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H(2s) formation in H^+-H and H-H collisions

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Cross sections for metastable H(2s) formation by electron capture in H^+-H(1s) collisions and by excitation in H(1s)-H(1s) collisions have been measured over the energy range 1.9–92 keV. A fast beam of H^+ ions or H(1s) atoms was passed through a tungsten-tube-furnace target which contained thermally dissociated hydrogen. Fast metastable H(2s) atoms formed by collisions in the target were detected downstream using electric field quenching and Lyman-α photon-counting techniques. The present values are normalized at 24.5 keV to the average value of three previous independent measurements of the H(2s) formation cross section in H^+-H collisions which agree to within 20%. Measured cross sections for both H^+-H and H-H collisions contain only one maximum, in contrast with certain theoretical predictions. For H^+-H the low-energy data are in good agreement with theoretical results based on a multistate molecular treatment of the collision. Above 75 keV the H^+-H data agree with the Born approximation cross sections. Close-coupling pseudostate predictions lack overall detailed agreement with the present results, although the maximum in the cross section is reproduced well. High-energy ($E > 15$ keV) coupled-state calculations using a scaled hydrogenic two-center expansion are in good accord with the data. For H-H collisions all theoretical treatments are in poor accord with the present experimental results. Above 40 keV the measured H-H cross section is inversely proportional to the impact energy. This E^{-1} energy dependence is in agreement with high-energy theoretical predictions. However, above 10 keV the Born approximation predicts structure in the cross section due to simultaneous excitation of the projectile and target which is not observed. The present results are compared with previous experimental determinations, and discrepancies are found to exist.

I. INTRODUCTION

The collisions H^+-H and H-H are the simplest ion-atom and atom-atom interactions. Since the wave functions of H_2 are known to a high degree of accuracy and those of H_2^+ and H are known exactly, experimental investigations of H^+-H and H-H collisions yield direct tests of different scattering approximations and serve as prototypical studies for the general ion-atom and atom-atom scattering problem. For these systems, H(2s) formation is the most basic and least complex well-defined inelastic event. Consequently, experimental measurements of H(2s) formation cross sections in these systems serve as a sensitive test of theory.

Five experimental measurements of the cross section for electron capture into the 2s state for H^+-H collisions have been published and many different calculations have been made in recent years. Bayfield,[1] using a furnace-target technique, has measured the cross section over the energy range 3–70 keV. Morgan *et al.*[2] and Chong and Fite,[3] using crossed-beam techniques, have performed measurements over the energy ranges 5–26 and 6–25 keV, respectively. Agreement exists among these three measurements except at low energy. At 6 keV the cross section as measured by Chong and Fite is a factor of 2 higher than the data of Morgan *et al.* and a factor of 3 higher than the result of Bayfield. Recently, Hill *et al.*[4] have measured the cross section over the energy range 1.5–25 keV. Their results are in good accord with the 5–26-keV data of Morgan *et al.* However, at low energies their values are considerably above those of Bayfield. Disagreement in the low-energy region is not restricted to experiment. At low energies, theoretical results are extremely sensitive to approximations made in the different scattering models. As a result, a long-standing problem has existed. Low-energy calculations of the cross section for H(2s) formation in H^+-H collisions differ considerably in both magnitude and energy dependence. At high energies the situation is also unclear. The data of Bayfield appear to approach the Born approximation at 70 keV, whereas normalization of the relative data of Ryding *et al.*[5] to the data of Bayfield at 43 keV gives results about twice the Born values for energies greater than 60 keV. Furthermore, although several theoretical calculations reproduce the maximum in the cross section at about 20 keV, they lie above the experimental data at higher energies.

For H-H collisions substantially less information is available. For H(2s) excitation, only six calculations and two experimental measurements have been performed. Morgan *et al.*[6] have measured the cross section over the energy range 10–24 keV. The measurements were performed using a modulated crossed-beam technique which allowed a direct determination of the H(2p) cross

section but required a subtraction of the $2p \rightarrow 1s$ field-free radiation in order to obtain the H(2s) cross section. As a result, the relative accuracy of the H(2s) measurements are limited to ±35%. Recent measurements by Hill *et al.*,[7] using a furnace-target technique, have yielded results with much smaller quoted relative uncertainties and have extended the energy range down to 2 keV. From a theoretical point of view the situation is quite unclear. All treatments of the H-H collisional excitation problem are in serious disagreement with experiment, both in absolute value and energy dependence. Similar calculations differ by as much as a factor of 10 at low energies. Inclusion of electron-exchange effects within the multistate impact-parameter formalism results in substantially worse agreement between theory and experiment.[6]

The paucity of data coupled with the lack of consistency of available information for these most basic ion-atom and atom-atom collisions have motivated us to measure the cross sections for the following processes:

$$H^+ + H(1s) \rightarrow H(2s) + H^+ \quad \text{(charge transfer)}, \quad (1)$$

$$H(1s) + H(1s) \rightarrow H(2s) + H(\Sigma) \quad \text{(excitation)}, \quad (2)$$

over the energy range 1.9–92 keV using a furnace-target technique. [H(Σ) means the target atom may be left in any excited state after the collision, including the continuum.] In addition to the fundamental importance of processes (1) and (2) it should be noted that these reactions are of significance in several areas of application including astrophysics and plasma physics.

II. APPARATUS AND PROCEDURE

The experimental arrangement used in the present study is shown in Fig. 1. The apparatus consisted of a neutralizer gas cell, a 75-cm drift region which contained a series of parallel plates across which an electric field could be applied, a hydrogen-atom target, an electric field quench region viewed by a solar blind channeltron, beam deflector plates, and a Faraday cup and secondary-electron-emission detector. The apparatus was maintained at a base pressure of $\lesssim 10^{-6}$ torr by five diffusion pumps. Part of the apparatus has been described previously by Morgan and Eriksen in the report of their work on H(2s) formation in collisions between protons and alkaline-earth atoms.[8] The major changes to the apparatus involved the incorporation of the neutralizer cell, drift region, and hydrogen-atom target.

In the case of H^+-H measurements the neutralizer cell was evacuated and the plates in the drift region grounded. For H-H measurements H_2, He, or Ar gas was admitted to the neutralizer cell in order to produce fast hydrogen atoms by electron-capture collisions. In this case an electric field of up to 8 kV/cm was applied over 40 cm in the drift region and over 3 cm just prior to the entrance of the target in order to sweep the remaining protons out of the beam and to quench the H(2s) component in the atom beam. The electric field also served to field-ionize highly excited states in the beam. The drift region was long enough to ensure radiative decay of $n \lesssim 7$ states in the beam before entry into the target. A series of tests, which involved varying by up to 50% the fractional population of Rydberg states in the beam by changing both the pressure and type of gas in the neutralizer and the electric field in the drift region,[9] demonstrated that the remaining small excited-state population in the atom beam did not affect the measured cross sections.

Target H atoms were produced by thermal dissociation of H_2. The target cell consisted of a 3-cm-long tungsten tube with a 0.75-mm-diameter entrance aperture and a 1.8-mm-diameter exit aperture. With a current of ~140 A flowing coaxially along the tube the temperature was measured with an optical pyrometer to be 2750 K (corrected for emissivity and window transmission). To investigate the effect of the magnetic fields produced by the current in the oven two experimental tests were performed. In the first test H(2s) atoms were produced by electron-capture collisions in the neutralizer cell and were allowed to pass unquenched through the drift region. The Lyman-α signal per unit incident beam was measured as a function of target-cell temperature with no target gas present. In the second test cross sections for H(2s) formation in H^+ and H^0 collisions with Ar gas were measured with the target cell hot and cold. Both tests demonstrated that the

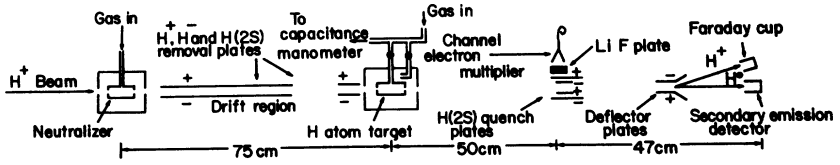

FIG. 1. Schematic diagram of the apparatus.

magnetic fields produced by the current in the oven did not appreciably quench the H(2s) atoms.

Using a 20-keV incident H^+ beam, the amount of molecular dissociation in the target was measured by the standard technique of monitoring the H^- intensity in the beam emerging from the target under single-collision conditions as a function of target temperature. The technique of using double-electron-capture collisions to measure molecular dissociation was originally developed by Lockwood *et al.*[10] The technique has been modified by Bayfield[11] to account for changes in cell conductance with temperature. The present experiment uses the method of Bayfield, which involves measuring the H^- yield for both hydrogen and argon targets and taking appropriate ratios to cancel out target-cell conductance effects. The dissociation fraction, defined by

$$f=\frac{N(\mathrm{H})}{N(\mathrm{H})+2N(\mathrm{H_2})}\,, \tag{3}$$

where $N(\mathrm{H})$ and $N(\mathrm{H_2})$ are the H and H_2 number densities in the hot target cell, was measured to be 0.88 ± 0.03. The manufacturer's specified minimum-purity level of the H_2 target gas was 99.99%.

During each series of cross-section determinations the density in the target was held fixed and monitored in a gas reservoir outside the target chamber by a capacitance manometer. Linearity of the Lyman-α signal with target density was experimentally verified over the full energy range of the measurements. Target-cell densities were chosen to produce $\lesssim$0.5% attenuation of the primary H^0 beam. The gas flow into the target could be bypassed and admitted instead into the region surrounding the target through a comparable conductance. The signal obtained in this manner was subtracted from the signal with the gas flowing directly into the target to give only the signal due to the target itself.

Fast H(2s) metastable atoms formed in the target were detected 50 cm downstream by observing the Lyman-α radiation induced by the application of an electric field of up to 1000 V/cm. [A detailed description of electric field quenching of H(2s) and the Lyman-α detection system used in the present experiment can be found in Ref. 8.] The electric-field-induced quench region consisted of two sets of parallel plates. The outer set served as image plates to reduce fringe fields. By studying the Lyman-α signal versus applied electric field, saturation of the signal was experimentally verified for incident beam energies below 30 keV, and no evidence of prequenching existed. Above 30 keV corrections of up to 20% were necessary to account for incomplete quenching within the field of view of the detector.[12] The detector was a CsI-coated channeltron operating in the pulse-counting mode. A LiF window with a grounded mesh on one surface was placed in front of the channeltron to provide a short-wavelength cutoff at 1080 Å. To eliminate the effect of polarization of the electric-field-induced Lyman-α emission the detector was placed at 54.7° with respect to the electric field direction.[8] To verify experimentally that beam scattering losses were negligible, the laboratory detector acceptance angle was changed from ±0.9° to ±1.3°, measured from the target entrance. No change in the measured cross sections was observed. Cascade contributions to the measured Lyman-α signal from $n\geq3$ states are estimated to be <10% at the higher energies and <5% at the lower energies, based on a n^{-3} extrapolation of experimental data for an H_2 target.[13]

After leaving the quench region the beam was charge separated in an electric field and the H^+ component measured with a biased Faraday cup. The H^0 component was measured with a secondary-electron-emission detector. In order to determine the neutral beam intensity the secondary-electron-emission coefficient for H^+, γ^+, was measured and the secondary-emission coefficient for H^0, γ^0, was obtained from the relation $\gamma^0/\gamma^+=1.11+0.001E$ (keV).[14,15]

To investigate the possibility of the presence of unknown errors in our data several changes were made to the apparatus during the course of the measurements. These included a new tungsten-tube furnace, a different source of H_2 target gas, a modified neutral detector, a different channeltron placed in both the 54.7° and 90° positions, and a modified Lyman-α quench region geometry. In all cases the cross sections obtained were within the error bars of the data presented in Sec. III.

If $\sigma^0(\mathrm{H})$ and $\sigma^+(\mathrm{H})$ are the cross sections for H(2s) formation for H^0 and H^+ impact on atomic hydrogen, and $\sigma^0(\mathrm{H_2})$ and $\sigma^+(\mathrm{H_2})$ are the cross sections for H(2s) formation for H^0 and H^+ impact on molecular hydrogen, we may perform the following analysis. Under single-collision conditions and with a fixed flow of gas into the furnace, the detected Lyman-α signal per unit incident H^+ beam (with a proportionality constant set equal to 1) is given by

$$S_h^+=\sigma^+(\mathrm{H})\Pi(\mathrm{H})+\sigma^+(\mathrm{H_2})\Pi_h(\mathrm{H_2}) \tag{4}$$

at 2750 K, and by

$$S_c^+=\sigma^+(\mathrm{H_2})\Pi_c(\mathrm{H_2}) \tag{5}$$

at 300 K, where $\Pi(\mathrm{H})$, $\Pi_h(\mathrm{H_2})$, and $\Pi_c(\mathrm{H_2})$ are the effective target thicknesses (atoms or molecules/cm^2) for H atoms, hot H_2 molecules, and cold H_2 molecules, respectively. Similarly, for an

incident H^0 beam,

$$S_h^0=\sigma^0(H)\Pi(H)+\sigma^0(H_2)\Pi_h(H_2) \quad (6)$$

and

$$S_c^0=\sigma^0(H_2)\Pi_c(H_2)\,. \quad (7)$$

From these equations it follows that

$$\sigma^0(H)=\left(\frac{S_h^0}{S_h^+}\right)\sigma^+(H)+\left(\frac{S_h^0}{S_h^+}-\frac{S_c^0}{S_c^+}\right)\left(\frac{1-f}{2f}\right)\sigma^+(H_2)\,, \quad (8)$$

where we have used the relation [see Eq. (3)]

$$\frac{\Pi_h(H_2)}{\Pi(H)}=\frac{N(H_2)}{N(H)}=\frac{1-f}{2f}\,. \quad (9)$$

The second term in Eq. (8) is the molecule correction term and was typically $\lesssim 3\%$ of the atom term. For H-H collisions the experimental procedure consisted of measuring S_h^+ and S_c^+ at 24.5 keV and S_h^0 and S_c^0 as a function of energy. Using the normalization cross-section values $\sigma^+(H)=3.43\times10^{-17}$ cm^2 and $\sigma^+(H_2)=2.45\times10^{-17}$ cm^2 at 24.5 keV the cross section $\sigma^0(H)$ was obtained as a function of incident energy from Eq. (8). The value of the normalization cross section $\sigma^+(H)$ is the average of three independent measurements[1,2,3] using dissimilar normalization procedures, which agree to within about 20% and contain quoted normalization uncertainties of $\lesssim \pm 30\%$. Based on this agreement we believe our normalization cross-section value provides a reliable calibration point. The value of $\sigma^+(H_2)$ is from Bayfield.[15] For H^+-H a similar procedure was used to obtain $\sigma^+(H)$ as a function of energy. The normalization procedure was chosen because of its experimental and methodical simplicity. The uncertainty in the absolute value of the present results is almost entirely dependent on only the value of the cross section $\sigma^+(H)$ at 24.5 keV since the H_2 correction is so small. In fact, the second term in Eq. (8) is smaller than the reproducibility of the data and can be neglected. In this case the experimental procedure for H-H collisions reduces to a measurement of S_h^+ at 24.5 keV and S_h^0 as a function of energy.

To provide a check on our apparatus and experimental procedure, cross sections for H(2s) formation in hot and cold Ar were measured for both H^+ and H^0 impact. In all cases the data were normalized to the H^+-Ar cross section measured by Bayfield at 24.5 keV.[15] For H^+-Ar our measured cross sections agree well with the data of Bayfield and Morgan *et al.*[2] at low energies (2–20 keV). At high energies our cross sections are generally higher than Bayfield's (~16% at 70 keV) but lower than those of Hughes *et al.*[16] (~18% at 80 keV). H^0-Ar measurements were carried out over the energy range 5–25 keV. Our results agree to within ~20% with the data of Sauers and Thomas[17] and Birely and McNeal.[18]

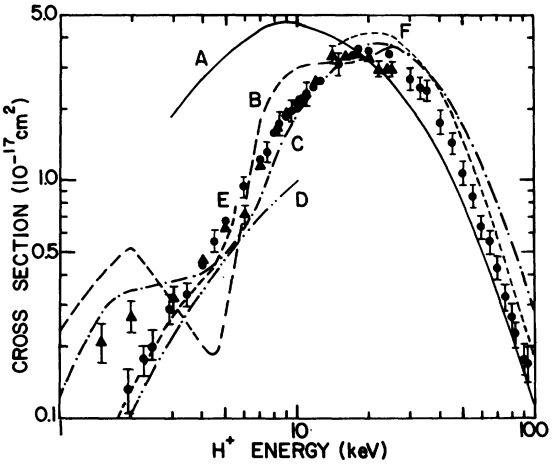

FIG. 2. Cross section $\sigma^+(H)$ for H(2s) formation by electron capture in H^++H(1s) collisions. Experiment: ●, present results; ▲, Ref. 4. Theory: curve A, Born approximation (Ref. 19); curve B, 4-state hydrogenic close-coupling calculation (Ref. 20); curve C, 7-state (4 hydrogenic and 3 pseudostates) close-coupling calculation (Ref. 21); curve D, 10-state molecular treatment using Coulomb trajectories (Ref. 22); curve E, 5-state molecular treatment using straight-line trajectories (Ref. 23); curve F, 34-state scaled, hydrogenic close-coupling calculation (Ref. 26).

III. RESULTS AND DISCUSSION

The present results for H^+-H collisions are shown in Fig. 2. Error bars represent relative errors and include differences in the value of the analyzed cross sections from different data runs. The data presented in the figures were taken over a period of several months, and many of the data points are the average of several values which typically agree to within ±8% for H^+-H and ±11% for H-H. Both $H^+(H^0)$ and $D^+(D^0)$ incident beams were used, and the cross sections agree when compared at the same velocity. Also shown in Fig. 2 are the recent results of Hill *et al.*[4] and various pertinent calculations.[19-23] Owing to the fundamental nature of the H^+-H collision, there have been a large number of theoretical treatments. However, for the sake of clarity, only a few are included in Fig. 2.[24] Also, we have chosen not to include the previous experimental results of Bayfield,[1] Morgan *et al.*,[2] Chong and Fite,[3] and Ryding *et al.*[5] The present results and those of Hill *et al.*[4] agree with the previous 5–26 keV measurements of Morgan *et al.*[2] For energies greater than 7 keV agreement also exists with the 3–70-keV measurements of Bayfield as well as the 6–25-keV results of Chong and Fite. At low energies the results of Bayfield

are substantially below our values. For energies greater than 70 keV the high-energy data of Ryding *et al.* when normalized to the result of Bayfield at 43 keV are about a factor of 2 higher than the present results. Below 3 keV the present results and those of Hill *et al.*[4] disagree. The low-energy data of Hill *et al.* appear to support the pseudostate close-coupling calculation of Cheshire *et al.*[21] (curve C). The inclusion of nonhydrogenic pseudostates provides a representation of the continuum in the wave-function expansion and attempts to simulate the molecular aspects of the collision at low energies and small internuclear separations. However, the present experimental results do not reproduce the change in slope of the close-coupled pseudostate curve near 3 keV and are instead in excellent agreement with the recent multistate molecular approach of Chidichimo-Frank and Piacentini[23] (curve E). The basis set employed in their calculation included the $1s\sigma_g$, $2p\sigma_u$, $2p\pi_u$, $3p\sigma_u$, and $3p\pi_u$ states of H_2^+, and the excitation of the H(2*s*) state is described by the two-state coupling $2p\sigma_u - 2p\pi_u - 3p\sigma_u$. For large internuclear separations the $3p\sigma_u$ orbital [which leads to H(2*s*) after the collision] and the $2p\pi_u$ orbital converge, and transitions occur via rotational coupling. The rotational-coupling coefficients used in this calculation were those calculated by Rosenthal[25] using exact matrix elements, and the resulting transition amplitudes were used to calculate the cross section within the straight-line-path eikonal approximation. As can be seen from Fig. 2 the structure in the hydrogenic coupled-state calculations[20] (curve B) at ~12 keV and ~4 keV is suppressed by the inclusion of pseudostates in the basis set[21] (curve C). However, a pronounced shoulder still remains at low energy which is not reproduced by the molecular calculations[22,23] (curves E and D) nor by the present experimental results. At high energies the present results for H^+-H are converging to the Born approximation values[19] (curve A), and good agreement is found for energies above 75 keV. On the other hand, the 4-state hydrogenic and 7-state hydrogenic plus pseudostate close-coupling cross sections are slow to converge and at 90 keV are high by a factor of 2. Recently Shakeshaft[26] has performed a high-energy (15–200 keV) coupled-state calculation (curve F) using a scaled hydrogenic basis set containing 34-state functions centered about each proton. One advantage of this basis set over a simpler hydrogenic one is that it provides a better representation of coupling to the continuum. With a small normalization change (theoretical results times 0.8), the scaled hydrogenic close-coupling results are in very good agreement with the present data.

The present results for H-H collisions are shown in Fig. 3 along with available theory,[27-31] the earlier crossed-beam measurements of Morgan *et al.*,[6] and the results of the concurrent experiment of Hill *et al.*[7] The crossed-beam results are normalized to the H(2*p*) formation cross section in H^+-H_2 collisions[32] and are subject to an overall uncertainty of up to 70% in the absolute value. As a result, the agreement with the present data is within combined experimental uncertainty. The data of Hill *et al.*[7] have been normalized at 16 keV to the absolute determination of the H(2*s*) formation cross section σ_{2s}(Ar) in H^+-Ar collisions measured by Andreev *et al.*[33] The present data are normalized at 24.5 keV to a value of σ^+(H) (see Sec. II) that agrees within 3% with the measurement of Bayfield,[1] who normalized his H^+-H data to his determination of σ_{2s}(Ar).[15] Over the energy range 10–30 keV the absolute values of σ_{2s}(Ar) as measured by Andreev *et al.* and Bayfield are in good agreement. In principle then, the normalization cross section chosen in the present experiment and in the work of Hill *et al.* are the same. Hill *et al.*[7] have used the same normalization procedure to obtain the cross section for H(2*s*) formation in H^0-H_2 collisions. At their normalization energy the cross section is about 35% lower than the result of Birely and McNeal[32] and unpublished data from our laboratory. Renormalizing the H-H data of Hill *et al.* upward by this amount yields agree-

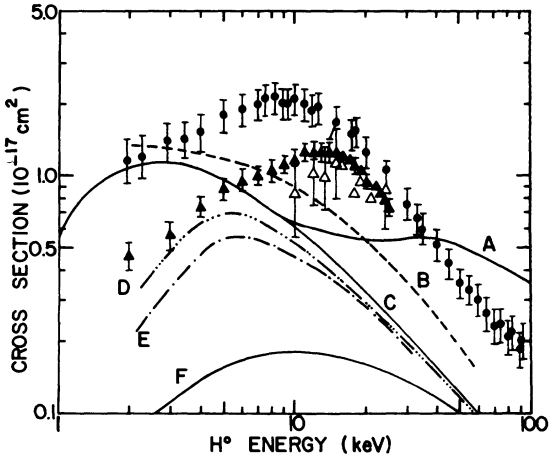

FIG. 3. Cross section σ^0(H) for H(2*s*) excitation in H(1*s*)+H(1*s*) collisions. Experiment: ●, present results; ▲, Ref. 7; △, Ref. 6. Theory: curve A, Born approximation (Ref. 27). Theory for H(1*s*)+H(1*s*) → H(2*s*)+H(1*s*): curve B, 2-state impact-parameter calculation with exchange (Ref. 28); curve C, Born approximation (Ref. 27); curve D, 4-state impact-parameter calculation without exchange (Ref. 29); curve E, eikonal distorted-wave Born approximation (Ref. 30); curve F, 4-state impact-parameter calculation with exchange (Ref. 31).

ment with the present results within combined relative experimental uncertainty down to 9 keV. However, this observation is not particularly satisfying, and the lack of agreement for H-H collisions is disturbing and inexplicable. Even so, it is clear from Fig. 3 that the available theoretical cross sections do not provide a good fit to the experimental data. Only curve A should be compared directly with experiment over the entire energy range since all other calculations are for the reaction $H(1s)+H(1s)\rightarrow H(2s)+H(1s)$. However, it is expected that below ~10 keV excitation of the target is negligible.[27] A likely cause for disagreement between theory and experiment at low energies is the neglect of the molecular aspects of the collision. It has been shown recently that, for the neutral collision system H-He, excitation to the $2p$ state of hydrogen proceeds via molecular coupling at low energies.[34] A multistate molecular calculation for H-H collisions is needed. An additional aspect of the H-H collision system which is not present in the one-electron H^+-H case is the possibility of electron exchange. Two recent calculations of the ionization cross section in $H(1s)+H(1s)$ collisions clearly demonstrate the importance of electron exchange below about 25 keV.[35,36] Ritchie[28] and Bottcher and Flannery[31] have included electron-exchange effects within the impact-parameter formalism for H(2s) formation. However, the results disagree by up to an order of magnitude. In one case (curve B) the impact-parameter approximation with electron-exchange effects included lies well above the impact-parameter calculation without exchange (curve D), whereas the other impact-parameter calculation with exchange effects included (curve F) lies well below the nonexchange result. It should be noted that the theoretical results represented by curve B in Fig. 3 do not include $2s$-$2p$ coupling. However, they do include the velocity dependence of the electron-exchange potential (due to exponential momentum factors associated with the traslational motion of the electrons) that has been neglected in the calculation of curve F. (We refer the reader to Ref. 30 for a comparative discussion of the assumptions associated with curves B and F.) A detailed study of the influence of the electron-exchange interaction in excitation collisions and of the methods used to include the requirements of the Pauli principle within scattering approximations is needed.

At high energies the present experimental results for H-H do not support the Born approximation prediction[27] (curve A) of pronounced structure in the H(2s) formation cross section due to simultaneous excitation of both target and projectile hydrogen atoms. The poor agreement at high energies is perhaps unexpected, since for H^+-H satisfactory agreement exists between the Born approximation and the present experimental results. However, there is no *a priori* reason to assume that electron correlation effects in double-excitation transitions will not greatly affect the cross sections. Also, for H-H collisions, theory predicts[27] that at high energies the H(2s) formation cross section falls off as the inverse of the impact energy since only short-range interactions are present. As can be seen from Fig. 3, for energies greater than 40 keV, the present results agree with this prediction.

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