Single-Electron Capture and Loss Cross Sections for 2–50-keV Hydrogen Atoms Incident upon Hydrogen and the Inert Gases

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The single-electron capture and loss cross sections $\sigma_{0,-1}$ and σ_{01} for 2–50-keV hydrogen atoms incident upon hydrogen molecules and inert-gas atoms have been measured directly by observing the growth of the fast-collision products (i.e., the fast primary H_1^0 which have changed charge) with the target-gas number density. In H_2, He, Ne, and Ar, the present values of $\sigma_{0,-1}$ and σ_{01} generally confirm previous measurements, with the notable exception that in H_2 the low-energy values of σ_{01} are as much as 30% smaller (at 2 keV) than the values of McClure. In Kr and Xe there are no previous measurements of $\sigma_{0,-1}$, while the only previously reported values of σ_{01} are smaller than the present values by as much as a factor of 3. For a hydrogen-gas target, the present measurements do not show any large subsidiary peaks. The use of an electrostatic field, prior to the collision cell, to quench possible excited states in the primary H_1^0 atom beam is shown to have only a very small effect upon the measured cross sections. Both σ_{01} and $\sigma_{0,-1}$ are found to increase in value with increasing atomic number for all the inert gases. The low-energy (adiabatic) region is extended down to 2 keV, where it is seen that σ_{01} rises exponentially with increasing relative velocity of the colliding particles.

INTRODUCTION

THE published values of the single-electron capture and loss cross sections $\sigma_{0,-1}$ and σ_{01} for fast hydrogen atoms show large differences which may be attributed to several factors. Differences in absolute values may arise from the difficulties of measurements of the target-gas number density.[1–3] These difficulties may partly be circumvented by measuring relative cross-section values and standardizing them against a well known cross-section value for each target gas.[4–6] Consideration of previous measurements of both σ_{01} and $\sigma_{0,-1}$ in H_2 gas shows, however, that the shapes of the cross-section–versus–energy curves obtained by different methods disagree. This indicated that difficulties, more serious than those associated with target calibration, exist. Curran and Donahue,[7] using the method of slow target-gas ion collection, obtained values of magnitude similar to those of Whittier,[8] who used the method of primary-beam attenuation in a transverse magnetic field. Stier and Barnett,[9] Fogel,[10,11] and Soloviev[12] have used the methods of primary-beam equilibration in the target gas and of target-gas ion collection to obtain values of both σ_{01} and $\sigma_{0,-1}$ which are different from those of other workers in both magnitude and energy dependence. The results of Curran and Donahue are the only ones to exhibit several small peaks superimposed upon the basic form of the cross-section curve.

A further method, namely, the observation of the growth of fast collision products with change in target-gas number density, has been used by the author[13] and by McClure.[14] At all energies from 4 to 120 keV, except in the vicinity of 6 keV, the values of McClure appear to be consistent with the values of Stier and Barnett.[9] However, the present investigation uses a method of cross-section measurement similar to that of McClure, but "integrated" particle-detection methods similar to those of Stier and Barnett, and obtains values of σ_{01} and $\sigma_{0,-1}$ which are substantially lower than their values at energies below 10 keV.

Measurements of σ_{01} and $\sigma_{0,-1}$ have been made for the five inert gases for which there are only a few previous values. The extension of the $\sigma_{0,-1}$ data in all gases from 10 keV down to 2 keV has permitted the dependence of the cross section upon relative velocity to be studied and fitted to an exponential dependence of the type used by Hasted[15] for other charge-transfer collisions in the adiabatic region.

In all the present measurements, however, all cross-section values are averages over an unknown population of excited states both in the incident hydrogen atoms and in the target particles after collision, and over

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unknown partial cross sections for electron loss from those excited states.

APPARATUS AND METHOD

The apparatus, the method of cross-section determination, and the extensive measurements made to establish the accuracy and validity of the cross-section values are similar to those previously reported.[6,13,16] The H_1^0 atoms were formed by electron capture by fast protons in the free-drift region between the beam image position of the 30-cm momentum-analyzing magnet and the electrostatic deflection plates prior to the collision cell. The neutralizing gases were hydrogen gas and the unknown residual gases in the vacuum system. H_1^0 atom flux densities of the order of 10^9 atoms/sec mm^2 were obtained. The H_1^0, H_1^+, and H_1^- currents were measured in an instrument[13] which could be used either as a Faraday cup, or a secondary-electron–emitting detector or a thermal response detector.

Relative cross-section values were determined from the slope of the linear portion of the graph of the growth of the fast-collision products versus collision-cell gas number density. Such graphs were obtained for every individual cross-section measurement. In no case did the charge-exchange currents exceed 1% of the primary-beam current.

The cross sections σ_{01} and $\sigma_{0,-1}$ were measured simultaneously at various energies in the range 2–50 keV. The relative values of σ_{01} and $\sigma_{0,-1}$ were standardized against the better known single-electron capture cross section σ_{10} for proton energies of 10 keV in each of the target gases, namely, 8.2×10^{-16} cm^2/mole in H_2, 9.5×10^{-17} cm^2/atom in He, 2.9×10^{-16} cm^2/atom in Ne, 9.9×10^{-16} cm^2/atom in Ar, 1.45×10^{-15} cm^2/atom in Kr, and 1.8×10^{-15} cm^2/atom in Xe.

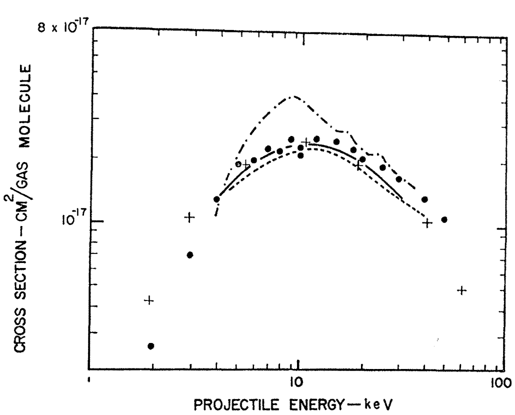

FIG. 1. The electron capture cross section $\sigma_{0,-1}$ for 2–50-keV hydrogen atoms incident upon hydrogen molecules is shown as a function of the energy of the incident hydrogen atoms. ● ● ● present values; —— Stier and Barnett (Ref. 9); – – – Fogel (Ref. 10); –·– Curran and Donahue (Ref. 7); +++ McClure (Ref. 14).

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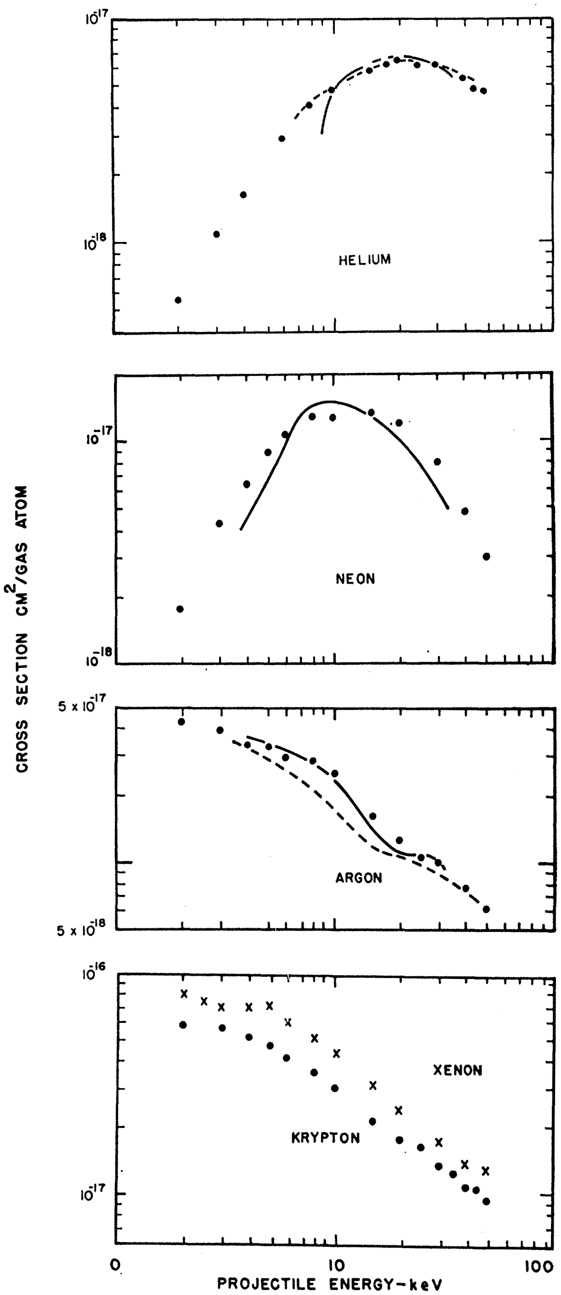

FIG. 2. The electron capture cross section $\sigma_{0,-1}$ for 2–50-keV hydrogen atoms incident upon rare-gas atoms is shown as a function of the energy of the incident hydrogen atoms. ● ● ● present values in He, Ne, Ar, and Kr; XXX present values in Xe; —— Stier and Barnett (Ref. 9); – – – Fogel (Ref. 10).

RESULTS

A. The Single-Electron Capture Cross Section $\sigma_{0,-1}$

Figure 1 shows that the present values provide little substantiation for the results of Curran.[7] There is no evidence of the fine structure nor of the enhanced values of the cross section in the region of the maximum from 5 to 15 keV. The relative accuracy of the magni-

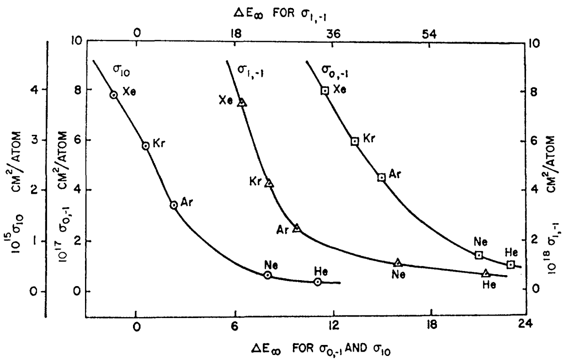

FIG. 3. Shows the maximum values σ_{max} of the electron capture cross sections σ_{10}, $\sigma_{0,-1}$, and $\sigma_{1,-1}$ as a function of the energy defect ΔE of the collision in each of the inert-gas target atoms.

tude of the present values is $\pm 5\%$. The proton beam, from which the $H_1{}^0$ atoms are formed, was momentum analysed by a 30-cm radius $\pi/2$ inflection-type magnet whose resolution was better than 0.8%. Cross-section values were determined every 0.5 keV within the energy range 2 to 10 keV. The present values become appreciably smaller than those of McClure at the lower energies.

Figure 2 shows the values obtained for $\sigma_{0,-1}$ in the rare gases. These results are in reasonable agreement with those of earlier workers.[9,10] The present measurements of $\sigma_{0,-1}$ show that, at all energies within the range 2–50 keV, the cross section increases as the atomic number of the target gas increases.

The data on argon indicate the presence of a small subsidiary maximum in $\sigma_{0,-1}$ at about 25 keV. The main maximum in $\sigma_{0,-1}$ in argon has been located by Hasted[17] below the minimum attainable energy of 2 keV in the present work, and it has been well identified with the maximum of the process $(00/-11)$.

$$H_1{}^0 + Ar \rightarrow H_1{}^- + Ar^+,$$

where all the particles are in their ground states both before and after the collision. The adiabatic criterion[17] may be applied to this collision process to indicate that if the incident $H_1{}^0$ atom is in an excited state, the energy defect ΔE of the collision is lowered and the maximum cross section occurs at a lower velocity than that of the ground-state collision. Similarly, if the Ar^+ ion is formed in an excited state, ΔE is raised and hence the maximum cross-section value occurs at a velocity higher than that of the ground-state collision. With the assumption that nearly all the incident $H_1{}^0$ atoms are in the ground state, the value of ΔE corresponding to the subsidiary maximum at about 25 keV places the excitation energy of the Ar target atom within 10% of its second ionization potential, i.e., the subsidiary maximum may be attributed to the process

$$H_1{}^0 + Ar \rightarrow H_1{}^- + Ar^{++} + e.$$

[17] J. B. Hasted and A. R. Lee, Proc. Phys. Soc. (London) **79**, 1049 (1962).

If considerations similar to those made above for Ar are made for Ne, Kr, and Xe, one expects subsidiary maxima to appear above the present upper energy limit of 50 keV in the case of Ne, and in the vicinity of 13 and 8 keV for Kr and Xe, respectively. However, the application of the adiabatic criterion to Kr and Xe data is less certain than for Ar because the inaccuracy in predicting the position of the main maxima[17] in Kr and Xe is much larger than in Ar. Nevertheless, the data of Fig. 2 show no maxima around 13 keV in Kr, while in Xe the appearance of a maxima at about 5 keV is in fair agreement with the predicted position.

The composite nature of electron capture cross sections has also been indicated by previous measurements[18] of $\sigma_{1,-1}$, which have indicated that the partial cross section for the process $(10/-13e)$ can be large compared with the cross section for the process $(10/-12)$. However, confirmation of such possibilities must come from coincidence-counting experiments on both primary and target particles.

It is well known that the adiabatic criterion gives no indication of absolute or relative values of the maxima in the cross-section functions. The present study on $\sigma_{0,-1}$ completes a series of measurements[6,18] on electron-capture cross sections σ_{10} and $\sigma_{1,-1}$ by hydrogen atoms and ions. Figure 3 shows the maximum values σ_{max} of the electron capture cross sections σ_{10}, σ_{0-1}, and $\sigma_{1,-1}$ in relation to the energy defect ΔE of the collision and to the atomic number Z of the target atom. It is seen that in all three kinds of capture processes, σ_{max} increases as either ΔE decreases or Z increases, and the curves for all three cases have a rough similarity in shape, each possessing a knee between Ar and Ne.

B. The Single-Electron Loss Cross Section σ_{01}

Figure 4 shows the present values of σ_{01} in H_2 above 10 keV to be in good agreement with the mean value of the results of other investigators which deviate up to $\pm 20\%$ from the mean. Most of the individual values possess nearly the same energy dependence above 10

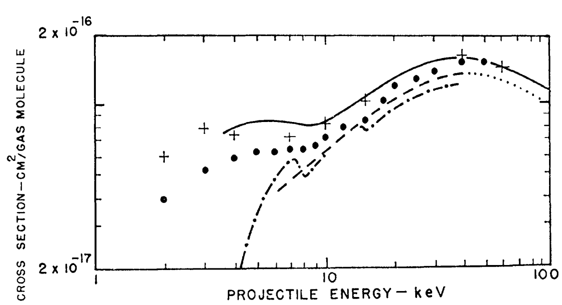

FIG. 4. The electron loss cross section σ_{01} is shown as a function of hydrogen atom energy for 2–50-keV hydrogen atoms incident upon hydrogen molecules. ● ● ● present values; —— Stier and Barnett (Ref. 9); - - - Fogel (Ref. 10); -·- Curran and Donahue (Ref. 7); ··· Ribe (Ref. 20); +++ McClure (Ref. 14).

[18] J. F. Williams, Phys. Rev. **150**, 7 (1966).

keV, which may indicate that the differences between values of the separate workers could arise from the difficulties[3,5] of determining absolute cross-section values as opposed to relative values. There is no support for the fine structure observed by Curran and Donahue.

Below 10 keV, there are two divergent groups of values, the rapidly decreasing values by Curran and Donahue and the relatively higher and steadier values of Stier and Barnett. The shape of the present curve definitely supports the general shape of the cross-section curve of Stier and Barnett, although the actual values are up to 25% smaller.

The possibility of the primary H_1^0 atoms being in excited states has been considered[14,19] as an explanation of the divergence of the above results. It is well known that electron capture by protons passing through gases results in the formation of hydrogen atoms with a population distribution over all excited states which decreases approximately as the inverse of the third power of the principal quantum number n. However, the method of formation of the primary H_1^0 atoms has varied greatly from the dissociation and electron capture by H_3^+ molecular ions in collision with a mercury-vapor target[10] to the charge equilibration of a proton beam in passing through a thick argon target.[9,20] It is unlikely that these methods would give the same population distribution over the H_1^0 excited states. Despite the different methods of formation of the primary H_1^0 atoms, all such atoms must pass through a transverse electric field before entering the collision cell in order to remove all charged particles from the primary beam. This external static electric field will cause the atom to experience an asymmetric electric field which mixes the wave functions of different atomic states with subsequent transitions to lower states. From the data of Bethe and Salpeter[21] it is seen that the mean lifetime of the metastable $2S_{1/2}$ state (for example) in a field of strength X (V/cm) is $(C/X)^2T_{2P}$, where T_{2P} is the lifetime of the $2P$ level and C is a constant for a given atomic state, equal to 475 for the $2S_{1/2}$ state. For states higher than the $2S$, the value of C decreases approximately as $n^{-4.5}$ and such states should all decay to the ground state in a time not greater than 10^{-8} sec for fields as low as 1 V/cm. At the highest energy used in the following experiments (10 keV), the time of flight from the center of the deflector plates to the collision cell was 3×10^{-8} sec, so there was only a small probability that an atom in an excited state, other than the $2S$ state, would undergo a collision within the collision cell.

While the population of this $2S$ state may be small compared with the ground state, the energy defects of

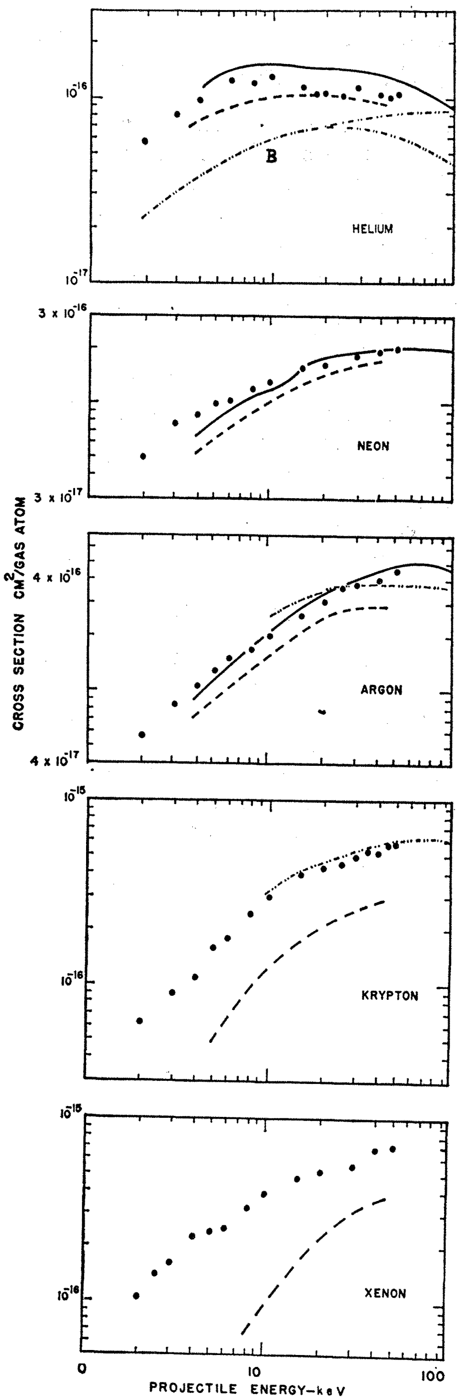

FIG. 5. The electron loss cross section σ_{01} is shown as a function of hydrogen atom energy for 2–50-keV hydrogen atoms incident upon inert gas target atoms. ● ● ● present value; —— Stier and Barnett (Ref. 9); – – – Fogel (Ref. 10); –·· – Soloviev (Ref. 12) (in Ar and Kr); –·· –B First Born approximation calculations by Bates and Williams (Ref. 22) for a helium target. The lower curve is the cross section for the collision in which the helium atom is left in its ground state while the upper curve is the sum of the partial cross sections in which the helium atom is left in the ground and all excited states.

[19] Y. M. Fogel, Usp. Fiz. Nauk 71, 243 (1960) [English transl.: Soviet Phys.: Usp. 3, 390 (1960)].

[20] F. L. Ribe, Phys. Rev. 83, 1217 (1951).

[21] H. Bethe and E. Salpeter, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1951), Vol. 35, p. 373.

the two reactions

$$H(1S)+H_2 \rightarrow H_1^+ + e + H_2 \ (\Delta E = 13.6 \text{ eV}),$$
$$H(2S)+H_2 \rightarrow H_1^+ + e + H_2 \ (\Delta E = 3.4 \text{ eV}),$$

are such that the second reaction may have the larger cross section at H_1 energies less than 10 keV. Hence a small $2S$-state population could have an appreciable effect upon the measured total cross section, which is an average over the relative populations of each of the H_1^0 states multiplied by the cross section for stripping of the electron from such states.

The deflection plates (P_1 and P_2 of Fig. 1 of Ref. 6) were positioned 5 cm from the entry canal of the collision chamber in order to obtain the lowest electric fields between the plates which would give the minimum quenching but still prevent all the charged particles from entering the collision cell. The fraction F of the $2S$ metastable state not quenched by the deflection field was calculated, after the manner of McClure,[14] from the expression

$$F = \exp[-(X/475)^2 1/T_{2P}],$$

where t is the time of flight through the deflection plates and T_{2P} is the lifetime of the $2P$ state. At H atom energies of 2, 3, 4, 5, 6, 8, and 10 keV, F was found to be 0.86, 0.72, 0.57, 0.48, 0.39, 0.31, and 0.20, respectively. At each of these energies σ_{01} was measured alternately with the $2S$ state population entirely quenched and then with the above expected fractions of the $2S$ state not quenched. Corresponding to each of these changes in the expected $2S$ state populations, there was an increase of from $(4\text{–}8)\pm2\%$ in σ_{01}. This change is smaller than that obtained by McClure, but it substantiates his result that a variable population by the electron capture of protons is much smaller, by at least an order of magnitude, than that required to afford an explanation of the discrepancies in the measured values of σ_{01}.

Figure 5 shows σ_{01} in the inert gases. There is generally better agreement between the present values and those of Stier and Barnett than with the values of Fogel. More recent measurements of Soloviev[12] in Ar and Kr agree well with the present values, which show a systematic increase of cross section with increasing atomic number of the target gas at all energies. This behavior is in marked contrast to that of Fogel's data, which show σ_{01} decreasing with increasing atomic number for H_1^0 energies less than about 20 keV. The present values extend the lower limit of energy of the measurements down to 2 keV such that there is a sufficient number of points to permit the investigation of the rise of cross section with energy within the adiabatic energy region. For all the inert gases there is a linear dependence of $\log\sigma$ upon the quantity $E^{-1/2}$, as predicted by the first Born approximation by Bates and Williams.[22]

ACKNOWLEDGMENTS

The author is indebted to Professor D. N. F. Dunbar for his comments both during the course of this work and during the preparation of this paper.

[22] D. R. Bates and A. Williams, Proc. Phys. Soc. (London) **A70**, 306 (1957).