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RESEARCH ESTABLISHMENT
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THE MEASUREMENT OF CHARGE TRANSFER CROSS SECTIONS FOR HIGH ENERGY (~1 MeV) PROTONS AND HYDROGEN ATOMS INCIDENT UPON HYDROGEN AND HELIUM GASES

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ABSTRACT

Measurements of the single and double electron capture cross sections for protons, σ_{10} and σ_{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 hydrogen and helium target gases are reported and compared with published theoretical estimates and experimental values. The present values of σ_{10} and σ_{01} agree well with the data of Barnett and Reynolds below 1 MeV. The extrapolation of the data of Barnett and Reynolds to pass through the single value at 10 MeV by Berkner is confirmed. The experimental values of σ_{01} agree with calculations that use the Born and free-collision approximations within the experimental uncertainty of ± 10 per cent. The values of σ_{1-1} decrease from 5.1 x 10^{-25} cm^2/mol at 0.4 MeV to 1.6 x 10^{-28} cm^2/mol at 1.0 MeV with an experimental uncertainty of up to a maximum of 60 per cent. These values are lower than, but exhibit a similar dependence upon proton energy to the first Born approximation calculations by Mittleman.

A detailed discussion is given of the experimental accuracy which was based upon a procedure previously devised by the author.

The cross sections were measured 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. This method is more accurate than the methods used by other workers. The apparatus was developed for use at pressures less than 10^{-8} mmHg by baking at 400 °C. It was designed for measuring a wide variety of charge changing collisions.

CONTENTS

1. INTRODUCTION

This report presents the measurements of the several charge–changing collision cross sections which have been made, and discusses the experimental apparatus, the method of cross section measurement, and those tests which have been made to determine the validity and experimental accuracy of the cross section values. Considerable details of the above aspects are given since this investigation was the first study of such charge–changing collisions at the Australian Atomic Energy Commission Research Establishment.

1.1 The Problems

Measurements were made of (a) the single electron loss cross section, σ_{01}, for fast hydrogen atoms incident upon hydrogen and helium targets and (b) the single and double electron capture cross sections, σ_{10} and σ_{1-1} respectively, for fast protons (~ 1 MeV) incident upon hydrogen and helium targets. These cross sections at energies less than 50 keV have all been measured previously by the author (Williams 1965, 1966a), who has given a discussion of other measurements before 1965.

The cross section for electron loss from fast hydrogen atoms in hydrogen and helium targets has been calculated previously by three methods: the semi–classical model of Bohr (1949), the "free–collision" (or impulse) approximation (Dmitriev and Nikolaev 1963), and the first Born approximation (Bates and Griffing 1955; Bates and Williams 1957). All of these models predict an E^{-1} energy dependence at high energies (that is where $Z e^2/hv << 1$). For a molecular hydrogen target this condition implies energies above a few hundred kilovolts. In this asymptotic region the Born and free–collision approximations give numerical values that agree with each other and with experiment (Barnett and Reynolds 1958; Berkner et al. 1964) to within 15 per cent. The only experimental values are those of Barnett and Reynolds from 0.25 to 1.0 MeV, and a single measurement for 20 MeV deuterium atoms made by Berkner et al. 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 accurate (Williams 1965) 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. The present work measures σ_{01} over the energy range 0.25 to 2.5 MeV to permit a comparison of experimental and theoretical values to be made over a wider energy range than previously possible.

Many calculations of cross sections for single electron capture by fast protons in gases, especially in atomic hydrogen, have been published. The various approaches have been discussed by Bates (1962), Bates and McCarrol (1962), Bransden and Cheshire (1963) Mapleton (1963), and Mittleman (1963). There is substantial disagreement on both the magnitude and the energy dependence of the published cross sections for energies about 1 MeV. The only experimental values above 0.25 MeV are those of Barnett and Reynolds (1958) from 0.25 to 1.0 MeV in molecular hydrogen and helium and those of Berkner et al. (1965) for 12.9 and 21 MeV deuterons in helium. The present work measures σ_{10} over the energy range 0.25 to 2.5 MeV and enables a comparison with existing data to be made over a larger energy range than previously possible as well as a comparison between the average energy dependence of protons and deuterons in the single electron capture process at high energies.

The double electron capture collision process, which can be represented by the equations:

$$H_1^+ + H_2 \rightarrow H_1^- + 2\,H_1^+ \quad , \qquad H_1^+ + He \rightarrow H_1^- + He^{++} \qquad (1) \text{ and } (2)$$

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. The collision is described by the notation of Hasted (1960), that is, a (10/−12) collision whose cross section is σ_{1-1}.

Before collision the hydrogen molecules are almost entirely in their ground vibrational level. The life-times of the electronically excited states are such that they would decay rapidly to the ground state while one may reasonably expect a Boltzmann distribution amongst its vibrationally excited levels which, at a temperature around 20 °C, implies that only 1 to 10^4 molecules would be expected to be in levels other than the ground vibrational (v = 0) level. The proton, of course, has no extra–nuclear electrons. Hence before the collision there are only two electrons present in the colliding system. By definition of a (10/−12) collision these two electrons must be transferred to the fast proton to form a negatively charged hydrogen ion during the collision. There are no free electrons after the collisions. Massey (1950) has shown that it is extremely unlikely that the H_1^- ion has any stable excited states. The almost stationary hydrogen molecule, after losing its two electrons, consists then of only two protons which are forced apart by virtue of their Coulomb repulsion.

A similar reasoning may be applied to reaction (2) above, except that the target helium atom loses its two electrons to form an alpha particle. Hence the reaction of the type (10/-12) for protons in either hydrogen or helium is extremely well defined and is suitable for both theoretical and experimental study.

Previous studies of this reaction (Fogel et al. 1959; McClure 1964; Williams 1965, 1966c), have been confined to the energy region centring around 20 keV for several reasons. First, power supplies in excess of 50 or 100 keV have not been both readily available and inexpensive. Secondly, the cross section σ_{1-1} appears (Williams 1965) to decrease roughly as the inverse eleventh power of the increasing relative velocity of the colliding particles once the peak of the cross section is reached, and hence the extremely small cross section values ($< 10^{-19}$ cm^2/mol) above 50 keV proton energy would be difficult to measure accurately. Thirdly, and most importantly, the (10/-12) process in hydrogen and the inert gases has been found to have its maximum value generally at energies less than 50 keV. Hence interest in the applicability of (10/-12) collisions to the adiabatic hypothesis (Hasted 1960; Williams 1966c) was confined to the energy region below about 50 keV.

Until very recently (Mittleman 1965) there had been no theoretical study of the double electron capture process in hydrogen. There have been two studies for helium. At low energies, less than 15 keV, Rosentsveig and Gerasimenko (1955) have used a perturbed stationary state (P.S.S.) method with little success in obtaining agreement with experimental values. Their predictions of energy dependence and absolute value were entirely different from the experimental values.

At high energies (above 100 keV) Gerasimenko and Rosentsveig (1956) have used the first Born approximation to predict values of σ_{1-1} which are several orders of magnitude higher than extrapolations of the experimental values (Bates and Griffing 1955). Possible reasons for the disagreement of their predictions with the extrapolated experimental values may lie (Williams 1965) in the facts that they (a) used non-orthogonal initial and final state wave functions, (b) neglected the change in translational motion of the two captured electrons, and (c) used a hydrogen-like wave function for the helium atom. It has been shown by many workers (Bates 1962) that each of these factors exerts an appreciable effect on the single electron capture cross section calculations in the first Born approximation, and so it is not unreasonable to expect a similar effect of these factors on the double electron capture cross section calculations.

The experimental and theoretical values can be compared only by a large extrapolation of either set of values, which is an unsatisfactory procedure. The need for experimental work in the energy region above 100 keV is clearly seen. In the present work, measurements of σ_{1-1} have been made in hydrogen gas for protons from 0.4 to 1.0 MeV. Measurements were attempted in helium gas but the cross section was too small to give reasonable statistics.

2. APPARATUS

2.1 General Description

The apparatus was arranged as shown in Figure 1. The primary proton beam emerged from the beam-defining aperture A_1. It then passed through collision cell C_1 which would be used as a charge-exchange cell to form neutral beams; the electrostatic field between plates P_1 and P_2 could then be used to remove the charged particles from this neutral beam. However the field between P_1 and P_2 was also used to check for the presence of background neutral particles in the primary proton beam. Then either a primary proton or a neutral atomic hydrogen beam could enter collision cell C_2. The electrostatic field between plates P_3 and P_4 separated the three charge states (H_1^+, H_1^0, and H_1^-) of the emergent beam; as there can be no mass change and a negligible momentum change of the atomic hydrogen beam, an electrostatic, rather than a magnetic, analysing field was sufficient for this experiment. The beam currents in the three charge states could then be detected simultaneously in various detectors.

Two bellows were provided on the system. The first bellows B_1 permitted the alignment of the collision cells with the beam defining canal. The second bellows B_2 permitted the alignment of any of the detectors with the undeflected beam axis or with any of the three charge state beams after analysis by field P_3P_4. It also permitted the movement of any detector from one beam axis to another without breaking the vacuum.

2.2 The Beam

The characteristics of the momentum-analysed beam were as follows. The protons were extracted from an electrodeless discharge type source, focused and accelerated to their final energy, E, and then momentum analysed by a 12-inch radius, 30° deflection magnet to pass through a seven-port switching magnet to form an approximately parallel beam through aperture A_1. Proton beam currents of 50 μA were readily obtained in an area of 2 mm diameter prior to aperture A_1, the current being limited only by the maximum permissible radiation dose at a distance of one metre from the aperture. The beam current stability at this position was usually better than 5 per cent. after several hours of operation.

The beam energy spread was negligible for the present purpose. Previous work (Williams 1966b) has shown that beam energy spreads up to 50 eV and mean beam energies up to 200 eV above the source extraction potential may exist in the beam after extraction from the source. Cook (1962) has shown that for more powerful discharges of up to 500 watts input of r.f. power, these beam energy spreads and mean energies may be three times as large as the above figures. However these variations are negligible when compared with the corona load fluctuations and the resolution of the main momentum-analyzing magnet for beam energies in the vicinity of 1 MeV.

The actual beam energy was calibrated absolutely against the following thresholds: the Li^7 (p,n)Be^7 at 1.882 MeV, the C^{13} (p,γ) at 1.75 MeV, the N^{15}(p,γ) at 0.898 MeV and 0.429 MeV; in each case the calibration was considered better than 3 per cent. which was sufficient for the present measurements.

2.3 The Collision Cell

The aperture A_1 served several purposes. It took the form of a canal 0.010 inch in diameter by 0.500 inch in length, accurately machined into a 0.5 inch diameter cylinder, which, in turn, was located at the centre of a 2 inch diameter aluminium water-cooled disc. First, this disc completely separated the vacuum system of the accelerator from that of the present experiment; the small canal had such a large pumping impedance that any fluctuations in the gas pressure of the accelerator would result in negligibly small increases in the ultimate pressure ($\sim 10^{-8}$ mmHg) of the present system. Secondly, it defined the beam diameter and placed an upper limit of 10^{-3} radians on the angular collimation of the beam. It was also an insulated aperture to permit measurement of the total beam current falling onto the aperture and, together with the beam current transmitted through the aperture, it enabled an estimate to be made of the beam current profile.

2.4 The Vacuum System

The collision chamber was mounted on top of an Edwards 6-inch mercury diffusion pump, complete with a liquid nitrogen trapped baffle and a plate valve. The pumping speed of this arrangement at the top of the plate valve (when fully open) was estimated from data supplied by Edwards to be of the order of 200 l/sec.

Complete working drawings of the apparatus are given in Figures 2(a) to 2(e). Stainless steel and copper gaskets were used exclusively in the construction to permit the baking of the apparatus up to 400 °C. The internal surface area of the vacuum system was of the order of 2.25 x 10^5 cm^2 which may be estimated (Dushman 1962) to contain up to 10 litres of absorbed gases. It is probable that the main portion of these absorbed gases would be removed after prolonged pumping at high temperatures, the remaining gases contributing an influx of gas to the vacuum system not greater than 100 l/sec at 5 x 10^{-8} mmHg.

The two collision cells were identical. In the initial work an entrance aperture diameter of 0.020 in. and an exit aperture diameter of 0.050 in. were used. The lowest residual gas pressure attained was of the order of 5 x 10^{-9} mmHg. If a maximum pressure of 7 x 10^{-4} mmHg* was required in the collision cell, then the quantity of gas flowing from the collision cell would be about 0.03 l/sec at 1 dyne cm^{-2}. Thus a pumping speed of 30 l/sec at 10^{-3} dyne cm^{-2} would be required to remove this target gas flow. This gas flow could be readily pumped by the above pumping system with an adequate allowance for the desorbed gas flow from the metal walls whilst an ultimate pressure of 5 x 10^{-7} mmHg was maintained throughout the system. The maximum pressure which could be maintained in both of the collision cells without any increase in the residual gas pressure would be about 10^{-5} mmHg.

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* This value was selected as it is the pressure equivalent to 1 dyne cm^{-2}, however it also fortuitously coincides approximately with the maximum pressure under which single collision conditions prevail in the collision cell (Williams 1965).

Each of the collision cells could be pumped separately by a 1 in pipe connected through a valve to the main 6 in. pump to permit a rapid pump-down time. Each cell was connected by a tube of 1 inch diameter and 2 inch length to a Metrovac ionization gauge, type 2D-19. There was no possibility of a localized low pressure in the vicinity of the ion gauge.

2.5 Construction Details (See Figures 2(a) to 2(e))

Great care was taken to align the four apertures of the collision cells on the one axis normal to the main 6 inch diameter flanges during construction. Three of them were bored with the one setting of the collision chamber on the milling machine. It was estimated that the four holes were aligned to better than 0.002 in. Considerable care was taken to have all flanges either perpendicular or parallel to one another as required, to facilitate the correct and easy alignment of the four apertures of the collision cells with the beam-defining canal A_5 and the detectors D.

2.6 Electrostatic Analyzers

The electrostatic analyzers took the simple form of two parallel plates, since only charge separation of the collision products was required. The plates P_1 and P_2 (see Figure 1) were spaced 0.04 in. apart while P_3 and P_4 were spaced 0.16 in. apart. Such spacings permitted the minimum voltages to be used (to reduce breakdown problems) and yet still produced sufficient field strengths to give the required separation of the three charge states of the hydrogen beam. A voltage of 30 kV across plates P_3 and P_4 separated the H_1^+ and H_1^0 beams by 1.5 cm at a distance of 30 cm from the centre of the deflection field.

The high voltage was derived from a 0-100 kV Hursant supply. The performance of the supply was determined by the C.S.I.R.O. High Voltage Laboratory to be as follows: At all voltages up to 100 kV and with any load current up to 100 μA, a sudden change ($<$ 1 sec) in the load current from zero to 100 μA changed the output voltage by less than 1 per cent. This performance was considerably better than that required for the present experiment.

2.7 Detectors

The detectors were of three types: (a) simple Faraday cups, (b) secondary electron emission detectors, and (c) surface barrier detectors. Types (a) and (b) permitted only total charge collection techniques while type (c) enabled single-particle counting techniques to be used. Types (b) and (c) responded to both charged and uncharged particles. The bellows system connecting the detectors to the vacuum system enabled any of the three types of detector to be used with any of the three charge states of the hydrogen beam. During measurements of, say, σ_{1-1}, the primary H_1^+ current ($\sim 10^{-6}$ A) was collected in a Faraday cup while the H_1^0 and H_1^- beams were counted separately by surface barrier detectors. Type (b) detector was used only for preliminary measurements.

The output from the surface barrier detector was fed into a low noise pre-amplifier, then into an amplifier, discriminator, and scaler as shown in Figure 3. The discriminator level was set with the aid of a multichannel analyzer. There were two separate detection and counting systems to enable the H_1^0 and H_1^- particles to be counted simultaneously.

3. EXPERIMENTAL METHOD

This method is based on that used previously by the author (Williams 1965). The composition of an atomic hydrogen beam, which has three possible charge states, as it passes through a target gas is given by the set of equations (Allison 1958):

$$\frac{d}{d(nl)} N_+ = -(\sigma_{10} + \sigma_{1-1}) . N_+ + \sigma_{01} . N_0 + \sigma_{-11} . N_- \tag{3a}$$

$$\frac{d}{d(nl)} N_0 = + \sigma_{10} . N_+ - (\sigma_{01} + \sigma_{0-1}) . N_0 + \sigma_{-10} . N_- \tag{3b}$$

$$\frac{d}{d(nl)} N_- = + \sigma_{1-1} . N_+ + \sigma_{0-1} . N_0 - (\sigma_{-11} + \sigma_{-10}) . N_- \quad , \tag{3c}$$

where N_+, N_0, and N_- are the numbers of protons, neutral hydrogen atoms, and negative hydrogen ions in the projectile beam at any point in the traverse of a target gas,

n is the number of target gas atoms/cm^3 ,

l is the beam path length in the target gas,

σ_{if} is the cross section for the collision in which a particle of charge ie is changed to a particle of charge fe .

Allison has fully discussed these equations and their solutions, from which it is readily shown, if the exponential solutions for the N's are expanded in polynomial form, that

$$\Delta N_f/N_i \cdot \Delta(n\,l) = \sigma_{if} + a_{if}\,(n\,l) + b_{if}\,(n\,l)^2 + \ldots\ldots + \text{higher powers of } (n\,l) \qquad (4)$$

where the a_{if} and b_{if} are functions of the six possible σ_{if} .

The method of measuring the cross section σ_{if} that is used in the present experiments readily follows from Equation 4. A study of the dependence of the growth of N_f, from a constant N_i , on the quantity (n l) shows it to be linear, that is, the change of charge of the primary atoms results only from single collisions, when $a_{if}\,(n\,l)/\sigma_{if} << 1$. Since this condition of linearity of $\Delta N_f/\Delta(n\,l)$ depends on the nature of the primary and target atoms and their relative velocity, each individual cross section has been measured by plotting $\Delta N_f/N_i$ against $\Delta\,(n\,l)$ to ensure the separation of the linear and parabolic components.

The N_i and N_f are readily measured absolutely by charge integration methods with electrometers. The absolute measurement of (n l) is made difficult because as n is varied within the collision cell both n and l vary in the vicinity of the entry and exit canals. Their measurement is complicated by the reduction (though very small) of the vacuum pumping speed by the baffling action of the electrostatic deflector plates P_1 and P_2 before the entry canal and P_3 and P_4 after the exit canal. For the present apparatus these complicating effects must be small for small values of (n l), and the assumption of a constant value for l is well supported by the relationship between the growth of N_f and (n l) (see Figure 4 for example), which was linear within 5 per cent. at low (n l) values. When the contributing errors to this 5 per cent. figure are considered (Section 4) it can be seen that l would vary by much less than 2 per cent.

The number, n, of gas atoms/cm^3 along the beam path was determined in two ways. First, only relative values, n', were measured with an ionization gauge (Metrovac type 2 D-19). From Equation 4,

$$\sigma_{if} = \frac{1}{N_i} \cdot \frac{\Delta N_f}{\Delta(n\,l)} \quad \text{to a first approximation,}$$

$$= \frac{1}{N_i} \cdot \frac{\Delta N_f}{\Delta n'} \cdot \frac{1}{C} \qquad (5)$$

where C, equal to $\Delta(n\,l)/\Delta n'$, is a calibration constant of the apparatus for each target gas and relates the true values of n to the relative values n'.

Figure 4 shows one of the best examples of the growth of N_f with (n l), in particular of the growth of H^0_1 particles from a 0.8 MeV H^+_1 beam incident upon a hydrogen gas target. Such a large number of experimental points was taken only during the early measurements of a given σ_{if} in each target gas. For all σ_{if} determinations, the ratio N_f/N_i did not exceed 10^{-3} and was generally very much smaller and n' was varied by at least a factor of 10. The error in the determination of the slope of the linear relationship of N_f/N_i to n' in Figure 4 is approximately 8 per cent., which is composed of (a) an error not greater than 5 per cent. in measuring N_i, N_f, and n', and (b) an error of not greater than 3 per cent. in fitting a straight line to the experimental data. The poor stability in both current and position of the primary beam was mainly responsible for the large scatter of the experimental points about the linear relationship.

The constant, C, in Equation 5, was determined by the method used by Fite (1960) and McClure (1963), namely the measurement of the conversion of H^+_1 to H^0_1 by single electron capture in each target gas used and assuming the cross section value for that process, σ_{10}, at a given energy. The energy of 0.8 MeV was selected for this standardization procedure because the majority of accuracy tests, described in Section 4, were performed at 0.8 MeV, giving most confidence to the present experimental results at that energy. The values of 6.0×10^{-22} cm^2/mol in hydrogen and 1.5×10^{-20} cm^2/atom in helium were selected as the standardization values of σ_{10}. If this value of σ_{10} should be shown to be in error in the future, all of the present σ_{if} values can be corrected accordingly. An alternative standardization procedure would be to position the present experimental curve such that the best agreement in shape over as wide an energy range as possible was obtained with the mean shape of Barnett and Reynolds' (1958) results. This procedure leads to the same results as the first procedure.

The above method of estimation of n was used only because a McLeod gauge was not available at the start of the project; however during the latter part of the project an opportunity arose to use a liquid N_2 trapped McLeod gauge which was currently being used for ionization gauge calibrations. The results of ten measurements of n by a McLeod gauge were within 10 per cent. of the values of n determined by the first procedure discussed above. It is felt that by far the greatest contribution to this 10 per cent. arises from beam instabilities, which cause the large errors in relative n measurements, rather than inaccuracies in measurement of pressure by the McLeod gauge. The errors caused by the pumping action of a liquid nitrogen trap on a McLeod gauge are estimated to be of the order of 3 per cent. for hydrogen (Ishii and Nakayama 1961; Meinke and Reich 1963). The effect of thermal transpiration upon the calibration is negligible for the geometry in use (Podgurski and Davis 1961).

4. EXPERIMENTAL ACCURACY AND VALIDITY

The accuracy and validity of the cross section measurements were investigated in the following experiments (Williams 1965), several of which were based on, or initiated by, considerations such as those given by Stier and Barnett (1956), Allison (1958), and Fogel (1960).

1. Tests performed at beam energies of 0.3 and 2.5 MeV (the lower and upper limits used in the present measurements) and 0.8 MeV (the standardization energy for the σ_{10} values) showed that:

(a) Each of the detectors D_0, D_1, and D_2 gave identical responses to the same beam.

(b) Variations, over the normal operating range, in either the target chamber or accelerator gas pressure or the analyzing electrostatic field had no effect upon the response of the detectors.

(c) Any scattering of the primary beam by collision with residual gas molecules along the beam path had a negligible broadening effect on the beam.

(d) Identical beam profiles were obtained at the D_1 and D_2 positions by reversing the direction of the electrostatic field $P_3 P_4$. Interchange of detectors had no detectable effect on the beam profile.

It was also shown that an increase in the collision cell gas pressure from 2×10^{-6} to 7.8×10^{-4} mmHg (the maximum value used in these measurements) while the gas pressure external to the cell was maintained below 1×10^{-6} mmHg produced negligible broadening of the primary H_1^+ ion beam.

2. The primary H_1^+ beam will always contain some H_1^0 and H_1^- particles originating from one or two-electron capture by the primary beam by collision with (a) the edges of the beam-defining apertures A_1 and A_2 or with the walls of the entry and exit canals, and with (b) the residual gas molecules along the beam path between the switching magnet and the entry aperture of the collision cell. The influence of effects such as (a) were minimized by using knife-edged apertures and a beam diameter considerably less than the aperture diameters. Also all apertures were aligned optically on assembly. Fortunately the numbers of H_1^0 and H_1^- particles so formed are independent of the collision gas cell pressure. Effectively all of the H_1^0 and H_1^- particles will pass through the collision cell because, in the present "single-collision" experiments, not more than 3 per cent. of the primary beam undergoes a charge-changing collision in the collision cell. Since a cross section is determined by measuring the linear rate of growth of collision products with pressure, the numbers of H_1^0 and H_1^- particles, which are produced in the above manner and which are pressure independent, will appear as a constant term which does not influence the slope of the relationship between collision product and pressure. For the present measurements this constant term was usually less than 10 per cent. of the total collision product current but occasionally for the smallest cross sections rose to 60 per cent.

Also when the values of (nl) of the target gas increase within the collision cell there will be an increase in (nl), which originates from the target gas, in the external vicinity of the entry and exit canals. This increase in (nl) will add to that within the cell but cause no error in the present cross sections when such values are only determined relatively. However for an absolute determination of the cross section, such "end effects" were determined by using a gas cell of a different geometrical length. This was readily achieved since any of the apertures A_5 to A_6 could be alternatively located at the end of a small cylindrical extension to the gas cell.

3. The impurity H_1^0 and H_1^- particles mentioned in paragraph (2) in their traverse through the collision cell may collide with the target gas atoms to give rise to linearly pressure dependent collision products which are indistinguishable from those formed from the true primary beam.

(a) The errors arising from the neutral atoms formed in the residual gas or on the edges of the beam-defining apertures, but not from the walls of the entry canal, are simply determined by electrostatic deflection of all charged particles from the beam before their entry to the gas cell. Thus the remaining "primary" beam is now a neutral atom (H_1^0) beam, from which the collision products H_1^+ and H_1^- may be observed as a function of the gas pressure. The fraction of such H_1^- impurities compared with those H_1^- ions produced from the H_1^+ ions, will thus depend upon (i) σ_{10} for H_1^+ ions in the residual gas of the vacuum system, and (ii) the relative values of σ_{1-1} and σ_{0-1} in a given target gas, whilst each cross section depends upon the projectile energies. This type of investigation has been made for every cross section measurement. Table 1 shows typical values of the detected currents for 0.8 MeV H_1^+ ions in hydrogen gas, for which the neutral atom impurities discussed above give rise to a negligible error in σ_{1-1}.

(b) The negative ions, formed in the primary beam before the electrostatic deflector plates, may lose electrons to form H_1^0 or H_1^+ particles. This error cannot be determined in the manner of paragraph (a) because the electric field will deflect the H_1^- ions. However the fraction of H_1^- ions present in the beam is so small (in Table 1 it is $\sim 10^{-10}$ of the primary H_1^+ beam) that, even if all these H_1^- ions changed charge to either H_1^0 or H_1^+ particles, the increase in the fractions of H_1^+ or H_1^0 particles would not be detectable.

(c) An estimate of any errors arising from those neutral atoms which might be formed on the entry aperture walls was attempted by increasing the diameter of the entry aperture from 0.01 in. to 0.02 in. and then to 0.05 in., for a beam diameter of approximately 0.01 in. The minimum gas pressure within the cell was unchanged by increasing the entry canal diameter since the cell had a separate pumping line. This increase of entry aperture diameter above 0.01 in. had no detectable effect upon the readings presented in Table 1. Therefore an upper limit may be placed upon the number of neutral particles formed on the entry canal walls, of 3 per cent. of the number of H_1^0 atoms formed by collision of the primary H_1^+ ions with the residual gas along the beam path. Less than 3 per cent. of these surface-formed H_1^0 atoms will suffer a collision with the target gas so that the fraction of H_1^- ions formed from these H_1^0 atoms will be negligible compared with that formed from the H_1^+ primary atoms in collision with the target gas at its minimum pressure.

5. RESULTS

Tables 2, 3, and 4 list the cross section values and their experimental uncertainty. Each value is the average of at least two separate measurements.

5.1 The Single Electron Capture Cross Section σ_{10}

5.1.1 Helium

Figure 5 compares the present determinations with previous experimental and theoretical values. At energies less than 1.0 MeV they agree well (within 15 per cent.) with the data of Barnett and Reynolds (1958). Berkner et al. (1965) who measured σ_{10} for 12.9 and 21.0 MeV deuterium ions, found that their two points coincided with the linear extrapolation of the data of Barnett and Reynolds, the line having an average energy dependence of about $E^{-5.6}$. The present data above 1 MeV indicate an extrapolation that passes below the values of Berkner et al.

The present values are in good agreement with an impulse approximation calculation by Bransden and Cheshire (1963), which also asymptotically approaches an $E^{-5.5}$ variation, and a Jackson-Schiff type first Born approximation (which includes a proton-nucleus interaction) by Mapleton (1963). The Oppenheimer-Brinkman-Kramers (O.B.K.) calculation (a first Born approximation but without a proton nucleus interaction) by Mapleton yields an E^{-6} asymptotic energy dependence that does not yet appear to be reached in the region below 10 MeV. For ease of comparison with experiment, Mittleman (1963) has produced a simple expression:

$$\sigma = a^2(2^{18}/5)(1.201)\cdot\pi^2 Z E^{-6}\, n_A(0)\,[1 + 0(Z/E)]\ \mathrm{cm}^2 ,$$

which is valid in the restricted energy region:

$$10 < E/Z^2 < 42 ,$$

where a is the Bohr radius, E is measured in units of 25 keV, and $n_A(0)$ is the electron density at the origin in atomic units [$n_A(0) = 3.60$ for He (Pekeris 1958)]. On the above criterion, Mittleman's calculation (giving essentially the O.B.K. result) is applicable in helium for proton energies in the range 1 to 4 MeV. This calculated curve is an order of magnitude above the experimental values at, or less than, 1 MeV and it is a factor of 2 above the results of Berkner et al. at 10 MeV. This calculation clearly over-estimates the true cross section values over the range of its validity.

5.1.2 Hydrogen

Figure 6 shows that the present values of σ_{10} agree well with the measurements of Barnett and Reynolds (1958) in the region below 1 MeV, where the cross section decreases approximately as $E^{-5.2}$. Above 1 MeV the present values indicate an $E^{-6.3}$ relationship. The first Born approximation calculations by Dalgarno and Griffing (1958) and by Jackson and Schiff (1953) have been selected as being representative of the many calculations (Bates 1962) which have included a proton-nucleus component in the interaction potential. 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. When σ_{10} is expressed in units of cm^2/mol, the former values agree well with the experimental values. Further comparison between the experimental and theoretical values is not made because (a) all existing predictions are for atomic hydrogen and (b) there are no experimental values for σ_{10} in atomic hydrogen for proton energies above 0.15 MeV to establish how closely a hydrogen molecule may approximate two hydrogen atoms for the purposes of charge transfer.

5.2 The Double Electron Capture Cross Section σ_{1-1}

Despite a large experimental uncertainty in the measurements of σ_{1-1} of up to a maximum of 60 per cent. at 1 MeV as shown in Figure 7, there is a definite preference for the first Born-approximation calculations of Mittleman (1965) when the calculations are modified to correct for non-orthogonality of the initial and final state wave functions rather than when they are not modified. Previous measurements of σ_{1-1} at energies less than 50 keV had indicated a preference for the lower calculated values. This tendency has been confirmed. Both the experimental and calculated values follow an E^{-9} energy relationship. The large experimental uncertainty was due primarily to low signal to noise ratio at high energies.

Measurements of σ_{1-1} were also attempted in helium gas, but the cross section at 0.5 MeV was too small to give reasonable counting statistics.

5.3 The Single Electron Loss Cross Section σ_{01}

The ratio of the electron loss cross section to the electron capture cross section, σ_{01}/σ_{10}, was determined by observing the charge distribution in the beam after passing through a gas target which was sufficiently thick for equilibrium to be established between the competing capture and loss processes. For energies greater than 0.5 MeV the equilibrium charge distribution was determined both with the proton beam incident on the neutralizer and with a neutral beam incident on the second gas cell. It was thus possible to confirm that the equilibrium charge distribution was independent of the initial charge state.

The stability of the beam from the van de Graaff accelerator deteriorated below 0.8 MeV, although at times the beam stability was about 5 per cent. at 0.5 MeV. In an attempt to obtain a beam with both low energy and good stability, diatomic and triatomic hydrogen ions were accelerated at energies from 0.8 to 1.6 MeV and dissociated in the neutralizer by impact with gas molecules (either H_2 or He). Emerging from the neutralizer were neutral atoms whose energy was one half (or one third in the case of primary H_3^+ ions) of the initial molecular ion energy. The electrostatic field P_1P_2 removed all H_1^+, H_1^-, and non-dissociated primary ions so that only neutral atomic particles were incident upon the second gas cell. Measurements by McClure (1963) at lower energies indicate that at about 1 MeV the quantity of fast H_2 molecules formed by electron capture is negligible. For both the measured equilibrium charge distribution and the single electron loss cross section σ_{01}, primary hydrogen atom beams, formed from either the single electron capture by 500 keV protons or the dissociation of 1 MeV hydrogen molecular ions, yielded results which were identical within the experimental accuracy of ±10 per cent.

The present experimental values of σ_{01} for hydrogen and helium are given in Figures 8 and 9 respectively. In both cases the measurements by Barnet and Reynolds (1958) below 1 MeV are confirmed, as is an extrapolation of their data, with an approximately E^{-1} energy dependence, to pass through the single values at 10 MeV (20 MeV deuterium atoms) by Berkner et al. (1964). In helium there is good agreement between the experimental values and the calculated values using the free-collision approximation (Dmitriev and Nikolaev 1963) and the first Born approximation (Bates and Williams 1957). In hydrogen the calculated values (Bates and Griffing 1955; Dmitriev and Nikolaev 1963) are in very good agreement with each other but they are only in fair agreement with the experimental values although it appears that above 10 MeV both theoretical and experimental values may have the same asymptotic variation.

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TABLE 1

COMPOSITION OF CHARGED AND NEUTRAL ION BEAMS PRODUCED BY A 0.8 MeV H_1^+ BEAM IN HYDROGEN AT TYPICAL TARGET GAS PRESSURES

Primary Particle	Target Gas Pressure (mmHg)	Detector Current (A)		
		H_1^+	H_1^0	H_1^-
Charged plus neutral	1.2×10^{-6}	3.1×10^{-6}	0.9×10^{-15}	1.8×10^{-19}
	7.3×10^{-4}	3.1×10^{-6}	1.8×10^{-13}	5.1×10^{-19}
Neutral only	1.2×10^{-6}	not detectable	4×10^{-16}	not detectable
	7.3×10^{-4}	4×10^{-19}	4×10^{-16}	” ”

TABLE 2

EXPERIMENTAL VALUES OF THE SINGLE ELECTRON CAPTURE CROSS SECTION, σ_{10}, FOR HYDROGEN AND HELIUM GASES

Proton Energy (MeV)	σ_{10}	
	H_2 (cm²/mol)	He (cm²/atom)
0.35		3.2×10^{-19}
0.43		1.5×10^{-19}
0.44	1.0×10^{-20}	–
0.5	5.1×10^{-21}	1.0×10^{-19}
0.6	1.9×10^{-21}	5.2×10^{-20}
0.7	8.9×10^{-22}	3.1×10^{-20}
0.8	4.7×10^{-22}	1.3×10^{-20}
0.88	2.3×10^{-22}	–
1.0	1.0×10^{-22}	5.0×10^{-21}
1.25	2.4×10^{-23}	1.6×10^{-23}
1.5	4.8×10^{-24}	5.0×10^{-22}
1.75	2.1×10^{-24}	2.0×10^{-22}
2.0	5.7×10^{-25}	6.5×10^{-23}
2.5	–	2.5×10^{-23}

TABLE 3

EXPERIMENTAL VALUES OF THE SINGLE ELECTRON LOSS CROSS SECTION, σ_{01}, FOR H_1^0 INCIDENT UPON HYDROGEN AND HELIUM GASES

Energy (MeV)	σ_{01}	
	H_2 (cm²/mol)	He (10^{-17} cm²/atom)
0.25	5.3	4.8
0.3	4.5	4.25
0.35	4.0	–
0.4	3.4	3.5
0.5	2.9	2.73
0.6	2.5	2.67
0.8	1.95	1.73
1.0	1.72	1.4
1.25	1.35	1.15
1.5	1.12	0.9
1.75	0.99	0.83
2.0	0.91	0.71
2.25	0.82	–
2.5	0.73	0.59

TABLE 4

EXPERIMENTAL VALUES OF THE DOUBLE ELECTRON CAPTURE CROSS SECTION, σ_{1-1} FOR H_1^+ INCIDENT UPON HYDROGEN GAS

Energy (MeV)	σ_{1-1} (cm²/mol)
0.4	5.1×10^{-25}
0.46	3.1×10^{-25}
0.48	8.6×10^{-26}
0.58	2.6×10^{-26}
0.63	1.8×10^{-26}
0.71	3.5×10^{-27}
0.8	3.2×10^{-27}
0.9	6.6×10^{-28}
0.94	2.1×10^{-28}
1.0	1.6×10^{-28}

LIST OF FIGURE CAPTIONS AND LEGEND

Figure 1 Schematic Representation of Collision Cells and Vacuum System

Figure 2 (a) Collision Chamber Seen in Vertical Section in the Plane Containing the Beam Axis

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Figure 2 (e) Beam Alignment Tube

Figure 3 Basic Circuit for Particle Counting

Figure 4 The Growth of H_1^0 Atoms from a 0.8 MeV H_1^+ Beam Incident upon a Molecular Hydrogen Target Gas

Figure 5 The Single Electron Capture Cross Section, σ_{10}, for H_1^+ Ions Incident upon Helium Gas in Units of cm^2/He Atom

- • • • Present experimental values
- — . . . — MI Mittleman (1963), first Born approximation
- ——— B C Bransden and Cheshire (1963), impulse approximation
- – . – . MA Mapleton (1963), O.B.K. approximation
- – – – { BR Barnett and Reynolds (1958), experimental; Mapleton (1963), first Born approximation with nucleus–nucleus interaction }
- □ B The 6 and 10 MeV values for primary deuterons by Berkner et al. (1965)

Figure 6 The Single Electron Capture Cross Section, σ_{10}, for H_1^+ Atoms Incident upon Hydrogen Gas in Units of cm^2/hydrogen Molecule

- • • • Present experimental data
- – – – Barnett and Reynolds (1958)
- — – —– Dalgarno and Griffing (1958)
- ——— Jackson and Schiff (1953)

Figure 7 The Double Electron Capture Cross Section, σ_{1-1}, for Protons Incident upon Hydrogen Gas in Units of cm^2/Hydrogen Molecule

- • • • Present experimental values
- —— Mittleman (1965), first Born approximation
- ——— Mittleman (1965), "modified" first Born approximation

(continued)

Figure 8 The Single Electron Loss Cross Section, σ_{01}, for Hydrogen Atoms Incident upon a Hydrogen Gaseous Target in Units of cm^2/Molecule.

- ● ● ● Present experimental values
- □ B Berkner et al. (1964) a single value for 20 MeV deuterium atoms
- — — — Barnett and Reynolds (1958)
- ——— Bates and Griffing (1958), first Born approximation Dmitriev and Nikolaev (1963), free-collision approximation.

Figure 9 The Single Electron Loss Cross Section, σ_{01}, for Hydrogen Atoms Incident upon a Helium Target in Units of cm^2/Atom

- ● ● ● Present experimental values
- □ B Berkner et al. (1964) – a single value for 20 MeV deuterium atoms
- – – – Barnett and Reynolds (1958)
- ——— Bates and Williams (1957) first Born approximation Dmitriev and Nikolaev (1963), free collision approximation.

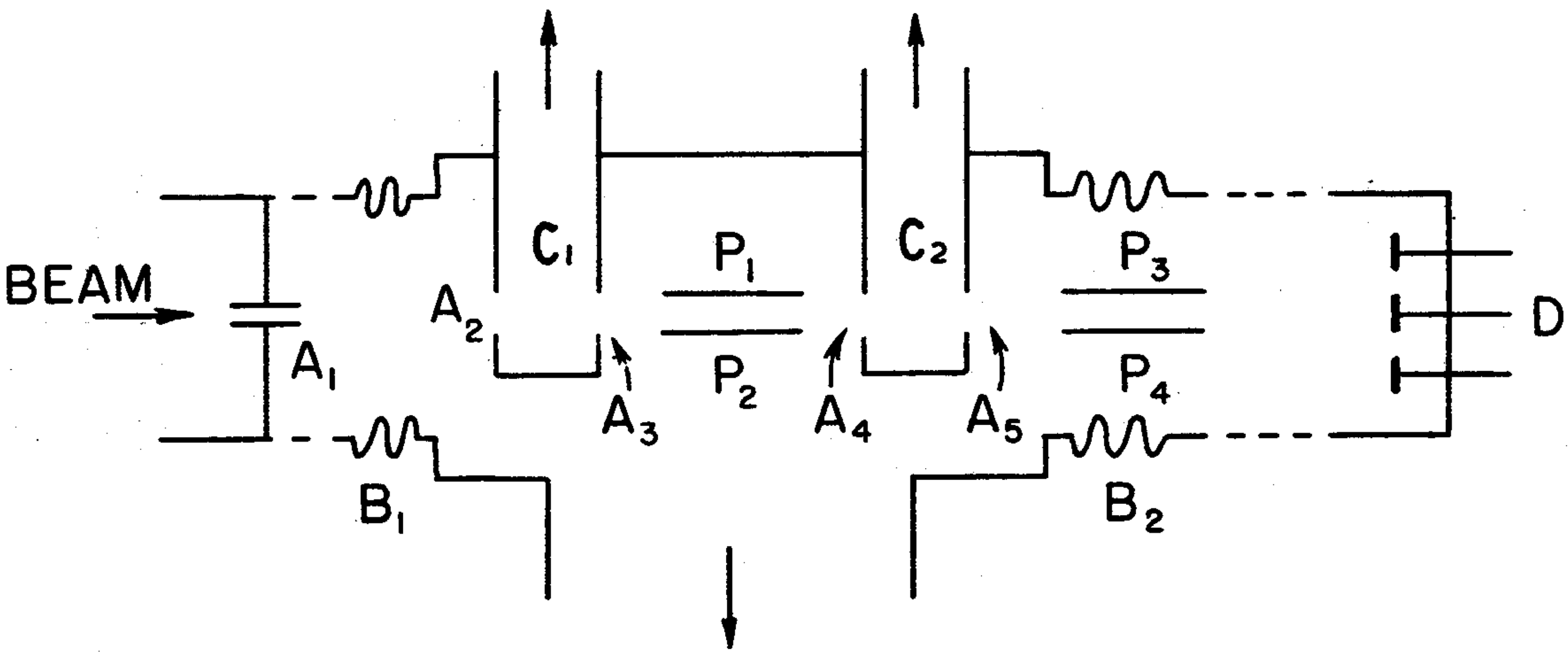

FIGURE 1. SCHEMATIC REPRESENTATION OF COLLISION CELLS AND VACUUM SYSTEM

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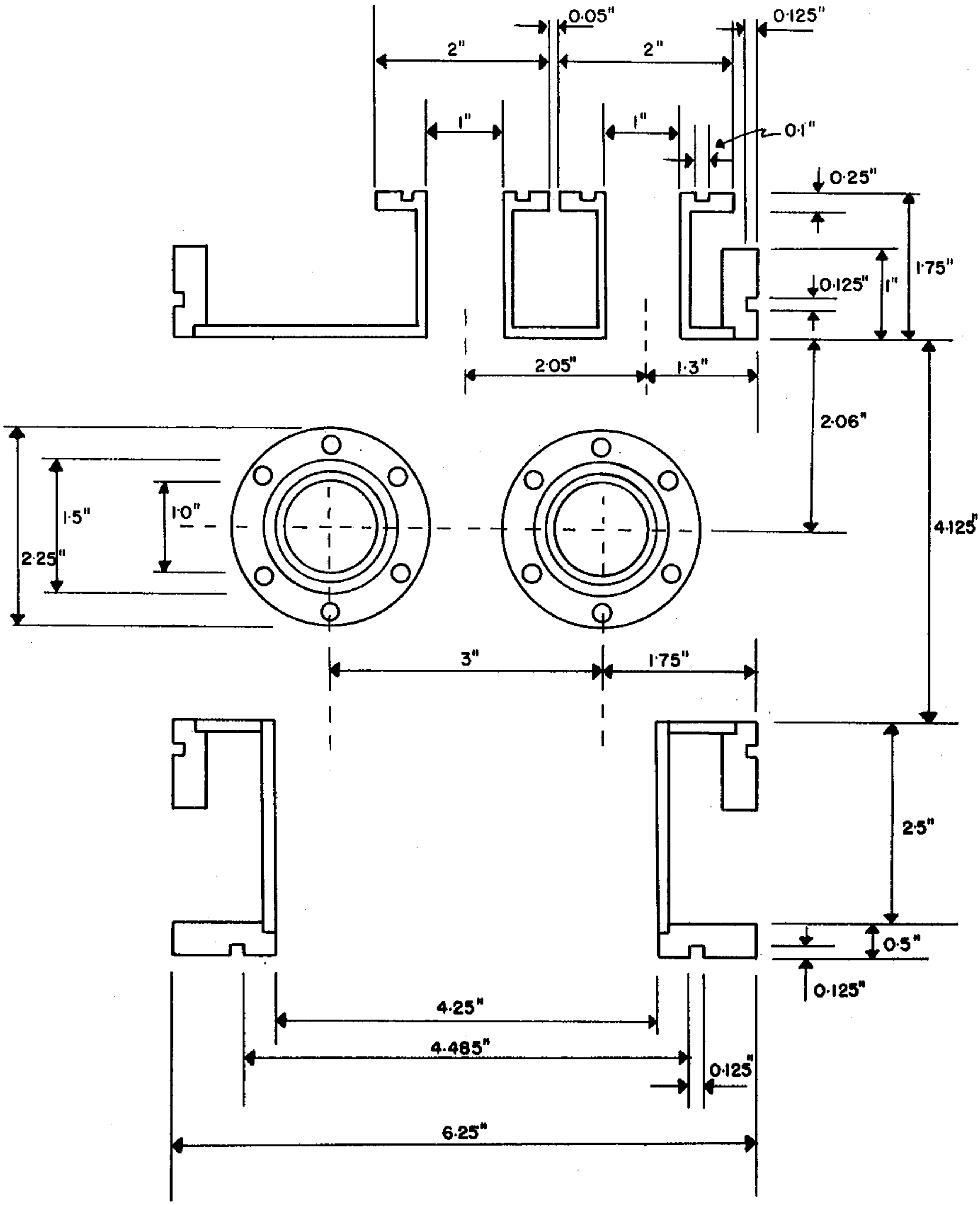

FIGURE 2(a) COLLISION CHAMBER SEEN IN VERTICAL SECTION IN THE PLANE CONTAINING THE BEAM AXIS

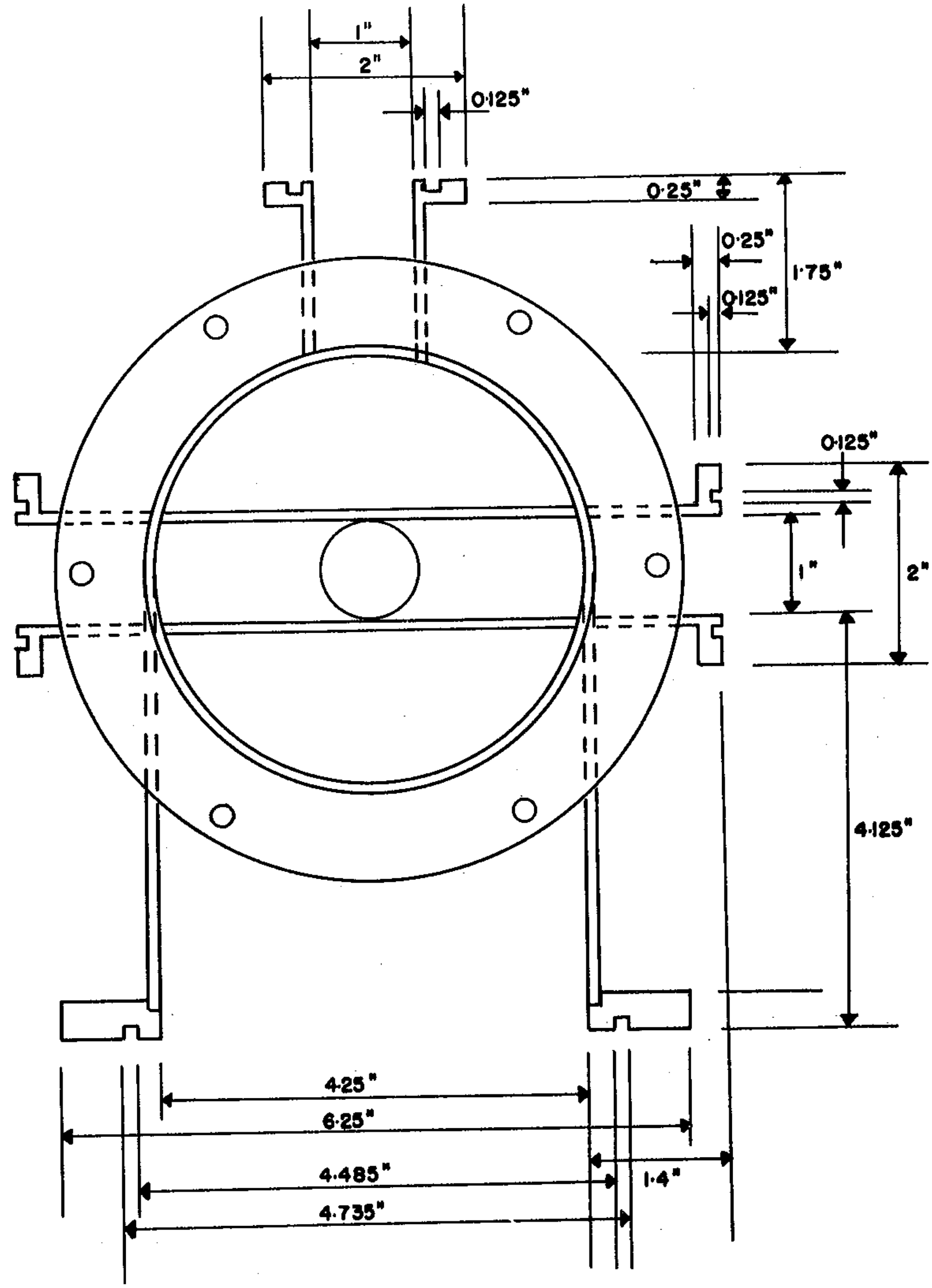

FIGURE 2(b) COLLISION CHAMBER SEEN IN VERTICAL SECTION NORMAL TO THE BEAM AXIS

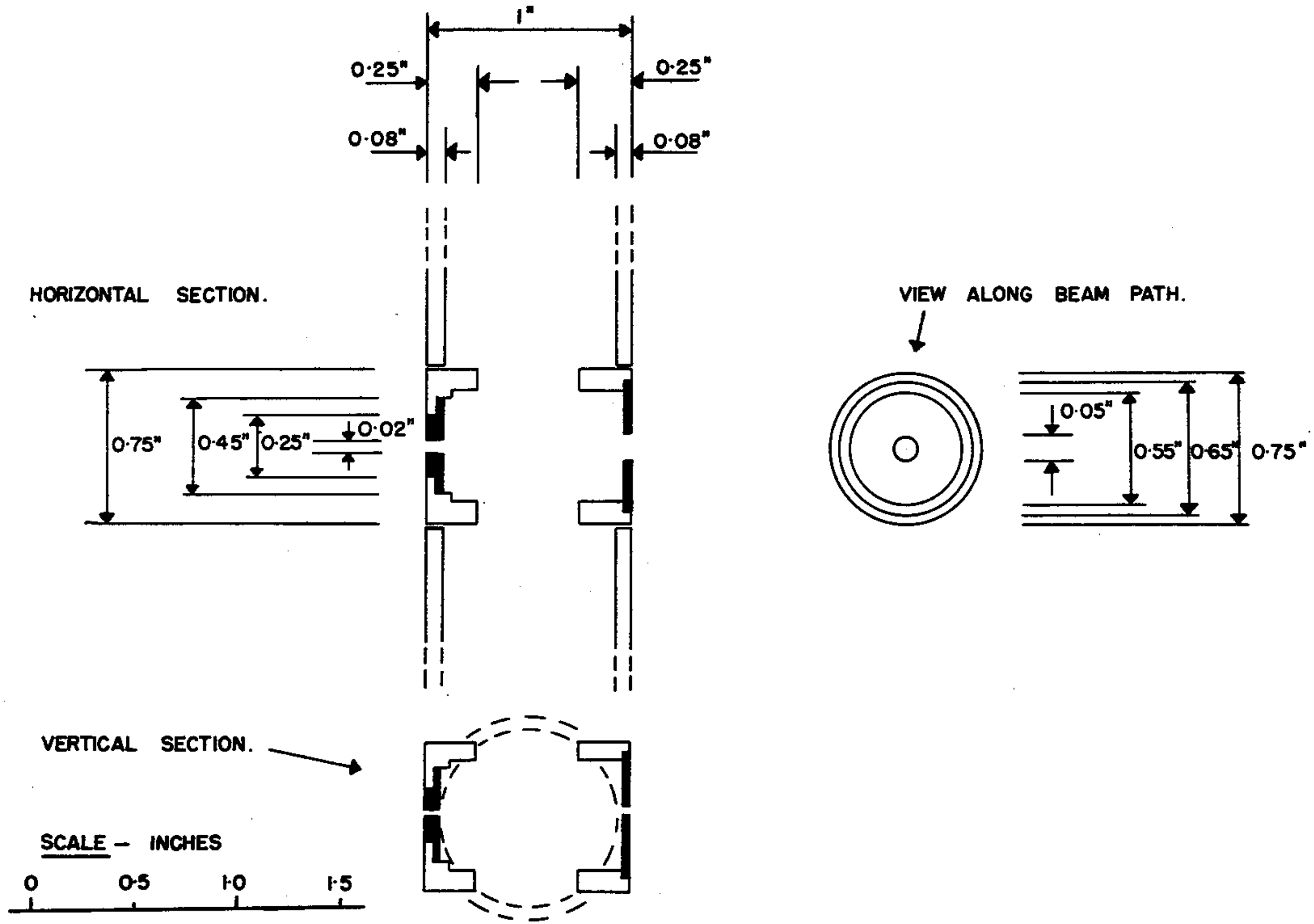

FIGURE 2(c) COLLISION CELL SEEN IN HORIZONTAL AND VERTICAL SECTIONS AND ALONG THE BEAM AXIS

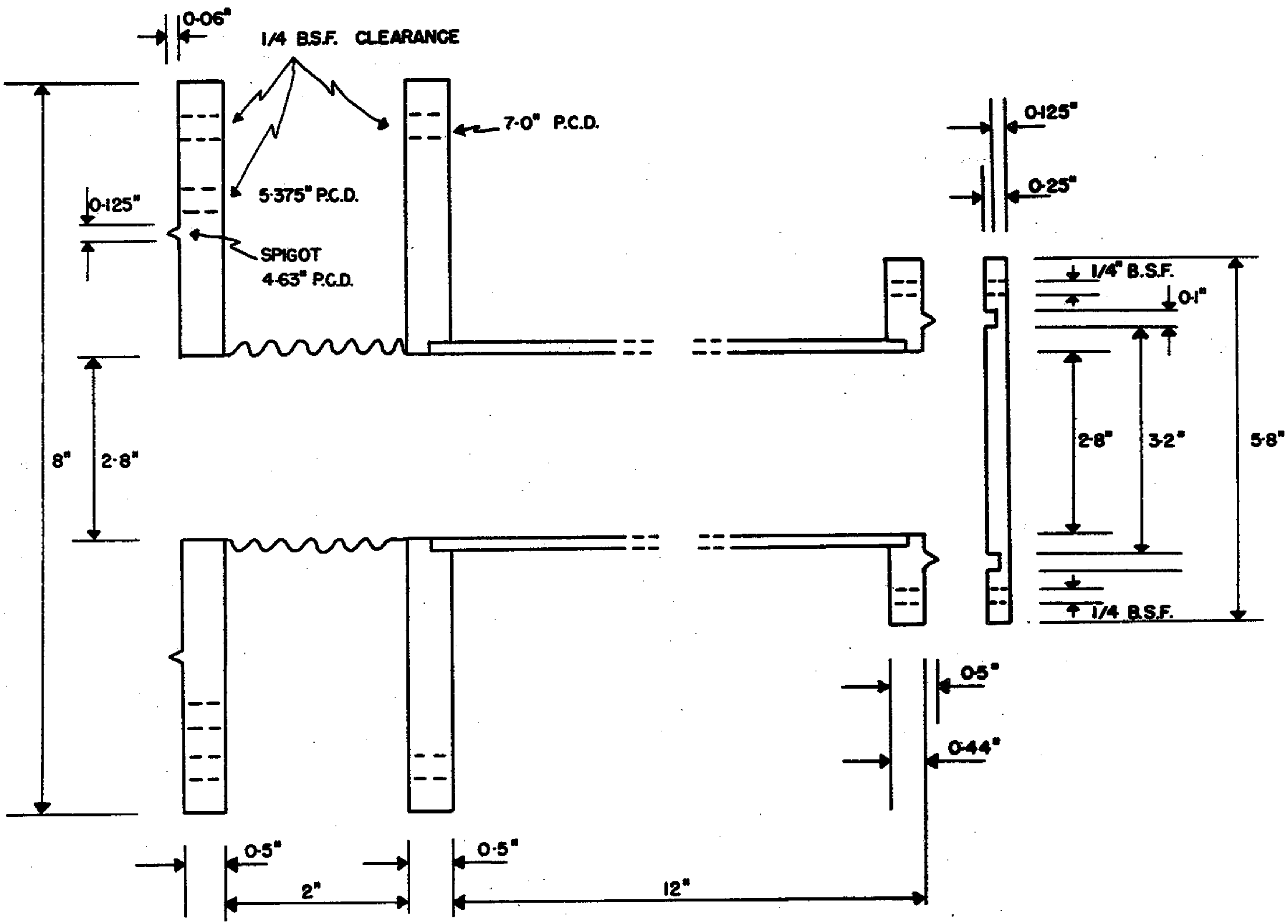

FIGURE 2(d) ANALYZER TUBE

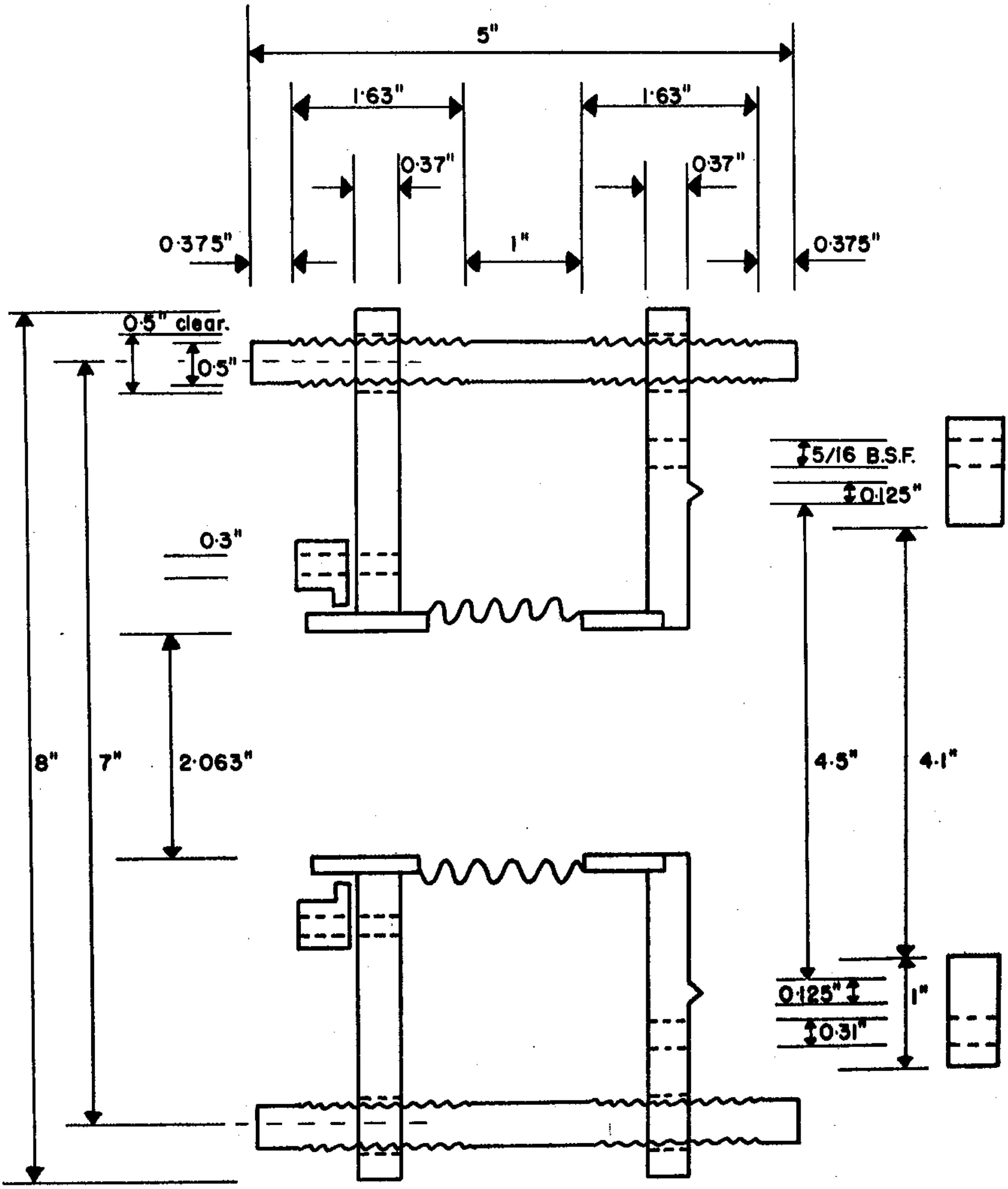

FIGURE 2(e) BEAM ALIGNMENT TUBE

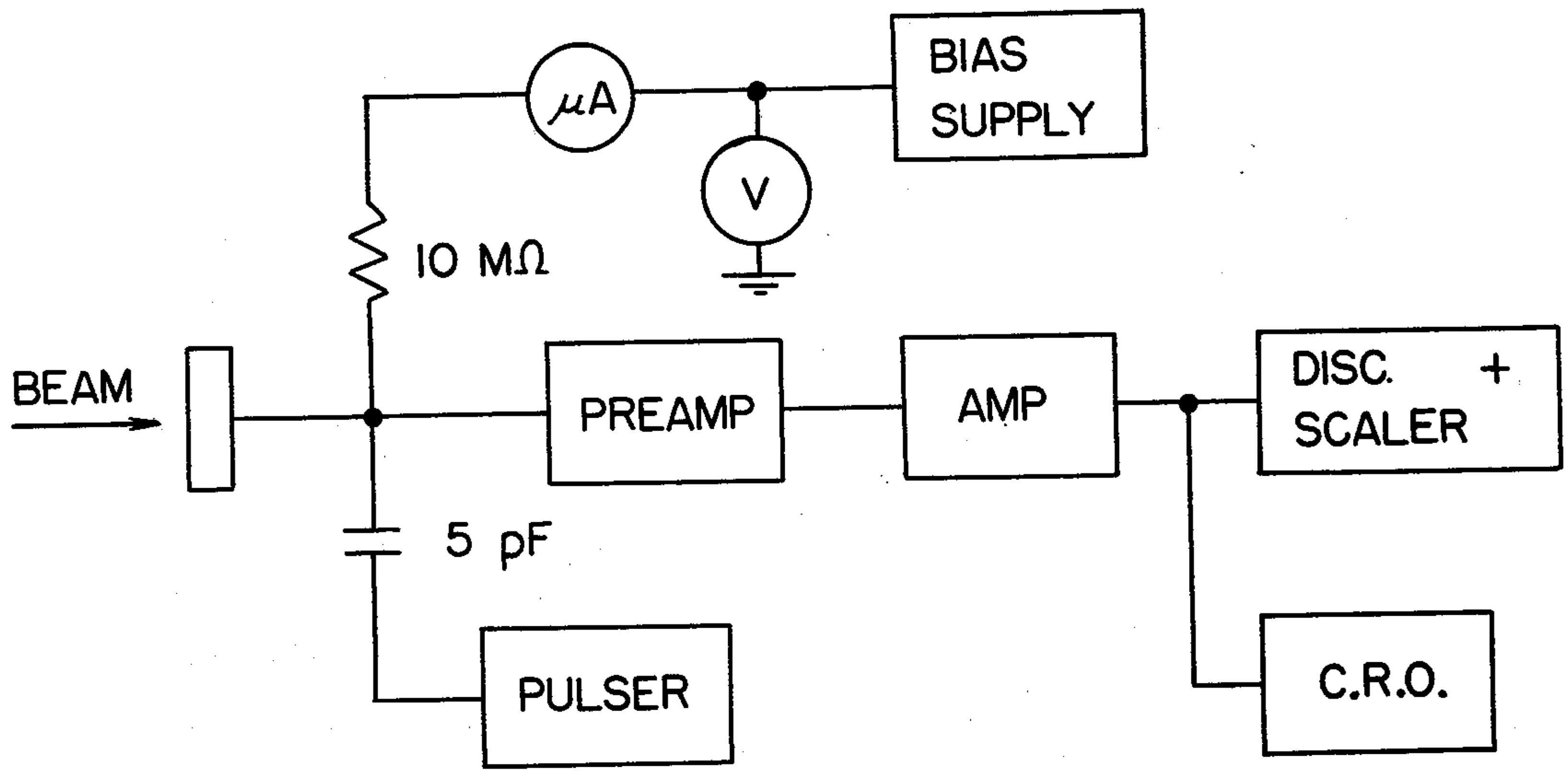

FIGURE 3. BASIC CIRCUIT FOR PARTICLE COUNTING

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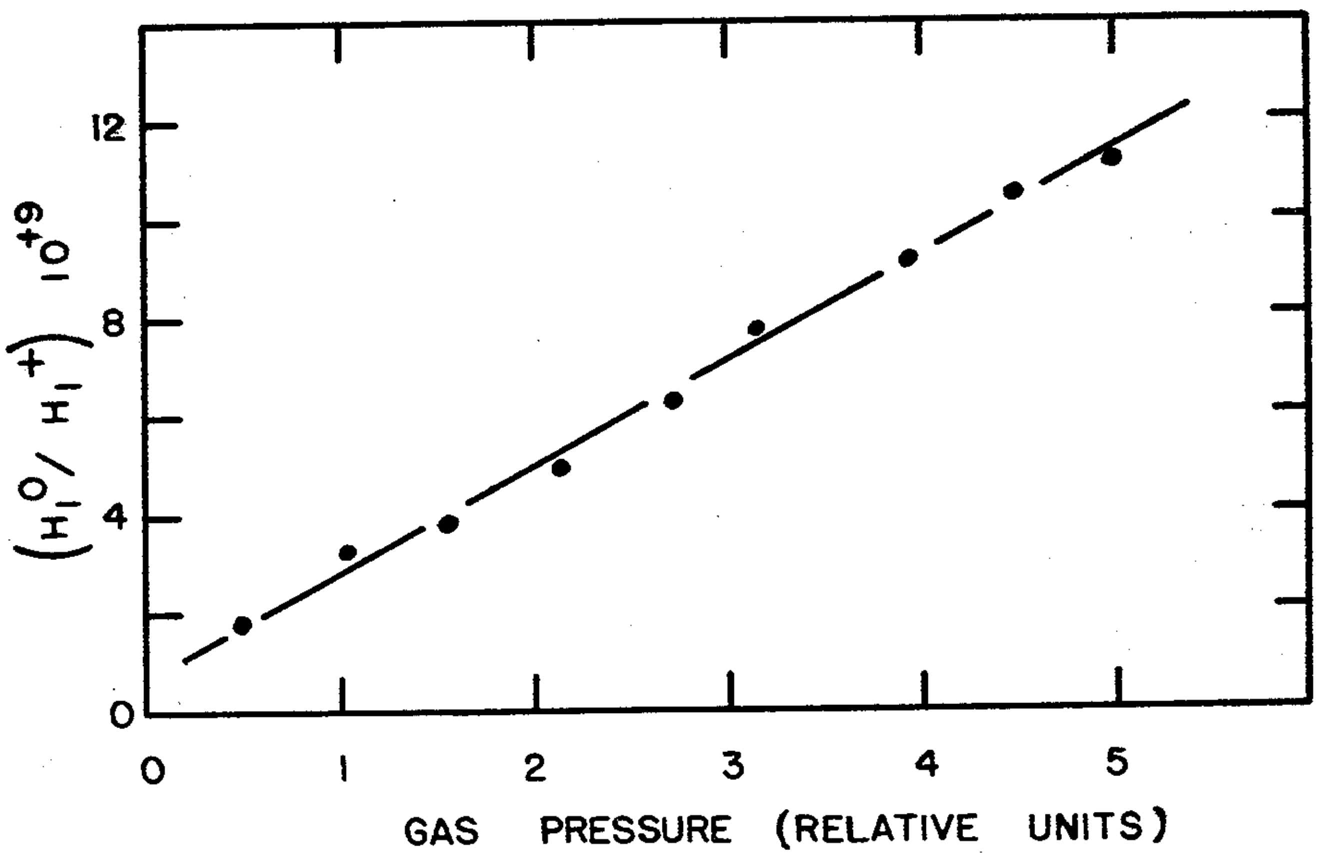

FIGURE 4. THE GROWTH OF H_1° ATOMS FROM A 0.8 MeV H_1^+ BEAM INCIDENT UPON A MOLECULAR HYDROGEN TARGET GAS

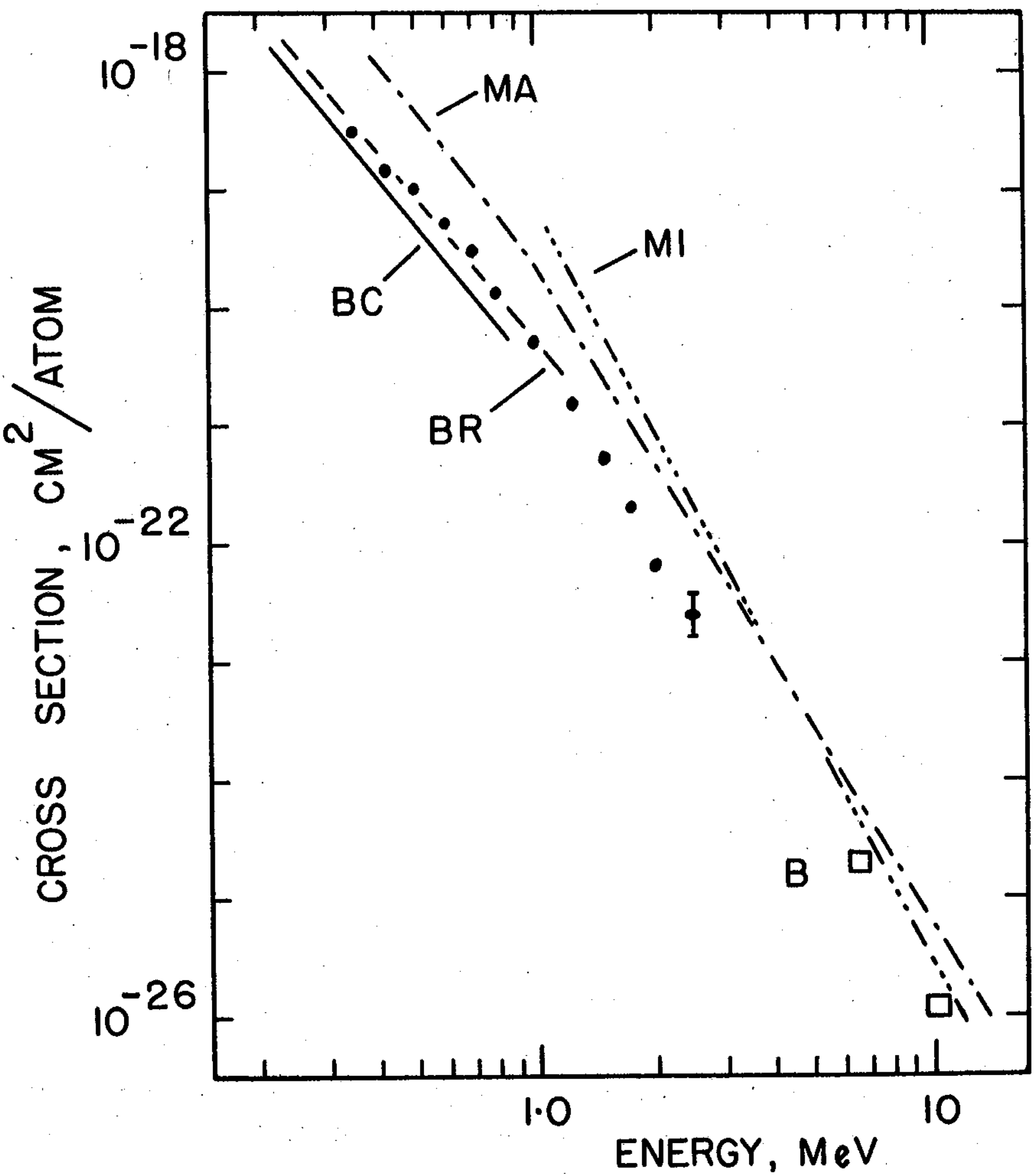

FIGURE 5. THE SINGLE ELECTRON CAPTURE CROSS SECTION σ_{10} FOR H_1^+ IONS INCIDENT UPON HELIUM GAS IN UNITS OF cm^2/He ATOM

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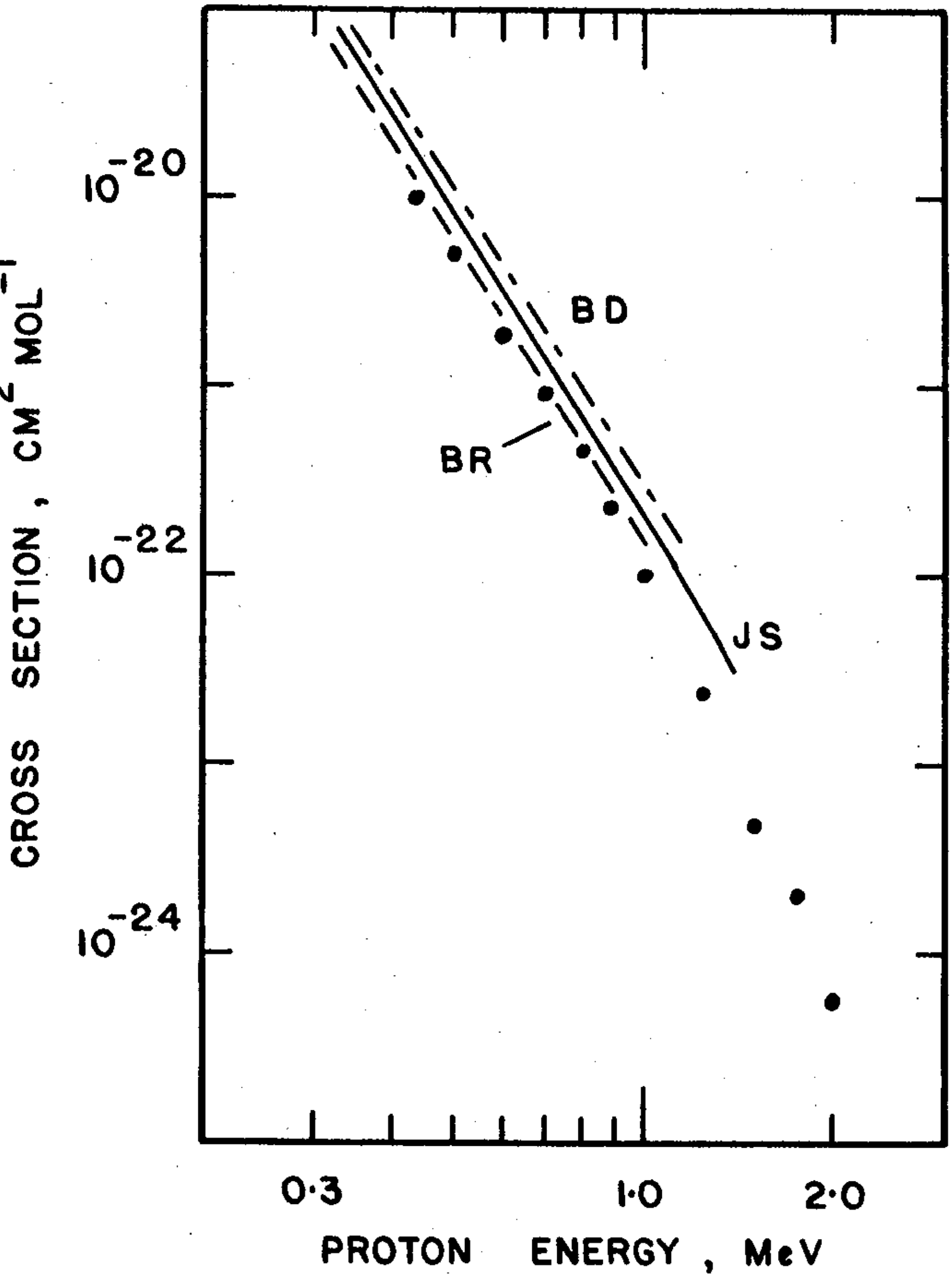

FIGURE 6. THE SINGLE ELECTRON CAPTURE CROSS SECTION σ_{10} FOR H_1^+ ATOMS INCIDENT UPON HYDROGEN GAS IN UNITS OF cm^2/HYDROGEN MOLECULE

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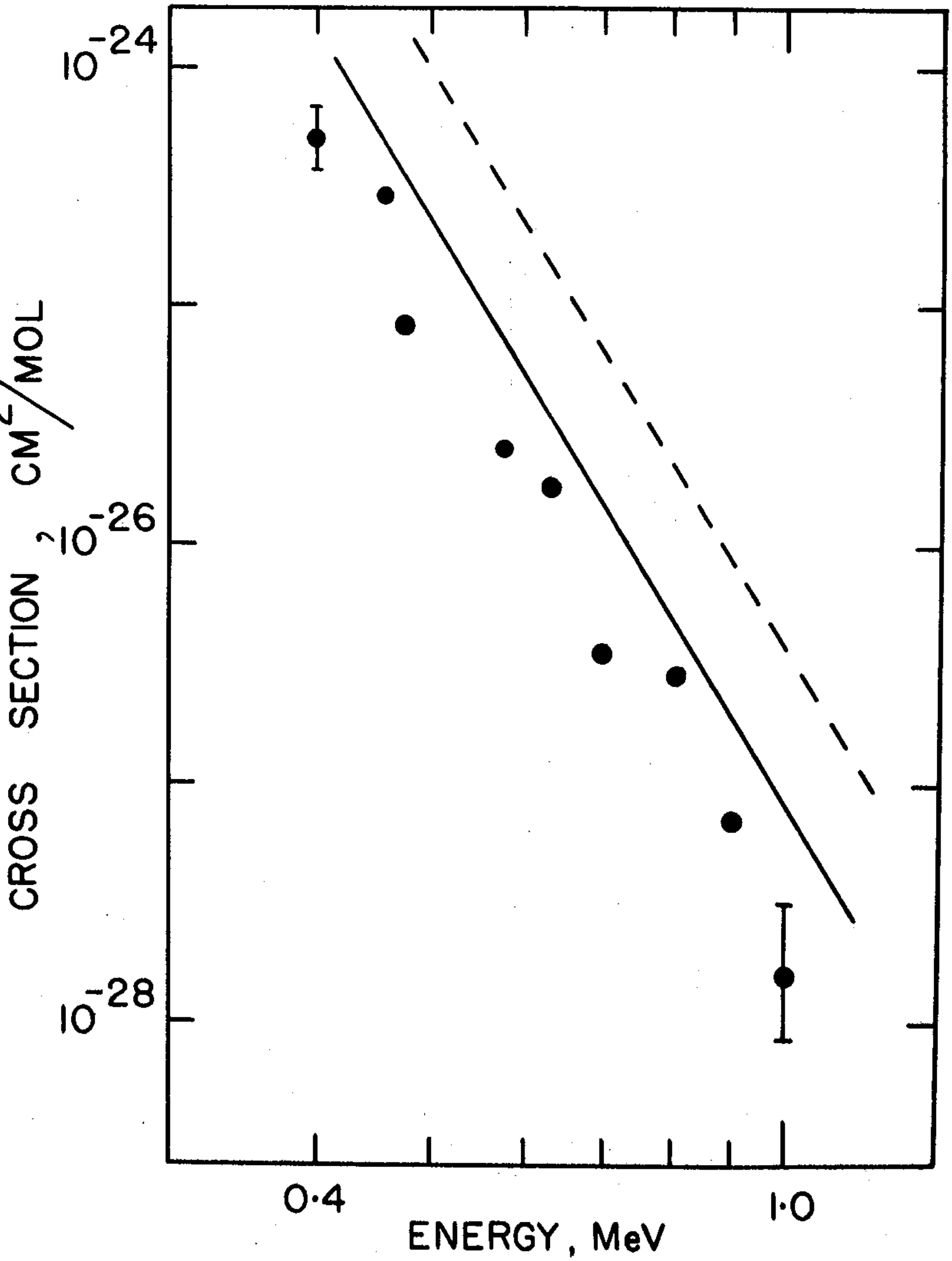

FIGURE 7. THE DOUBLE ELECTRON CAPTURE CROSS SECTION σ_{1-1} FOR PROTONS INCIDENT UPON HYDROGEN GAS IN UNITS OF cm^2/HYDROGEN MOLECULE

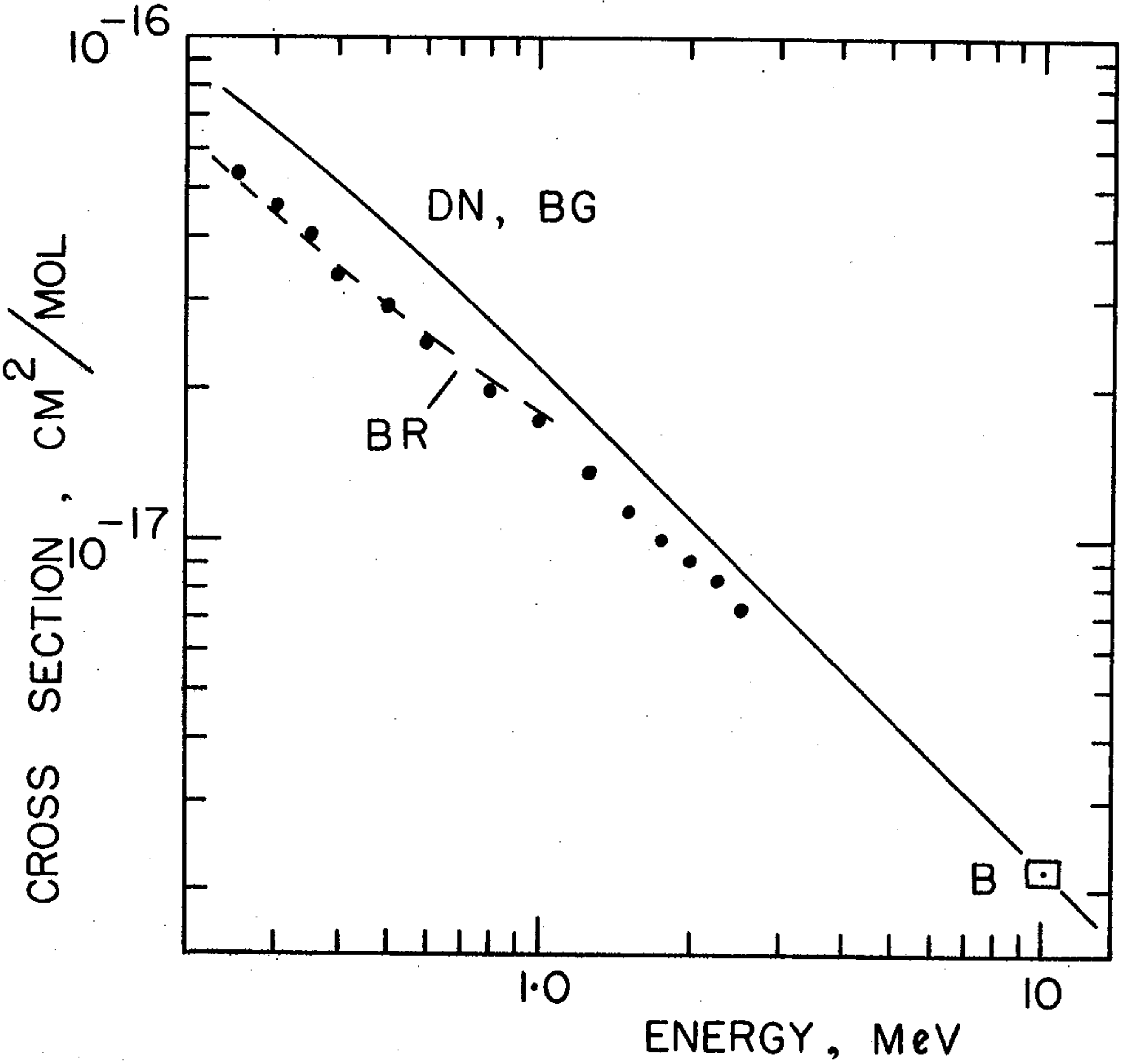

FIGURE 8. THE SINGLE ELECTRON LOSS CROSS SECTION σ_{01} FOR HYDROGEN ATOMS INCIDENT UPON A HYDROGEN GASEOUS TARGET IN UNITS OF cm^2/MOLECULE

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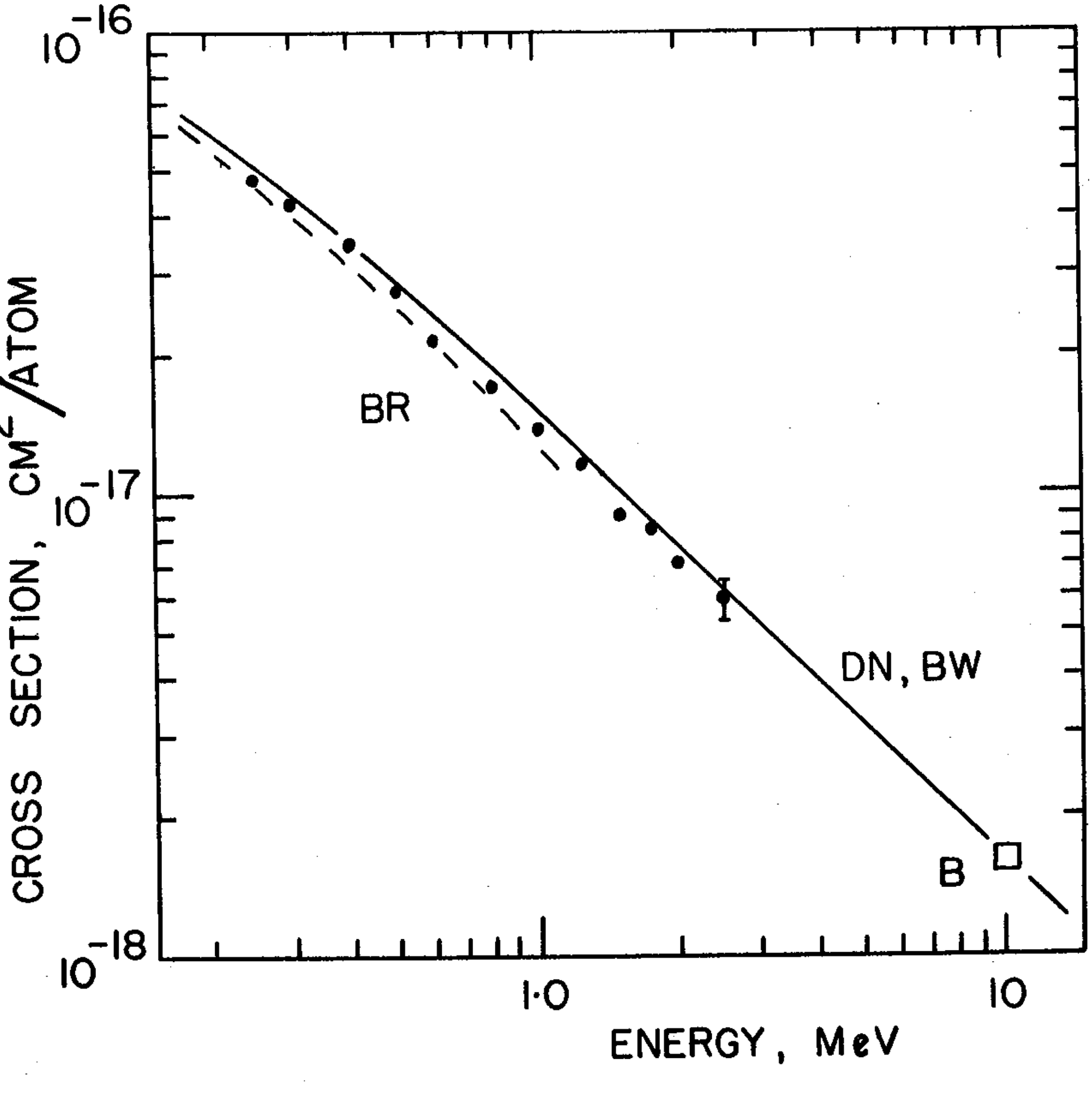

FIGURE 9. THE SINGLE ELECTRON LOSS CROSS SECTION σ_{01} FOR HYDROGEN ATOMS INCIDENT UPON A HELIUM TARGET IN UNITS OF cm^2/ATOM