Single and double detachment from H^-

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Absolute cross sections for single and double detachment from H^- following electron impact have been measured over a range of collision energies from the thresholds to 170 eV. The measurements were made using a magnetic storage ring. The ions in the ring were merged with a monoenergetic electron beam and neutral and positively charged fragments were detected. We cover larger energy ranges than in many of the previous experiments, and this is the first time both single and double detachment have been measured simultaneously. This allows us to present accurate ratios between the single and double detachment cross sections. On the basis of these ratio measurements we discuss possible mechanisms leading to double detachment.

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I. INTRODUCTION

The structure and dynamics of negative ions differ intrinsically from those of atoms and positive ions due principally to the short-range nature of the force that binds the outermost electron. The simplest negative ion, H^-, is a prototypical bound three-body system that has been the focus of numerous experimental and theoretical investigations. Of particular interest are the structure of this ion and the dynamics involved in its interaction with electromagnetic radiation and other particles. Reactions involving the production and destruction of the H^- ion, for example, determine the characteristics of many astrophysical and terrestrial plasmas. Many studies to date involve the fundamentally important problem of how the H^- ion interacts with a photon. In photodetachment experiments on H^-, one (or both) of the bound electrons is liberated following the absorption of a photon [1–4]. Correlations between the two electrons play an important role in determining the threshold behavior of the cross section and in the formation and decay of doubly excited states that manifest themselves as resonance structures in the cross sections.

Of equal fundamental importance is the problem of how an H^- ion interacts with a free electron. In the process of electron-impact detachment from H^-, one or both of the bound electrons are ejected following a collision with a free electron. This process exhibits interesting features that are not present in photodetachment. For example, in the initial state of both single and double detachment the free electron experiences a repulsive Coulomb force as it approaches the H^- ion. The final continuum state interactions are also more complex than those found in photodetachment due to the fact that the free electron is not absorbed but rather scattered by the ion. In single detachment, the three-particle continuum state consists of the detached and scattered electrons moving in the field of an H atom. For double detachment, the H^- ion is completely fragmented into its constituents. In the Coulomb four-body continuum state, two detached electrons and the scattered electron move in the field of a proton. At threshold, strong correlations develop between the interacting particles in the final state. The additional complexities associated with electron-impact detachment from H^- make it very difficult to calculate cross sections, particularly for the double detachment process. *Ab initio* calculations of cross sections for electron-induced detachment over a large range of collision energies, including the threshold regions, do not exist. For this reason, it is particularly important to have accurate measurements of the cross sections for both single and double detachment.

In this article, we report on measurements of electron-impact detachment from the H^- ion over the collision energy range 0–170 eV. This range encompasses the thresholds for the single (σ_0) and double (σ_+) detachment processes

$$H^- + e^- \rightarrow H + 2e^- \tag{1}$$

and

$$H^- + e^- \rightarrow H^+ + 3e^-. \tag{2}$$

There have been several previous measurements of electron impact cross sections for single detachment from H^-. Early experiments include the work of Tisone and Branscomb [5], Dance, Harrison, and Randell [6], and Peart, Walton, and Dolder [7]. All of these experiments involved the use of a crossed beam apparatus. More recently, Andersen *et al.* [8] used a storage ring to merge an electron beam with a beam of D^- ions. The focus of this measurement,

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which covered the energy range from threshold to about 20 eV, was the threshold behavior and a search for resonances that had been previously reported by Peart and Dolder [9].

Ab initio calculations of the single detachment cross section over small energy regions have been reported in the literature, although no single calculation covering the entire energy range of the current experimental study has yet been reported. A theoretical treatment of this problem is difficult due to the strong electron correlation present in both the initial and final states. Most of the early calculations concentrated on the asymptotic region of the cross section curve, where the high energy of the electrons makes it possible to apply variations of the Born approximation. The Born-type calculations, however, differ appreciably, especially near the peak of the cross section. The low-energy region near threshold is even more difficult to treat due to the correlations that develop between the slow moving electrons. Recent calculations by Pindzola [10], Rost [11], and Robicheaux [12] have investigated threshold behavior in the case of electron-induced single detachment from H^-. Investigations of double detachment from H^- have proceeded in parallel with those of single detachment. The early crossed beam measurement of Peart, Walton, and Dolder [13] stimulated Born-approximation calculations by Tweed [14]. Later, measurements by Defrance, Claeys, and Brouillard [15] and Yu *et al.* [16] showed that a large discrepancy existed between these measurements and those of Peart, Walton, and Dolder [13]. Calculations of the double detachment cross section have not been forthcoming, again due to the fact that electron correlation plays a major role in determining the structure and dynamics of the collision process. This is particularly true at low electron energies where correlations have time to develop.

II. EXPERIMENT

The present experiment was performed at the magnetic storage ring CRYRING, situated at the Manne Siegbahn Laboratory in Stockholm. Details of the experimental procedures can be found in the recent publication by Fritioff *et al.* [17]. In the present experiment, H^- ions were produced in a Cs sputter ion source [18], extracted at 10 keV, and then injected into the ring. Within the ring the ions were accelerated to an energy of 5 MeV. On each orbit around the ring the ion beam was collinearly merged with a beam of electrons. These electrons served two purposes. First, they were used to enhance the quality of the ion beam by reducing its emittance and hence its transverse and longitudinal velocity distribution. Thus, phase-space cooling of the ions by the velocity-matched electrons improves the energy resolution of the measurements. The longitudinal and transverse electron temperatures were 0.2 and 2 meV, respectively. Second, by varying the laboratory-frame electron-beam energy, the electrons were used as collision partners in the study of the electron-impact detachment of the H^- ions in the beam. One of the principal advantages of the merged beam technique is that very low collision energies can be accessed in the center-of-mass frame. This feature has allowed us to investigate detachment cross sections all the way down to the single detachment threshold. This is technically more difficult to do if a crossed beam geometry is employed, due to problems associated with producing and controlling very low energy electrons in the laboratory frame.

A typical ring cycle consisted of the following phases: injection, acceleration, cooling, measurement, and the dumping of the beam. During the cooling phase, the H^- ions interacted with the velocity-matched electrons, which served to cool the ions. The measurement phase lasted 2 s, and within this time the electron beam was switched on and a voltage ramp was applied to the electron gun. The additional energy of the electrons was continuously changed as the voltage on the ramp rose from zero to the maximum value.

In addition to electron impact on H^-, the electrons ionized particles in the residual gas in the ring. These slow positive ions are trapped in the potential well of the electron beam. Charge exchange between the H^- ions of the beam and the trapped positive ions can produce H atoms and H^+ ions that could be recorded in the detectors used in the cross section measurements. This contribution to the background was determined in a separate measurement. In this measurement the electron beam was turned off for 0.2 s, directly after the cooling phase. During this period the interaction region is swept clean of trapped positive ions. Thereafter the electron beam is turned on again, but now in a chopped mode. The electron beam is turned on and off by an analog switch, 10 ms on and 10 ms off. The same voltage ramp as before is applied to the electron gun. The background contribution from trapped ions in the cooler is determined by comparing the results from the unchopped and the chopped measurements. The shapes of the cross sections were the same for the single and the double detachment measurements. Our chopped beam measurements showed that in the unchopped measurements, 6% and 13% of the signal were caused by trapped ions for single and double detachment, respectively.

The collision energy is the center-of-mass energy E_{cm}, which is essentially the energy of the electrons as seen by the moving ions, and is given by

$$E_{\mathrm{cm}} = (\sqrt{E_e} - \sqrt{E_{\mathrm{cool}}})^2, \tag{3}$$

where E_e and E_{cool} are the electron laboratory-frame energies corresponding to a particular collision energy and the cooling energy, respectively. In order to derive this equation, the reduced mass is approximated by the electron mass and the center-of-mass velocity is taken to be the same as the detuning velocity, $v_d = |\overline{v_e} - \overline{v_{\mathrm{ion}}}|$. The latter approximation is valid, since the reaction threshold energy in the center-of-mass frame is much greater than the electron temperature.

The ion beam current was measured in order to determine an absolute cross section. A sensitive current transformer was used for this purpose.

The production of residual H atoms was monitored to determine the single detachment cross section. These particles were detected with a surface barrier detector (SBD) placed 3.5 m downstream of the interaction region. For the double detachment process, the production of the H^+ ions

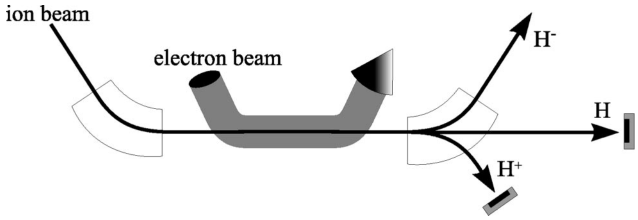

FIG. 1. A schematic of the merged beam apparatus showing the interaction region and the downstream detection region. H atoms and H^+ ions were detected in the single detachment and double detachment measurements, respectively.

was detected by a second SBD. Figure 1 shows a schematic of the interaction and detection regions. The efficiency for the collection and detection of both the H and H^+ particles was determined to be unity. In order to establish an absolute scale, it was also necessary to measure the electron beam current and determine the interaction geometry. The diameter of the cooled ion beam was much smaller than that of the electron beam, and the electron density was constant along the path of the ion beam. The major source of background in the measurements of both single and double detachment cross sections originated in collisions of the H^- ion with the residual gas. This source was minimized by maintaining the ring at a residual pressure of 10^{-11} mbar.

III. DATA ANALYSIS

The cross sections for single and double detachment are related to the measured quantities according to

$$\sigma = \frac{dN}{dt}\frac{1}{I_i}\frac{v_i e}{v_d n_e l}, \tag{4}$$

where dN/dt is the electron impact detachment reaction rate, I_i the ion current, v_i the velocity of the ions, e the elementary charge, v_d the detuning velocity, n_e the electron density, and l the length of the interaction region [17].

Two corrections have to be made in the analysis of the cross section data. First, the center-of-mass energy of the electrons must be corrected for the effect of the space charge of the electron beam. Second, due to the geometry of the cooler, the electrons interact with the ions in both the straight section of the cooler and also in the regions where the electron beam is merged into and out of the ion beam. The relative velocity of the two beams is larger here. As a result, the signal at a given electron energy will have a contribution from collisions that occur at a slightly higher center-of-mass energy [17,19].

We estimate that the total systematic uncertainty in the measurements of both the single and double detachment cross sections is 11.5%. The contributions arise from the uncertainties in the measured quantities shown in Eq. (4).

IV. RESULTS AND DISCUSSION

Figure 2 shows the cross sections that have been measured in the present work. Figure 2(a) is the cross section for single detachment from H^- over the energy range from threshold to 170 eV. The thresholds for both the single and double detachment cross sections were not well defined. This is due to the fact that tunneling occurs when the electron energy is below the height of the Coulomb barrier associated with the repulsive interaction between the incident electron and the H^- ion in the initial states. The classical model developed by Vejby-Christensen *et al.* [20] has been used to determine a model threshold of 2.8 eV, in the case of single detachment. Changes in the range of the fit to the data from 3 to 10 eV produced variations in the model threshold from 2.6 to 3.0 eV. The best fit is shown in the inset of Fig. 2(a). This energy is in excess of the electron affinity of the H atom, which is 0.75 eV. The balance is due to the work done by the incident electron in surmounting the electrostatic barrier. The magnitude and shape of the cross section shown in Fig. 2(a) agree well with that of Peart, Walton, and Dolder [7] and with that of Andersen *et al.* [8] over the limited energy range of their experiment. There is a disagreement, however, with the early work of Tisone and Branscomb [5].

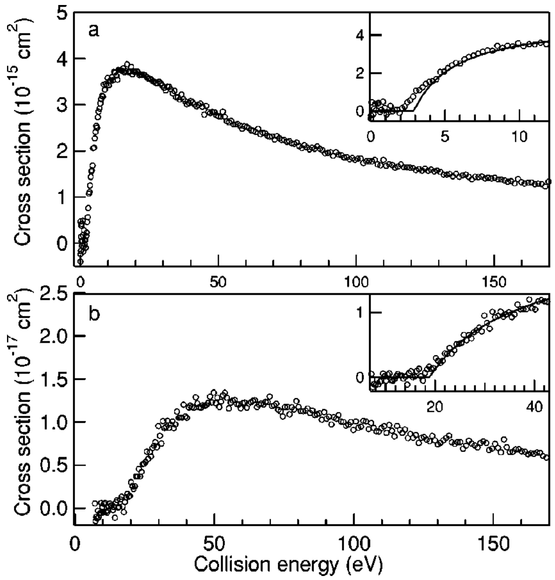

FIG. 2. The absolute cross sections for (a) single and (b) double detachment in e^-–H^- collisions. The insets show the threshold regions in more detail, including the classical over-the-barrier fits. The statistical error bars are comparable to the spread of the data points.

In Fig. 2(b) we show the cross section for double detachment from H^- over the range from threshold to 170 eV. The classical model fit yields a threshold value of 18.7(5) eV, compared to the binding energy of 14.36 eV. This value is the electron affinity of H plus the ionization energy of H. The fit is shown in the inset of Fig. 2(b). The magnitude and shape of the cross section agree reasonably well with that of Yu *et al.* [16] over the common energy range. The magnitude of the present cross section is 16% higher than that of Yu *et al.* [16] over the common energy range, but the difference is just within the combined uncertainty limits of the two measurements.

As a result of measuring both the single and double detachment cross sections with the same merged beam apparatus, we are able to determine reliable values for the ratio of

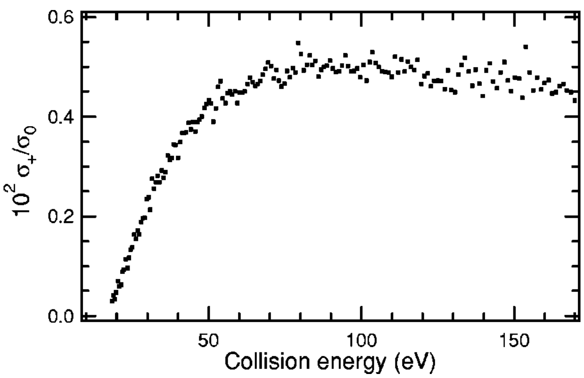

FIG. 3. The ratio of the double-to-single detachment cross sections (σ_+/σ_0).

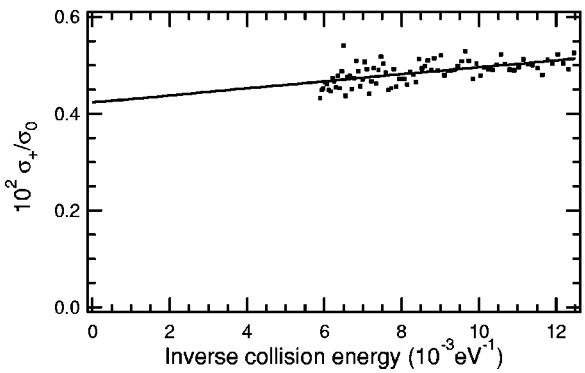

FIG. 4. The ratio of the double-to-single detachment cross sections (σ_+/σ_0) vs the inverse collision energy.

the two cross sections over the range from the double detachment threshold to 170 eV. It should be pointed out that the merged beam apparatus used in the present experiment enabled us to access the lower energy region of the cross section more readily than could be done with a crossed beam apparatus, such as the one used by Yu *et al.* [16]. Figure 3 shows the energy dependence of the ratio of the double-to-single detachment cross sections σ_+/σ_0. The statistical uncertainty in the ratio is 10% at 20 eV, 7% at 50 eV, and 7.5% at the end of the energy ramp. The uncertainty of the double-to-single detachment ratio due to systematic errors is small since these errors will cancel out when dividing σ_+ with σ_0. The uncertainty in the ion current could, in principle, contribute to a small uncertainty in the ratio since the ion current was higher in the double detachment measurement than in the single detachment measurement. Hence, we estimate that the uncertainty in the ratio due to systematic errors is less than 4%.

We will now discuss the possible mechanisms for double detachment in the case of electron impact on the two-electron H^- ion. This discussion is based on the theoretical treatment of the double ionization of the He atom by both photons and charged particles by Wang *et al.* [21]. The detachment of the outer electron in the isoelectronic systems He and H^- is initiated by the ejection of the inner electron in a collision with the incident electron. It was found that two distinct mechanisms played a major role in the double ionization of He. In one process, the detached inner electron interacts with the outer electron on its way out of the atom, leading to the ejection of the outer electron. At high incident electron energies, this interelectronic scattering process is expected to scale approximately as the inverse of the energy of the detached electron, which in turn depends on the energy of the incident electron. It has been shown that by integrating over the energy distribution of the detached electron, there is also an inverse dependence in terms of the energy of the incident electron [21]. In the case of electron impact detachment, another mechanism may also play a role. The incident electron may detach the inner electron and then scatter to detach the outer electron. This process is presumably more important for electrons than other charged particles.

Another important mechanism is the shake process, which has been discussed in the context of the detachment of B^- by Andersen *et al.* [22]. If a high-energy incident electron rapidly ejects the inner electron of H^- in a collision, the outer loosely bound electron experiences a sudden change in potential. The wave function for the remaining electron in the H atom has to relax to the new eigenstates of the altered potential. Since these states include continuum states, the second electron can be “shaken off” in the relaxation process. This shake-off process is independent of the incident electron energy at high energies. In the asymptotic energy limit the contribution from the previously described interelectronic scattering process, which has an approximate inverse dependence in the energy of the incident electron, becomes zero and only the contribution from shake off remains. Wang *et al.* [21] have shown that the ratio between double and single ionization cross sections for He becomes constant in the asymptotic limit of the energy of the incident particle. For charged particle projectiles, Wang *et al.* [21] predict this ratio to be about 0.25%, a number that has been verified experimentally. The value of the asymptotic ratio might be expected to be a little different for the H^- ion than for the He atom due to the enhanced strength of the electron-electron interaction relative to the electron-nucleus interaction in the ion. One might also expect differences in the overlap with the continuum in the shake process. The difference between the asymptotic ratios for He and H^- is, however, expected to be relatively small.

The ratio of the cross sections for double detachment and single detachment as a function of the energy of the incident electron is plotted in Fig. 3. It can be seen that over the range from about 80–170 eV, the data appears to have an approximate inverse dependence on electron energy. In Fig. 4 we have plotted the same data as a function of the inverse of the energy of the incident electron. By extrapolating the linear fit to the data we can determine the intercept on the ratio axis. This procedure yields a ratio of 0.42(5)%. It appears, however, that the data may not have a pure linear dependence in the energy range 80–170 eV. By changing the range of the fit to 130–170 eV we obtain a somewhat lower ratio intercept of 0.37(5)%, although the values agree within the quoted uncertainties. Since it is possible that we have not reached the energy above which the data has a linear energy dependence, the best we can do with the present data is quote an upper limit on the asymptotic ratio of the double-to-single detachment cross sections of about 0.4%. This value is comparable to, but somewhat higher than the predicted theoretical value of ~0.25% for He. It is, of course, conceivable that double detachment via the mechanism of double scattering of the incident electron is important and leads to a larger value for the asymptotic ratio. The ratio data reported by Yu *et al.* [16] covers a much larger range of energies, from the double detachment threshold to 2000 eV. However, the single detachment cross section used in the ratio determined by Yu *et al.* [16] was not measured in the experiment. The single detachment cross sections used in the ratio were experimental values from Peart, Walton, and Dolder [7], up to an energy of about 1000 eV, and theoretical values from Inokuti and Kim [23] over the range of 1000–2000 eV. If the ratio data of Yu *et al.* [16] is extrapolated to the asymptotic limit, one obtains a ratio of the double-to-single detachment cross sections of ~0.3%–0.4%, comparable to the value determined in the present experiment. If, however, the data from about 400–1000 eV, which appear linear, are used in the extrapolation, the asymptotic ratio is somewhat lower at ~0.25%. The measured asymptotic ratio can be used to estimate the shake-off probability, as demonstrated by Andersen *et al.* [22] in the case of the electron impact detachment of B^-. The measured ratio of 0.4% corresponds to a shake-off probability of about 8% for the outer electron in the H^- ion. This value is about an order of magnitude smaller than the corresponding probability for double detachment in the case of B^-. A shake-off probability of 8% for the H^- ion seems quite high, and would probably be lowered if a more sophisticated calculation were performed.

V. CONCLUSION

We have measured the electron impact cross sections for single and double detachment from the H^- ion over an energy range from the thresholds to 170 eV using the same merged beam apparatus for both measurements. Our result for the single detachment cross section agrees with previous measurements of the same cross section that were performed either using a crossed beam apparatus or a merged beam apparatus. In the case of the double detachment cross section, no merged beam experiments have been previously performed, but the present result agrees, within the combined uncertainties, with the previous experiment by Yu *et al.* [16]. We also present the ratios of the double-to-single detachment over a range of energies from the threshold of double detachment to 170 eV.

We have also discussed possible mechanisms leading to double detachment. Theory predicts that at sufficiently high electron energies the energy dependence of the cross section ratio should scale approximately as the inverse of the energy dependence, and in the asymptotic limit the ratio should be constant. We obtained an intercept on the ratio axis of about 0.4%, which is a little higher than the asymptotic value theoretically predicted for charged particles on the isoelectronic He atom. Possible sources of the small discrepancy are discussed.

Reliable *ab initio* calculations of the cross sections for single or double detachment over an extended range of electron collision energies do not presently exist. It is hoped that the present work will stimulate activity in this field.

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[1] H. C. Bryant *et al.*, Phys. Rev. Lett. **38**, 228 (1977).
[2] P. G. Harris *et al.*, Phys. Rev. A **42**, 6443 (1990).
[3] J. B. Donahue *et al.*, Phys. Rev. Lett. **48**, 1538 (1982).
[4] P. Balling *et al.*, Phys. Rev. Lett. **77**, 2905 (1996).
[5] G. Tisone and L. M. Branscomb, Phys. Rev. Lett. **17**, 236 (1966).
[6] D. F. Dance, M. F. A. Harrison, and R. D. Randell, Proc. R. Soc. London, Ser. A **299**, 525 (1967).
[7] B. Peart, D. S. Walton, and K. T. Dolder, J. Phys. B **3**, 1346 (1970).
[8] L. H. Andersen, D. Mathur, H. T. Schmidt, and L. Vejby-Christensen, Phys. Rev. Lett. **74**, 892 (1995).
[9] B. Peart and K. T. Dolder, J. Phys. B **6**, 1497 (1973).
[10] M. S. Pindzola, Phys. Rev. A **54**, 3671 (1996).
[11] J. M. Rost, Phys. Rev. Lett. **82**, 1652 (1999).
[12] F. Robicheaux, Phys. Rev. Lett. **82**, 707 (1999).
[13] B. Peart, D. S. Walton, and K. T. Dolder, J. Phys. B **4**, 88 (1971).
[14] R. J. Tweed, J. Phys. B **6**, 270 (1973).
[15] P. Defrance, W. Claeys, and F. Brouillard, J. Phys. B **15**, 3509 (1982).
[16] D. J. Yu, S. Rachafi, J. Jureta, and P. Defrance, J. Phys. B **25**, 4593 (1992).
[17] K. Fritioff *et al.*, Phys. Rev. A **68**, 012712 (2003).
[18] R. Middleton, Nucl. Instrum. Methods Phys. Res. **214**, 139 (1983).
[19] A. Lampert *et al.*, Phys. Rev. A **53**, 1413 (1996).
[20] L. Vejby-Christensen *et al.*, Phys. Rev. A **53**, 2371 (1996).
[21] J. Wang, J. H. McGuire, J. Burgdorfer, and Y. Qiu, Phys. Rev. A **54**, 613 (1996).
[22] L. H. Andersen, M. J. Jensen, H. B. Pedersen, L. Vejby-Christensen, and N. Djuric, Phys. Rev. A **58**, 2819 (1998).
[23] M. Inokuti and Y.-K. Kim, Phys. Rev. **173**, 154 (1968).