\frac{1}{2}] J=1$ states). The solid line is a fit to the data.

axial magnetic field of up to 250 G. The gas pressure, the discharge current, the discharge voltage, and the applied magnetic field were all variable in this work. We have previously discussed the operation of this source with the applied magnetic field in detail [17].

Within the discharge, the high-energy electrons (energies approximately equal to the discharge voltage in eV) create both excited states of atoms and atomic ions. Excited states are also produced by energetic secondary electrons following the ionizing collisions. Molecular ions are formed principally by associative ionization

where Ne^* is in a sufficiently high Rydberg state ($n > 5$), or Eq. (5) could occur at the pressures used here if both atoms are in the metastable state. Subsequently, slow electrons dissociatively recombine with molecular ions to form fast neutral product atoms. The effect of the axial magnetic field is to constrain the discharge, by constricting both fast and slow

TABLE II. Yield as a function of discharge voltage.

Pressure 15 mTorr	Magnetic field 106 G		Discharge current 80 mA		
Discharge voltage	State	Energy (cm^{-1})	Relative intensity	Width (m/sec)	
39 V	no DR				
52 V	$3s[\frac{3}{2}]$	$J=2$ 134 044	80	900	
	$3s'[\frac{1}{2}]$	$J=0$ 134 821			
	$4p[\frac{1}{2}]$	$J=0$ 163 403	26	25	
	$3p[\frac{1}{2}]$	$J=1$ 148 260	83	900	
58 V	$3s[\frac{3}{2}]$	$J=2$ 134 044	416	900	
	$3s'[\frac{1}{2}]$	$J=0$ 134 821			
	$3p'[\frac{3}{2}]$	$J=1$ 150 774	1093	300	
65 V	$3s[\frac{3}{2}]$	$J=2$ 134 044	156	900	
	$3s'[\frac{1}{2}]$	$J=0$ 134 821			
	$3p[\frac{5}{2}]$	$J=3$ 149 659	1128	500	
	$3p'[\frac{3}{2}]$	$J=1$ 150 774	3298	300	
71 V	$3p'[\frac{1}{2}]$	$J=0$ 152 972	1900	200	
78 V	$3p'[\frac{1}{2}]$	$J=0$ 152 972	677	200	
	$4s[\frac{3}{2}]$	$J=2$ 158 603	541	65	
91 V	no DR				
104 V	no DR				

electrons to a narrow region along the axis in alignment with the filament and the exit slit of the discharge. Thus excitation, ionization, and DR all occur close to the axis of the discharge, which increases the observed yield of metastable atoms very effectively (by a factor of 10–100). With discharge voltage, pressure, and magnetic field constant, the yield of our source is a linear function of discharge current over a wide range of currents, from ≈ 50 mA to over ≈ 300 mA. Sweep plates located downstream from the source maintain an electric field of about 250 V/cm which removes all charged particles and long-lived Rydberg states from the atom beam. The long-lived Rydberg states are removed from the beam by field ionization. The detector consists of a tungsten catcher plate from which electrons are collected and detected by a Channeltron electron multiplier. Our system is only sensitive to atoms in the metastable state, which have sufficient electronic energy to eject electrons from the catcher plate. Charged products and atoms in the Rydberg state are removed from the beam at the sweep plates, and the flight time ($> 200 \mu\text{sec}$) is long enough for all other species to radiate either to the metastable state or the ground state. In this apparatus the flight path length is 1.227 m, and the chopper open time is $75 \mu\text{sec}$.

IV. TREATMENT OF THE EXPERIMENTAL DATA

Our DR data were obtained in the apparatus described above under a variety of source conditions with pressures ranging from 5 to 15 mTorr, discharge voltages ranging from 20 to 120 V, and magnetic fields ranging from 0 to 250 G. The experimental data were fitted, using the least-squares criteria, to the function of Eq. (4). The free parameters in the

(a)

(b)

FIG. 4. The dependence of the state produced on the discharge voltage. Note the dependence of the $3p$ states on voltage in (a). (b) illustrates the yield of the $3s$ and $3s'$ states as a function of discharge voltage (see text).

fit were c , v_p , t_0 , and a function used to describe the background. The background functions used were either a constant, a quadratic, or a Maxwellian. t_0 is the midpoint of the chopper open time. Note that the velocity of the DR transitions was not a free parameter. When more than one DR final state was in the data to be fitted, Eq. (4) was summed over all possible lines in the data. If the identification of the DR product state was not obvious, each possibility was tried, and the least-squares criterion was used to identify the state.

V. DISCUSSION OF OBSERVED PRODUCT STATES

We have previously reported on our observations of the ground-state products of DR of Ne_2^+ [14]. This report will deal with final-state products other than the ground state. A list of the states used to fit the data is shown in Table I.

Figure 1 shows a time distribution with only the Maxwellian thermal peak. The peak of this distribution occurs in channel 33 which corresponds to a most probable velocity

(a)

(b)

FIG. 5. An illustration of the fact that the DR product state atoms can dominate the time spectra. (b) illustrates the fit to the data. These data were fit with two DR product states with velocities of 4140 and 2770 m/sec. These velocities correspond to DR to the $3s$, $3s'$ states and the $3p'[\frac{3}{2}]$ state.

V_p of 930 m/sec and a temperature of ≈ 1000 K. The time channel width on all data shown in this report is $40 \mu\text{sec}$, and t_0 is at channel zero. The chopper open time is $\approx 75 \mu\text{sec}$. We assume an error of less than ± 1 channel in our ability to determine the time at which the chopper opens.

We studied the final product states produced as a function of discharge voltage, magnetic field, and discharge pressure. We found that by varying the source operating conditions, we were able to change the final state of the product atoms. This variation in final product state was largest in the case of varying discharge voltage.

Figure 2(A) shows a time spectrum that exhibits DR. The small peak near channel 10 is due to DR which produces superthermal atoms that arrive at the detector before the Maxwellian distribution. Figure 2(B) is an expansion of the region around the DR peak. The data were fit to two DR product states with velocities of 4173 m/sec (channel 7), a weighted average of the velocities of the $3s[\frac{3}{2}] J=2$ and

(a)

(b)

FIG. 6. The yield of the thermal (a) metastable atoms and the (b) $3s$, $3s'$ direct product state atoms as a function of the magnetic field surrounding the source. The discharge voltage was 120 V, and the pressure was 10 mTorr.

$3s'[\frac{1}{2}] J=0$ product states, and 2855 m/sec (channel 11), a weighted average of the velocities of the $3p[\frac{3}{2}] J=3$, the $3p[\frac{3}{2}] J=2$, and the $3p[\frac{3}{2}] J=1$ states. The source conditions for these data were a magnetic field of 16 G, a source pressure of 17 mTorr, and a discharge voltage of 74 V.

Figures 3(A) and 3(B) show an example of the effects of changing source conditions. In this spectrum, the $3s$ states are no longer evident, and the averaged sum of the $3p$ states is much larger than in the spectrum discussed above. The data were fitted to a DR product line with a velocity of 2855 m/sec, the weighted average of the $3p$ states. The discharge conditions were a magnetic field of 27 G, a source pressure of 9 mTorr, and a discharge voltage of 100 V.

A. DR of Ne_2^+ as a function of discharge voltage

Table II gives the results of varying the discharge voltage, with fixed values of magnetic field (106 G), pressure (15 mTorr), and discharge current (80 mA). These data are illus-

(a)

(b)

FIG. 7. The yield of the thermal (a) metastable atom time distribution and the $3s$, $3s'$ state DR product atom peak (b) as a function of source pressure. The dashed line in (b) is a fit of a straight line to the data. Note that in (a) the yield saturates due to quenching of the metastable state atoms in the source.

trated in Figs. 4(A) and 4(B). Figure 4(A) shows the energies of the various states produced at various discharge voltages. It shows that $3s$ states are produced over a range of discharge voltages (with the pressure, and magnetic field used), and that $4s$ and $4p$ states are produced over a much narrower range of voltages. Figure 4(A) also shows that, because the energy changes, the $3p(J)$ state produced varies with the discharge voltage.

The yield of the $3s$ state as a function of discharge voltage is shown in Fig. 4(B). The yield peaks sharply at ≈ 60 V and drops to zero for discharge voltages less ≈ 5 V and voltages greater than ≈ 70 V. Figures 5(A) and 5(B) are examples of the data used in Table II.

B. DR of Ne_2^+ as a function of axial magnetic field

The axial magnetic field surrounding the source constrains the discharge, by constricting both fast and slow elec-

FIG. 8. The dependence on the width of the DR direct product velocity distribution peak as a function of kinetic energy released.

trons to a narrow region along the axis, in alignment with the filament and the exit slit of the discharge. Thus excitation, ionization, and DR all occur close to the axis, which increases the observed yield of metastable atoms very effectively (by a factor of 10–100). In addition, increasing pressure increases both the yields of metastables and of associative ionization, reaction (5). We studied the yield of DR product atoms as a function of the axial magnetic field surrounding the source, with discharge voltages of 40, 50, and 120 V. At discharge voltages of 50 and 120 V, a pressure of 10 mTorr was used; however, at 40 V, a pressure of 15 mTorr was used in order to produce sufficient intensity of the DR final states. The data shown in Figs. 6(A) and 6(B) were taken at 10 mTorr and 120 V and indicate that the yield of thermal metastable atoms and the DR products both increase with magnetic field and saturate at magnetic fields of about 150 G for discharge voltages of 120 V. These data show that at the values of discharge voltage we have studied, the DR peak yield is proportional to the yield of metastable atoms at their most probable velocity in the thermal distribution. The ratio of DR product atoms to thermal metastable atoms ranges from $\approx 1\%$ at a discharge voltage of 40 V to $\approx 3\%$ at 120 V. These observations indicate that the Ne_2^+ ions, that subsequently undergo DR to give the product atoms, are indeed formed by reaction (5) in our discharge.

C. DR of Ne_2^+ as a function of source pressure

We measured the yield of the $(n+1)s$ state DR products as a function of source pressure with a discharge voltage of 100 V and a magnetic field of 53 and 106 G. Figures 7(A) and 7(B) are the data taken with a magnetic field of 106 G on the source. Figure 7(A) shows that the yield of thermal atoms saturates at pressures of ≈ 15 mTorr. At this pressure the thermal atoms are quenched by associative ionization, [see reaction (5)]. Figure 7(B) shows that the yield of DR products is roughly a linear function of pressure from 4 to 25 mTorr. The dashed line in Fig. 7(B) is a least-squares fit of a straight line to the data. The primary production of Ne^* con-

FIG. 9. This figure shows the data (circles), a thermal distribution without DR used for background determination (squares), and a fit to the data (lines). The DR peaks are the $4p[\frac{3}{2}] J=1$ and the $4p[\frac{1}{2}] J=0$ product states.

tinues to increase with pressure but they are quenched by reaction (5), which continues to produce the Ne_2^+ that yield DR products.

D. Width of the DR peaks as a function of the kinetic energy released

We have reported that in the case of Ar_2^+ [12] the energy width of the DR product peaks increased with the kinetic energy released. We find a similar behavior in our studies of neon. The width of the DR peaks corresponding to the $3s$ and $3s'$ states was approximately what would be expected if the velocity distribution of the parent Ne_2^+ molecular ions was the same as the velocity distribution of the thermal metastable atoms from our source. We find that for states in the product atom higher in energy than the $3s$ and $3s'$ states, thus with smaller kinetic energy released, the linewidth is considerably narrower. Figure 8 shows this relationship. As in the case of argon [12], such states are produced only under specific discharge conditions. If the width of the DR product line were not smaller than the width of the thermal distribution, it would not be observable. We still have no explanation for this phenomenon. The time-of-flight observation of these states may select only events that occur from ions with a slow and narrow velocity distribution, and therefore the observed small width. The narrow velocity widths of these higher state channels imply that those Ne_2^+ ions whose DR contribute to these narrow peaks must have a subthermal population. This narrowing is not understood but may result from a combination of slower DR rates for these states and collisional quenching of the intermediate Ne_2^{**} states with higher thermal velocities and thus shorter mean free lifetimes.

E. Binding energy of Ne_2^+

Product state peaks that are observable because of their narrow widths can be used to determine the dissociation energy of Ne_2^+ . One of these is the $4p[\frac{1}{2}] J=0$ state which

appears at channel 70 in Fig. 9. This peak has a velocity of 439 m/sec and a width of 28 m/sec, assuming a dissociation energy of $D_0 = 1.30$ eV and electron energy $E_e = 0$.

In a TOF experiment, the velocity resolution is inversely proportional to t^2 , so a slow DR product state peak allows an accurate measurement of its velocity. The velocity initially used in the fitting procedure is determined using the assumed value of D_0 in Eq. (2), since t_0 , the midpoint of the chopper open time, is a free parameter in the fit. The difference between the value obtained from the fit and 0 may be used to determine the true velocity of the DR peak. This value may then be used, with Eq. (2), to find the correct value of D_0 .

We have observed the $4p[\frac{1}{2}] J=0$ state with six different sets of operating conditions in our source and have used these measurements in a new determination of the dissociation energy of the molecular ion obtaining $D_0 = 1.26 \pm 0.02$ eV. From a single measurement using the $4p[3/2] J=1$ DR peak that appears at channel 47 in Fig. 9, a value of $D_0 = 1.265$ eV was obtained. The most accurate previous determination was $D_0 = 1.20 \pm 0.08$ eV reported by Ciurylo *et al.* [18], who obtained the velocities of superthermal DR products by fitting the Doppler broadened profiles of emission lines in a plasma afterglow. Similar techniques used earlier by Frommhold and Biondi [4] gave a value of $D_0 = 1.35 \pm 0.07$ eV. Schneider and Cohen [19] obtained a theoretical estimate of $D_0 = 1.13$ eV.

VI. CONCLUSION

We have observed 19 final product states of the DR reaction of Ne_2^+ . We have previously [14] reported the unexpected observation of ground-state products. Our results do not disagree with the earlier investigations [4–9] that used optical methods and were not able to detect the $3s$, $3s'$, and ground-state products, but we have added new important product states. We found that, as in our studies of argon, the width of the product state lines depends on the electronic energy of the product state becoming very narrow for the higher states. This aspect is not presently understood. We also observed that the DR product state distribution depends to some extent on the voltage across the glow discharge, the pressure, and the magnetic field. The time-of-flight method of studying the final products of DR offers better velocity resolution for the higher-energy product states than other methods and has been used to obtain an accurate value of the Ne_2^+ dissociation energy $D_0 = 1.26 \pm 0.02$ eV.

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