Dissociative Recombination of HeH^+ at Large Center-of-Mass Energies

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Dissociative recombination of HeH^+ has been studied for the HeH^+ ion stored in a storage ring. A strong recombination resonance having a peak at around 20 eV center-of-mass energy has been found for the first time in addition to the known recombination at around 0 eV center-of-mass energy. The strength of the recombination rate changes depending on the storage time of HeH^+ ions.

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HeH^+ is the simplest two-electron heternuclear molecule and is attracting much interest due to its unique features. In astrophysical research, HeH^+ has been expected to be detectable in planetary nebulas [1], as HeH^+ can be synthesized by helium and hydrogen, the two most abundant elements in the universe, and the destruction rate of HeH^+ is low. However, the results of a search for HeH^+ in the planetary nebula NGC7027 [2] suggest that the HeH^+ density is much lower than expectations based on the analysis of the molecular process. As for the e-HeH^+ dissociative recombination (DR), it has been believed that the cross section is negligibly small due to the absence of any evidence for potential-energy-curve crossings of neutral dissociating molecular states with the ground state of HeH^+ [3]. A few years ago, however, Yousif and Mitchell [4] showed that the HeH^+ recombination cross section is not small at low energies, using a merged-beam technique. They also found that the cross section depends strongly on the extraction potential of the ion source. That means the excited HeH^+ contained in the ion beam greatly modifies the cross section. Although their results suggest that the HeH^+ ground state was obtained at low extraction potential, it is not completely clear that contributions from excited ion states can be ruled out. A new method to remove the indeterminate mixture of internal excited states of the molecular HeH^+ emerging from ion sources was recently proposed by Datz and Larsson [5]. They showed from their analysis of the radiative lifetime of HeH^+ that vibrationally excited HeH^+ ions can be cooled in a short time by storage of accelerated HeH^+ ions in a ring system. With a few exceptions, the vibrational lifetimes for HeH^+ are of the order of tens of ms or less according to their calculations. So, if one can store HeH^+ ions sufficiently longer than these times, the ions are expected to be mostly in the ground state. Another advantage of the storage ring technique is the much higher luminosity compared with single-pass experiments [5], allowing studies on various kinds of molecular ions with low intensities. On the other hand, the DR of molecular ions so far reported occurs mainly around 0 eV center-of-mass (c.m.) energy and we have no knowledge of any experimental DR results at large c.m. energies, although for the dissociative excitation process, resonances have been found at high energies (20 and 26 eV for $e+HeH^+$ [4]).

In this Letter, we describe the first experimental study of HeH^+ recombination in a storage ring and report the surprising finding that there exists a large recombination resonance having a peak at around $E_{\text{c.m.}} \sim 20$ eV in addition to the known peak at $E_{\text{c.m.}} \sim 0$ eV.

An investigation was performed by using the storage ring TARN II and its associated electron cooler [6]. $^4HeH^+$ ions were produced from a He and H_2 gas mixture in a pulsed Penning ion source and accelerated to 9.53 meV in the injector cyclotron. The beam was then multiturn injected into the storage ring after being momentum analyzed to $\Delta p/p \sim 10^{-3}$. The flight time of the beam from the ion source to the injection point of the ring is approximately 50 μs. The multiturn injection took about 100 μs and the number of stacked particles was of the order of 10^6 particles. The beam circulated in the ring with a 78 m circumference at a frequency of 0.245 MHz and merged with an electron beam. The electron beam, with a diameter of 5 cm and a current of 0.2 or 0.3 A, was guided by a solenoid field of 400 G. The lifetime of the beam in the ring was about 1.5 s at an average vacuum pressure of 1.3×10^{-10} Torr.

The experimental setup has been described in our previous paper [7]. The neutral atoms produced in the electron cooler were detected by a solid-state detector (SSD) with a diameter of 34 mm installed in the vacuum extension of the cooler straight section outside the dipole magnet with a field of 2.46 kG. The length of the electron beam in the merging section is 1.5 m. The distance between the center of the electron beam and the detector is 4.5 m, which corresponds to the flight time of neutral particles of 230 ns. The neutral-beam position and size were checked with a position-sensitive solid-state detector at the same position of the SSD. The creation rate of neutral atoms was measured between times T_i and T_f while changing the electron energy in small steps. Here we refer to $T_i - T_f$ as the "time window." The time measurement was started at the time of firing the ion source and pulsing the bump magnets for the injection system.

The stored beam was kicked out and then renewed at every injection period. The neutral atoms are produced from the reactions

$$HeH^+ + e \rightarrow H + He^+ + e\,, \quad (1)$$

$$\rightarrow H^+ + He + e\,, \quad (2)$$

$$\rightarrow H + He\,. \quad (3)$$

The neutral particles produced by these reactions were identified as three peaks corresponding to particles with $\frac{1}{5}$, $\frac{4}{5}$, and full beam energy, respectively. For each counting interval, the neutral-beam count and the electron acceleration voltage were stored simultaneously in a computer with the information on the time window. The dc electron-beam current was confirmed to be constant at a fixed gun-anode voltage by reading the collector current during experiments. The ion-beam current was monitored by measuring the background neutral rate off the resonance which was mainly created from the interaction of the beam with residual gas. The ion-beam intensity was almost constant during measurements. The absolute value of the circulating beam current was, however, too low to measure with a Permalloy current transformer.

The actual energy of the electrons at an acceleration voltage V_c is given by [7]

$$E_e = (V_c + V_s)e\,, \quad (4)$$

where V_s is the space-charge potential in the electron beam and e is the electron charge. V_s is given by

$$V_s = -KI_e/E_e^{1/2}\,, \quad (5)$$

where I_e is the electron current and K is a constant which depends on the degree of space-charge compensation and the radial distance measured from the electron-beam axis.

The rate of neutral (He+H) atoms formed in the electron cooler for different time windows is shown in Fig. 1 as a function of the electron acceleration voltage. The central sharp peak for each spectrum comes from the usual DR process, which is located at zero detuning energy ($E_{\text{c.m.}}=0$). Here we refer to this peak as DR-A. We can calculate the constant K in Eq. (5) from the beam energy, the electron current, and the electron acceleration voltage corresponding to this peak. By using this value, electron energies on the axis were calibrated in an absolute way. For the DR-A peak, weak shoulders appear for the delayed time window. New DR maxima were observed for electron velocities both slower and faster than the ion-beam velocity almost symmetrically with respect to the central peak. Here we refer to these peaks as DR-B. Each peak appears to comprise two gross components. The first component is located in the c.m. energy of around 20 eV and the second component forms a shoulder at around 30 eV. Although the electron energy was further scanned up to 150 eV c.m., no prominent resonances were found. The ratio of the peak yield of DR-A to that of DR-B changes with the variation of the time window. For the measurement just after the injection, the peak DR-A is stronger than the peak DR-B, while for the one after a long-time storage, the relation is reversed. Such a situation can be more clearly seen in Fig. 2, where the relative peak heights are plotted as a function of storage time. In this figure, the peak intensity at each datum was normalized by the constant background off the resonance which reflects the ion-beam intensity.

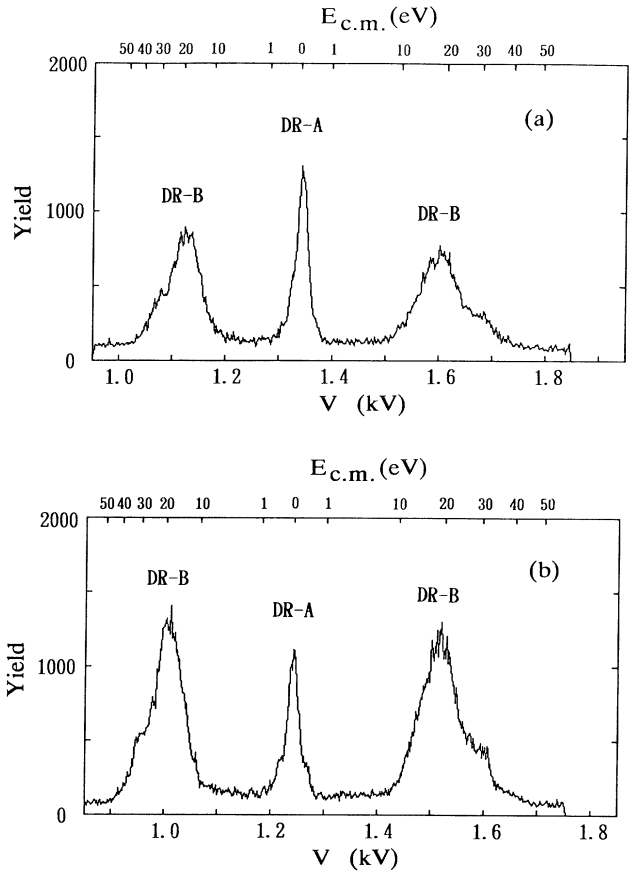

FIG. 1. Yields of neutral He+H atoms formed in the electron cooler as a function of electron acceleration voltage. The c.m. energy scale is also shown. The time window (see the text) is (a) 0–30 ms and (b) 0.92–1.47 s and the electron current is (a) 0.3 A and (b) 0.2 A.

For DR-A, the rapid decrease of the rate in a short time appears to show the quench of the excited-state population and the increase of the ground-state population with storage time, just like the decrease of cross sections from high- to low-extraction conditions of the ion source observed in Ref. [4]. The shoulder which appears for the measurements after the long-time storage also supports this view. The shoulder corresponds to a c.m. energy of about 0.1 eV, which almost agrees with the result of Ref. [4]. However, the deep resonance in the 20–30 meV region reported in Ref. [4] has not been observed. This

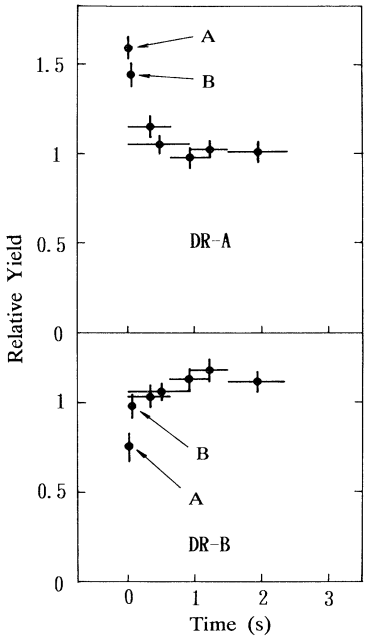

FIG. 2. Relative yields of DR-*A* and DR-*B* (low-electron-energy side) peaks as a function of storage time. Horizontal bars represent the width of the time window. The data indicated by *A* and *B* correspond to the time windows 0–3 ms and 0–30 ms, respectively. The data are normalized to unity at the yield of the DR-*A* peak for the time window of 1.5–2.33 s.

might be obscured by the partly modified velocity distribution of the ion beam due to the drag force of the electron cooling. The drag force has a peak at the relative energy of about 0.3 meV and is effective to the order of 30 meV [6]. The cooling time can be estimated to be about 3 s by using the cooling data measured for the proton beam [6] and taking account of the dependence on the electron velocity and current and on the ion mass and charge.

The DR-*B* peak could be explained as the dissociation through two-electron excited states which form Rydberg manifolds converging to the first and more highly excited states of HeH^+. Potential energies for the ground and excited $^1\Sigma$ and $^3\Sigma$ states of HeH^+ have been studied by Michels [8] and Green *et al.* [9]. The ground state of HeH^+ correlates to $He(1s^2)+H^+$ and the first excited $^1\Sigma$ state correlates to $He^+(1s)+H(1s)$ in the limit of separated atoms. According to the Franck-Condon principle, the transition energy from the ground state with an internuclear distance of about 1.5 bohrs to the first excited $^1\Sigma$ state can be estimated to be about 25 eV from the potential-energy curves as can be seen in Fig. 3. The high-energy tail of the first component which has a peak at about 20 eV extends to this energy region. The low-energy side of this component spreads down to about 10 eV. This limit appears to come from the transition to the $^2\Sigma^+$ two-electron excited state which dissociates to $H(1s)$ and $He^*(1s2s)$ [10], taking account of the finite width of the internuclear distance. Thus the first component is formed through the two-electron excited states converging to the first excited state of HeH^+. Those are not only $^2\Sigma^+$ states, but also quartet states and Π states; many states could contribute to the dissociation. This is likely to be the reason that the cross section in DR-*B* becomes large in spite of the high incident energy. On the other hand, the transition energy from the ground state to the lowest state of the second excited $^1\Sigma$ group which correlates to $He^*(1s2l)+H^+$ and $He^+(1s)+H^*(2l)$ in the limit of separated atoms is about 33 eV (see Fig. 3). This energy approximately corresponds to the maximum energy of the second component of the experimental data. In the same way as the discussion on the first component, the lower-energy side of the second component is likely to originate from transitions to the series of the HeH system which energetically lie between the limits of $He^+(1s)+H(1s)$ and $He^*(1s2l)+H^+$ and dissociate to $He^*(1s2l)+H^*(nl)$ or $He^*(1snl)+H^*(2l)$.

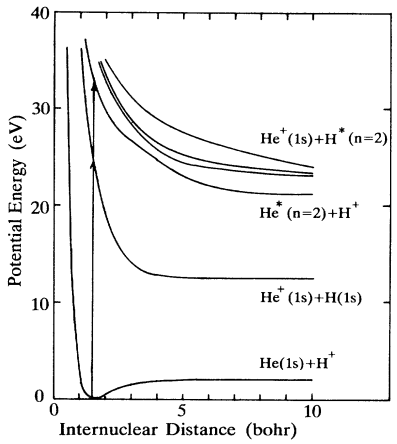

FIG. 3. Potential energy curves for the ground and excited $^1\Sigma$ states of HeH^+ (Ref. [9]).

The relative energy between the He and H pair for DR-*B* after dissociation is much higher than that for DR-*A* due to the steep change of final potential energies for DR-*B*. Experimentally, the present high beam energy facilitates the simultaneous detection of both particles despite such a large relative velocity.

As can be seen in Fig. 2, the rate of the DR-*B* peak decreases when the storage time decreases to zero. The cross section for this process is considered to depend on the Franck-Condon overlap between the initial vibrational wave function and the final dissociative one. For the steeply decreasing final potential, the wave function oscillates violently as the internuclear distance is apart from the classical turning point. On the other hand, if the HeH^+ initially stays at a highly excited vibrational state and has a diffusive and nodal wave function, the overlap between the initial and final wave functions is expected to

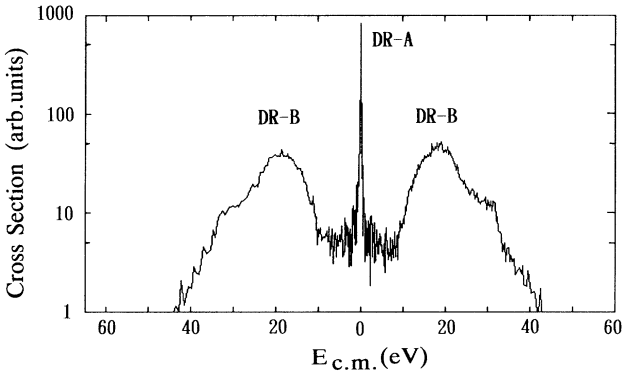

FIG. 4. Relative cross sections of the HeH^+ DR reaction corresponding to the spectrum in Fig. 1(b) as a function of c.m. energy. Right and left halves of the spectrum correspond to the regions $V_e > V_i$ and $V_e < V_i$, respectively. The background has already been subtracted.

be poor, resulting in a smaller transition probability. Such an effect can explain the decrease of the DR-B peak at the short storage time, if the vibrationally excited-state population contained initially in the beam is still unrelaxed.

The DR of quasimetastable excited states of HeH^+ has been reported under the high-extraction condition of the ion source in Ref. [4]. These states already have large internal energies and only a little energy is needed for the DR process. Therefore it seems that these states contribute much to the DR-A peak, but little to the DR-B peak which occurs at electron energies more than 10 eV. This can also explain the variation of the DR-A and -B peaks with time, if the beam initially contains a certain fraction of metastable states and they decay in a moderately short time.

In order to study the relative cross sections for the DR-A and -B processes, the data in Fig. 1(b) were transformed to relative cross sections. The cross section for the process under study is determined using the expression

$$\sigma = (C_n e^2 / I_i I_e L) |V_i V_e / (V_i - V_e)| F , \quad (6)$$

where I_i, I_e, V_i, and V_e are the ion and electron currents and velocities, respectively, L is the length of the collision region, F the effective collision area, and C_n the signal count rate. Relative cross sections were obtained as shown in Fig. 4, assuming a constant F value and neglecting the energy widths of the beams. For DR-A, the data for $E_{\text{c.m.}} > 0.01$ eV are plotted. As can be seen in the figure, the cross sections of DR-B are smaller than those of DR-A by about 1 order of magnitude. If we assume the cross sections of DR-A are of the order of 10^{-15} cm^2 as observed for the low-extraction condition in Ref. [4], the cross sections of DR-B are of the order of 10^{-16} cm^2.

This Letter reports a marked recombination resonance of HeH^+ at large c.m. energies. Apparently more work is required before a satisfying explanation can be given for this phenomenon. We hope that these results will help to stimulate further research for HeH^+ both experimentally and theoretically.

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