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Distributions in Energy and Angle of Electrons Ejected from Molecular Hydrogen by 0.3–1.5-MeV Protons*

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Cross sections for the ejection of electrons, differential in electron energy and angle of emission, have been measured for energetic protons on molecular hydrogen. Electron-energy distributions are presented for ten angles between 20 and 130 deg for each of five proton energies between 0.3 and 1.5 MeV. The results are compared with calculations based on binary-encounter theory, the Born approximation, and the Faddeev equations. Qualitative agreement with the latter provides supporting evidence for the inclusion of a long-range electron-proton interaction in the collision theory for proton energies as high as 1.5 MeV.

I. INTRODUCTION

Measurements of cross sections, differential in electron energy and emission angle, for the ejection of electrons in ion-molecule collisions provide information concerning the ionization process. Comparison of these cross sections to theoretical ones is a test of the reliability and limitations of the theoretical treatments used to describe the ionization process and, hence, provides information concerning the relative importance of various types of interactions which enter into a complete description of ionization by fast charged particles. Several different theoretical approaches have been used to calculate double-differential electron-ejection cross sections[1–5] for incident protons. In general, the nature of these calculations restricts the range of validity of the results to apply to protons with velocities greater than several times the velocity of the electron in the first Bohr orbit of the hydrogen atom. Previous measurements of double-differential cross sections for ejection of electrons from molecular hydrogen were limited to protons with a maximum energy of 0.3 MeV,[5–7] in which case the velocity of the proton was only about 3.5 times the velocity of the electron in the first Bohr orbit of hydrogen. In our work, we have extended the proton energy range to 1.5 MeV in order to obtain cross sections which may be compared more reliably to values calculated using high-energy approximations.

II. EXPERIMENTAL METHOD

The apparatus and experimental technique used for our measurements have been described in detail previously[8] and will be discussed here only briefly. The proton beam from a Van de Graaff generator was energy analyzed, collimated, and passed through a differentially pumped target cell before being collected in a Faraday cup. Electrons ejected from the target gas were energy analyzed by a cylindrical-mirror electrostatic analyzer and detected by a continuous-channel electron multiplier. The electrostatic analyzer had an energy resolution of 3.5% and was collimated to accept electrons ejected in a given direction with an angular spread of approximately 5 deg. The electron-energy spectra were measured by recording the number of electrons transmitted by the electrostatic analyzer with a given analyzing voltage for a preset number of protons passing through the target cell. The analyzer voltage was automatically advanced and the process repeated. Data points were obtained at intervals of either 2 or 4 eV with each point representing the electron yield for approximately 10^{12} protons. The energy spectra of ejected electrons were normally obtained at ten different angles from 20 to 130 deg.

In order to reduce the background count rate due to scattered electrons, the deflection plate of the electrostatic analyzer was made of a grid of fine wire. In this way, electrons which enter the analyzer with energies greater than the energy being transmitted would pass through, rather than be scattered from, the reflecting element of the analyzer. This reduction in the background count rate was important in measuring the smaller cross sections reported in this work. Knife-edge collimation and precise alignment of the apparatus with respect to the proton beam were also necessary to reduce the number of scattered electrons.

III. DATA REDUCTION AND ERROR ANALYSIS

Absolute cross sections, differential in ejected-electron energy ϵ, and emission angle θ, were obtained from the raw data by means of the following expression:

$$\sigma(\epsilon,\theta)=\frac{N_e e^{\alpha(\epsilon)Px}}{N_p P dS \Delta E\, 3.23\times 10^{16}}\ \mathrm{cm^2/eV\,sr\,molecule}\ , \qquad (1)$$

where N_e is the number of electrons counted for N_p protons; P is the target gas pressure in Torr; ΔE is the energy spread of the electrons transmitted by the electrostatic analyzer, dS is the product of solid angle subtended by the analyzer and the proton-path length observed within this solid angle; $\alpha(\epsilon)$ is the absorption coefficient for electrons of energy ϵ in the target gas; and x is the effective-path length of the target gas through which the ejected electrons must pass before being detected. Only a brief discussion of the parameters used in Eq. (1) will be presented here since they are discussed in detail in Ref. 8. The target gas pressure was measured with a capacitance manometer and was maintained constant to within a few percent by means of an automatic pressure controller. Target pressures of 0.006 and 0.012 Torr were used in these measurements which together with the physical size of the target region resulted in a small probability that ejected electrons would interact with the target gas before being detected. The term $e^{\alpha(\epsilon)Px}$ in Eq. (1) was included to account for the probability that electrons, primarily very low-energy electrons, may be scattered by the target gas before they reached the detector. The absorption coefficients used in this work were taken from Normand[9] and from Golden *et al.*[10] The uncertainty in the calculated cross sections due to uncertainty in the correction for electron scattering may be as large as 15% for electron energies of a few electron volts (the electron scattering cross-sections peak at a few electron volts), however, for electron energies greater than 30 eV, the uncertainty in the measured cross section due to scattering of ejected electrons is negligible.

When calculating the product of the solid angle subtended by the electrostatic analyzer and the path length of the proton through the target gas, it was necessary to account for variations in gas density along the proton track which result from differentially pumping the target cell. A computer program was developed to give a first-order correction to the quantity dS due to this density variation. The uncertainty in the product of solid angle and path length resulting from this calculation is considered to be less than 10%. This program was also used to obtain the effective path length of target gas, x, through which the electron must pass in reaching the detector.

The energy calibration and energy resolution of the electrostatic analyzer were obtained by measurements of the line shapes and positions of Auger electrons detected for gas targets containing carbon, nitrogen, oxygen, neon, and argon. Particular lines used for the energy calibration were the $2p$-$3p3p$ line of argon at 206 ± 2 eV,[11] the $KL_{2,3}L_{2,3}$ lines of neon at 805 ± 0.5 eV,[12] as well as the position of the unresolved K-Auger spectrum of molecular nitrogen and of cargon and oxygen obtained from carbon monoxide gas which were compared to measurements by Moddeman.[13] For electron energies greater than 1000 eV, the energy calibration was determined by means of an electron gun. The electron energies reported in the present work are estimated to have an uncertainty of less than 1.0%.

The over-all uncertainty in the absolute magnitude of the measured cross sections due to uncertainties associated with the determination of the solid angle, target gas pressure, effects of density variation along the proton path, correction for

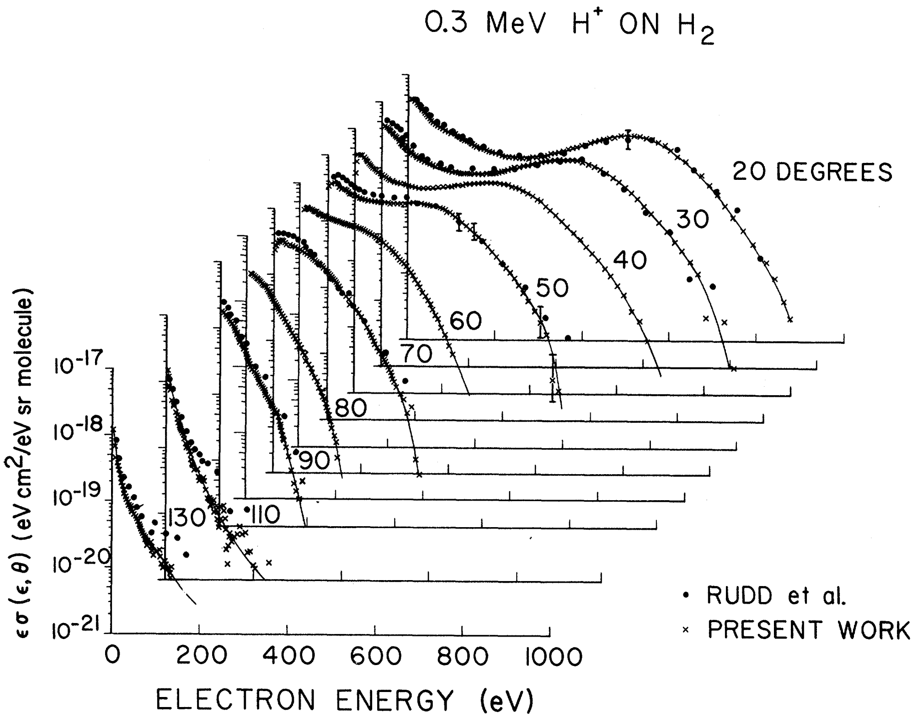

FIG. 1. Cross sections, differential in electron energy and emission angle, for ejection of electrons by 0.3-MeV protons on molecular hydrogen. Previous measurements are from Ref. 7.

scattering of ejected electrons by the target gas, etc., is estimated to be less than 25% for electron energies greater than 5 eV. However, where the cross sections are very small, the signal to background ratio is small and the uncertainty in the measured cross section may become as large as a factor of 2. Representative error bars are shown in several of the figures to illustrate the uncertainties in the absolute values of the measured cross sections. Uncertainties quoted in this paper are understood to be one standard deviation.

IV. EXPERIMENTAL RESULTS

Cross sections, differential in electron energy and emission angle for ejection of electrons from molecular hydrogen, are shown in Figs. 1–5 for incident proton energies of 0.3, 0.5, 0.75, 1.0 and 1.5 MeV, respectively. These cross sections were multiplied by the respective electron energy before being plotted in order to reduce the range of the ordinate necessary to display the results.[14]

The results shown in Fig. 1 for 0.3-MeV proton-impact energy are more accurate for low-energy electron emission (energies less than 30 eV) than those previously reported[8] and cross sections have been added for emission angles of 60 and 80 deg. The accuracy of the data for ejection of low-energy electrons has been improved by using better magnetic field nullification techniques and improved data reduction procedures. The cross sections reported in this work should be accurate to 25% for electron energies as low as 5 eV. These cross sections are in good agreement throughout most of the range of electron energy and ejection angles with the results previously reported by Rudd *et al.*[7] These are shown in Fig. 1. There are some discrepancies; i.e., for electron energies less than approximately 50 eV and emission angles of 30, 50, and 70 deg, the cross sections reported by Rudd *et al.* are somewhat larger than our values. At the back angles, and for small cross sections where much larger uncertainties prevail, there is as much as a factor of 4 difference between the results presented here and those reported by Rudd *et al.* However, even in these regions, there is agreement within the combined experimental uncertainties assigned to the cross sections.

Two distinct features are evident in the energy distributions shown in Figs. 1–5; these are a peak in the cross section for low-energy electron ejection and a second peak observed for forward angles and higher energy electrons. The low-energy peak is associated with the distant collisions in which a small amount of energy is transferred to the hydrogen molecule whereas the high-energy peak results from binary collisions between the proton and an atomic electron.

As the proton energy is increased from 0.3 to 1.5 MeV, the two peaks separate. The best illustration of this separation is seen in the 20-deg spectrum from 1.5-MeV proton-impact energy shown in Fig. 5. In this example, the cross sec-

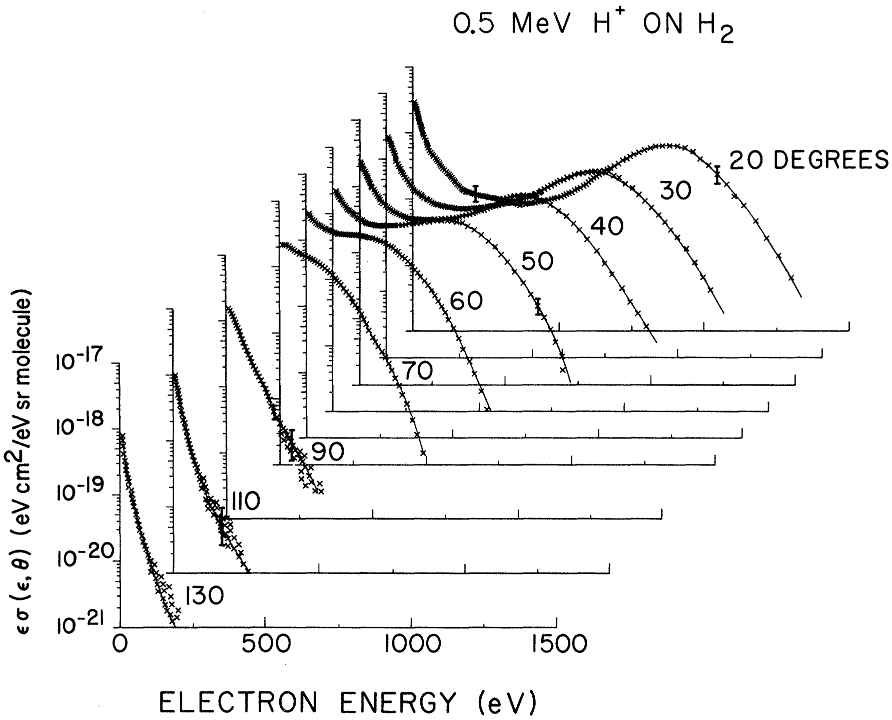

FIG. 2. Cross sections, differential in electron energy and emission angle, for ejection of electrons by 0.5-MeV protons on molecular hydrogen.

tions in the valley between the peaks are nearly two orders of magnitude smaller than the high-energy peak. For these small cross sections, the signal count rate and background count rate were nearly equal which resulted in rather large statistical uncertainties in the measured cross sections. Error bars indicate the total, random and systematic, uncertainties estimated for these measurements.

The comparison of double-differential electron-emission cross sections to theoretical results has become a useful tool in understanding details of the collision process. Comparisons of cross sections calculated using the Born approximation to the measurements for 0.3-MeV protons indicated the importance of a long-range interaction between the outgoing proton and the ejected electron.[1,15] This interaction was suggested to account for the enhance-

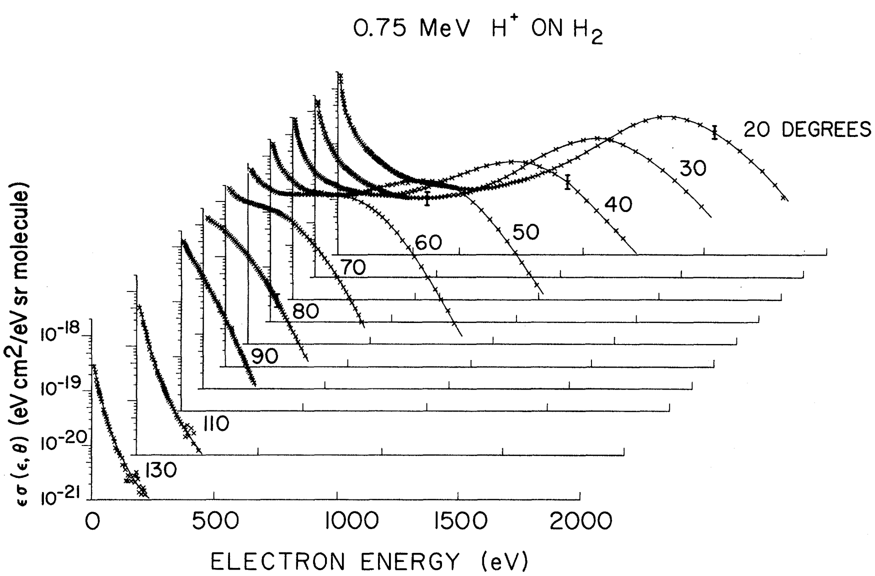

FIG. 3. Cross sections, differential in electron energy and emission angle, for ejection of electrons by 0.75-MeV protons on molecular hydrogen.

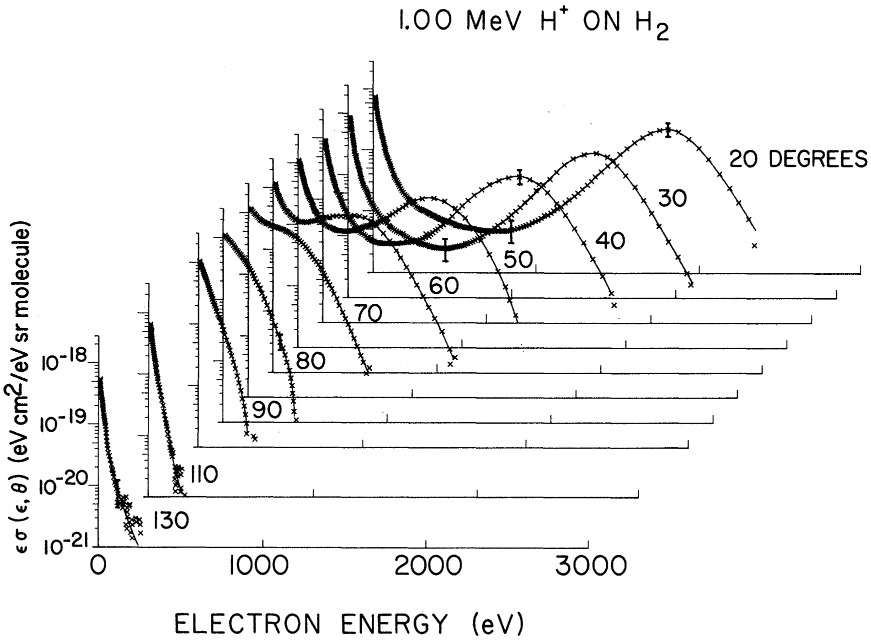

FIG. 4. Cross sections, differential in electron energy and emission angle, for ejection of electrons by 1.0-MeV protons on molecular hydrogen.

ment of the emission cross sections at small angles. The effects of such a long-range interaction have been investigated by Macek[16] using Faddeev's equations and by Salin[17] using a velocity-dependent potential and the Born approximation. Both calculations show an increase in the small-angle cross sections which brings about qualitatively better agreement with experimental values, however, quantitative agreement was not realized. Due to lack of high-energy results, comparisons of calculated and measured cross sections have been restricted to proton energies of at most 0.3 MeV and Macek has cautioned[16] that first-order approximations, as used in his work, may not be adequate to describe details such as the energy and angular distributions of the ejected electrons for proton energies as low as 0.3 MeV.

Macek has recently extended his calculations[18] to proton energies of 1.5 MeV for comparison with our present measurements and the results are shown in Figs. 7 and 8. We have plotted cross sections for selected electron energies in Fig. 6 which were obtained in our measurements for 1.0-MeV protons on molecular hydrogen along with the results of the Born approximation[19] and Macek's calculation based on Faddeev's equations.[16] The cross

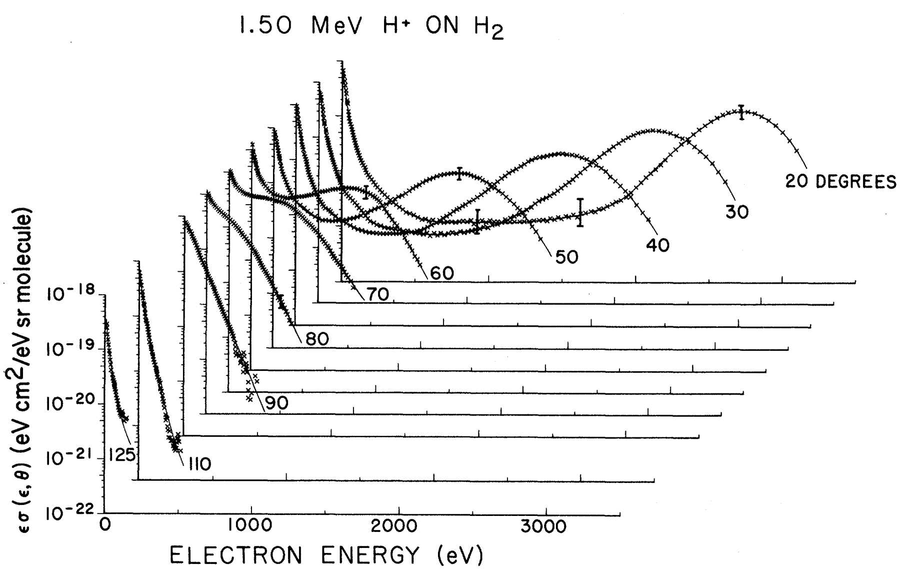

FIG. 5. Cross sections, differential in electron energy and emission angle, for ejection by 1.5-MeV protons on molecular hydrogen.

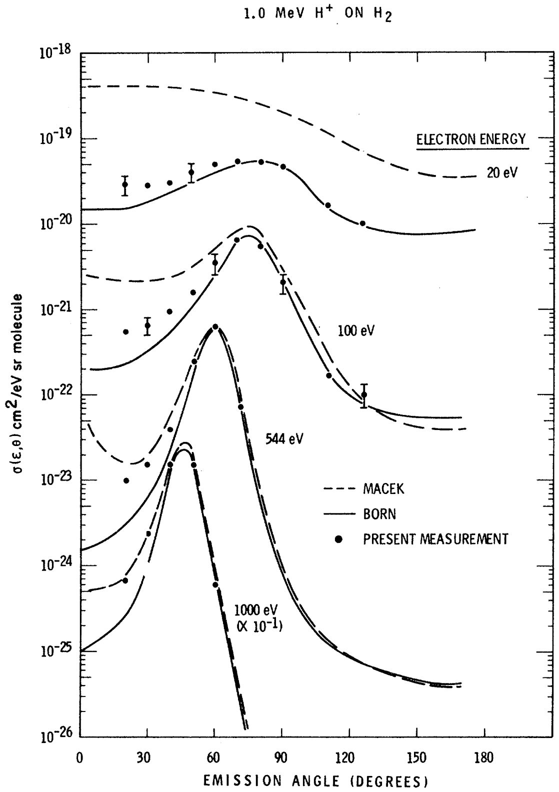

FIG. 6. Angular distributions for ejection of electrons of selected energies from molecular hydrogen by 1.0-MeV protons. Theoretical results: See Refs. 16 and 19.

sections calculated by Macek are restricted to ejected-electron energies greater than several rydbergs by approximations used in his development of Faddeev's equations. Therefore, the discrepancies indicated in Fig. 6 between his calculation and measured cross sections for the ejection of 20-eV electrons are not surprising. For ejected electrons of high energy, agreement between Macek's results and the measured cross sections improves, with close agreement being obtained for electron energies near 1000 eV. On the other hand, cross sections calculated using the Born approximation, appear to be a nearly constant factor smaller than the measured values with no improvement in the agreement for higher-electron energies. The largest discrepancy between the measured cross sections and those calculated using the Born approximation occur for small angles and, for 1.0-MeV proton-impact energy, for electron energies near 544 eV. This discrepancy is associated with the long-range proton-electron interaction which is strongest when the incident proton and ejected electron have the same velocity, a condition which is met by 1.0-MeV protons and 544-eV ejected electrons. For electron energies greater than 100 eV and emission angles larger than approximately 50 deg, cross sections based on the Born approximation and on Macek's development of the Faddeev equations are in good agreement with each other and with measured values. In Fig. 7 are shown the results of measured and calculated cross sections for ejection of electrons with velocities equal to the incident proton energy for the range of proton energies studied in our work. The results of Rudd *et al.*[7] for 0.3-MeV proton impact are included in order to indicate the close agreement between these independent measurements. As the proton energy increases, the results of the Macek theory approach the experimental values with close agreement being

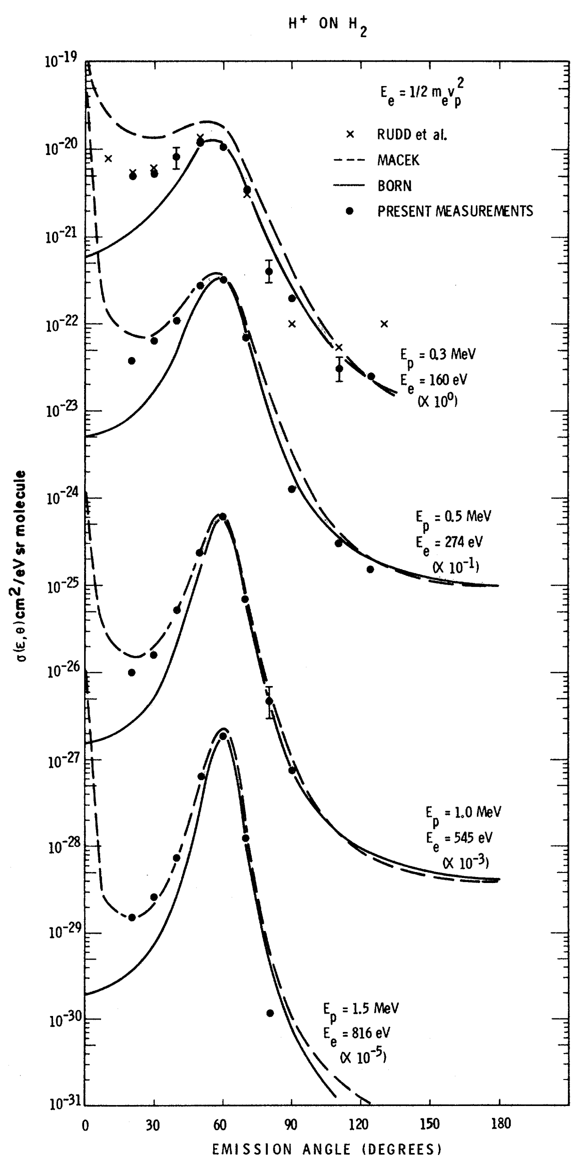

FIG. 7. Angular distribution for ejection of electrons with velocities equal to the incident proton velocity. Previous measurements: See Ref. 7. Theoretical results: See Refs. 16 and 19.

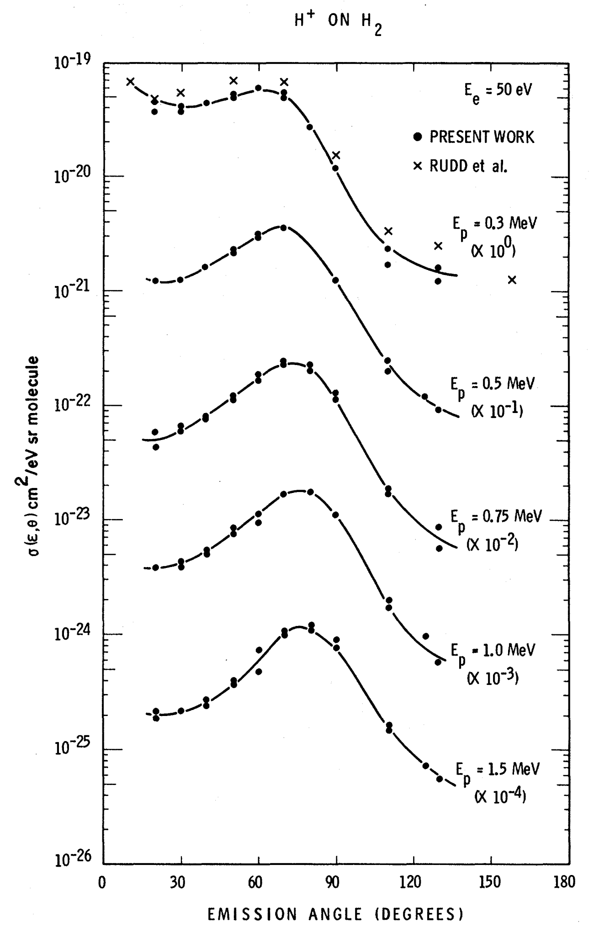

FIG. 8. Angular distributions of cross sections for ejection of 50-eV electrons from molecular hydrogen by several different proton energies. Previous measurements: See Ref. 7.

observed for 1.5-MeV protons. Both the measured cross sections and those calculated by Macek tend toward the Born cross section as the proton energy increases; however, the effect of the long-range electron-proton interaction is still very much in evidence for proton energies as high as 1.5 MeV.

The shape of the angular distribution of electrons ejected with low energy (electrons with velocity small compared to the proton velocity) is found to be nearly independent of the proton energy. Cross sections for ejection of 50-eV electrons from molecular hydrogen by proton energies from 0.3 to 1.5 meV are shown in Fig. 8. Similarly, the shape of the angular distributions for electrons with velocities greater than the proton velocity are nearly independent of the proton energy; Fig. 9 shows, as an example, the distributions for electrons whose velocity is $\sqrt{2}$ times the proton velocity. There is a small variation in the shape of the small-angle portion of the curves which is accounted for by the decreasing effect of the long-range electron-proton interaction as the proton energy increases. The diminishing importance of the long-range interaction with increasing proton energy has been observed at lower proton energies by Crooks and Rudd[20] in their electron spectra recorded for electrons ejected in the direction of the proton beam. The principal difference in the shapes of the angular distributions for ejection of low-energy electrons (Fig. 8) and for ejection of high-energy electrons (Fig. 9) is in the cross sections for electron emission into large angles. The cross sections for low-energy electron emission are much greater for large angles than is the case for high-energy electrons. This

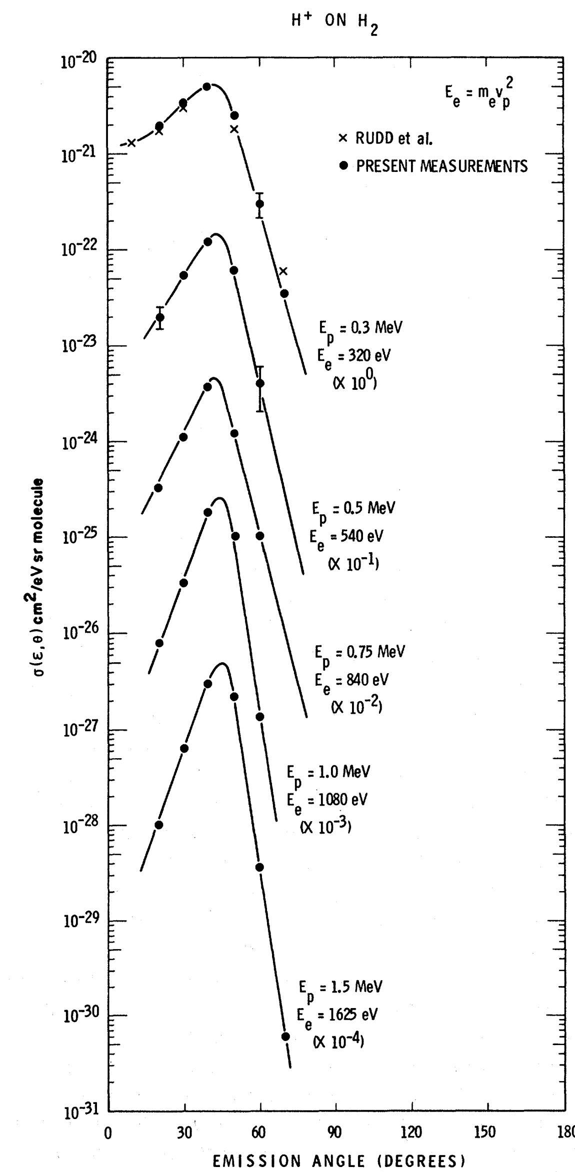

FIG. 9. Angular distributions for ejection of fast electrons from molecular hydrogen for several different proton energies. Previous measurements: See Ref. 7.

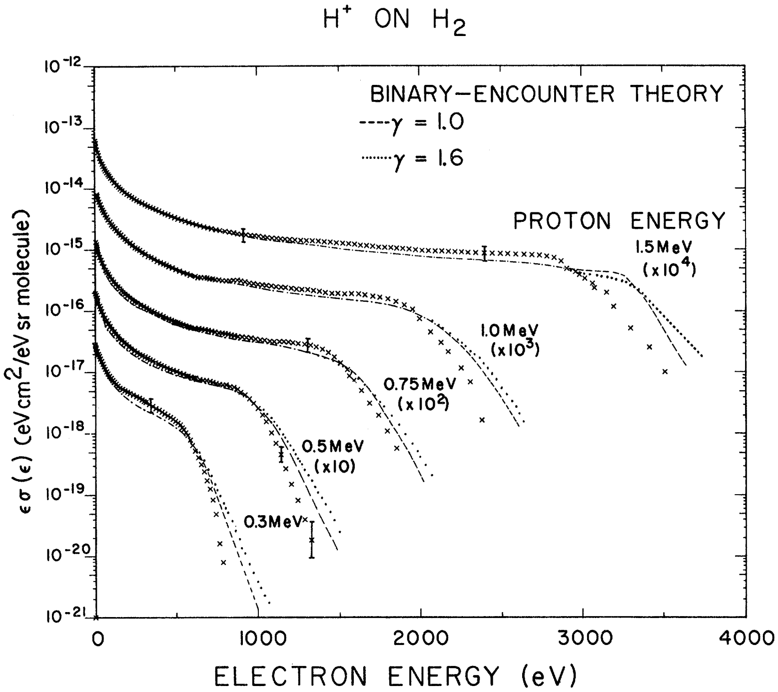

FIG. 10. Cross sections, differential in electron energy, for ejection of electrons from molecular hydrogen for incident proton energies of 0.3, 0.5, 0.75, 1.0, and 1.5 MeV. Previous measurements: See Ref. 7. The calculated results were obtained using a program of Rudd, Gregoire, and Crooks (Ref. 23).

enhancement has been attributed to the effects of distortion of the target atom by the impulsive interaction with the passing proton.[21,22]

Cross sections differential only in electron energy may be obtained from our double-differential cross sections by integration with respect to the emission angle. These differential cross sections are displayed in Fig. 10 along with cross sections calculated from an analytical expression developed by Rudd, Gregoire, and Crooks.[23] This calculation, which utilizes binary-encounter theory based on the Thomas-Gerjuoy-Vriens equations,[24–26] involves a semiclassical treatment in which the cross sections for electron ejection are calculated taking into account the relative velocity between the incident proton and the bound electron. The velocity distribution of the bound electrons is assumed to be isotropic and is deduced quantum mechanically from a Fock distribution. Other parameters in the calculation include the ionization potential of the molecule and the ratio γ of the orbital kinetic energy to the ionization potential. This ratio is commonly taken to be 1 and, indeed, can be shown to be unity for atomic hydrogen by use of the virial theorem. For larger atoms and for molecules, there is some question as to the proper value to be used for γ. Robinson[27] has suggested the use of Slater's rules to obtain an estimate of the orbital kinetic energy and. in this way, depending on the assumptions made, a value of 1.6 or 2.56 can be obtained as an estimate of γ for molecular hydrogen. The determination of γ for molecular hydrogen has been discussed by Rudd and Gregoire[28] and from their work, one is led to use $\gamma = 1.6$ for the best agreement between measured and calculated cross sections when the entire energy range of the ejected electron is considered. The two calculations based on binary-encounter theory shown in Fig. 10 are for $\gamma = 1.0$ and $\gamma = 1.6$. For the proton-energy range covered in our work and on the compressed logarithmic scale used in Fig. 10, the difference in the calculated values due to using different values of γ is negligible except for the extreme high-energy end and the extreme low-energy end of each electron spectrum. At the high-energy end of the spectra, calculations with $\gamma = 1.0$ result in cross sections that are in somewhat better agreement with measured values than corresponding cross sections calculated using $\gamma = 1.6$; this result was also noted by Rudd and Gregoire[28] in their comparisons for low-energy proton impact. The cross sections for ejection of very-low-energy electrons are shown on an expanded scale in Fig. 11 along with binary-encounter cross sections obtained using $\gamma = 1.6$. Had $\gamma = 1.0$ been used instead of $\gamma = 1.6$ in this calculation, the computed values would be smaller by approximately 20% at 2 eV and approximately 8% at 100 eV. From the results shown in Fig. 11, one can conclude that as the proton energy increases, the accuracy of binary-encounter theory in predicting low-energy electron-emission cross sections decreases. One important effect of the failure to predict these low-energy cross sections will be discussed later; that is, the tendency of binary-en-

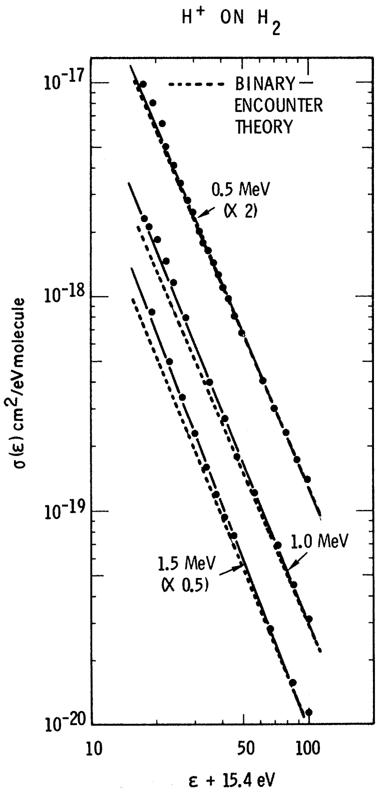

FIG. 11. Cross sections, differential in electron energy, for ejection of low-energy electrons from molecular hydrogen by incident protons with energies of 0.5, 1.0, and 1.5 MeV. The calculated values were obtained using a program of Rudd, Gregoire, and Crooks (Ref. 23).

counter theory to yield total ionization cross sections which fall off more rapidly with increasing proton energy than do measured cross sections or those calculated using the Born approximation.

In general, the agreement between measured cross sections and those calculated from binary-encounter theory is quite good. With the exception of the extreme high-energy end of the electron spectra, calculated and measured values agree to within 30%. The ease of calculation and the reliability of the results make binary-encounter theory a very useful tool for calculating the energy spectrum of electrons ejected in high-energy collisions.

If the measured double-differential cross sections are integrated with respect to both angle and energy, the result is the total cross section for ionization of molecular hydrogen by fast protons. In Fig. 12, the total ionization cross sections obtained from our measurements are compared with values reported by other investigators. The ionization cross sections reported by Rudd *et al.*[7] and by Kuyatt and Jorgensen[5] were measured in the same manner as ours; i.e., integration of double-differential electron-ejection cross sections. Rudd *et al.* obtained absolute values directly whereas Kuyatt and Jorgensen normalized their results at 50 keV to a previously measured ionization cross section. Cross sections reported by Afrosimov *et al.*[29] and by Hooper *et al.*[30] were obtained by a technique involving total charge measurements. The dashed line in Fig. 12 is the result of the Born calculation of Bates and Griffing[31] for atomic-hydrogen cross sections scaled to molecular hydrogen according to the procedure discussed by Hooper *et al.* We have also included the results of binary-encounter calculations which were obtained by integrating the curves shown in Fig. 10. The ionization cross sections obtained from our measurements are in agreement to within approximately 20% with the results of Hooper *et al.* This agreement with previous work which utilizes an entirely different experimental technique lends confidence to the absolute calibration used in arriving at the double-differential cross sections reported in this paper. This comparison is, however, primarily a test of our cross sections for ejection of low-energy electrons since the total ionization cross section is strongly dependent on relatively large differential cross sections for these electrons. The influence of the low-energy portion of the emission spectrum is apparent when the cross sections calculated from binary-encounter theory are considered. As was shown in Fig. 11, the binary-encounter cross sections underestimate the measured values for low-energy electron emission by increasingly larger amounts as the proton energy is increased and this is directly correlated with total ionization cross sections which fall off more rapid-

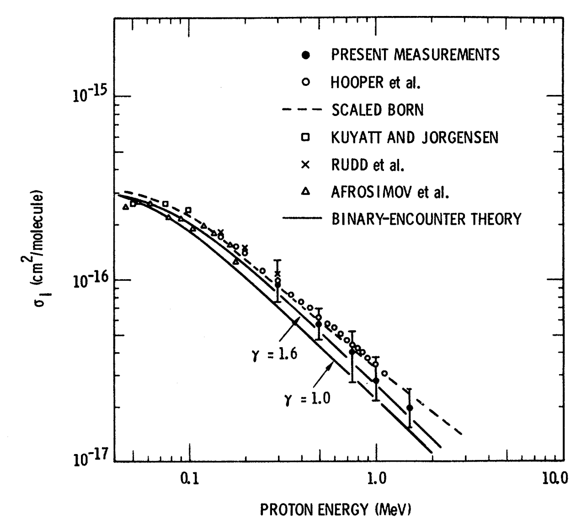

FIG. 12. Total ionization cross sections for protons on molecular hydrogen. Previous measurements: See Refs. 30, 7, and 29. Calculated cross sections: Scaled Born (see Ref. 30), and binary-encounter theory (see Ref. 23).

ly with proton energy than is expected based on both measured cross sections and those calculated using the Born approximation. It is also apparent that the cross sections we have obtained for 1.0- and 1.5-MeV protons decrease more rapidly with increasing proton energy than the Born cross sections. It is not clear whether this trend is significant or whether the uncertainty in our cross sections for electron energies less than 5 eV is leading to an underestimation of the ionization cross sections. In any case, the agreement between our measurements and the scaled Born calculation is within the experimental uncertainties of our results.

V. SUMMARY

A comparison of measured and calculated angular distributions of ejected electrons indicates that the effect of the long-range electron-proton interaction is evident for proton energies as high as 1.5 MeV. The Macek theory, which includes this interaction, agrees very well with our measurements for protons of sufficiently high energy and correspondingly high-energy electron ejection. Although the Born approximation is expected to become more reliable as the proton energy increases, Born calculations, which do not include the long-range electron-proton interaction, are still found to underestimate the small-angle cross sections for proton energies as high as 1.5 MeV. This discrepancy, which is largest for ejection of electrons into small angles with velocities equal to the proton velocity, however, decreases from a factor of 5 at 0.3-MeV to a factor of 3 for 1.5-MeV protons for ejection of electrons at 20 deg with respect to the forward direction of the proton beam. With the exception of the small angles, the measured double-differential cross sections are in close agreement with values calculated using the Born approximation for ejected-electron energies from 20 to 2000 eV and for proton energies from 0.3 to 1.5 MeV.

Cross sections, differential in electron energy, are in close agreement throughout most of the energy range of the ejected electrons with those calculated using binary-encounter theory. The largest discrepancies are at the extreme high-energy end of the electron spectra. However, in this region, experimental uncertainties are too large to allow a definitive test of the theory. The total ionization cross sections obtained in this work agree within experimental uncertainties with previously measured results and with cross sections calculated using the Born approximation.

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