Electron-Impact Detachment of D^-: Near-Threshold Behavior and the Nonexistence of D^{2-} Resonances

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D^- has been stored in the heavy-ion storage ring ASTRID and merged with the electron beam from an electron cooler. The cross section for electron-impact detachment of D^- was measured for relative energies from 0 to 20 eV. No evidence is found for the earlier reported resonances that were ascribed to short-lived H^{2-} states. We present a simple classical "reaction-zone" model which essentially reproduces the data except in the region close to the threshold where the cross section is small.

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We have studied electron-impact detachment of the negative-hydrogen isotope D^-:

$$e^- + D^- \rightarrow e^- + e^- + D^0. \tag{1}$$

We address two problems that are of contemporary interest in atomic physics: (1) The cross-section behavior near threshold, which is of basic interest from a theoretical point of view and important for all classes of experiments which determine threshold energies. Experimental data for electron-impact detachment are reported here in the low-energy region from threshold (0.75 eV) to 20 eV. (2) We have reinvestigated an earlier claim of resonances in the detachment cross section that might be associated with triply excited states of D^{2-}. Such resonances would be very spectacular and challenging for theoretical models since the resonances would represent states possessing an extreme amount of electron correlation.

Detachment of negative ions by photons results in two outgoing particles and the cross-section behavior near threshold has been studied intensively (see, e.g., Ref. [1] and references therein). The Wigner threshold law [2] has been found to describe the behavior of the cross section a few meV above threshold. For impact by electrons, there are no experimental data available in the energy region around threshold and the situation is more complex since there are three particles in the final state. The well known Wannier theory [3] is not applicable since the residual is a neutral atom rather than a positive ion. We know of only one work that deals theoretically with the energy dependence of the electron-impact detachment cross section near threshold. Hart, Gray, and Guier [4] made use of general properties of the solutions to the time-independent three-particle Schrödinger equation valid outside a reaction zone. At threshold, the cross-section behavior becomes independent of the short-range interaction and Hart, Gray, and Guier calculated the cross sections using Green's functions. With a reaction-zone radius of $4a_0$, they derived the expression [correct rendition of their Eq. (28a)]

$$\sigma \propto (E - E_{\rm th})^{9/4} \exp\left[-16.4/\sqrt{(E - E_{\rm th})}\right], \tag{2}$$

which was expected to be valid at energies much smaller than 0.4 eV above threshold. There are a number of second Born approximation calculations [5], but the validity in the region around threshold is doubtful. Here we present a simple classical model that reproduces our data over a wide energy range. The approach is similar to that employed to account for reaction cross sections in nucleus-nucleus scattering [6].

In recent years, there have been a number of papers discussing the possible existence of doubly charged atomic negative ions [7–12]. Experiments have indicated that ions such as H^{2-}, O^{2-}, F^{2-}, and Cl^{2-} exist in free space with lifetimes of the order of 10^{-6} to 10^{-15} s. The most extreme case is H^{2-}, which belongs to the three-electron isoelectronic series H^{2-}, He^-, Li, Be^+, etc. The electron-electron repulsion in H^{2-} is much more important than in other members of the isoelectronic series, which makes the system much more "fragile" and complicated to treat theoretically.

Walton, Peart, and Dolder [13–15] reported on two pertinent resonances in the collision $e^- + H^- \rightarrow e^- + e^- + H^0$ at 14.5 and 17.2 eV, respectively, each with a width of about 1 eV (equivalent lifetime of $\sim 10^{-15}$ s). These resonances were attributed to short-lived states of H^{2-}. The existence of such states at an energy slightly *above* the threshold for complete breakup (0.75 + 13.6 eV) of the four-particle system was indeed exceptional The states were attributed to the $(2s)^2 2p(^2P^0)$ and $(2p)^3(^2P^0, {}^2D^0)$ configurations by Taylor and Thomas [16,17] for the low- and high-energy resonances, respectively. Despite this agreement between early theory and experiment Robicheaux, Wood, and Greene [9] have recently questioned the existence of such resonance states. Since the work of Taylor and Thomas, the capability of handling electron correlation nonperturbatively has improved dramatically. Robicheaux, Wood, and Greene presented two independent types of *ab initio* calculations for the three electrons in the field of the proton and neither showed any evidence for the earlier proposed resonances. It was also argued [9] that the observed cross-section

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variation violated unitarity of the scattering matrix or flux conservation.

The measurements by Peart, Dolder, and Walton have not been confirmed by any experimental group for more than twenty years. In the present work, we have performed new, independent, high-resolution measurements of the electron-negative hydrogen ion scattering cross section in the energy region where the two resonances were seen earlier. No structure related to the existence of short-lived H^{2-} resonance states was observed.

The present experiment was carried out at the Aarhus Storage Ring Denmark (ASTRID) [18]. The ring is 40 m in perimeter and has two 45° bending magnets in each of the four corners, as shown in Fig. 1. The D^- ions were produced in a conventional duoplasmatron ion source and injected into the ring at an energy of 150 keV. The ion current was typically $\sim 5\ \mu A$. In the ring, the ions were accelerated to an energy of either 2 or 4 MeV at which energies the ions were merged with an almost monoenergetic beam of electrons from the electron cooler (a description of the electron-cooler device was given by Andersen, Bolko, and Kvistgaard [19]). Injection and acceleration were accomplished in about 5 s. After acceleration, the D^- beam had a storage lifetime of about 2 s at the average ring pressure of $\sim 3 \times 10^{-11}$ mbar. In the present work, we used the heavy hydrogen isotope D^- instead of H^-, since D^- is easier to store and accelerate in the storage ring. The difference in the electron affinity

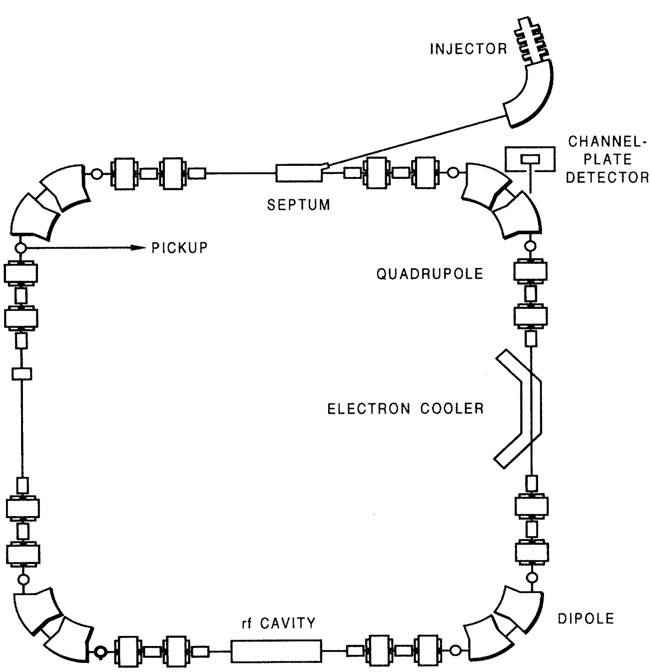

FIG. 1. Schematic drawing of the storage ring ASTRID. The electron cooler is used as the electron target (interaction length of 85 cm).

for these two isotopes is known to be only $\sim 4 \times 10^{-4}$ eV [20] and the electronic properties of D^- are essentially identical to those of H^-.

We studied the detachment reaction (1) at different relative energies E, obtained by tuning the energy of the electrons. Behind the 45° bending magnet following the electron-cooler device, the neutral D^0 particles were counted by a 25 mm diameter channel-plate detector (see Fig. 1). Experimentally, we determined a "rate coefficient" given by

$$\langle v\sigma \rangle = \frac{N(D^0) - N_0(D^0)}{N(D^-)} \frac{v_i}{l \varepsilon \rho_e}, \tag{3}$$

where v_i is the ion velocity, l (= 85 cm) is the effective length of the interaction section, ε is the ion-detection efficiency, and ρ_e is the electron density. $N(D^0)$ is the yield of neutral particles measured by the detector, and $N_0(D^0)$ is the contribution from collisions with the background gas and a small contribution due to electron-impact detachment in the toroid region where the two beams were brought into merging conditions. $N(D^-)$ is the number of beam particles per second passing through the interaction region. $N_0(D^0)$ was easily measured at $E = 0$, since no detachment signal from the interaction region was present below the threshold. The relative electron-beam energy was modulated at 25 Hz between 0 (cooling) and energy E (measuring), whereby $N_0(D^0)$ and $N(D^0)$ were obtained. Relative measurements were obtained by using $N_0(D^0)$ as a measure of the beam current since this number is proportional to the stored ion current for a constant pressure in the ring.

The absolute rate coefficient was obtained from

$$\langle v\sigma \rangle = (k - k_B) \frac{L}{l} \frac{1}{\rho_e}, \tag{4}$$

where L is the perimeter of the ring, k is the total decay rate, and k_B is the background-decay rate primarily due to collisions with the background gas. The decay rates were found from the measured storage lifetimes τ ($k = 1/\tau$). The total decay rate k changed from about 0.65 s^{-1} at $E = 20$ eV relative energy to $\sim 0.5\ s^{-1}$ at $E = 0$. The experimental uncertainty on the absolute rate coefficient is estimated to be $\pm 30\%$.

The energy resolution in the experiment is determined by the velocity distribution of the electrons which is given by [19]

$$f(v) = \left(\frac{m}{2\pi k T_\perp}\right) e^{-m v_\perp^2 / 2kT_\perp} \sqrt{\left(\frac{m}{2\pi k T_\parallel}\right)}\, e^{-m(v_\parallel - \Delta)^2/2kT_\parallel}, \tag{5}$$

where m is the electron mass, $v_\perp$ and $v_\parallel$ are the electron-velocity components perpendicular and parallel to the ion-beam direction, respectively, and Δ is the detuning velocity that defines the relative energy ($\frac{1}{2} m \Delta^2$). It was found that the two temperatures were $kT_\perp = 0.15$ eV and $kT_\parallel = 0.001$ eV with an unexpanded electron beam in the present electron cooler [19]. Measurements of the cooling force on D^+ ions stored in ASTRID with the same electron beam indicated a transverse temperature $kT_\perp$ of about 0.1 eV. To improve the electron-beam temperature, we expanded the electron beam adiabatically in the present measurement. The magnetic field in the cooler was reduced from 1 kG to 200 G prior to the interaction region. This resulted in a fivefold reduction of the transverse-energy spread [21] which became $\sim$0.02 eV sufficient to detect any $\sim$1 eV wide resonance structure in the cross section.

Figure 2 shows the rate coefficient as a function of the relative energy. Negative (positive) energies correspond to the situation where the electrons move slower (faster) than the ions. The slight asymmetry that is observed in the spectrum is related to the change in the detachment contribution from the regions in the toroids where the beams gradually merge and demerge and where the relative energy is finite even at the cooling condition. The contribution from the toroid regions was at the maximum estimated to be 10%–20%. A small change in the vacuum conditions during the measurement caused by the change in the electron current and energy also generated a small asymmetry in the data. The cross section normalized according to Eq. (4) is shown in Fig. 3. It was obtained by dividing $\langle v\sigma\rangle$ by the relative velocity v.

To understand the general behavior of the cross section, we made a simple classical model calculation. We assumed that the incoming electron experienced a purely repulsive Coulomb potential. The distance of closest approach $D(\rho)$ as a function of the impact parameter ρ is then given by

$$D(\rho) = \tfrac{1}{2}D_0 + \sqrt{\left(\tfrac{1}{2}D_0\right)^2 + \rho^2}, \qquad (6)$$

where D_0 (in a.u.) is $27.2/E$, E being the electron energy in the ion-rest frame in eV. It is assumed that detachment takes place with a constant probability p if the electron gets inside a certain reaction radius R. The cross section can then be expressed as

$$\sigma = 2\pi \int_0^\infty \rho\, d\rho \times \begin{cases} p & [\text{for } D(\rho) \le R] \\ 0 & [\text{for } D(\rho) > R] \end{cases}$$
$$= p\pi R^2 \max[0, (1 - U_c/E)], \qquad (7)$$

where $U_c = 27.2/(R/a_0)$ (in units of eV). As seen in Fig. 3, this classical "reaction model" reproduces the general behavior of the data remarkably well when the reaction radius R is equal to $15a_0$ and $p = 20\%$. Note that p is just a scaling factor which has no influence on the shape of the cross section. The value obtained for R corresponds to the distance where the spatial probability distribution function for the loosely bound electron in D^- becomes vanishingly small (see, e.g., Ref. [8]) and thus appears reasonable. At high energy, the model that predicts a constant cross section of $p\pi R^2$ breaks down. The cross section is known to scale as $\ln E/E$ in the Bethe approximation [22].

The value obtained for R yields a cutoff energy $U_c = 1.8$ eV, which is significantly larger than the threshold energy of 0.75 eV. The electron cloud does not end abruptly at $r = R$ and within the classical reaction model detachment at energies below U_c is associated with impact parameters for which $D(\rho) > R$. Our data do not allow a rigorous test of the threshold theory by Hart, Gray, and Guier [4]. Yet, in order to be consistent with the experimental data, the cross section has to be practically zero in the region of energy where the theory is expected to be pertinent due to the strong energy dependence of Eq. (2). The theoretical cross section increases by more

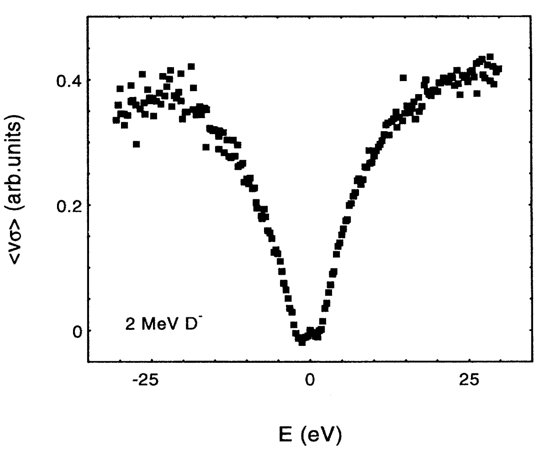

FIG. 2. Rate coefficient $\langle v\sigma\rangle$ as a function of the relative energy ($E < 0$ for $v_e < v_i$ and $E > 0$ for $v_e > v_i$, where v_e and v_i are the electron and ion velocity, respectively). The ion-beam energy was 2 MeV; the electron density at $E = 0$ was 7×10^5 cm^{-3}.

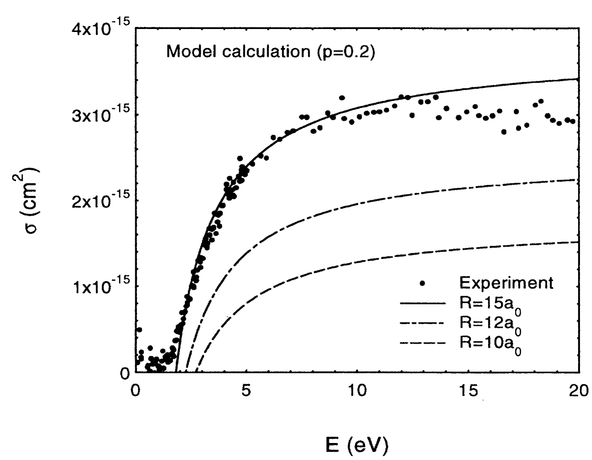

FIG. 3. Electron-impact detachment cross section of D^- as a function of energy. The model calculation is compared with the experimental data for three different values of reaction radius R.

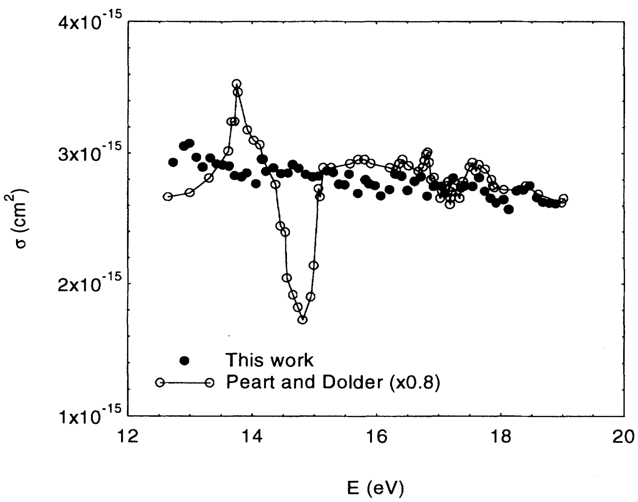

FIG. 4. The detachment cross section as a function of energy for D^- in the region where resonances had been reported. Our data are compared to H^- data of Peart and Dolder [15].

than 7 orders of magnitude when the energy increases from 0.1 to 0.2 eV above threshold. Taking the energy distribution of the electrons into account, this yields an increase in the measured cross section of more than 2 orders of magnitude for the same energy change. The fact that the cross section is small (within ~1 eV) above the threshold can be understood classically. The incident electron first loses energy to come close enough to release the bound electron.

To determine whether there are resonance structures in the cross section as seen by Walton, Peart, and Dolder [13–15], we scanned through the 13–19 eV energy region several times and at two different ion energies. The results are shown in Fig. 4 together with the data of Peart and Dolder (read from Fig. 2 of Ref. [14]). There is clearly no structure in our data that supports the existence of short-lived ($\sim 10^{-15}$ s) resonances in D^{2-}. This is in agreement with the recent theoretical work of Robicheaux, Wood, and Greene [9].

In conclusion, the cross section for electron-impact detachment of D^- has been measured from threshold to 20 eV. A simple classical model, which takes into account the Coulomb repulsion in the incoming channel, reproduces the data well. The earlier postulated resonances associated with doubly charged negative ions [15] were not observed. This indicates that the stabilization method [16,17] was not a reliable indicator of resonant structure in the full ionization continuum.

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