Search for H²⁻ resonances in the detachment of H⁻ by electron impact with a high-resolution cooler ring

T. Tanabe, I. Katayama, H. Kamegaya, K. Chida, T. Watanabe, Y. Arakaki, and M. Yoshizawa Institute for Nuclear Study, University of Tokyo, Tanashi, Tokyo 188, Japan

> Y. Haruyama and M. Saito Kyoto Prefectural University, Kyoto 606, Japan

> > T. Honma

Cyclotron and Radioisotope Center, Tohoku University, Sendai 980, Japan

K. Hosono and K. Hatanaka Research Center for Nuclear Physics, Osaka University, Ibaraki 567, Japan

F. J. Currell

University of Electro-Communications, Chofu, Tokyo 182, Japan

K. Noda

National Institute of Radiological Sciences, Anagawa, Chiba 260, Japan (Received 20 May 1996)

Electron-impact detachment of H^- was studied with the TARN II storage ring (at the Institute for Nuclear Study, University of Tokyo) and the associated high-resolution electron beam. The relative cross sections were measured for the relative energies between electron and ion from 0 to 60 eV in search of the H^{2-} resonances that were reported earlier. No evidence was found for the existence of these resonance states. [S1050-2947(96)03711-0]

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INTRODUCTION

A quarter of a century ago, Walton, Peart, and Dolder found two resonances in the collision $H^- + e \rightarrow H^0 + 2e$ at incident electron energies of 14.5 eV [1,2] and 17.2 eV [3] with widths of about 1 and 0.4 eV, respectively. These resonances are located at energies slightly above threshold for the complete breakup of the four-particle system consisting of a proton and three electrons (see Fig. 1) and were attributed to the formation of H^{2-} with a lifetime of the order of 10^{-15} s. They employed an inclined ion- and electron-beam method in their experiments with an energy resolution of 0.25 eV [2]. Taylor and Thomas [4,5] theoretically classified the 14.5- and 17.2-eV resonances as having $(2s)^2(2p)[^2P^o]$ and $(2p)^{3}[^{2}P^{o}]$ configurations using the stabilization method. On the other hand, Robicheaux, Wood, and Greene [6] recently reported two independent types of *ab initio* calculation for three electrons in the field of a proton. Neither calculation shows any evidence of the existence of a resonant state of H^{2-} contributing to $e+H^{-}$ inelastic scattering, at energies above the three-electron escape threshold. Their calculations suggest that previous experimental and theoretical studies of the system were in erroneous agreement in suggesting that two ${}^{2}P^{o}$ resonances exist in H^{2-} above the three-electron escape threshold. Recently, Andersen and coworkers [7,8] studied the collision $D^- + e \rightarrow D^0 + 2e$ with a storage ring technique and concluded that no structure related to the existence of short-lived H²⁻ resonance states was observed. However, the electron affinity of atomic hydrogen

is not exactly the same as that of atomic deuterium, although the difference is small (4×10^{-4}) [9]. Even this minor difference may not be ignored for a subtle and fragile system like H^{2-} due to the large amount of repulsion. It would be desirable to conduct other independent experiments on this sub-



FIG. 1. Energy diagram of hydrogen. The H^{2-} resonances proposed by Walton, Peart, and Dolder [1-3] are also shown.

4069

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FIG. 2. Schematic diagram of the TARN II cooler ring. Neutral particles produced in the cooler straight section are detected by a neutral particle detector.

ject. In the present work, we have reinvestigated the same $H^- + e \rightarrow H^0 + 2e$ collision as the earlier experiment, but with a different experimental technique using a storage ring and an ultracold electron beam.

EXPERIMENTAL APPARATUS

The investigation was performed by using the TARN II storage ring and its associated electron cooler at the Institute for Nuclear Study, University of Tokyo [10]. The H⁻ ions were produced in a pulsed Penning ion source and accelerated to 13.8 MeV in the injector cyclotron. The beam was then multiturn injected into the storage ring with a circumference of 78 m, as shown in Fig. 2. The number of stored ions was of the order of 10^7 particles. The ion beam circulated in the ring at a frequency of 0.65 MHz and merged with an electron beam with a length of 1.5 m and a diameter of 5.2 cm guided by a solenoid field of 0.35 kG. The electron beam current was 0.32 A, which corresponds to a density of 1.8×10^7 cm⁻³ at an energy of 7.5 keV, equivalent to the velocity of H⁻ ions. The beam lifetime during which the intensity decreased to 1/e of its original value was about 3 s at an average vacuum pressure of 1×10^{-10} mbar. The stored ions were cooled both longitudinally and transversely to a diameter of 1 mm within a time of less than 1 s. The cooled beam profile was monitored with a nondestructive residual gas ionization beam profile monitor [11]. The electron beam was produced from a heated cathode with a diameter of 1.4 cm in a solenoid field of 4.8 kG and then adiabatically expanded to a diameter of 5.2 cm while the field was gradually reduced to 0.35 kG. Thus the electron beam entered into the cooling solenoid after being enlarged by a factor of about 14 in cross-sectional area. Since the transverse electron temperature is proportional to the solenoid field [12], we can expect a much lower temperature than the initial temperature of about 100 meV corresponding to the cathode temperature of 1200 K. The expected values for the transverse and longitudinal temperature in the comoving system with electrons are approximately given by [12,13],

$$\delta E_{\perp} = \frac{B_{\text{cool}}}{B_{\text{gun}}} \left(k T_{\text{cath}} \right), \tag{1}$$

$$\delta E_{\parallel} = \frac{(kT_{\text{cath}})^2}{4E_e} + 2m_e c^2 r_e \left(\frac{4\pi n_e}{3}\right)^{1/3},\tag{2}$$

where kT_{cath} is the energy spread of the electron beam in the laboratory system originated from the heated cathode. B_{gun} and $B_{\rm cool}$ are magnetic fields in the electron gun and the cooling region, respectively, E_e is the electron cooling energy, m_e is the electron mass, r_e is the classical electron radius, and n_e is the electron density. The expected transverse energy spread estimated by Eq. (1) is $\delta E_{\perp} = 7$ meV for the conditions mentioned above. Such a low temperature was actually confirmed in our previous experiments [14]. For the longitudinal energy spread, the first term in Eq. (2) results from the transformation of the thermal energy to a frame of reference traveling at the cooling velocity. For our conditions, the first term has a value of 0.3 μ eV at the electron cooling energy of 7.5 keV. On the other hand, the second term in Eq. (2) results from the Coulomb field of the surrounding electrons and random relaxation motion, and is not changed during acceleration. This term contributes 0.12 meV to the longitudinal energy spread and is dominant in comparison with the first term. After all, the longitudinal temperature is much less than the transverse temperature. The resolution at the cooling energy in which relative energy $E_{\rm rel} \sim 0$ is determined mainly by δE_{\perp} . However, the overall resolution at the detuned condition depends on the relative energy between electron and ion beams. The actual energy resolution at the relative energy of $E_{\rm rel}$ is given by [15]

$$\delta E = \delta E_{\parallel} + \delta E_{\perp} \pm 2\sqrt{E_{\rm rel}\delta E_{\parallel}}.$$
 (3)

If we assume $\delta E_{\perp} = 7$ meV and $\delta E_{\parallel} = 0.12$ meV, we have $\delta E \sim 0.1$ eV at $E_{rel} = 15$ eV. Thus, the resolution is sufficient to detect the structure of resonances with the width of 1 and 0.4 eV.

The neutral atoms produced in the electron cooler were detected by a solid-state detector (SSD) with a diameter of 46 mm installed in the vacuum extension of the cooler straight section outside the dipole magnet (see Fig. 2). The distance between the center of the electron beam and the detector is 4.5 m. There are neutral hydrogen atoms arising from the collisions with the residual gas molecules as well as those produced from the electron-H⁻ collisions. The amount of such background neutral atoms can easily be estimated from the yield at zero relative energy, because detachment from electron-H⁻ collision does not occur below the threshold of 0.75 eV. The ratio of the H⁰ production rate at $E_{\rm rel} \sim 0$ and $\sim 40 \text{ eV}$ is about 1:2. This means that half of the yield at $E_{\rm rel} \sim 40$ eV comes from the background process. Absolute cross sections were hard to determine, because the circulating ion current was too low to measure with a dc-current transformer.

The experiments were performed by measuring the rate of H^0 production as a function of electron acceleration voltage. The injected ions were first stored for 2 s, which is more than



FIG. 3. Relative rate coefficients as a function of relative energy. Positive (negative) energy means that the electron velocity is faster (slower) than the ion velocity.

sufficient to allow both longitudinal and transverse cooling of the ion beam. After this period, the electron acceleration voltage was stepped up (or down) to a measuring voltage and switched back and forth between measuring voltage and cooling voltage at a frequency of 20 Hz in order to avoid an energy shift of the ions due to the drag force of the electrons. The yields at the measuring voltage were normalized to those at the cooling voltage, since the latter yield is proportional to the stored ion current for a constant vacuum pressure in the ring. Since the rising (or falling) time of the electron acceleration voltage is about 0.1 ms, the counting was suppressed during the transient time. The electron acceleration voltage was scanned over an energy range corresponding to relative energies between 0 and 60 eV in steps of 2-4 V.

RESULTS AND DISCUSSION

Figure 3 shows the rate coefficients $\langle v \sigma \rangle$ as a function of the relative energy. Positive (negative) energies indicate that the electrons are faster (slower) than ions. As expected from the results of the electron-impact detachment of D⁻ [7], a dip is observed near threshold. The spectrum is almost symmetric with respect to zero relative energy. As can be seen in the figure, no structures can be observed at the expected resonance energies of 14.5 and 17.2 eV.

In order to search for the resonances in more detail, we scanned the electron energy more finely in the energy region where two resonances were found earlier. The relative cross section was obtained by dividing $\langle v \sigma \rangle$ by the relative velocity v, neglecting the energy spread of the beams. The results are shown in Fig. 4 together with the data of Peart and Dolder [3]. Since the absolute cross sections were not measured in this experiment, the present data were scaled in magnitude and normalized to earlier data in order to facilitate comparison. As can be seen in the figure, there is no definite structure in the present data. This is in agreement with recent experimental results of Andersen and co-workers [7,8] on D⁻ and also the theoretical works of Robicheaux, Wood, and Greene [6].

Here we consider the difference in experimental condi-



FIG. 4. Comparison of H^- detachment cross sections as a function of relative energy. Absolute values of present data (filled squares) are normalized to those of Peart and Dolder [3].

tions between the inclined-beam method and the mergedbeam cooler-ring method, because the appearance of resonances seems to be sensitive to experimental conditions and the resonance system can easily be distorted with external fields. The inclined-beam method sacrifices energy range and resolution and, because the beams interact only over a short path, it yields smaller signals [2]. The collision geometry is well defined, however, in the inclined-beam method. On the other hand, the cooler-ring method allows measurements over a wide energy range with high resolution. Since it is easy to scan a wide energy range, we can readily observe a symmetrical spectrum with respect to the zero relative energy. This symmetry confirms that the measurements were performed under correct experimental conditions. Furthermore, the storage-ring method gives higher luminosity, because the beam intensities are much higher and the beams interact over a long path. A big difference between the cooler-ring method and the inclined-beam method is the presence of the solenoid field that confines the electron beam in the cooler-ring case. The direction of the magnetic field is, however, parallel to the ion-beam movement, which does not yield any $v \times B$ motional electric field to a first approximation. The optimization of the cooling requires the electronand ion-beam axes to be aligned within about 1 mrad. As the electrons move spiraling along the magnetic field lines, the alignment errors yield a transverse magnetic field for the ions, resulting in an equivalent motional electric field. The divergence angle of the ion beam can be estimated to be ~ 0.1 mrad from the observed beam width of ~ 1 mm and an amplitude function of the storage ring. Since the alignment error can be assumed as the same order as the divergence angle, a resultant transverse component of the solenoid field produces a motional electric field of ~ 2 V/cm for the present experimental conditions. The space charge of the electron beam forms a parabolic potential distribution in the radial direction. The electron cooling condition is optimal at the bottom of the parabola. As the ion-beam flux has a finite width, a small displacement of ions from the electron-beam axis can exist even for the well-aligned beam. Thus, an elecThe merged-beam experiment we have performed differs from inclined- and crossed-beam experiments in another way. In our experiment, there is a motional electric field due to the dipole magnet passed by the H⁰ atoms on their way to the detector (see Fig. 2). This field will ionize states of excited H* with a high value of principal quantum number *n*. The strength of a dipole field of 1.3 kG provides an estimate of the maximum quantum number (n_{max} =10), which the hydrogen atoms can have and still be able to pass through the analyzer unaffected to reach the detector according to [17],

$$n_{\rm max} = (6.2 \times 10^{10} q^3 / \nu_i B)^{1/4}, \tag{4}$$

where q is the initial charge on the ion before recombination, v_i is the ion velocity in m/s, and B is the magnetic-field

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strength in *T*. Above the triple escape threshold at 14.35 eV incident electron energy, there are an infinite number of double-escape continua, such as H(n) + e + e, that are energetically open. If hydrogen atoms formed through the resonances are in highly excited Rydberg states with n > 10, they cannot reach the detector, because they are ionized and deflected by the dipole magnet.

In summary, the earlier proposed resonances related to doubly charged negative hydrogen ions were not observed in the cooler-ring experiments. The reason for the discrepancy between the earlier experiment and the present one remains unclear, but it is true that there are some differences in experimental conditions.

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