

Measurement of Charge-Transfer Cross Sections for 0.25- to 2.5-MeV Protons and Hydrogen Atoms Incident upon Hydrogen and Helium Gases

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Measurements of the single- and double-electron-capture cross section for protons, σ_{10} and $\sigma_{1,-1}$, respectively, and the single-electron-loss cross section for hydrogen atoms σ_{01} within the energy range 0.25 to 2.5 MeV for molecular-hydrogen and atomic-helium target gases have been made by the method of observing the rate of growth with target-gas number density of the fast-collision products from an originally pure primary beam. The present values of σ_{10} and σ_{01} agree within 10% with the data of Barnett and Reynolds below 1 MeV and confirm the extrapolation of their data which passes through the single measurements of σ_{10} and σ_{01} for 12.9- and 21-MeV deuterons and 20-MeV deuterium atoms by Berkner, except for σ_{10} in He where the extrapolation of the present data passes below the values of Berkner. The experimental values of σ_{01} agree with calculated values of σ_{01} derived from the Born and free-collision approximations within the experimental uncertainty of $\pm 10\%$. The values of $\sigma_{1,-1}$ in molecular hydrogen decrease from 5.1×10^{-26} cm²/molecule at 0.4 MeV to 1.6×10^{-28} cm²/molecule at 1 MeV with an experimental uncertainty of up to 60%. These values are lower than the first-Born-approximation calculations by Mittleman.

1. INTRODUCTION

At high energies, the single-electron-capture cross section σ_{10} , and the single-electron-loss cross section σ_{01} , for protons and hydrogen atoms, respectively, incident upon hydrogen and helium gases have been measured by Barnett and Reynolds¹ from 0.25 to 1.0 MeV and Berkner^{2,3} for 12.9- and 21-MeV deuterons and 20-MeV deuterium atoms. Barnett and Reynolds determined σ_{01} by measuring the attenuation of a beam of hydrogen atoms in a gaseous region across which a transverse electric field was maintained, and then determined σ_{10} by measuring the equilibrium ratio of hydrogen atoms to protons for the passage of fast hydrogen ions through a thick gaseous target. Berkner *et al.* determined σ_{01} and σ_{10} by measuring at several gas pressures the ratio of the incident primary beam current to the current of those primary beam particles which had changed charge in a single collision. Such methods of cross-section measurement are fundamentally less accurate⁴ than the method used in the present study, namely the measurement of the rate of growth with target gas number density of the fast collision products from an originally single-charge-state beam.

σ_{01} has been calculated by the use of three methods: the first Born approximation,^{5,6} the semiclassical model of Bohr,⁷ and the "free-collision" (or impulse) approxi-

mation.⁸ All of these models predict an E^{-1} energy dependence at high energies where $e^2Z/hv \ll 1$. For a molecular-hydrogen target this condition implies energies above a few hundred kilovolts, in which region the theoretical calculations agree with each other within 10%. If the data of Barnett and Reynolds are extrapolated with an E^{-1} energy dependence in accordance with the calculated relationship, the extrapolation passes through the 20-MeV determination by Berkner. The present investigation measures σ_{01} over the energy range 0.3 to 2.5 MeV to seek confirmation of the extrapolation and the calculated values.

Discussions of the many approaches and calculations of σ_{10} for fast protons incident upon atomic hydrogen and helium have been given by Bates,^{9,10} Mittleman,¹¹ Mapleton,¹² and Bransden.¹³ There is substantial disagreement between the various theoretical predictions and also between the theoretical and experimental values. In atomic hydrogen there are no experimental values above 0.15 MeV, while the theoretical predictions extend to much higher energies; however in molecular hydrogen the experimental values extend up to 1.0 MeV, but there are no theoretical predictions with the exception of the work of Tuan and Gerjuoy¹⁴ who calculated the ratio of the single-electron-capture cross section for protons in atomic hydrogen to that in molecular hydrogen. The present measurements of σ_{10} in molecular hydrogen have been made over the range of proton energies from 0.3 to 2.5 MeV to check pre-

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¹ C. F. Barnett and H. K. Reynolds, *Phys. Rev.* **109**, 355 (1958).

² K. H. Berkner, S. N. Kaplan, and R. V. Pyle, *Phys. Rev.* **134**, A1461 (1964).

³ K. H. Berkner, S. N. Kaplan, G. A. Paulikas, and R. V. Pyle, *Phys. Rev.* **140**, A729 (1965).

⁴ M. H. Mittleman, *Phys. Rev.* **137**, A1 (1965).

⁵ D. R. Bates and A. Williams, *Proc. Phys. Soc. (London)* **A70**, 306 (1957).

⁶ D. R. Bates and G. W. Griffing, *Proc. Phys. Soc. (London)* **A68**, 90 (1965).

⁷ N. Bohr, *The Penetration of Atomic Particles through Matter* (J. Komm. Hos. E. Munksgaard, Copenhagen, 1949).

⁸ I. S. Dmitriev and V. S. Nikolaev, *Zh. Eksperim. i Teor. Fiz.*, **44**, 660 (1963) [English transl.: *Soviet Phys.—JETP* **17**, 447 (1963)].

⁹ D. R. Bates and R. McCarroll, *Advan. Phys.* **11**, 39 (1962).

¹⁰ D. R. Bates, *Atomic and Molecular Processes* (Academic Press Inc., London, 1962).

¹¹ M. H. Mittleman, *Proc. Phys. Soc. (London)* **81**, 633 (1963).

¹² R. A. Mapleton, *Phys. Rev.* **130**, 1839 (1963).

¹³ B. H. Bransden and I. M. Cheshire, *Proc. Phys. Soc. (London)* **81**, 820 (1963).

¹⁴ T. F. Tuan and E. Gerjuoy, *Phys. Rev.* **117**, 756 (1960).

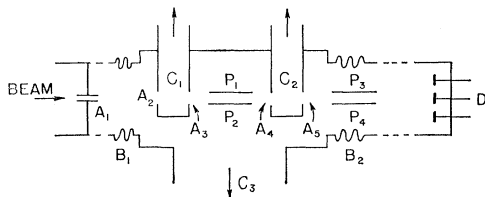
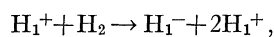


FIG. 1. Schematic representation of the apparatus. A_1 is a cylindrical canal, 0.010 in. in diameter and 0.50 in. in length, machined into an aluminium disk which separates the present vacuum system from that of the Van de Graaff. A_2 , A_3 , A_4 , and A_5 are circular apertures with knife edges of diameters, 0.010, 0.020, 0.015, and 0.025 in., respectively; P_1P_2 and P_3P_4 are parallel electrostatic deflection plates of separations 0.060 and 0.12 in. respectively, and lengths 1.0 and 1.5 in., respectively. Bellows B_1 and B_2 allow alignment of the apertures A_2 to A_5 with aperture A_1 and alignment of the detectors, D , with the beam axis; C_1 and C_2 are each connected through valves to a diffusion pump, ionization gauge, and gas leak. C_3 is connected to a liquid-nitrogen trapped-mercury diffusion pump of speed 650 liters/sec. The residual gas pressure is about 10^{-8} mm Hg. The detectors D are a Faraday cup, and two surface-barrier detectors each separated by 1 in. and approximately 14 in. from the center of P_3P_4 . The complete system was constructed from stainless steel and used copper gaskets for baking at temperature up to 400°C . The target-gas handling system was also bakeable.

vious values up to 1.0 MeV and extend the energy range up to 2.5 MeV.

The double-electron-capture collision process, which can be represented by the equation



is one of the few basic collision processes which are not complicated by the presence of excited states in any of the colliding particles, both before and after the collision. It is thus particularly suitable for a comparison between experimental and theoretical values of the cross section $\sigma_{1,-1}$. However, previous experimental values¹⁵ are confined to the energy region below 50 keV in which the theoretical predictions⁴ are expected to be poor because of the nature of the Born approximation. We have made experimental measurements of $\sigma_{1,-1}$ over the energy range 0.4 to 1.0 MeV.

2. METHOD

Figure 1 is a schematic representation of the apparatus. The beam from the Van de Graaff accelerator of the Australian Atomic Energy Commission was collimated by an aperture, prior to canal A_1 , and by the canal A_1 to a semiangle of divergence of approximately 10^{-3} rad to produce a proton current of up to several microamperes in the collision cell. The beam-energy spread was negligible and the beam energy was calibrated to 3% against the $\text{Li}^7(p,n)$ threshold at 1.881 MeV and the $\text{N}^{15}(p,\gamma)$ thresholds at 0.429 and 0.898 MeV.

Neutral hydrogen-atom beams were formed within cell C_1 either by single-electron capture by protons or by the dissociation of H_2^+ molecular ions since the relative probability of formation of H_2^0 is negligible¹⁶

¹⁵ J. F. Williams, Phys. Rev. **150**, 7 (1966).

¹⁶ D. R. Sweetman, Proc. Roy. Soc. (London) **A256**, 416 (1960).

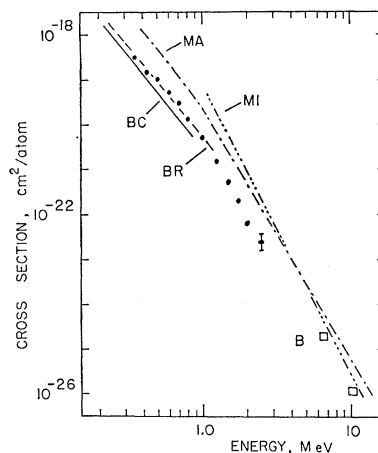


FIG. 2. The single-electron-capture cross section σ_{10} for H_1^+ incident upon helium gas in units of $\text{cm}^2/(\text{He atom})$. \cdots present experimental values. \cdots MI, Mittleman, first Born approximation (Ref. 11). \cdots BC, Bransden, impulse approximation (Ref. 13). \cdots MA, Mapleton, O.B.K. approximation (Ref. 12). \cdots BR [Barnett and Reynolds, experimental (Ref. 1); Mapleton, first Born approximation with nucleus-nucleus interaction (Ref. 12)]. \square B, the 6- and 10-MeV values for primary deuterons by Berkner (Ref. 2).

above 0.5 MeV. The electrostatic fields P_1P_2 and P_3P_4 were used for several purposes: to check the presence of background neutrals in the primary proton beam, to remove the charged particles from the primary neutral-atom beam, and to provide a charge separation of the fast collision products.

The particles H_1^+ , H_1^0 , and H_1^- were detected either by a Faraday cup, or by a secondary-electron-emission detector together with a Cary electrometer, or by 20-mm-diam. surface-barrier detectors using standard single-particle counting techniques.

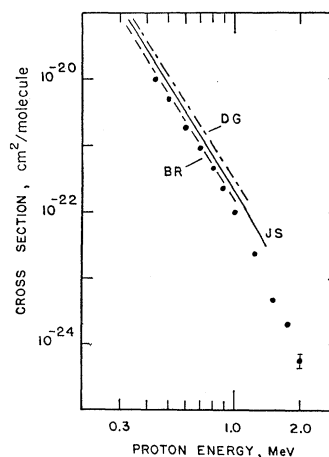


FIG. 3. The single-electron-capture cross section σ_{10} for H_1^+ incident upon hydrogen gas. Units of σ_{10} are $\text{cm}^2/(\text{hydrogen molecule})$. \cdots , present experimental data; \cdots Barnett and Reynolds (Ref. 1); \cdots Dalgarno and Griffing (Ref. 21); \cdots Jackson and Schiff (Ref. 22). Both of the theoretical predictions are for atomic hydrogen targets and the values have been multiplied by 2.

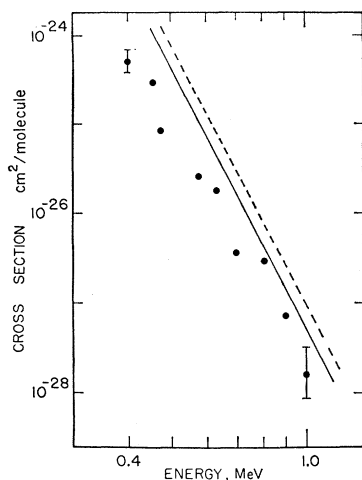


FIG. 4. The double-electron-capture cross section σ_{1-1} for protons incident upon hydrogen gas. Units are $\text{cm}^2/(\text{hydrogen molecule})$. \cdots , present experimental values; $---$, Mittleman, first Born approximation (Ref. 4); $—$, Mittleman, "modified" first Born approximation (Ref. 4).

The method of cross-section measurement, namely, determination of the rate of growth with target-gas number density of the fast collision products from an originally pure primary beam, the experimental accuracy, and the validity of the measurements were determined in a manner used previously¹⁷ and are given in detail elsewhere.¹⁸ Error bars are given in Figs. 2 to 6. The present cross-section values are absolute values with due allowance made for the effects of "pumping" by a liquid-nitrogen trapped McLeod gauge¹⁹ and of

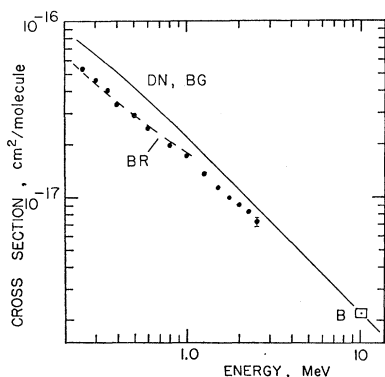


FIG. 5. The single-electron-loss cross section σ_{01} for hydrogen atoms incident upon a hydrogen gaseous target in units of $\text{cm}^2/\text{molecule}$. \cdots , present experimental values; \square , Berkner *et al.*—a single value for 20-MeV deuterium atoms (Ref. 3); $---$, Barnett and Reynolds (Ref. 1); $—$, Bates and Griffing, first Born approximation (Ref. 5); $---$, Dmitriev and Nikolaev, free-collision approximation (Ref. 7). The theoretical values are for atomic hydrogen and have been multiplied by 2 in this figure.

¹⁷ J. F. Williams and D. N. F. Dunbar, *Phys. Rev.* **147**, 62 (1966).

¹⁸ J. F. Williams, Australian Atomic Energy Commission Internal Report, 1966 (unpublished) (available from author).

¹⁹ H. Ishii and K. Nakayama, *Transitions of the Eighth National Vacuum Symposium* (Pergamon Press, Inc., New York, 1961), p. 519.

thermal transpiration,²⁰ which are however only of the order of 2% for hydrogen and helium for the present geometry.

3. RESULTS

A. The Single-Electron-Capture Cross Section, σ_{10}

It is seen in Fig. 2 for a He target gas that below 1 MeV, the present values are in good agreement with the previous experimental values of Barnett and Reynolds,¹ and they show an average dependence upon energy which is similar to that shown by the impulse-approximation calculations by Bransden and Cheshire,¹³ and by the first-Born-approximation calculations (which include a proton-nucleus interaction) by Mapleton.¹² The results from 1 to 2.5 MeV follow approximately an $E^{-11/2}$ energy relationship which, if extrapolated, passes slightly below the values measured by Berkner *et al.*³ for 12.9- and 21.0-MeV deuterons. One hesitates to draw inferences between the similarity of D_1^+ and H_1^+ for electron capture from the limited energy range of the data. A first Born approximation, which excludes the proton-nucleus interaction in the manner of Oppenheimer-Brinkman-Kramers, by Mapleton¹² yields an E^{-6} asymptotic energy dependence. The values of the cross section calculated by Mapleton, as well as those of a similar calculation by Mittleman,¹¹ are more than a factor of 2 larger than the experimental values.

Figure 3 shows that the present values of σ_{10} in molecular hydrogen agree well with the measurements of Barnett and Reynolds in the region below 1 MeV, where the cross section decreases approximately as $E^{-5.3}$. Above 1 MeV, the cross section then decreases more quickly than $E^{-5.3}$. The first-Born-approximation calculations by Dalgarno and Griffing²¹ and by Jackson

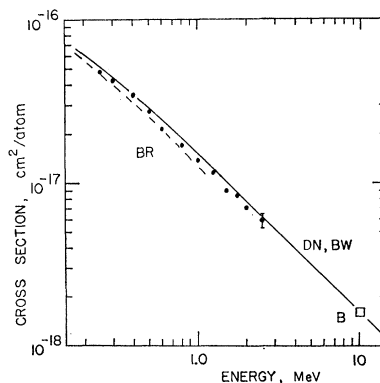


FIG. 6. The single-electron-loss cross section σ_{01} for hydrogen atoms incident upon a helium target. Units of cm^2/atom . \cdots , present experimental values; \square , Berkner *et al.*—a single value for 20-MeV deuterium atoms (Ref. 3); $---$, Barnett and Reynolds (Ref. 1); $—$, Bates and Williams, first Born approximation (Ref. 4); $---$, Dmitriev and Nikolaev, free-collision approximation (Ref. 7).

²⁰ H. H. Podgurski and F. N. Davis, *J. Phys. Chem.* **65**, 1343 (1961).

²¹ A. Dalgarno and G. W. Griffing, *Proc. Roy. Soc. (London)* **A248**, 415 (1958).

and Schiff²² have been selected as being representative of the many calculations¹¹ which have included a proton-nucleus component in the interaction potential for an atomic-hydrogen target. Their values are the sum of the partial cross sections for capture into all states of the atom and for capture into the ground state only, respectively. Assuming that a hydrogen molecule behaves effectively as two isolated hydrogen atoms for the purposes of charge transfer, the theoretical values of σ_{10} (multiplied by 2) in atomic hydrogen are higher than the experimental values. Tuan and Gerjuoy¹⁴ have pointed out that, even for fast collisions, a molecule does not behave like two isolated atoms. However for energies less than 0.4 MeV the capture cross section in molecular hydrogen σ_m is approximately twice that in atomic hydrogen σ_a only because of accidental compensation of interference effects from the two atoms in the molecule. At the extreme high-energy limit they have shown that $\frac{1}{2}\sigma_m = (1.2 \text{ to } 1.4)\sigma_a$. A comparison of such predictions with the experimental values in molecular hydrogen is doubtful because of (a) the uncertainty of the molecular wave functions and of (b) the uncertainty of the theoretical value of σ_a when there are no experimental values in atomic hydrogen to determine the correct interaction potential for use in the Born-approximation calculations of σ_a .

B. The Double-Electron-Capture Cross Section, σ_{1-1}

The large experimental uncertainty of up to 60%, in the measurements of σ_{1-1} as shown by error bars in Fig. 4, was due primarily to a low signal-to-noise ratio. The theoretical values of Mittleman⁴ have been extrapolated above 0.625 MeV. Previous measurements^{15,23}

²² J. D. Jackson and H. Schiff, Phys. Rev. **89**, 359 (1953).

²³ G. W. McClure, Phys. Rev. **132**, 1636 (1963).

of σ_{1-1} at low energies have indicated a preference for the modified-Born-approximation values (modified to correct for nonorthogonality of the wave functions) rather than the unmodified-Born-approximation values. The present experimental values are appreciably lower than the predicted values but both sets of values appear to be converging to the same limit above 1 MeV.

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

The present experimental values of σ_{01} in hydrogen and helium are given in Figs. 5 and 6. In both gases the previous measurements by Barnett and Reynolds¹ below 1 MeV are confirmed within 10%, as is an extrapolation of their data with an approximately E^{-1} energy dependence to pass through the single value of Berkner *et al.*² for 20-MeV deuterium atoms. In helium there is good agreement between the experimental values and the values calculated from the free-collision approximation⁸ and the first Born approximation.^{5,6} The calculated values^{6,8} for atomic hydrogen are in good agreement with one another but they are only in fair agreement with the experimental values in molecular hydrogen, although it appears that above 10 MeV both sets of values may have the same asymptotic dependence.

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