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PHYSICAL REVIEW A VOLUME 7, NUMBER 5 MAY 1973

Electron Emission in H_2^+-H_2 Collisions from 0.6 to 1.5 MeV*

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Cross sections, differential in emission energy and angle, have been measured for the ejection of electrons in collisions of H_2^+ ions with molecular hydrogen. Incident-ion energies studied were 0.6, 1.0, and 1.5 MeV and electron emission was measured over the laboratory angular range of 20°–125°. Electron-emission cross sections are compared with measurements for incident protons having the same velocity as the H_2^+ ions. Electron distributions attributable to dissociative ionization of the H_2^+ ion are integrated to obtain the total cross section for dissociative ionization. A comparison is made between measured differential cross sections for scattering the bound electron of the incident ion and cross sections for scattering of incident free electrons.

I. INTRODUCTION

Experimental electron-emission cross sections, differential in ejected electron energy and angle, provide sensitive tests of the reliability and limitations of theoretical treatments of the ionization process. Cross sections for electron emission by protons have been measured for proton energies from 0.05 to 2.0 MeV for a wide range of gas targets.[1–7] These measured cross sections, when compared with theoretical predictions, have provided information concerning the relative importance of various interactions which enter into a complete description of ionization by fast charged particles.[8–12] Very few measurements of doubly differential electron-emission cross sections have been made, however, for incident particles which possess an electronic or molecular structure in themselves. Cacak[13] has measured electron distributions of Ne-Ne and Ar-Ar collisions for ion energies of 50–300 keV and autoionization Auger electron emission has been studied for Ar, Ne, He, and H_2^+ impact.[14–16] To our knowledge, the autoionization studies with H_2^+ impact are the only published measurements of electron distributions resulting from an incident molecular ion.[14]

Electron distributions from molecular-ion impact can be used to obtain information regarding collisional dissociation of the incident ion as well as ionization of the target. When fast molecular-hydrogen ions (H_2^+) collide with a target molecule any of several modes of dissociation of the incident ion are possible:

$$H_2^+ \rightarrow H + H^+, \tag{1}$$

$$H_2^+ \rightarrow H^+ + H^+ + e, \tag{2}$$

$$H_2^+ + e \rightarrow H + H, \tag{3}$$

and each reaction may be accompanied by excitation and/or ionization of the target atom or molecule.

McClure and Peek have recently critically reviewed the extensive published work on dissociative collisions of heavy particles, especially H_2^+

dissociation.[17]

All previous experiments on H_2^+ dissociation have involved detecting and analyzing the heavy fragments resulting from the collision. The present work reports the measurement of cross sections for electron emission for molecular-hydrogen ions colliding with a molecular-hydrogen gas target. Incident-ion energies ranged from 0.6 to 1.5 MeV and electrons were detected at angles from 20° to 125°. A comparison of these electron spectra with similar spectra obtained from proton impact was made and we attributed the observed differences to the electrons lost from the H_2^+ as a consequence of collisional ionization [reaction (2)]. These electrons are emitted preferentially in the forward direction ($\theta_{lab} \leq 90°$) and with a velocity near that of the incident ion. By integrating with respect to emission angle, the energy distributions for electrons from collisional ionization of H_2^+ are obtained and integration over electron energy then provides a total electron yield which is compared with measurements of the cross section for reaction (2). Angular distributions of electrons resulting from dissociative ionization are compared with measurements of elastic scattering for incident electrons velocity-equivalent to the incident molecular-hydrogen ions of the present work.

II. INSTRUMENTATION AND MEASUREMENT TECHNIQUE

A detailed description of the measurement technique and errors involved has been presented in an earlier publication.[4] The experiment consists of passing a collimated ion beam through a differentially pumped gas target cell. Electrons ejected from the interaction region exit through a slit in the target cell, are energy analyzed by a cylindrical-mirror electrostatic analyzer with an energy resolution of 3.5%, and are detected by a continuous-channel electron multiplier. Magnetic fields in the region of the target cell are reduced to a few milligauss by three mutually perpendicular sets of Helmholtz coils.

The ion-beam source was a 2-MV Van de Graaff employing a conventional rf discharge to produce the ions from hydrogen gas. It is well known that this type of source produces H_2^+ ions in a distribution of vibrational states[18,19] and that the initial vibrational-state distribution can have a substantial effect on the observed dissociation cross sections of low-energy H_2^+ ions.[19,20] However, Barnett and Ray[18] observed that there was no change in the observed cross sections with variation of ion source parameters for high ion-beam energies ($40 < E < 200$ keV). Also, Williams and Dunbar observed that variations in ion-source gas pressure and magnetic field around the rf source produced only a small change in the observed dissociation cross sections at 50-keV ion energy.[19] No systematic study was made of the effect of source parameter variations on the observed electron distributions reported here; however, it was observed that routine adjustments of the ion source and beam focusing elements during data runs produced no apparent abnormalities in the spectra. We have assumed that the relative insensitivity of dissociation cross sections to ion-source operating parameters observed for the range 50–200 keV will hold also for the higher ion energies covered in the present work.

Cross sections for electron emission are derived from measurement of the incident-ion beam intensity, electron countrate, solid angle of acceptance and energy resolution of the electron energy analyzer, electron detection efficiency, target gas density, and path length of the ion beam through the target gas. A small correction is applied to account for the small fraction of low-energy electrons which are scattered by the target gas before they can be detected. Absorption coefficients of Normand[21] are used for this purpose. Propagation of the estimated uncertainties in the quantities which are used to determine the absolute cross section results in a total uncertainty of ± 20% in the absolute cross sections reported in this paper. Experience has shown the precision of the data to be better than about ± 10%.

III. RESULTS AND DISCUSSION

Cross sections, differential in ejected-electron energy and angle, are shown in Figs. 1, 2, and 3 for incident H_2^+ ion energies of 0.6, 1.0, and 1.5 MeV, respectively.[22] These cross sections are characterized by three main features (a) large cross section for ejection of low-energy electrons, (b) a broad high-energy peak which is most prominent for small emission angles and fast incident ions and is attributed to direct interactions between the incident ion and an electron bound in the target molecule, and (c) a broad peak at an electron energy which corresponds to an electron having nearly the same velocity as the incident molecular ion. The first two features are also characteristic of spectra resulting from proton impact.[1–7] We associate the third feature with electrons which are released in the ionization of the incident molecular ion, i.e., the electrons resulting from reaction (2), and we will refer to this feature as the dissociative ionization electron distribution. The energy of the maximum of the dissociative ionization electron distribution is less than the energy of the velocity-equivalent electron by 20–25 eV. The energy necessary for a Franck–Condon transition from the ground vibrational state of H_2^+ to the dissociative ionized state $H^+ + H^+ + e^-$ is 27–29 eV.[23] The difference between the observed

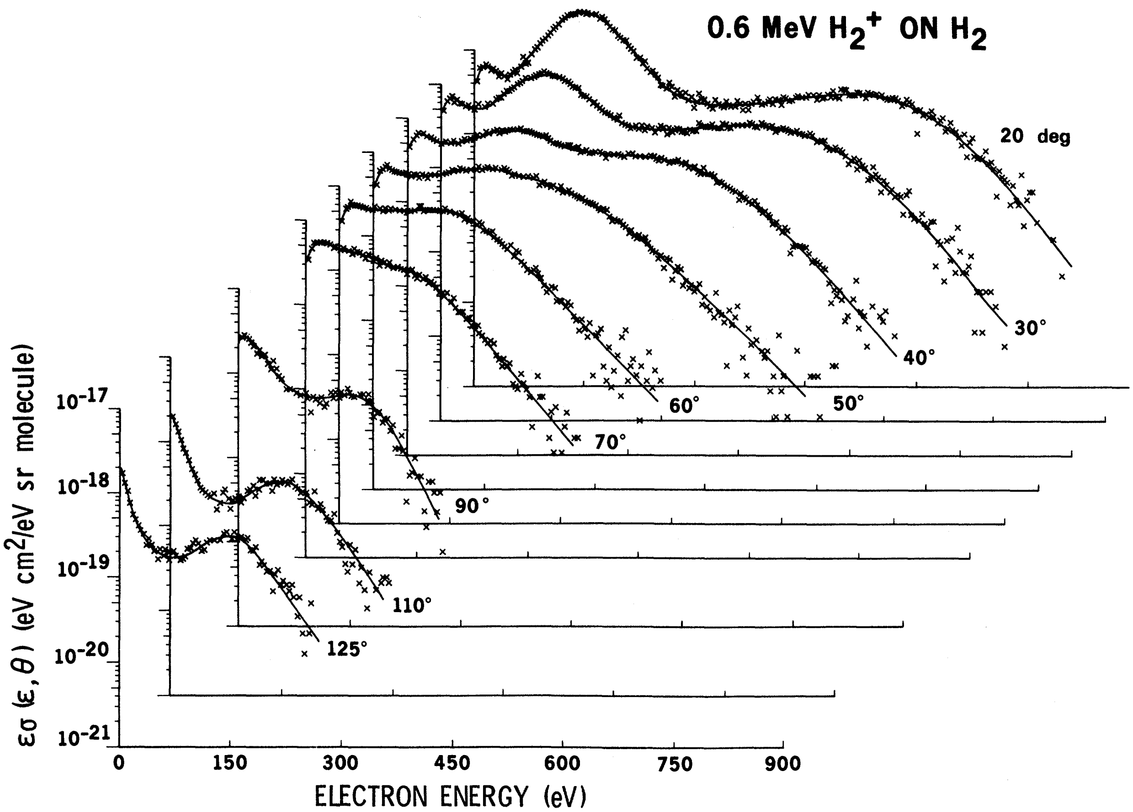

FIG. 1. Cross sections, differential in electron energy and emission angle, for electron emission following collisions of 0.6-MeV H_2^+ ions with molecular hydrogen.

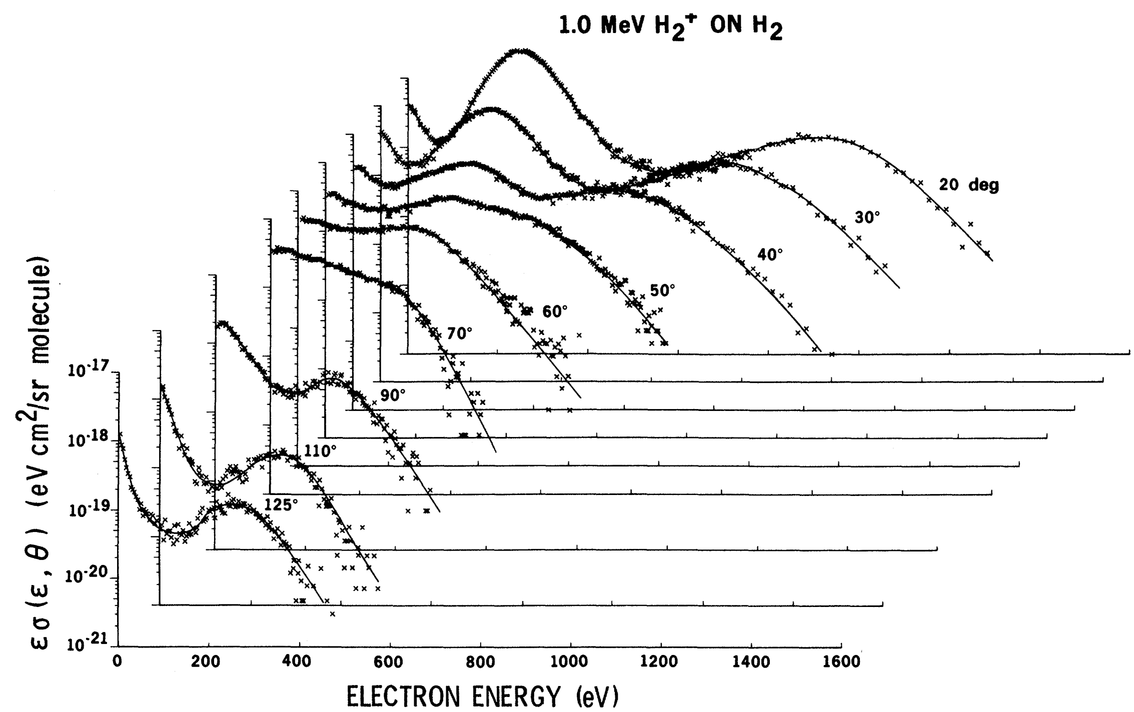

FIG. 2. Cross sections, differential in electron energy and emission angle, for electron emission following collisions of 1.0-MeV H_2^+ ions with molecular hydrogen.

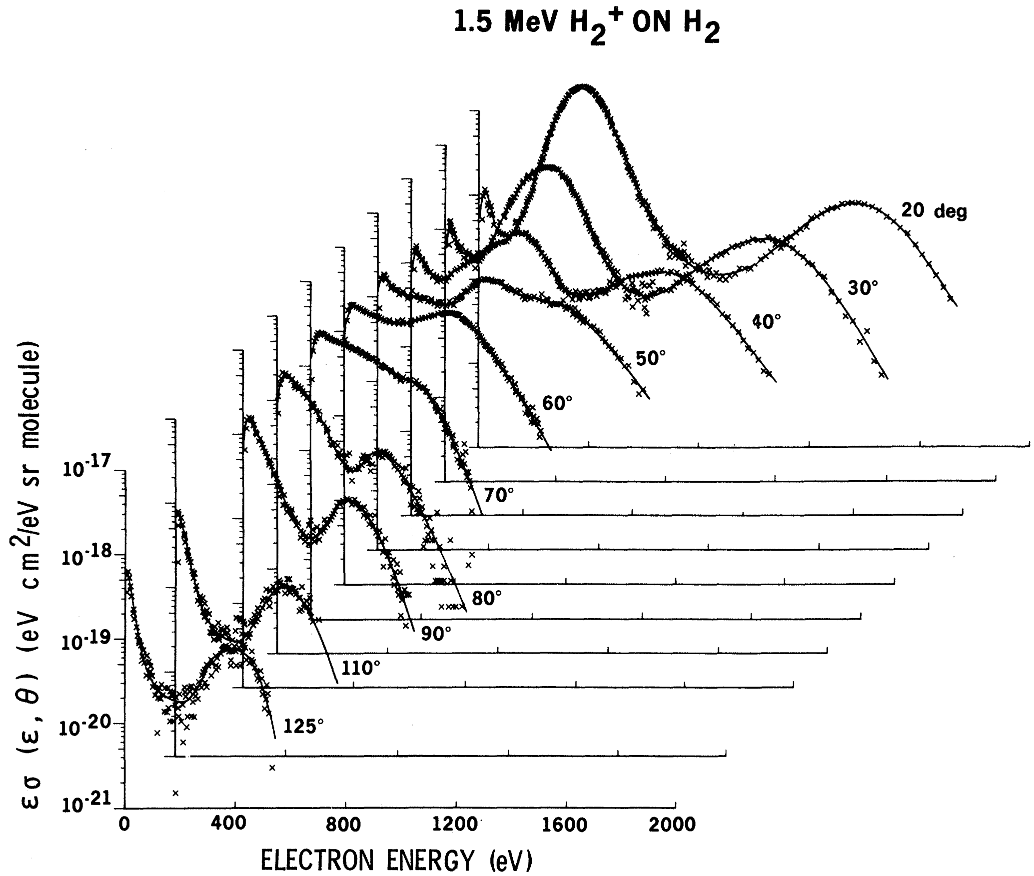

FIG. 3. Cross sections, differential in electron energy and emission angle, for electron emission following collisions of 1.5-MeV H_2^+ ions with molecular hydrogen.

shift and the dissociative ionization energy may be attributed, in part, to vibrational excitation of the incident H_2^+.

The width of the dissociative ionization electron distribution may arise from several sources: (a) the electrons in both the incident H_2^+ ion and the H_2 target have an initial distribution of velocities, (b) the incident ions may have varying degrees of vibrational excitation, and (c) the target may be left in an excited and/or ionized state. Except for ionization of the target, these three sources introduce small kinetic-energy changes and qualitatively can account for the broadening of the electron peak. At certain angles, however, the width of the dissociative ionization electron distribution is anomalously large, for example, at 40° in Figs. 2 and 3, and the shape is noticeably asymmetric with an enhanced yield of lower-energy electrons. This feature possibly arises from ionization of the target molecule simultaneously with the collisional ionization of the incident ion.

Electron-emission cross sections differential in electron energy, after integration with respect to emission angle, are shown in Fig. 4 for H_2^+ ions

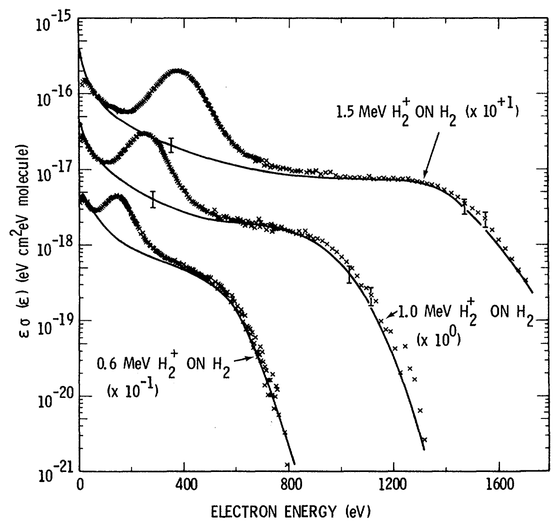

FIG. 4. Energy spectra, after integration over emission angle, of electrons ejected in H_2^+-H_2 collisions. The solid lines are twice the cross sections for electron emission for incident protons of the same velocity. The representative error bars (±20%) indicate the total uncertainty in the absolute magnitudes.

of energies 0.6, 1.0, and 1.5 MeV. For comparison, cross sections arbitrarily multiplied by 2, for electron ejection by equivalent-velocity protons, are shown as solid lines in Fig. 4. For high-energy electrons, the cross sections are very nearly in the ratio 2:1. Furthermore, kinematics require that the high-energy electrons be produced only from a collision with heavier particles in the reaction. Therefore, if proton-proton correlation is unimportant in the ionization of the target by incident H_2^+ one should expect the 2:1 ratio of yields since there are two protons per unit charge in the incident H_2^+ ion. The error bars shown in Fig. 4 are ±20% and indicate the uncertainty in the absolute magnitudes.

The actual ratios of the cross sections for electron ejection by H_2^+ and H^+ ions as a function of ejected-electron energy for the three ion velocities are presented in Fig. 5. We observe that the ratio of cross sections for ejection of electrons with energy greater than 750 eV is 2.0±0.3 for the three ion velocities. The error quoted and also indicated in Fig. 5 (±14%) is the rms folding of the precision in the two experiments which were used in determining the ratio. Data points shown in Fig. 5 are representative and were selected only for convenience in plotting; they do not exhaust the total data available.

The ratio of cross sections for ejection of low-energy electrons, $E \leqslant 100$ eV, is less than 2, suggesting proton-proton correlation may be important for these predominantly distant collisions.

On the basis of the comparisons presented in Figs. 4 and 5 we assumed that the energy distribution for electron ejection from the target by the heavy constituents of the H_2^+ ions has the same form as for proton ejection and is given by twice the latter. The ejected-electron distribution for target ionization by incident heavy particles can then be subtracted and the electron distribution which remains after this subtraction is assumed to be characteristic of H_2^+ ionization. This distribution was integrated with respect to electron energy to give a total collisional ionization cross section for H_2^+ on H_2. The results of this analysis are shown in Fig. 6 with other measurements of the cross section for dissociative ionization of H_2^+ ions which are based on the collection of heavy dissociation fragments. Excellent agreement is obtained between the present measurements and those of Sweetman[24] and Pivovar *et al.*[25] Berkner *et al.*[26] used the Born approximation to calculate the cross section for reaction (2) for 20-MeV ions in vibrational levels V = 0 through 18 and also for a vibrational-level population distribution calculated from the Franck–Condon principle. The broken line in Fig. 6 is the E^{-1} extrapolation of their result at 20 MeV for the Franck–Condon distribution of levels.

Electron spectra for three different emission angles and for 1.0-MeV H_2^+ ion impact are compared in Fig. 7 with similar spectra for incident

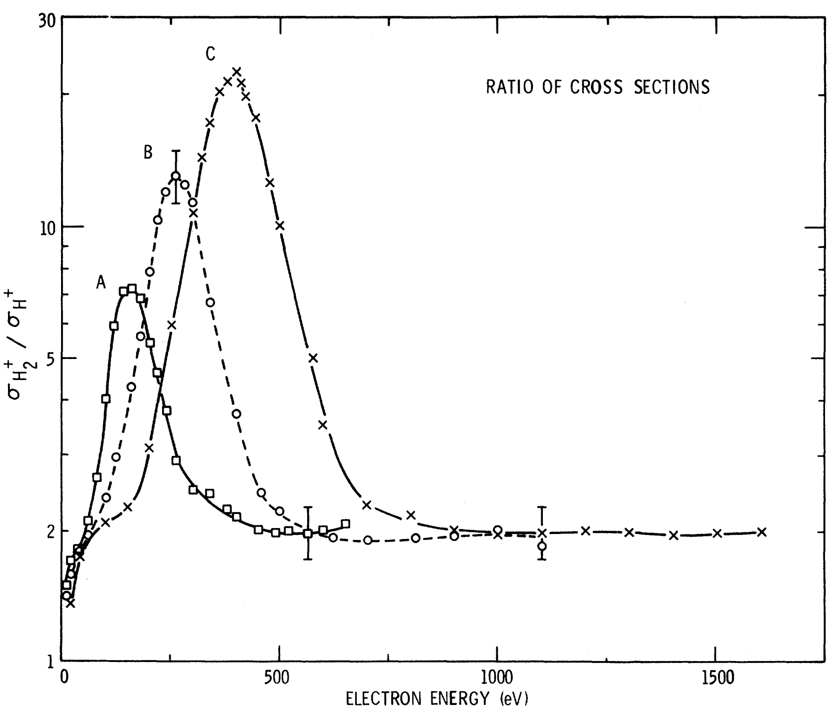

FIG. 5. Ratio of electron-emission cross sections for incident H_2^+ ions to those for incident protons of equal velocity. A, B, and C, are, respectively, for 0.6-, 1.0-, and 1.5-MeV H_2^+ ion energies. The error bars are ±14% and are the rms folded errors in the precision of the two experiments involved.

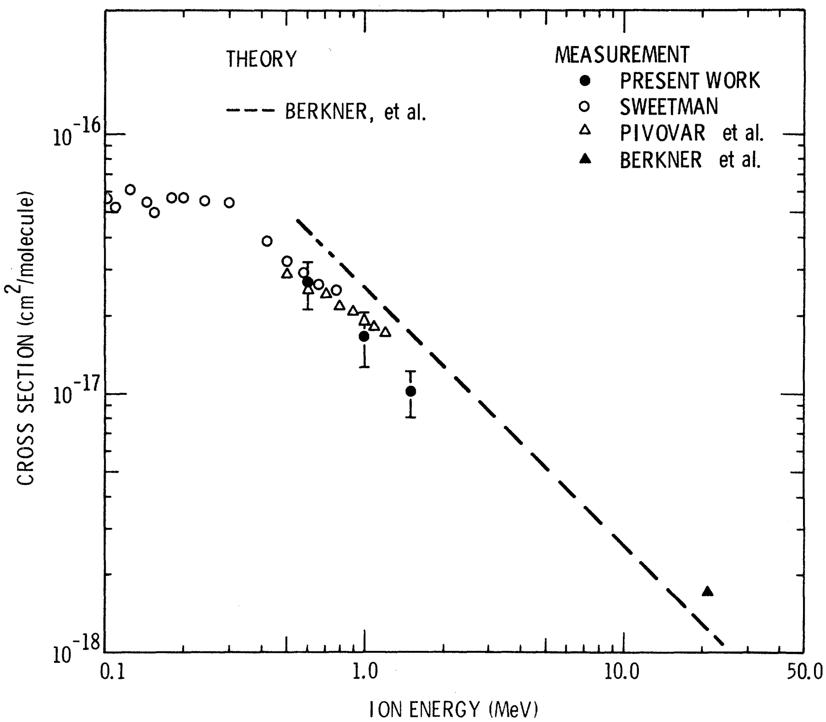

FIG. 6. Cross sections for ionization of H_2^+ in collisions with H_2. Previous experimental values: (circles, Ref. 24; triangles Ref. 25; shaded triangles, Ref. 26. Theoretical result: dashed line, Ref. 26.

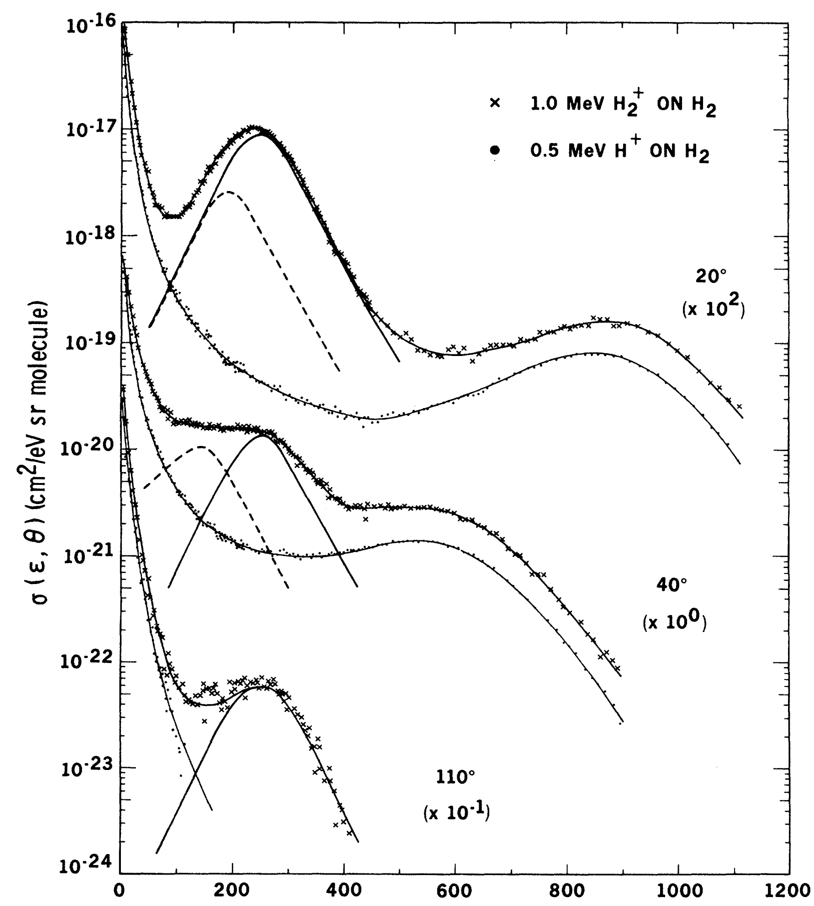

FIG. 7. Electron-emission spectra for three different angles for 1.0-MeV H_2^+ on H_2. Emission spectra for equivalent-velocity protons are also shown for comparison. The solid line under each peak indicates the angularly independent electron distribution and the dashed line indicates the angularly dependent electron distribution.

protons of the same velocity. For large angles the dissociative ionization electron distribution is nearly symmetric in shape, whereas at small angles a shoulder is evident on the low-energy side of the distribution. If one fits a Gaussian curve to the peak in the spectra at an energy corresponding to the peak energy for large-scale (110°) scattering, then one can separate the distribution into two components: one which shows no energy variation as a function of angle (indicated by solid lines in Fig. 7), and a second component which exhibits a definite energy dependence with emission angle (indicated by dotted lines in Fig. 7).

For the purpose of identification in this paper, the component of the dissociative ionization electron distributions showing no energy shift with angle is called *angularly independent*, and the component distribution exhibiting a dependence of electron energy for a variation in ejection angle is called the *angularly dependent*. We interpret the angularly dependent energy distributions as arising from the interaction of the incident ion with the target molecule, resulting in ionization of the incident ion with simultaneous ionization of the target molecule. On the other hand, we interpret the angularly independent energy distributions as arising when the target molecule undergoes ionization without simultaneous ionization of the incident ion. The scattering of the electron resulting from dissociative ionization of the incident H_2^+ ion at MeV energies is found to be similar to the scattering of incident free electrons of the same velocity. This behavior is not surprising, since the kinetic energy of the incident velocity-equivalent electron is more than an order of magnitude larger than its binding energy in the molecular ion.

We have multiplied by 2 the energy and angular distributions for electron ejection for proton ionization of H_2[6] and subtracted the result from the energy and angular distributions measured for H_2^+ impact and, as mentioned above, attributed the difference to electrons from the ionization of the H_2^+. The shape of the angularly independent energy distributions was assumed to be symmetric and was obtained by folding over the high-energy half of the distributions as measured for emission angles greater than 90°. Angularly independent energy distributions for the three H_2^+ ion energies are shown in Fig. 8. The curves A, B, and C are for 20° electron emission and indicate the relative magnitude and width variations for ion energies of 0.6, 1.0, and 1.5 MeV, respectively. It is not clear why the width of the distributions should increase with increasing ion energy.

By calculating angularly independent energy distributions like those in Fig. 8 for all angles one obtains doubly differential electron yields for the ionization of the incident H_2^+ ion presumably without simultaneous ionization of the target.

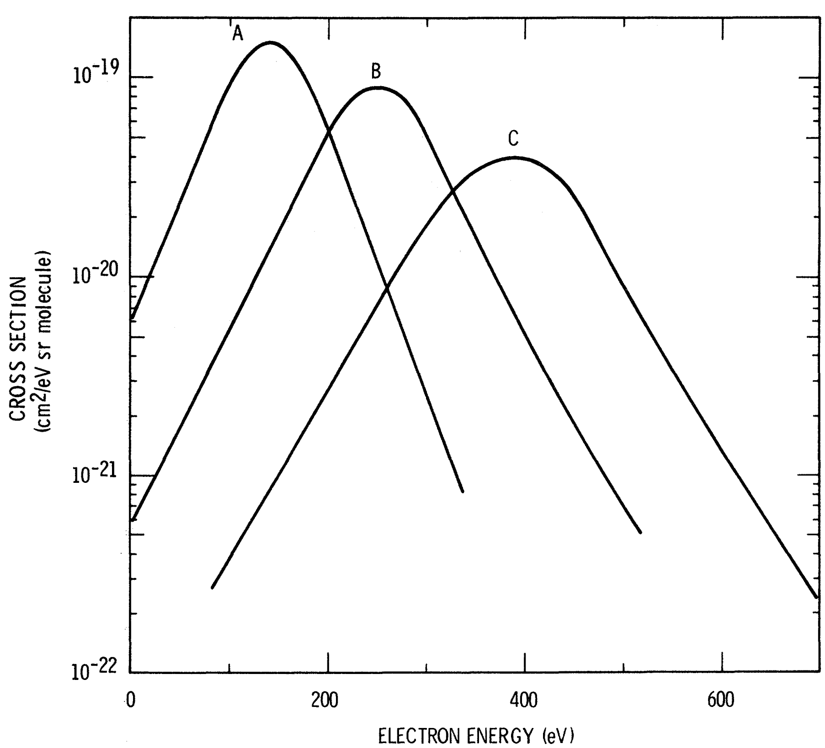

FIG. 8. Angularly independent energy distributions of electrons emitted at 20° from H_2^+ ionization. Distributions labeled A, B, and C result from incident H_2^+ ion energies of 0.6 1.0, and 1.5-MeV, respectively.

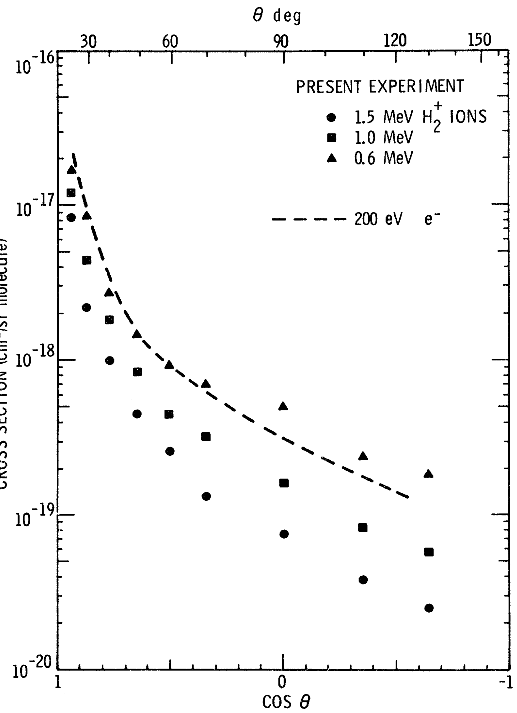

FIG. 9. Angular distributions for electrons exhibiting no energy dependence on angle. Other results are for elastic scattering of free electrons by H_2 (see Ref. 27).

These cross sections for collisions which may impart a relatively small amount of excitation energy to the target molecule can then be integrated with respect to electron energy to obtain yields differential only in emission angle. Angular distributions obtained in this manner are plotted in Fig. 9. Recall that our usage of "angularly independent" refers to the observation that the shape and peak energy of these component distributions do not change with angle. The area under the distributions, however, does vary with angle and it is this variation which is shown in Fig. 9 as the angular distribution. These cross sections compare well with both theoretical and experimental angular distributions for the scattering from molecular hydrogen on incident free electrons of similar velocities.[27]

We now turn our attention to the angularly dependent component of the dissociative ionization electron distributions shown as dashed lines in the examples in Fig. 7. We have analyzed the data by successively subtracting from the doubly differential cross sections for electron ejection, first the contribution from the two incident protons, and then the angularly independent energy distribution. The result is doubly differential yields for electrons ejected when presumably the target is simultaneously ionized along with the incident H_2^+ ion. An example of these doubly differential yields is shown in Fig. 10 for 1.0-MeV H_2^+ ions. The energies of the maxima of the ejected-electron distributions very nearly follow a cosine-squared angular dependence, which suggests that these electrons arise from essentially a binary encounter between the incident ion's electron and an electron of the target molecule.[28]

By integrating distributions like those in Fig. 10 with respect to electron energy, one obtains the differential cross sections for electrons ejected with significantly reduced energy (see Fig. 11). The uncertainty is quite large in obtaining these

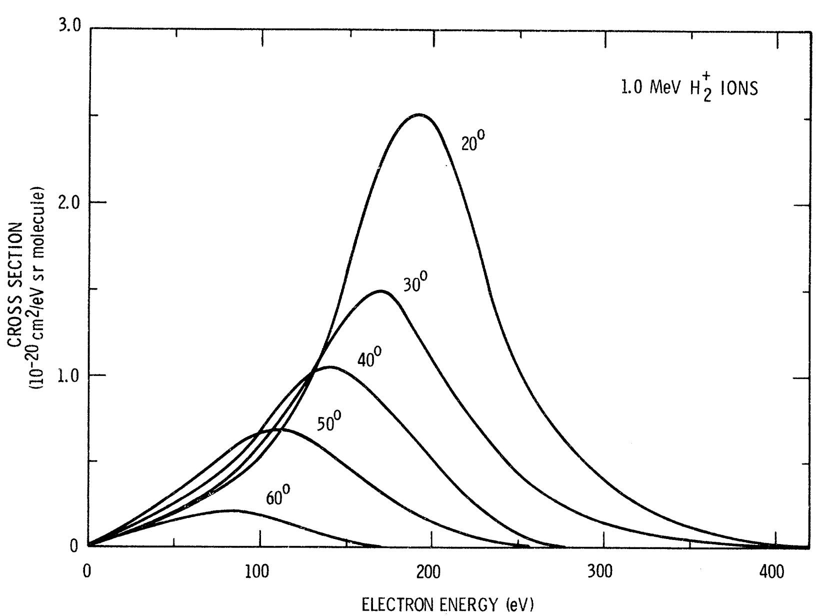

FIG. 10. Doubly differential cross sections for electrons exhibiting an energy dependence on angle and which are assumed to arise from simultaneous ionization of the target molecule and the incident H_2^+ ion.

angular distributions, owing both to uncertainties in the shape of the angularly independent energy distributions used in the unfolding process and to the fact that the angularly dependent energy distributions occur in the spectrum between the very large yield of low-energy electrons ejected from the target molecule by the incident heavy particles and the angularly independent part of the energy distributions of scattered electrons. The error bars indicated in Fig. 11 are estimates of the uncertainties associated with unfolding the distributions in order to obtain the angularly dependent contribution. Although the only reliable determination possible with this procedure was for emission angles up to and including 50°, there does appear to be evidence for such electrons at angles as large as 125°. Because of the large uncertainties, the points in Fig. 11 for large angles, $\theta \geqslant 60°$, are useful only as a guide in extrapolating the data from small angles to larger angles. No direct experimental or theoretical work on angular distributions for ionization induced by incident electrons is available for comparison with these results.

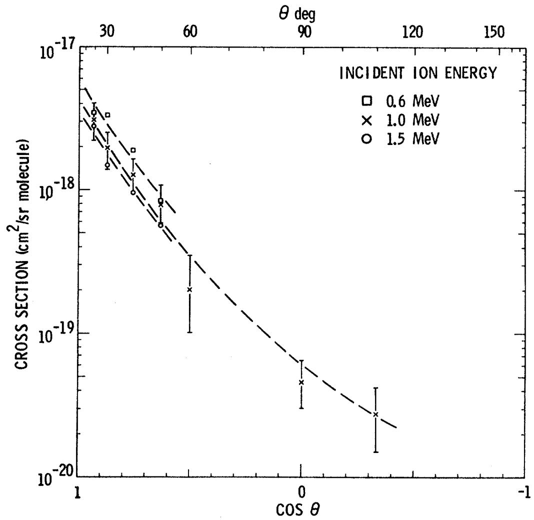

FIG. 11. Angular distributions for electrons exhibiting an energy dependence on angle.

IV. CONCLUSIONS

The dissociative ionization cross section for H_2^+ arrived at by analyzing the emitted electrons agrees well with measurements that detect the heavy particles. For H_2^+ ion energies of 0.6–1.5 MeV, the effective cross section for ejection of electrons of high energy from H_2 is twice that for protons of the same velocity. This implies that proton-proton correlation is relatively unimportant for these energies. However, for ejection of low-energy electrons—i.e., distant or glancing collisions—the ratio is less than 2.

The angular distributions of electrons resulting from collisional dissociation of the H_2^+ ion are similar to those for incident electrons of equivalent velocity. Differential cross sections for electrons exhibiting no energy variation with angle of emission compare well with measured elastic scattering cross sections for incident equivalent-velocity electrons.

In summary, the cross sections for electron emission following H_2^+ impact on H_2 targets at 0.6–1.5 MeV are found to be quantitatively similar to electrons ejected for the sum of the three incident particles acting independently.

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Charge Transfer in Proton-Hydrogen Collisions by the Faddeev Approach

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An approximate form of the Faddeev equations has been applied to the charge-transfer process in proton-hydrogen collisions. The effect of coupling with the direct channel has been taken into account. This formalism satisfies the unitarity condition below the breakup threshold. Results for the ground-state capture cross section have been obtained with and without the inclusion of the effect of proton-proton interactions. The experimental findings for the capture cross section in the energy range 1–150 keV lie in between our two sets of results. At high incident energies our results with and without the proton-proton interaction have approached the Jackson–Schiff and Brinkman–Kramers cross sections, respectively. The differential cross sections for the capture in the forward direction have been compared with the corresponding results of other theorists. Results for the elastic cross section have also been obtained.

I. INTRODUCTION

The problem of electron transfer in the proton-hydrogen (H^+, H) system has drawn the special attention of many theorists since this is one of the simplest type of rearrangement collisions. Several experimental investigations[1] have also been carried out on this problem. Brinkman and Kramers (BK)[2] have investigated this problem in the first-order Born approximation, neglecting the proton-proton interaction. The neglect of this interaction is justifiable since the ion-nucleus interaction can be removed by Canonical transformation (cf. Wick's comment in Ref. 3). Jackson and Schiff (JS)[3] and Bates and Dalgarno,[4] who have reconsidered the problem after many years, have shown that the effect of the proton-proton interaction when taken in the first Born approximation reduces the cross section considerably in better agreement with experiment. Unexpectedly, this reduction does not become vanishingly small even at high energies. In the high-energy limit, the JS cross section approaches the BK cross section multiplied by 0.661. These results have encouraged further investigations into the actual nature of the cross section. One may also visualize the problem in an impact-parameter treatment. The proton-proton interaction in this formalism affects the transition amplitude by introducing a phase factor, and thus the transition probability cannot be affected. Sil[5] has developed a variational method which has been applied to the problem in the impact-parameter formalism. He has concluded that when the Schrödinger equation is solved exactly the proton-proton interaction would not contribute to the capture cross section. Dalgarno and Yadav[6] have made calculations for the capture cross section using the perturbed-stationary-states method of Bates, Massey, and Stewart.[7] Further work by Drisko,[8] who has included the proton-proton interaction, has shown that the electron-transfer cross section obtained by taking higher-order Born series up to the third-order term does not converge to the BK results. Prodhan and Tripathy[9] have also obtained similar results using the impulse approximation. The work of McCarrol and Salin[10] has also reached a similar conclusion. Geltman[11] has suggested a formalism to study the electron-transfer problem by including the proton-proton interaction in the unperturbed part of the problem. For this inclusion, the plane wave in the final state is replaced by a Coulomb wave. Their results for the capture cross section lie between the corresponding BK and JS values for all energies. In the high-energy limit, their value of the cross section approaches 0.8 times the BK value. Gallaher and Wilets[12] have used a Sturmian basis in their close-coupling approximation for the low-incident-energy region and have obtained good agreement with the experimental findings. In their investigations on the proton-hydrogen collision, Cheshire, Gallaher, and Taylor[13] have also used the close-coupling method. They have retained $1s$, $2s$, $2p$ hydrogenic states and three orthogonal pseudostates are then added.