Stripping in Collisions between Hydrogen Molecules below 500 eV*

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The cross sections for the production of forward scattered molecular ions in collisions of H_2 on H_2 and D_2 on D_2 have been measured at lab kinetic energies between 50 and 500 eV. The cross section for the H_2 reaction ranges from 2.2×10^{-17} cm^2 at 500 eV to 5.0×10^{-19} cm^2 at 50 eV. The D_2 cross section varies from 1.7×10^{-17} cm^2 to 3.8×10^{-19} cm^2 in the same energy range. The angular dependence of the ions was found to be highly peaked in the forward direction for energies above 100 eV. The cross sections for H_2 and D_2, when plotted versus incident velocity, are found to approach the same values at the highest velocities. The two cross sections diverge toward lower velocities as they approach their respective cutoffs, where each process becomes energetically impossible. These cross sections have been found to fit a theoretical model based on ideas first presented by Demkov and Komarov, in which ionization is presumed to take place via a series of pseudocrossings of the initial state into the continuum of the adiabatic states of the collision complex. For the interaction of two hydrogen molecules, the internuclear separation of each molecule is assumed fixed and the adiabatic states are defined in terms of the distance between the two molecular centers (R). Best agreement between the observed cross sections and those predicted from the model are obtained assuming the transition region to be centered around $R = 0.43$ Å, 16.5 eV above the energy of the ground state at large R. For the purpose of monitoring the neutral-beam intensity, the secondary-electron emission coefficients (γ) were measured for H_2 and D_2 impinging on a brass surface. The γ values were found to vary from 0.77 and 0.65 at 500 to 0.045 and 0.020 at 50 eV for H_2 and D_2, respectively.

I. INTRODUCTION

Ionization in collisions between neutral particles at low (<300-eV lab) energies has received increased interest because of its potential use in detecting the neutral outflux from magnetically confined plasmas. Neutrals formed by charge-exchange collisions between the trapped ions and thermal neutrals in the background gas leave the magnetic confinement system carrying information as to the ion distribution within the plasma. A fraction of these neutrals may be ionized by passage through a gas cell external to the plasma. The ions formed in these "stripping" reactions may then be analyzed as to energy and identity using conventional methods.

Until recently, the use of this method in fusion research has been limited primarily because the cross sections for hydrogen stripping reactions were unknown. In 1968, Fleischmann and Tuckfield[1] measured the efficiency for stripping atomic hydrogen in various gases (not including H_2) at projectile energies between 50 and 300 eV. Other experiments[2,3] with atomic hydrogen involved measuring the total negative charge (presumed to be electrons) produced in a collision chamber rather than detecting the fast ions. These measurements cannot be applied directly to the calibration of a neutral stripping cell owing to the uncertainties in both the fraction of the total charge corresponding to ionization of the atomic hydrogen projectiles and the fraction of those protons formed which would leave the exit aperture of a gas cell.

To date, the only data pertinent to the problem of detecting low-energy hydrogen molecules have been the cross sections for total negative-charge production in the $He + H_2$ reaction measured by Utterback.[4] Assuming that no negative ions are produced and that H_2 is ionized rather than He, Utterback's measurements may be used to estimate the efficiency for stripping H_2 in a helium-gas cell. These assumptions are not unrealistic, but two factors may limit the practical use of the $He + H_2$ data. The more important one is that the angular distribution of the H_2^+ ions is not known, causing an uncertainty in the fraction of the ions formed which leave the cell to be analyzed. The other problem is that the use of helium (or any other gas besides hydrogen) in a stripping cell may contaminate the source of hydrogen molecules being studied. Obviously, these difficulties could be avoided if the cross section for stripping H_2 in H_2 (as opposed to the cross section for total negative-charge production) were known. This process has previously been studied only for energies above 6 keV.[5]

This report describes such an investigation of stripping in collisions between hydrogen molecules at lab kinetic energies between 50 and 500 eV. An experiment has been performed to measure the cross sections for the production of molecular ions in the processes

$$\underline{H_2} + H_2 \rightarrow \underline{H_2}^+ + H_2 + e,$$

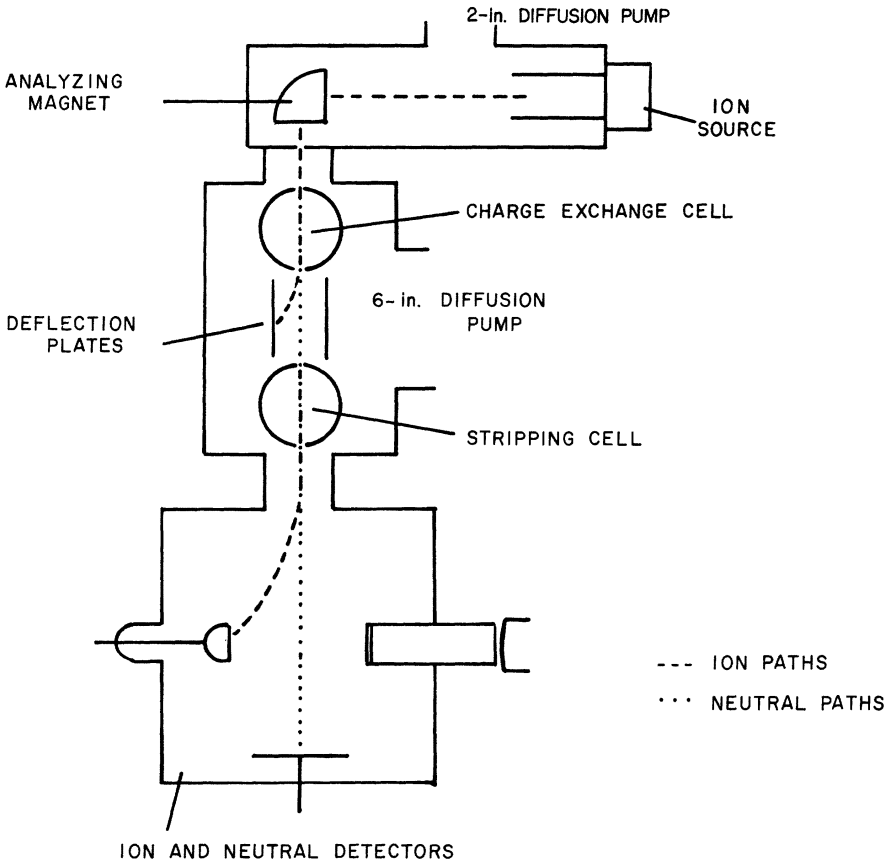

FIG. 1. Schematic diagram of experiment.

$$\underline{D}_2 + D_2 \rightarrow \underline{D}_2^+ + D_2 + e,$$

where the underlined particles are energetic. In addition to its possible application in fusion work, the deuterium reaction was also studied, since any relationship found between the cross sections for the two isotopes might be used to test the result obtained by Demkov and Komarov[6] which stands as the only general theoretical treatment of ionization in neutral-neutral collisions.

II. EXPERIMENT

The experimental arrangement is shown in Fig. 1. A beam of nearly monoenergetic H_2 (or D_2) molecules is produced by passing a mass analyzed H_2^+ (or D_2^+) ion beam through a cell containing the parent gas. Due to the high cross section for charge exchange, as many as 10% of the ions are converted to fast neutrals. The charged component of the beam leaving the charge-exchange cell (CXC) is removed as the beam passes through a transverse sweeping field, leaving a pure neutral beam. The neutrals then enter the stripping cell (SC) containing H_2 (or D_2) in which the reactions of interest take place. The fast ions formed in small-angle stripping reactions pass out of the cell's exit aperture and are deflected into a large-aperture ion detector where they are counted. The unstripped neutrals leaving the cell are monitored by a secondary-electron-emission detector placed on the beam axis a short distance behind the ion detector.

Three reactions producing fast ions in the stripping cell are possible:

$$\text{(i)}\quad H_2 + H_2 \rightarrow \underline{H}_2^+ + H_2 + e,$$

$$\text{(ii)}\quad H_2 + H_2 \rightarrow \underline{H}^+ + \underline{H} + H_2 + e,$$

$$\text{(iii)}\quad H_2 + H_2 \rightarrow \underline{H}^+ + \underline{H}^+ + H_2 + e.$$

Neglecting energy defects (i.e., if we consider the above reactions at kinetic energies much greater than that required to make them go), reaction (i) produces ions at the primary neutral beam energy, whereas ions produced in reactions (ii) and (iii) have one-half the primary energy. A simple retarding field energy analyzer placed near the exit aperture of the SC was used to determine the energy of the ions produced in stripping collisions at 500 eV. It was found that a positive bias of around 450 V was required to reduce the ion

signal at all, ruling out reactions (ii) and (iii) at 500 eV. Similar results were obtained at energies as low as 200 eV where focusing effects in the biased grid system limited its use. Due to their larger energy defects, reactions (ii) and (iii) are expected to be even less significant at energies below 200 eV.

The ion beam is produced in an ion source and lens system very similar in design to that of Utterback and Miller[7] and will not be further described. The H_2^+ ions were isolated from the primary ion beam by passage through a 90° sector magnet. The mass-2 component corresponded to an available current of around 10^{-10} A with an energy spread of a few electron volts and was directed into the input aperture of the CXC.

The charge-exchange cell was supported in the beam path by a tube through which the reaction gas was supplied. The tube passed through the vacuum wall, permitting alignment of the cell's axis along the beam direction from outside the main vacuum chamber. The cell pressure was measured using an ion gauge mounted on top of the tube. No pressure correction was required since at the low gas-flow rate used, the drop across the $\frac{7}{8}$-in. i.d. tube was negligible. Due to the $\frac{1}{16}$-in. input aperture and the $\frac{1}{8}$-in. exit aperture (both circular), the pressure in the main vacuum chamber could be maintained as 5×10^{-6} Torr at cell pressures up to 10^{-3} Torr. The cell measured 5.5 cm between apertures.

A major problem was to determine I_n, the equivalent neutral current produced in the CXC which ultimately passed through the stripping cell. The first attempt to determine I_n involved measuring the fast-ion current emerging from the CXC and then calculating the total neutral current produced using the known charge-exchange cross section.[8,9] The fraction of these neutrals which passed through the SC could be calculated knowing the shape of the beam and the size of the input aperture. This method proved to be very unsatisfactory because the direction and shape of the ion beam emerging from the CXC could not be determined accurately or consistently in the presence of the electric field needed to separate the neutrals and ions.

It was found that the most direct way to determine I_n was to measure the secondary-electron current released when the neutral beam impinged on a brass plate located after the γ stripping cell. Although the secondary-emission coefficient γ was expected to vary rapidly with energy, its variation over the range of energy spread of the neutral beam is negligible and thus I_s, the secondary-electron current, is related to I_n as follows:

$$I_n = I_s/\gamma. \tag{2.1}$$

Because the exit aperture of the stripping cell was made larger than the input aperture to allow for beam divergence, the equivalent neutral current striking the brass plate is equal to I_n.

At low energies γ has been measured for helium,[10] atomic hydrogen,[1] and molecular nitrogen[7] but γ for H_2 has been measured only at energies above 1 keV. For this reason, a separate experiment was performed to measure γ for molecular hydrogen and deuterium over the energy range 50–500 eV.

γ was determined by a simultaneous measurement of the slow-ion current produced in the charge-exchange cell (each slow ion corresponds to the production of a fast neutral) and the electron current released from the secondary-emission detector (SED). The SED plate was biased about 9 V negatively with respect to ground to ensure that none of the secondary electrons could recombine with the plate. Also, the stripping cell was removed during the γ measurement to allow all the fast neutrals to strike the SED. γ is given by $\gamma = I_{\mathrm{SED}}/I_{\mathrm{slow}}$, where I_{SED} is the positive current to the SED and I_{slow} is the current of slow ions produced in the charge-exchange cell.

I_{slow} was determined with an electrode system mounted within the charge-exchange cell as shown in Fig. 2. The slow ions were formed in the weak electric field caused by biasing the cylindrical cage positively (0 to 25 V) with respect to the rectangular ion-collection electrode which was referenced to ground through an electrometer. The cage and collection electrode were made of 90% transparency electromesh to reduce stray electron current due to fast ions and neutrals striking their surfaces. The total current to the collection grid I as a function of cell pressure P and cage bias V is given by

$$I(V,P) = I_{\mathrm{slow}}(V,P) + I_{\mathrm{scat}}(P) + I_{\mathrm{def}}(V),$$

where I_{scat} is the current of fast ions elastically scattered to the grid and I_{def} is the current of fast ions deflected to the grid by the bias voltage. Since I_{scat} is independent of the bias voltage and $I_{\mathrm{slow}} = I_{\mathrm{def}} = 0$ at zero bias, $I_{\mathrm{scat}}(P) = I(0,P)$. Assuming that I_{slow} and I_{scat} are negligible compared to $I_{\mathrm{def}}(V)$ with no gas leak (e.g., at the background pressure in the cell 10^{-5} Torr) yields $I_{\mathrm{def}}(V) = I(V,0)$. The total slow-ion current produced at a given pressure in the charge-exchange cell should be independent of V at voltages above that necessary to force all the slow ions to the collector grid. Indeed, I_{slow} was found to saturate at a bias voltage of about 5 V at all beam energies in the range investigated. An example of data taken with a beam energy of 200 eV and a cell pressure

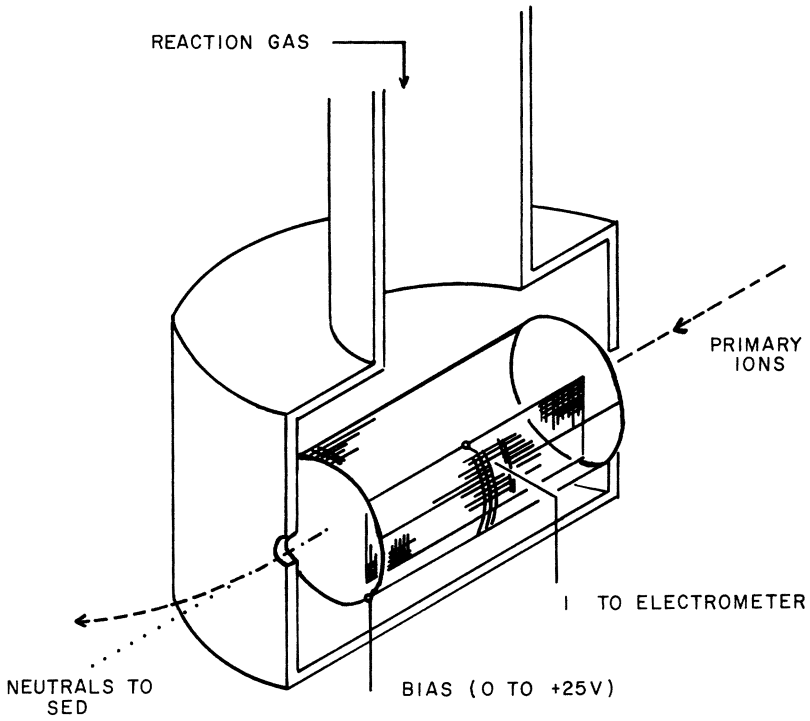

FIG. 2. Section view of charge-exchange cell showing electrodes used for γ measurement.

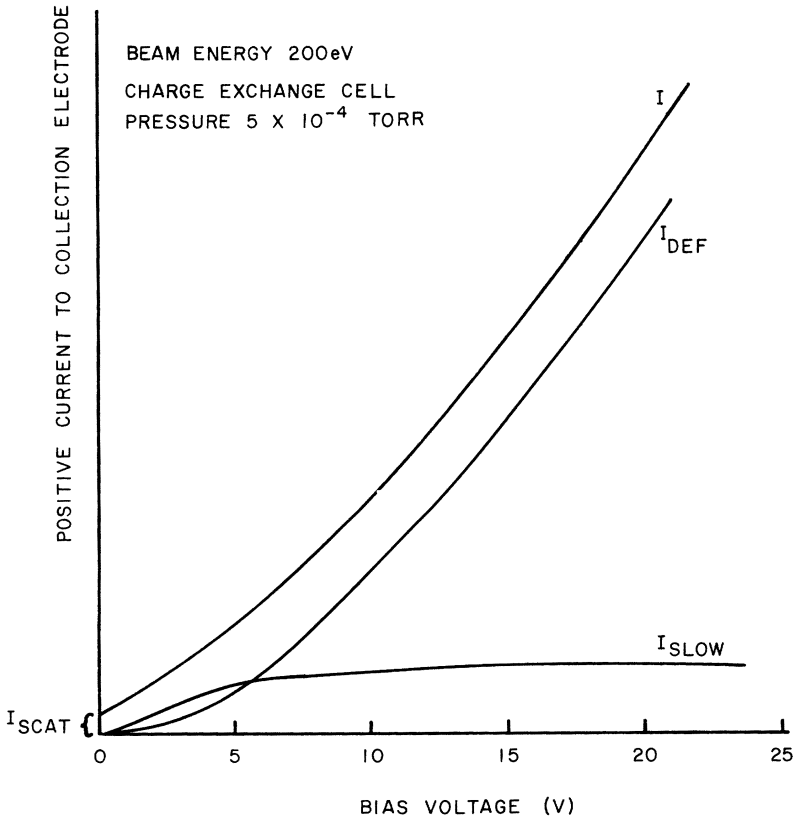

FIG. 3. Components of total current to collection electrode in charge-exchange cell vs cage bias.

of 1×10^{-4} Torr is shown in Fig. 3. γ is calculated from the relation $\gamma = I_{SED}/I_{slow}$ for H_2 and D_2 at energies between 50 and 500 eV and is presented later.

In the analysis described above to determine γ it was assumed that probability for charge exchange is at least as great as the probability for an ion or neutral to be elastically scattered at an angle large enough to prevent it from leaving the cell. Since the charge-exchange probability was about 0.02 under the conditions used for the γ measurement, the probability for a second-order process (charge exchange *and* scattering) would be 4×10^{-4} or less and could be ignored. This assumption was verified by observing the attenuation of the neutral beam through the stripping cell containing gas at the same density as used in the charge-exchange cell during the γ measurement. The scattering probability was observed to be roughly one-half the charge-exchange probability at all energies investigated.

It should be mentioned that there are three reactions possible which would produce fast ground-state neutrals in the charge-exchange cell. They are

(iv) $\underline{H}_2^+ + H_2 \rightarrow \underline{H}_2 + H_2^+$ (resonant charge exchange),

(v) $\underline{H}_2^+ + H_2 \rightarrow \underline{H}^+ + \underline{H} + H_2$ (simple dissociation),

(vi) $\underline{H}_2^+ + H_2 \rightarrow \underline{H} + \underline{H} + H_2^+$ (dissociative charge transfer).

Reaction (iv) is assumed to predominate. Reaction (v) is endothermic by 2.8 eV and produces both H and H^+ at one-half the primary ion energy. This reaction has been observed in this laboratory in the drift region between the ion source and the momentum analyzer. No accurate measurement of the cross section for this reaction was made but an upper limit of one-twentieth of the cross section for reaction (iv) at 200 eV was determined. This seems compatible with McClure's measurements above 1 keV.[5] Reaction (vi) is endothermic by 4.72 eV and should be less probable than reaction (v) at low energies. Therefore, the neutral beam is assumed to contain a maximum of 5% atomic hydrogen at one-half the molecular hydrogen energy.

The question of possible electronic excitation in the neutral beam can be easily dealt with. According to the Franck-Condon principle, excited electronic states of H_2^+ are not stable against dissociation.[11] Only excited vibrational and rotational states having energies up to a few electron volts above the ground state are possible in the primary ion beam. This limits the energy defects in charge-exchange reactions producing electronically excited neutral molecules to a minimum of about 10 eV. According to the Massey criterion,[12-14] the cross sections for such large energy-defect processes are quite low compared to that for near-resonant processes. Therefore, the beam will be assumed to be in the ground electronic state.

Owing to the difference in the nuclear potential minima of H_2^+ and H_2, electron ionization of H_2 may produce H_2^+ preferentially in the third or fourth vibrational quantum levels.[15] Simple electron transfer between the fast vibrationally excited ion and the ground-state neutrals in the charge-exchange cell could then produce fast neutrals in excited vibrational states with very little energy exchange. Because resonant (zero energy-defect) reactions are not generally preferred over reactions involving very small (0.1 eV) energy defects at collision energies above a few tens of electron volts,[16,17] the resonant process producing a ground-state neutral molecule is not necessarily favored. McClure[5] has suggested that the large nuclear separation of vibrationally excited H_2^+ would produce H_2 in high vibrational levels when undergoing charge transfer. Because details of the vibrational levels are not involved, these arguments apply to both H_2 and D_2. It therefore seems reasonable that excited vibrational states exist in the neutral beams used in this work.

The stripping cell differed in construction from the charge-exchange cell only in that its entrance and exit apertures have been enlarged to $\frac{1}{8}$ and $\frac{3}{16}$ in., respectively. The arrangements for gas leak, pressure measurement, and alignment were identical with those used with the CXC.

The ions produced in small-angle stripping reactions which passed through the SC exit aperture were counted by an ion detector constructed on designs developed by previous workers.[18-20] Positive ions entering the detector region are accelerated through 25 kV and impinge on a brass target (mounted 1 in. off axis to allow the free passage of neutrals to the SED behind the ion detector) where they release, on the average, four or five electrons each. These electrons are then accelerated through the same potential to a grounded scintillator, giving rise to a light signal detected by a RCA 4518 photomultiplier situated outside the vacuum system where it could be shielded from stray magnetic fields. The photomultiplier output pulses, examined with a 400-channel pulse-height analyzer, were found to form a broad peak centered around an amplitude corresponding to the release of about six electrons from the photocathode. This peak was superimposed on the usual

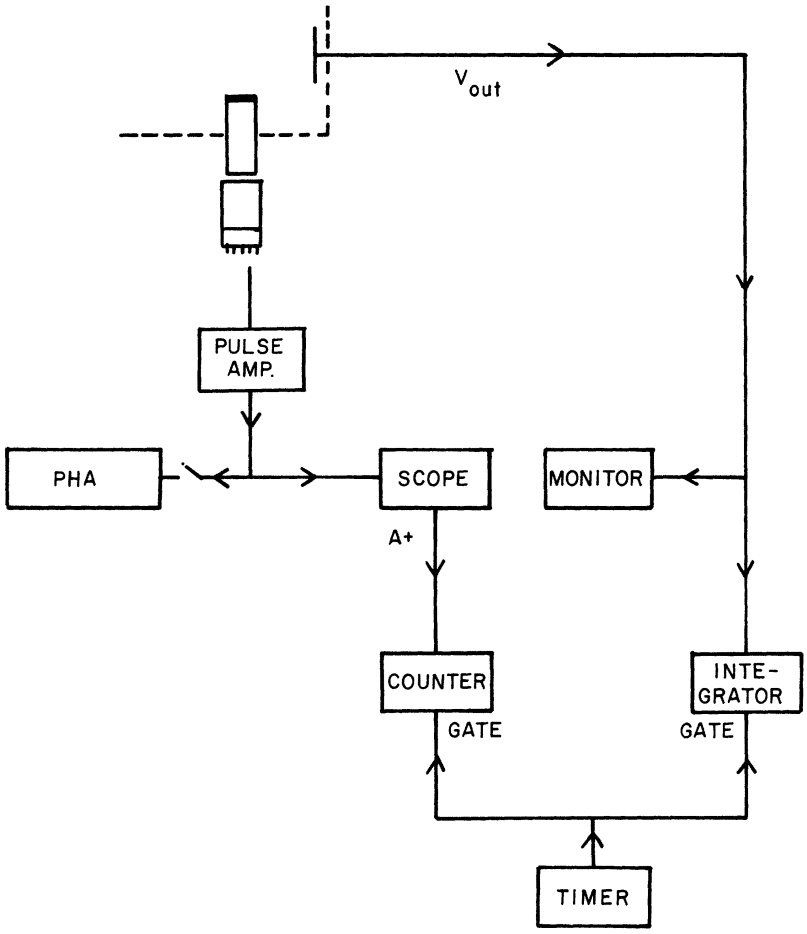

FIG. 4. Block diagram of data-gathering system.

Poisson noise distribution owing to thermally emitted electrons. Those pulses above an amplitude corresponding to just over five photoelectrons were counted, yielding a counting efficiency (ϵ) of 60% for H_2^+ and 55% for D_2^+ with a noise level of 40/sec. These efficiencies were found to be constant over the entire ion energy range under consideration. Ion rates smaller than 40/sec were not encountered in the stripping experiments and consequently, the detector was not improved by employing a lower noise (and more expensive) photomultiplier tube.

A block diagram illustrating the data-gathering system is shown in Fig. 4. The photomultiplier output, amplified by a factor of 50, was monitored with a Tektronix model 454 oscilloscope which was also used as a discriminator. Pulses from the A+ output of the scope, one for each pulse above the adjustable trigger level were counted. Both the counter and an integrator to measure the charge released from the SED were gated by a single timer unit. The stripping cross sections (σ) were calculated with the basic formula

$$\sigma = \frac{1}{\epsilon n l} \frac{I_i - \theta}{I_n}, \tag{2.2}$$

where I_i, θ, and I_n are the equivalent ion, noise, and neutral currents, respectively, each averaged over the 10-sec integration time. ϵ is the ion detection efficiency, n is the target gas density in the SC, and l is the SC length.

Equation (2.2) was corrected for stripping reactions in the background gas by taking measurements with and without gas in the SC. Gas effusing from the SC apertures increased the effective scattering length of the cell beyond its actual physical length of 5.5 cm. To determine the extent of the scattering region, a pair of electrostatic deflection plates was installed in the stripping cell so that ions formed inside the stripping cell proper could be removed from the total ion signal. A comparison of the stripping signals obtained with and without deflection voltage and target gas indicated that the ratio of the number of ions produced in the exit-aperture region to the number produced inside the cell was 0.15. The quantity l in Eq. (2.2) was therefore increased to 6.3 cm.

A rough measure of the angular distribution of the stripping product ions was obtained as follows. All ions created in the background or in the exit-aperture region at angles up to 15° from the beam axis are counted with maximum efficiency due to the large acceptance aperture of the ion detector. The collimating effect of the exit aperture allows the detection of only those ions formed at much smaller angles inside the stripping cell. A recent investigation of the $H + H_2$ stripping reaction[21] showed that the angular distribution of H^+ ions produced was highly peaked in the forward direction down to the lowest energy considered (500 eV). Therefore, for high energies at which ions are produced primarily in the forward direction, the ratio of the signal produced inside the cell to that from the background (assumed to be hydrogen) should be constant as a function of energy until, going towards lower energy, large-angle events become more probable and the resulting loss of signal from the stripping cell causes this ratio to decrease. This ratio was found to be constant for both isotopes down to around 100 eV indicating a very sharply peaked angular distribution in the forward direction. Below 100 eV, this ratio was found to decrease indicating a loss of one-half the ions formed at 50 eV. Several processes which might possibly come into play at low energies would appear to decrease this ratio, and thus an upper limit to the loss at 50 eV may be taken to be 50%.

Before presenting the results of this experiment in Sec. IV, a simple theoretical model which exhibits the basic features of stripping reactions will be presented in Sec. III.

III. THEORY OF DEMKOV AND KOMAROV AND THE ISOTOPE EFFECT IT CONTAINS

The Landau-Zener formula (LZ) has been used on various occasions to describe inelastic collisions for low nuclear velocities at which the reactions may be considered nearly adiabatic. The formula is applicable when the reaction of interest involves a single transition near the pseudocrossing of two adiabatic states of the quasimolecule

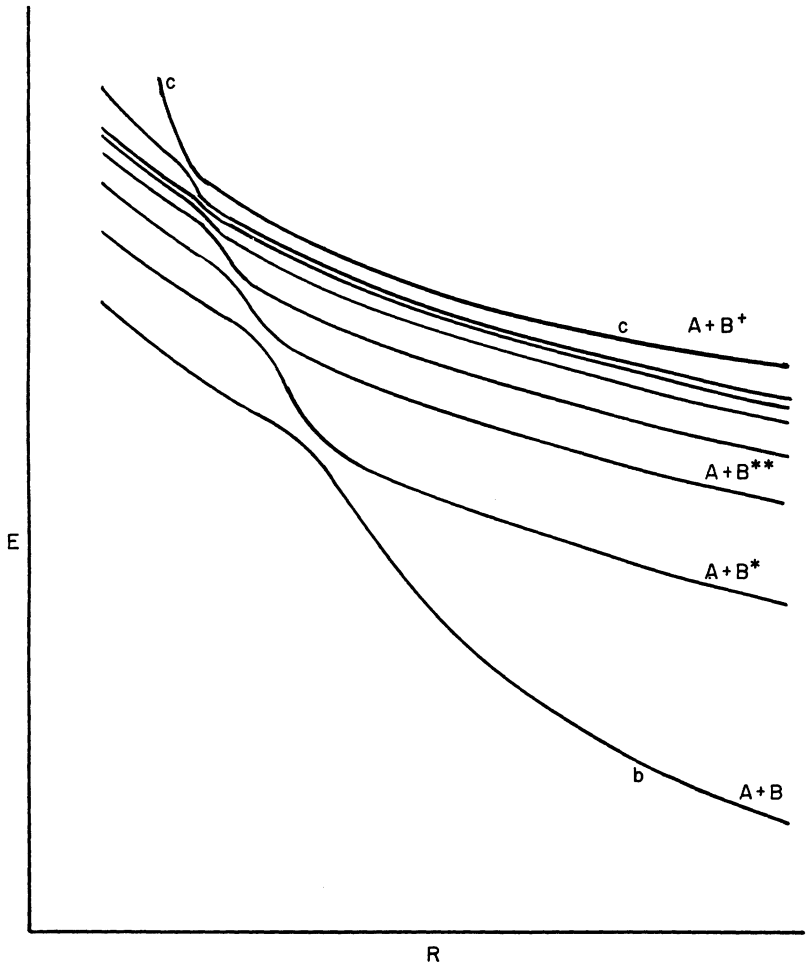

FIG. 5. Pseudocrossing of repulsive initial state (b) through the excited states (of same symmetry) into the continuum (c).

appropriate to the collision partners. However, ionization in the collision of two ground-state neutrals requires transitions up through the complete range of excited states into the continuum of the quasimolecule.

Demkov and Komarov[6] dealt with this case by assuming that an excited state (or the ground state) is highly repulsive and, to first order, pushes up through the higher excited states into the continuum, causing a series of pseudocrossings, through which ionization takes place. The pseudocrossing region (Fig. 5) is, for simplicity, taken to be centered at an energy U above the ground state at nuclear separation R_0. Applying the LZ formula to each pseudocrossing and utilizing the properties of the highly excited Rydberg states common to any molecule, the probability P for ionization was found to be

$$P = P_0 e^{-2LE/\hbar v}, \tag{3.1}$$

where the constant $2L$ is the interval of nuclear separation in the pseudocrossing region for which the energy levels of the highly excited states changes by one-half. E is the energy separation between level n and the continuum with n being that level above which all states may accurately be described as an electron in the screened Coulomb field of the nucleus. P_0 is the probability for the system to reach the level n and v is the nuclear radial velocity at R_0. Effectively then, Demkov and Komarov have shown that the contribution to the total ionization probability from the infinity of highly excited states does converge and retains the exponential dependence on inverse velocity as contained in the LZ formula.

If one now assumes that the initial state (presumed here to be the ground state) is responsible for the pseudocrossing series, the contribution from each of the finite number of pseudocrossings between states below n may be combined to calculate P_0 in Eq. (3.1). A straightforward application of the LZ formula yields

$$P_0 = 2e^{-\phi/v}, \tag{3.2}$$

where ϕ is a constant representing the combined effect of all states below n and therefore is dependent upon the exact nature of the colliding particles. The factor of 2 in (3.2) results from the fact that state n may be reached through either of its two adjacent states in the pseudocrossing region. Thus Eq. (3.1) may be written simply as

$$P = 2e^{-\Delta/v} \tag{3.3}$$

with

$$\Delta = \phi + 2LE/\hbar. \tag{3.4}$$

The cross section for ionization may now be calculated by integrating P, given in Eq. (3.3), over all impact parameters (ρ):

$$\sigma(T) = 2\pi \int_0^\infty P(v(T,\rho))\,\rho\, d\rho, \tag{3.5}$$

where T is the initial kinetic energy of relative motion in the c.m. frame. Using (3.3), Eq. (3.5) becomes

$$\sigma(T) = 4\pi \int_0^{\rho_{\max}} \rho\, e^{-\Delta/v(T,\rho)}\, d\rho, \tag{3.6}$$

where

$$v(T,\rho) = \{(2/\mu)T[(1-\rho^2/R_0^2) - U]\}^{1/2}. \tag{3.7}$$

$\rho_{\max}$ is that value of impact parameter for which $v = 0$ and μ is the reduced mass.

Performing the integration, Eq. (2.6) reduces to

$$\sigma(T) = 2\pi\Delta^2 R_0^2 (\mu/2T)[e^{-\eta}/\eta^2 - e^{-\eta}/\eta - E_i(-\eta)], \tag{3.8}$$

with η defined by

$$\eta = \Delta[(2/\mu)(T-U)]^{-1/2}, \tag{3.9}$$

where

$$E_i(-\eta) = -\int_\eta^\infty dx\, e^{-x}/x.$$

To examine the isotope effect contained in Eq. (3.8), we must compare the value of σ predicted for A on B with the value predicted for A' on B' at the same initial relative velocity v_i, where the primed particles differ from the unprimed particles in mass only. The factor $\mu/2T$ is simply I/v_i^2 and has been assumed constant in our comparison. The expression for η in Eq. (3.7) may be rewritten as

$$\eta = \Delta(v_i^2 - 2U/\mu), \tag{3.10}$$

where the same value of Δ and U is required for the description of either collision. Here, the mass dependence of η is clear and the resulting isotope effect may be easily handled using Eqs. (3.8) and (3.10). The isotope effect may be understood from another point of view by noting that Δ/η is the velocity in the pseudocrossing region for a head-on collision ($\rho = 0$). Equation (3.10) states that for equal incident velocities, colliding systems differing in reduced mass reach the pseudocrossing region with unequal velocities after surmounting the repulsive potential difference U. Because the transition probability depends exponentially on v, the isotope effect becomes considerable at c.m. kinetic energies near U (threshold).

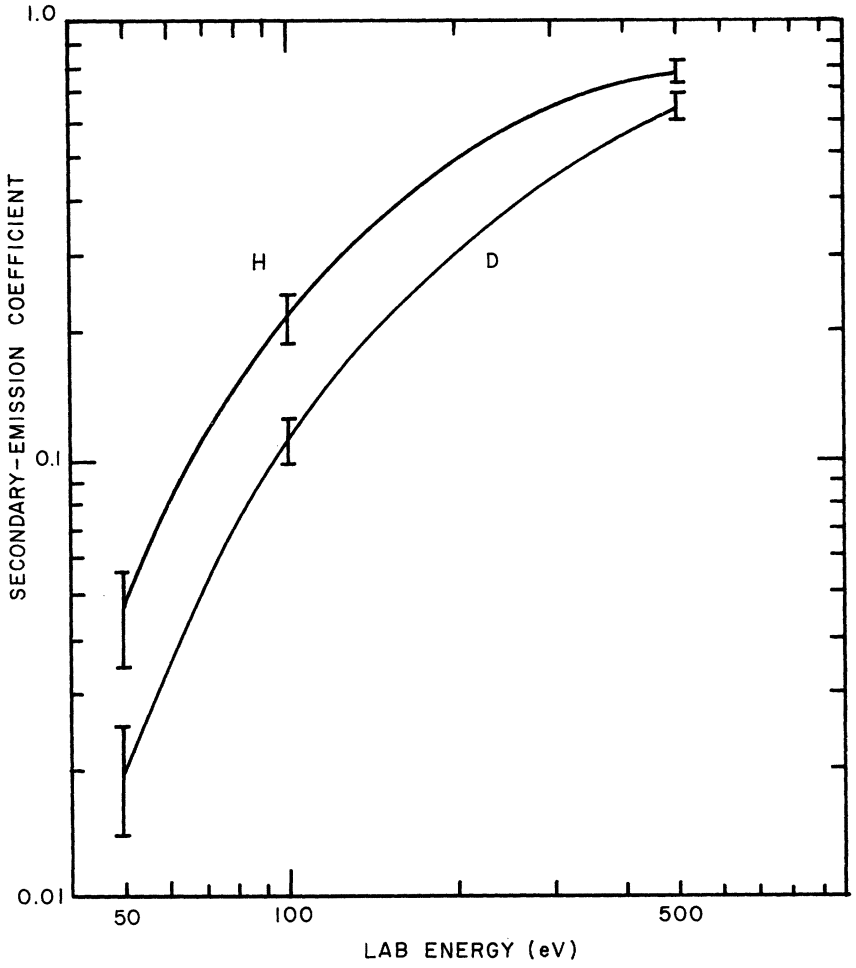

FIG. 6. Secondary-emission coefficient γ for H_2 and D_2 incident on brass vs lab energy.

IV. RESULTS AND ANALYSIS

A. Results

The secondary-emission coefficient γ, for H_2 and D_2 impinging on brass, required for the calibration of the neutral detector, is shown as a function of energy in Fig. 6. The error bars indicate one standard deviation on either side of the mean for a set of ten measurements at 500, 100, and 50 eV. A smooth line has been drawn to connect the mean values at each energy.

The cross sections for producing fast molecular ions via the processes

$$H_2 + H_2 \rightarrow H_2^+ + H_2 + e,$$

$$D_2 + D_2 \rightarrow D_2^+ + D_2 + e,$$

the basic experimental results of this investigation, were obtained by subtracting the background stripping contributions. The cross sections were measured at 10-eV intervals between 500 and 50 eV. Since no reproducible structure was found in the σ data, most data runs involved measurements at the selected energies of 500, 300, 200, 100, 75, and 50 eV. The cross sections are shown as functions of lab energy in Fig. 7, where the error bars represent the sum of the uncertainties in γ and that introduced by the scatter in the raw stripping data. The mean values of σ for both reactions at the selected energies given above are displayed in Table I. The uncertainty in the absolute magnitude of the cross sections arises from several sources (listed in Table II) and totals to about 70%.

The σ data of Fig. 7 have been replotted versus inverse velocity in Fig. 8 (solid lines) to facilitate the discussion in Sec. IV B. The dashed lines show the data with maximum correction for signal lost in large-angle events. The vertical separation between the solid and dashed curves indicates the maximum fraction of ions lost in the stripping cell. Later, arguments based on the theoretical stripping model in Sec. III will be used to show that the angular loss must be small and the solid curves in Fig. 8 are closest to the truth.

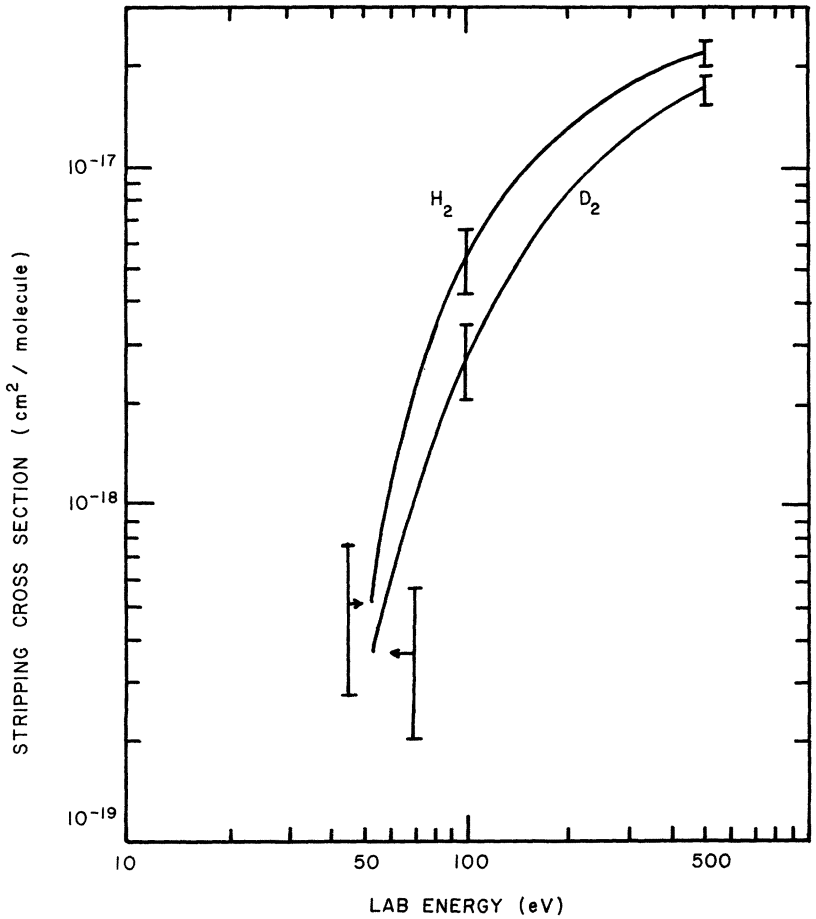

FIG. 7. Stripping cross sections for H_2 on H_2 and D_2 on D_2 vs lab energy.

Figure 9 shows the present H_2 cross-section data of Fig. 7 plotted along with values for the same reaction at energies between 6 and 100 keV, obtained by McClure.[5] No other measurements of the cross section for stripping molecular hydrogen are known.

B. Analysis of Results

First, the similarity in the shapes of the γ and σ curves (Figs. 6 and 7) is not surprising in light of the corresponding similarities in the energy dependences of γ and σ for secondary emission and stripping with positive-ion projectiles reported by Massey and Burhop.[22] The more-rapid fall-off of the σ curve relative to that of γ is expected owing to the larger energy defect in the stripping reaction. Further, the greater yield from H_2 relative to D_2 (at a given energy) in both processes is due to the higher velocity of the lighter isotope. Isotope effects in the stripping reactions will now be examined more fully with emphasis on the quantitative model developed in Sec. III.

TABLE I. Mean values of the molecular hydrogen stripping cross sections at selected energies.

Lab energy (eV)	$\sigma(H_2)$ ($\times 10^{-17}$ cm^2)	$\sigma(D_2)$ ($\times 10^{-17}$ cm^2)
500	2.2	1.7
300	1.9	1.3
200	1.4	0.87
100	0.57	0.27
75	0.28	0.13
50	0.050	0.038

Inverse velocity values of 18.5 and 25.5×10^{-7} sec cm^{-1} for H_2 and D_2, respectively, are the points at which the c.m. energy is just equal to the ionization potential of the hydrogen molecule (see Fig. 8). Therefore, at inverse velocities above these threshold points, the σ for the corresponding symmetric stripping reaction must be zero from energy considerations alone. It is reasonable to expect the cross sections for the two isotopes to be quite close at low inverse velocities (i.e., at c.m. energies well above the ionization threshold) and, as inverse velocity increases, the σ for H_2 to fall more rapidly, since its threshold is encountered first. Recalling that this same qualitative feature was shown to be present in the theoretical model developed in Sec. III, the model

TABLE II. Estimated uncertainties in the absolute values of quantities used for determining the stripping cross sections.

Quantity and symbol	Estimated uncertainty
Secondary-emission coefficient (γ)	30%
Stripping path length (l)	15%
Target gas density (n)	10%
Ion detection efficiency (ϵ)	10%

was examined quantitatively by choosing the parameters in the expression for the cross section so as to provide a fit with the observed values at selected energies. Equation (3.8) contains three undetermined parameters: R_0, U, and Δ. R_0, the nuclear separation at which the pseudocrossings occur, does not affect the velocity dependence of σ and will be used, after selecting U_0 and Δ, to scale σ to the proper magnitude. U is the energy of the pseudocrossing region relative to the initial state of the quasimolecule (assumed to be the ground state) and Δ is a constant related to the exact nature of the adiabatic wave functions for the system. Note that in the limits $V_i^2 \gg 2U_0/\mu$ and $V_i > \Delta$, the velocity dependence of σ is given by

$$\sigma \sim e^{-\Delta/v_i}, \tag{4.1}$$

indicating that a plot of $\ln\sigma$ vs $1/v_i$ should become linear for large v_i. This is often the case in inelastic heavy-particle collisions and explains the use of the inverse velocity variable in Fig. 8. The parameters Δ and U were chosen as follows: U was chosen arbitrarily and, by trial and error, Δ was chosen to yield the observed ratio

$$\sigma(H_2,\ 500\ \text{eV})/\sigma(H_2,\ 50\ \text{eV}) = 44. \tag{4.2}$$

This value of Δ was then used to compare the cal-

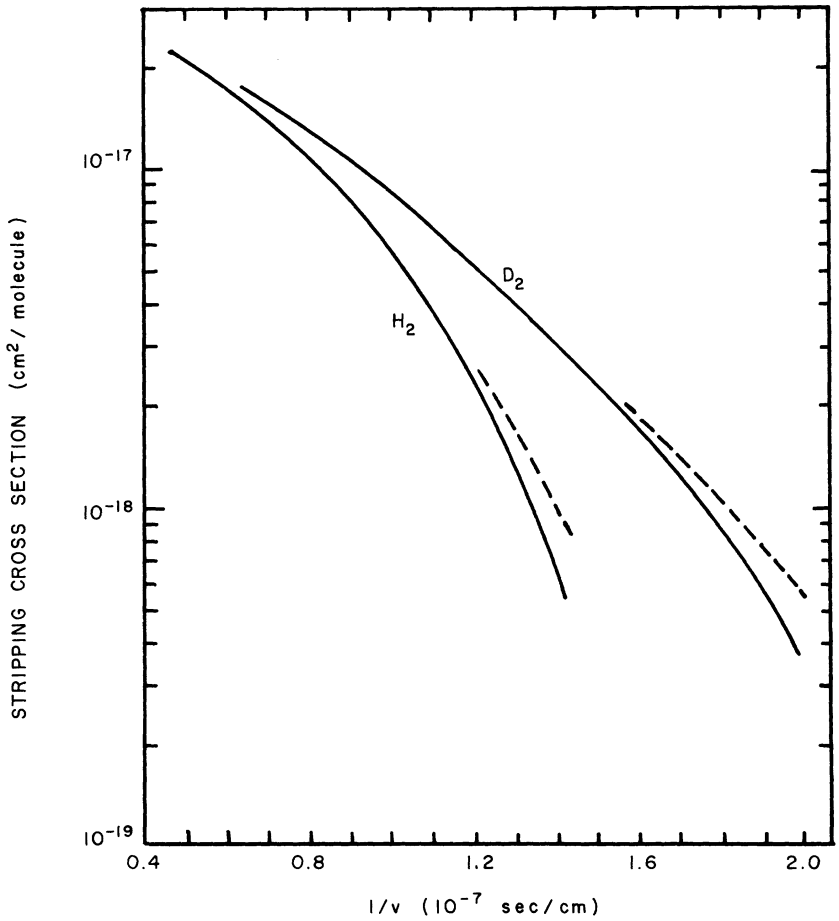

FIG. 8. Stripping cross sections for H_2 on H_2 and D_2 on D_2 vs inverse velocity.

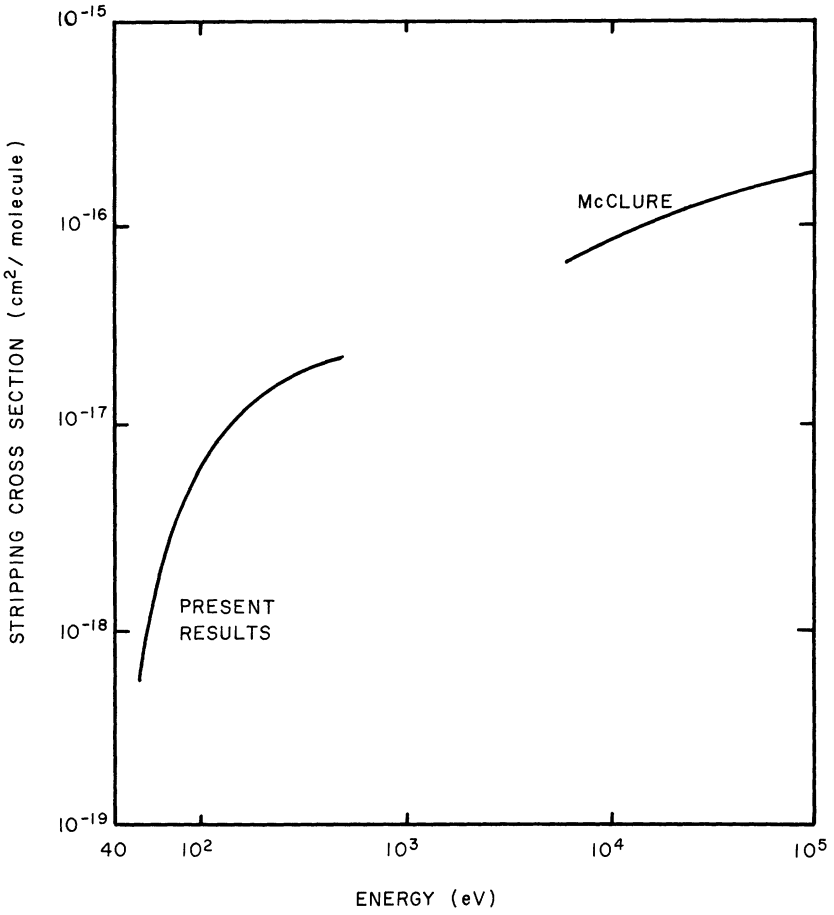

FIG. 9. Stripping cross section as a function of energy. Present results for H_2 on H_2 compared with those of McClure.

culated value to the following observed value of the ratio:

$$\sigma(D_2,\ 100\ \text{eV})/\sigma(H_2,\ 50\ \text{eV}) = 5.4\,. \qquad (4.3)$$

If in disagreement, a new value for U was selected and the process repeated. The proper ratios were obtained with parameter values of $U = 16.5$ eV and $\Delta = 1.10\times10^{7}$ cm sec.$^{-1}$ R_0 was then chosen so that σ coincided with twice the measured value of $\sigma(H_2,\ 500\ \text{eV})$. The factor of 2 is required because Eq. (3.8) predicts the total ionization cross section (target ionization plus stripping) which, due to symmetry, equals twice the stripping cross section. The experimental values of σ and the fit to Eq. (3.8) are shown in Fig. 10 where the fitting points are marked with arrows.

The above procedure was followed to fit the stripping data corrected for scattering losses (dashed curves, Fig. 8). The fit was obtained with the parameter values $\Delta = 1.01\times10^{7}$cm sec^{-1} and $U = 14.7$ eV. This fit compared with that for the uncorrected data above serves to show the sensitivity of the stripping model to the adjustable parameters.

It should be mentioned that to define properly the adiabatic states describing the interaction between two molecules, one must in general specify such parameters as the internuclear separation of each molecule, the angles defining their relative orientation, and finally, the distance between the two molecular centers (R). The use of the single parameter R, implies that the individual internuclear separations remain fixed during the collision and that the adiabatic states used represent averages over-all possible orientations.[23] It is not at all clear that this one-parameter description of the collision between two hydrogen molecules is justified at the separation (at crossover) of 0.43 Å suggested by the stripping model since this is less than the 0.76 Å spacing between the atoms in the isolated molecules. One would expect the need for more parameters in describing four hydrogen atoms in such close proximity. However, the general nature of the cross sections predicted by this simple model appear quite reasonable, which suggests that the model may be found useful for predicting isotope effects in other inelastic collisions.

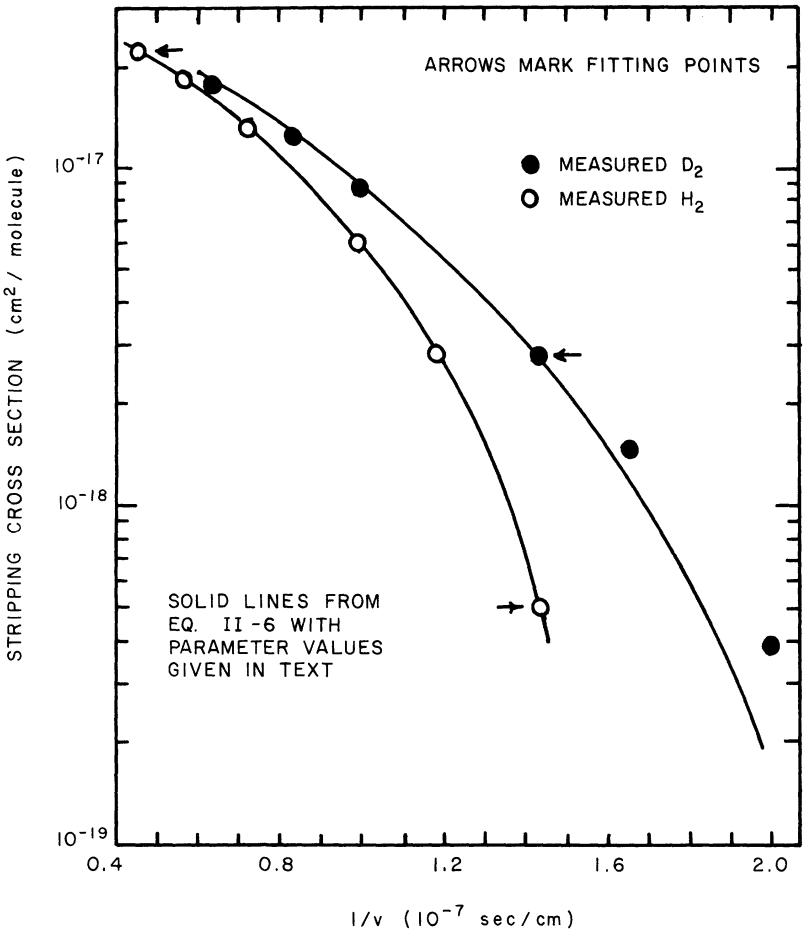

FIG. 10. Comparison of theoretical and experimental stripping cross sections for H_2 on H_2 and D_2 on D_2.

V. SUMMARY

The cross sections for stripping both the mass-2 and mass-4 isotopes of molecular hydrogen in their parent gases have been measured for projectile kinetic energies between 50 and 500 eV. The cross sections were measured by detecting the fast ions emerging from the collision chamber, a technique different from that used in all but one of the previous experiments dealing with ionization in neutral-neutral collisions at low energies. The single previous experiment using this technique involved no attempt to determine the magnitude of the signal lost due to large-angle reaction products. This effect has been given some attention in this work; the angular loss was found to be negligible at energies above 100 eV and within reasonable limits at the lowest energies. The results of this work therefore provide both the data necessary for the calibration of a molecular hydrogen neutral detector and values for the total stripping cross sections.

It is sometimes generally assumed that the cross sections for two collision processes differing only in the isotope identity of the reactants should be equal when scaled to the same relative velocity. However, a significant difference in the cross sections for stripping H_2 on H_2 and D_2 on D_2 at low relative velocities has been observed in this work. This isotope effect has been explained semiquantitatively using a model based on ideas first presented by Demkov. The theory involves an extension of the Landau-Zener formula to include transitions into the continuum of the adiabatic states of the collision complex. Regardless of the exact mechanism involved, the theory indicates that an isotope effect should occour at low velocities in any heavy-particle collision process having a large energy defect.

Although it has not been recognized in the literature, the velocity dependence of many of the known cross sections for ionization in low-energy collisions between neutrals closely follows that predicted by Demkov at the highest velocities in the adiabatic region. However, at low velocities near threshold these cross sections are found to drop below the values indicated in Demkov's theory. The isotope effect is another feature absent in

Demkov's results. In addition to containing an isotope effect, the model presented in this work accounts for the behavior of the ionization cross section near threshold and blends smoothly into agreement with the simple exponential dependence on inverse velocity predicted by Demkov at the higher velocities. One is therefore led to conclude that the theory presented in Sec. III may prove to be a valuable tool for the study of inelastic collisions at low energies.

In the course of this work the secondary-electron-emission coefficients have been measured for H_2 and D_2 impinging on a brass surface with kinetic energies between 50 and 500 eV. These results may be applied directly to determine the intensity of a ground-state molecular hydrogen beam provided it has a known energy distribution. These measurements should greatly simplify future experiments with molecular hydrogen beams.

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