# Energy and Angular Dependence of the Differential Cross Section for Production of Electrons by 50–100 keV Protons in Hydrogen Gas*†

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(Received 28 August 1962)

Hydrogen gas in a rotatable scattering chamber was bombarded by 50-, 75-, and 100-keV protons. The resulting secondary electrons were analyzed in both direction and energy by a slit system and a cylindrical electrostatic analyzer and counted by an electron multiplier tube with suitable electronics. Relative values of the differential cross section for ejection of secondary electrons were measured for 4- to 300-eV electrons at angles of 23°, 45°, 67.5°, 90°, 112.5°, 135°, and 152° from the proton direction. Absolute values for the differential cross sections were obtained by integration of the 50-keV results over all angles and energies and normalizing to the total ionization cross section measured by Schwirzke. As a function of electron energy, at a fixed angle, the differential cross sections show a broad peak at 4 to 8 eV with a monotonic decrease at higher electron energies. As a function of angle, for fixed electron energy, most of the differential cross sections are largest at 23°, drop off rapidly to about 100°, and are then relatively constant. The differential cross sections have been integrated in various ways to obtain cross sections differential only in energy and in angle, total cross sections for ionization, average energies of the ejected electrons, and the stopping cross section due to ionization. Comparisons are made with other experimental results and with Born approximation calculations.

## I. INTRODUCTION

ALTHOUGH the ionization of gases by fast protons is a well-known process and is an important method of energy loss, very little information is available on the energy and angular distribution of the ejected (secondary) electrons. Blauth[1] has measured the energy distribution of secondary electrons produced by 8.8-, 11.8-, and 49-keV protons in several gases, including hydrogen. Only electrons ejected at an angle of 54.5° with respect to the proton beam were investigated. Berry[2] has measured the energy distribution of secondary electrons produced by 0.30- to 3.0-keV hydrogen, nitrogen, argon, and helium ions and atoms in the parent gas. Only electrons ejected at an angle of 90° with respect to the incident beam were investigated. Moe and Petsch[3] have measured the energy spectrum of secondary electrons produced by 0.1–0.9 keV $K^+$ ions in Ar, Ne, and Kr. In this case, electrons ejected at 0° and 90° were investigated. None of these measurements show the angular distribution of the ejected electrons, nor do they give cross sections for the production of electrons in a given energy and angular range.

In the investigation reported here, energy distributions of secondary electrons produced by 50–100 keV protons in hydrogen gas were measured at angles of 23°, 45°, 67.5°, 90°, 112.5°, 135°, and 152° from the incident proton beam direction. The experimental conditions were sufficiently precise to enable accurate relative values of the ionization cross section, differential in both energy and angle, to be obtained. Absolute values of the cross sections were determined by integrating the 50-keV results over all angles and energies of ejection and normalizing to the total ionization cross section measured by Schwirzke.[4]

## II. EXPERIMENTAL METHOD

A schematic diagram of the apparatus used for the determination of energy and angular distributions is shown in Fig. 1. A magnetically analyzed, nearly parallel, beam of protons from the Nebraska Cockcroft-Walton accelerator was collimated by two circular apertures. These apertures had knife edges so as to present a very small surface area for scattering of the beam, and the second was biased to prevent the escape of secondary electrons. The proton beam then traversed a scattering chamber containing hydrogen gas at a pressure of about $10^{-3}$ Torr and was collected in a small Faraday cup biased at +67.5 V to prevent the escape of secondary electrons. The Faraday cup and associated electrical lead were surrounded with a grounded shield to eliminate disturbance of the secondary electrons which were to be measured. Two pairs of electrostatic deflection plates were inserted between the two beam collimating apertures, allowing small changes in the proton beam direction to be made in both the horizontal and vertical plane, so that the proton beam could be accurately centered in the Faraday cup. The proton current was monitored with a galvanometer and integrated with a precision $1.003\pm0.005\,\mu$F polystyrene dielectric capacitor connected between the input and output of a Philbrick USA-3 operational amplifier.

The scattering chamber used has been described by Cook.[5] A unique sliding vacuum seal allowed the scattering chamber to be rotated over a wide angular range

* Supported in part by the U. S. Atomic Energy Commission.
† This report is based in part on a thesis submitted by C. E. Kuyatt to the University of Nebraska in partial fulfillment of the requirements for the Ph.D. degree.
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[1] E. Blauth, Z. Physik **147**, 228 (1957).
[2] H. W. Berry, ARL Technical Note 60-144, 1960 (unpublished); Phys. Rev. **121**, 1714 (1961).
[3] D. Moe and O. Petsch, Phys. Rev. **110**, 1358 (1958); **115**, 349 (1959).
[4] F. Schwirzke, Z. Physik **157**, 510 (1960).
[5] C. J. Cook, Rev. Sci. Instr. **26**, 92 (1955).

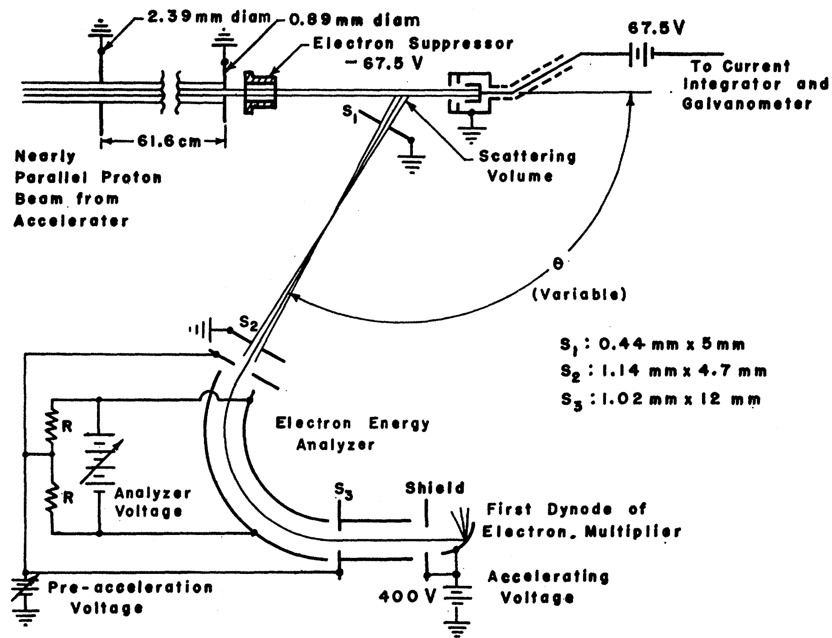


FIG. 1. Schematic diagram of the apparatus.

with respect to the proton entrance port. The angular range of secondary electrons available to the electron exit port was 23° to 152°, measured from the incident proton beam. Measurements at smaller angles were prevented by the Faraday cup used to collect the proton beam. Replacement of the natural rubber diaphragms used in the original scattering chamber with Butyl rubber diaphragms allowed a pressure of $5\text{–}10\times10^{-6}$ Torr to be obtained in the scattering chamber, when pumped by a VMF-20 diffusion pump.

Two gold slits, shown in Fig. 1, were used to select electrons in a narrow angular range. The maximum angle that electrons could make with the center line of the slit system was 0.335°. Accurate measurements of the size and spacing of the slits were made. Calculations using the results of Herb *et al.*[6] gave an effective solid angle of $2.48\pm0.002\times10^{-4}$ sr and an effective path length of scattering of $0.480\pm0.004$ mm when the slit system was at 90° with respect to the proton beam. Electrons emerging from the slit system were analyzed in energy by a 127° cylindrical focusing electrostatic analyzer similar to the one described by Hughes and McMillen.[7] The radii of the cylindrical plates were made equal to 5.00 and 6.00 cm. The analyzer is shown schematically in Fig. 1. An electron gun which provided sharply focused beams of 5 to 1000 eV was used for testing. The magnetic field in the region of the gun and analyzer was reduced to 5 mG or less with Helmholtz coils. The measured relationship between electron energy $E$ in electron volts and the voltage between the analyzer plates $V_a$ is $V_a=(0.360\pm0.004)E$. The calculated value[8] is $V_a=[2\ln(r_b/r_a)]E=0.365E$, where $r_b$=radius of outer cylinder and $r_a$=radius of inner cylinder, and agrees well with the measured value.

The resolution of the analyzer may be defined in several ways. When the "line shape" of the analyzer is determined experimentally, it is convenient and customary to measure the energy width $\Delta E_{1/2}$ between the half-maximum points and call $\Delta E_{1/2}/E$ the corresponding resolution, where $E$ is the mean energy transmitted by the analyzer. Another definition was used by Rudberg[9] and Van Atta[10] in calculating the theoretical resolution of the 127° analyzer. They calculate the base or total width of the energy distribution accepted by the analyzer, and since their results did not agree, an independent calculation was made by us. The result is essentially in agreement with Rudberg:

$$\Delta E_{\text{base}}/E=4\alpha^2/3+[(h_1+h_2)/2x]^2+(w_1+w_2)/r_0, \quad (1)$$

where $\alpha$ is the maximum angle the incoming electrons in the plane of the analyzer can make with the normal to the entrance slit, $h_1$ is the height of the first defining slit, $h_2$ is the height of the second defining slit, $r_0$ is the mean radius of the analyzer, $x$ is the distance between the first and second defining slits, $w_1$ is the width of the analyzer entrance slit, and $w_2$ is the width of the analyzer exit slit.

A third definition of resolution is of particular interest when measuring cross sections and will be called the effective resolution $\Delta E/E$. Assume that a beam of electrons with a "white" energy distribution enters the

[6] R. G. Herb, D. W. Kerst, D. B. Parkinson, and G. J. Plain, Phys. Rev. **55**, 998 (1939).

[7] A. L. Hughes and J. H. McMillen, Phys. Rev. **34**, 291 (1929); **39**, 585 (1932).

[8] A. L. Hughes and V. Rojansky, Phys. Rev. **34**, 284 (1929).

[9] E. Rudberg, Proc. Roy. Soc. (London) **A129**, 628 (1930).

[10] L. C. Van Atta, Phys. Rev. **38**, 876 (1931).

analyzer, where $j$ is the current per unit energy interval. Then, the current transmitted by the analyzer is $j\tau\Delta E$ where $\tau$ is the effective transmission of the analyzer, that is, the maximum transmission of the analyzer for a monoenergetic electron beam.

The relationship between the three resolutions is not obvious since it depends on the detailed line shape. Kuyatt and Rudd[11] have shown that if the first two terms in Eq. (1) are small compared to the third term, then $\Delta E_{1/2}/E$ and $\Delta E/E$ are very nearly the same and equal to $w_{\max}/r_0$ where $w_{\max}$ is the larger of $w_1$ and $w_2$. The effective transmission is 1.0 if $w_1 \leq w_2$ and equal to $w_2/w_1$ if $w_1 > w_2$. Using very general assumptions they find that $\tau\Delta E/E$ is equal to $w_2/r_0$ and does not depend on the size of the entrance slit nor on the distribution of electrons in position or direction at the entrance slit. The first two terms on the right-hand side of Eq. (1) produce a percentage energy shift one-third as large as their contribution to base resolution. For the analyzer and slits used in this research the effective resolution is 2.1%, the effective transmission is 88%, and the energy shift is negligible. The corresponding base resolution is 4.0%.

Tests of the analyzer were made using slits of somewhat different dimensions than the gold slits finally used. The value obtained from measurements at several energies was $\Delta E_{1/2}/E = (2.25 \pm 0.25)\%$, in good agreement with the calculated value of 2.2%.

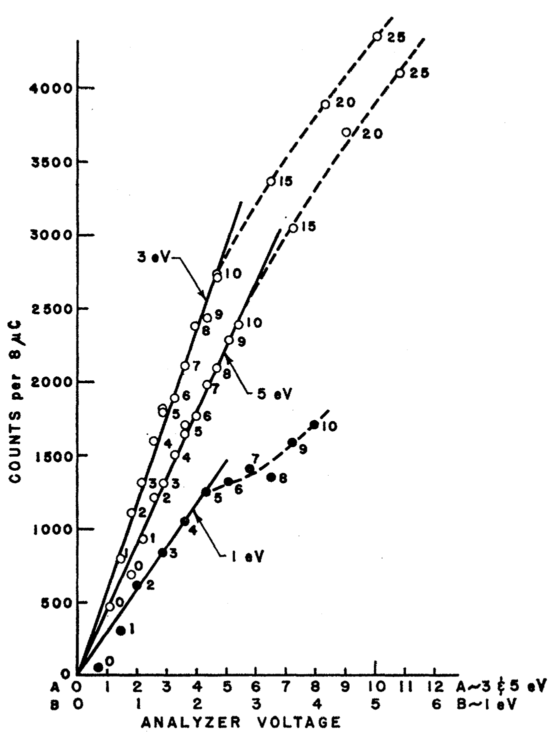


FIG. 2. Effect of preacceleration on the resolution of the analyzer. Electrons of 1, 3, and 5 eV, produced by 8 $\mu$C of 100-keV protons in hydrogen gas, are analyzed using different combinations of preacceleration and analyzer voltage. A straight line through the origin is expected. The small figures on the curves show the amount of preacceleration in volts.

Provision was made for accelerating electrons just before entering the analyzer. The accelerating voltage was placed on a slit just after and slightly larger than the analyzer entrance slit. The accelerating slit, the analyzer exit slit, and the midpoint of the analyzer voltage supply were connected together as shown in Fig. 1. In tests using the electron gun the electrons were accelerated to at least 20 eV before entering the analyzer. Tests were made for electrons with energy down to 5 eV. Hughes and McMillen[7] made measurements on electrons down to at least 1 eV, but had to accelerate the electrons to at least 40 eV for the analyzer to function. Rudberg[9,12] could not obtain satisfactory operation of his analyzer with electrons of less than about 70 eV. Mohr and Nicoll,[13] using a bakeable analyzer trapped with liquid air, analyzed electrons of energy down to 16.3 eV without accelerating, but usually accelerated to 30 eV, allowing measurements of electrons to 3.4 eV.

The analyzer described here is believed to operate well for electrons down to at least 3 eV because of the following features not found in the other analyzers: (1) The ambient magnetic field, mainly due to the earth's field, is annulled by Helmholtz coils to within 1 mG. (2) All slits used with the analyzer are made of gold and the analyzer plates are gold plated. In this way, contact differences of potential are minimized and the production of insulated layers on the slits and plates is retarded. These insulating layers can become charged and produce extraneous electric fields. (3) Due to the high energy and angular resolution the electron current in the slit system and analyzer was very small and reduced any possible effects of charging.

A sensitive test of analyzer operation was made by observing electrons of a given energy from hydrogen gas with different combinations of accelerating and analyzer voltage. The number of counts produced by 8 $\mu$C of protons for each combination was plotted as a function of analyzer voltage and is shown in Fig. 2. A straight line should be obtained since the energy range of electrons accepted by the analyzer is directly proportional to the analyzer voltage. Tests were made on 1-, 3-, and 5-eV electrons. The results show the predicted straight-line behavior when the accelerating voltage is not too high. For an acceleration of 10 V, used in all measurements reported here, there is negligible effect on electrons of 3 eV and above, and a 30% reduction of 1-eV electrons.

The analyzer was also tested for the effects of contact difference of potential between its various parts, including the electron collimating slits. With an accelera-

[11] C. E. Kuyatt and M. E. Rudd (to be published).

[12] E. Rudberg, Proc. Roy. Soc. (London) **A130**, 182 (1930).

[13] C. B. O. Mohr and F. H. Nicoll, Proc. Roy. Soc. (London) **A138**, 229 (1932); **A144**, 596 (1934). F. H. Nicoll and C. B. O. Mohr, *ibid.* **A142**, 320 (1933).

tion of 10 V, the number of counts per 8 $\mu$C of protons was measured as a function of electron energy in the analyzer. Readings were taken for a range of voltages both above and below the analyzer voltage which was expected to correspond to zero energy electrons. For voltages below this value the number of counts dropped sharply to zero, consistent with the resolution of the analyzer which was 0.2 eV, and showed that the energy scale was not displaced by more than 0.1 eV. This test also demonstrated the absence of extraneous electrons produced by secondary emission or reflection.

A ten-stage electron multiplier was used to count single electrons from the analyzer. It had the outstanding advantage of a very low background counting rate, about 4 to 10 counts per minute. The multiplier was prepared from a DuMont 6292 photomultiplier tube essentially as described by Koyama and Connally.[14] The base of the tube was first removed and the wiring done directly on the wires emerging from the glass envelope. The photocathode was then cut off with a glass saw about $\frac{1}{2}$ in. from the flat end, washed successively with distilled water, absolute ethyl alcohol, and trichloroethylene, and immediately mounted in the vacuum system with an O-ring seal.

The base of the tube was removed because spurious pulses were produced, probably by dielectric breakdown, in both the base of the 6292 and in DuMont mica-bakelite sockets. For nearly complete elimination of spurious pulses it was found necessary to spray the wiring with Krylon and flow dry nitrogen through the wiring space. The tube was operated at 370 V per dynode stage and produced an average current gain of about $6\times10^4$ when electrons from the electrostatic analyzer were accelerated to 400 eV before striking the first dynode. Since the first dynode was required to be at 400 V above ground, the anode of the multiplier was required to be at 3900 V above ground, necessitating a high quality capacitor to block the dc level from the signal. The high voltage power supply must be free from transients and have a very low ripple since either will be part of the signal and may contribute spurious pulses or overload the amplifier. The power supply used consisted of a transformer-rectifier-filter supplying 51 85A2 glow tubes in series, with a ripple of 6 mV rms. An additional *RC* filter was required.

By careful wiring an anode capacitance of 10 pF was achieved, giving average pulse heights of about 1 mV for single electrons. Standard electronics were used to amplify and count the pulses. A differential pulse-height distribution similar to that of Koyama and Connally[14] was obtained. Integral counting curves showed that nearly all single electron pulses were counted. To maintain the gain of the multiplier at a sufficiently high value, it was found necessary to shine an infrared lamp on the base of the tube at all times except when making measurements.

[14] K. Koyama and R. E. Connally, Rev. Sci. Instr. **28**, 833 (1957).

Some careful measurements which have been made of the efficiency of counting electrons with an electron multiplier showed an efficiency of 65 to 75% for carefully prepared tubes.[15] No direct measurements of efficiency were made for the multiplier used for the measurements reported here, but the normalization of the results, described later, gave an efficiency of 27% which is not unreasonable in view of the exposure of the dynode surfaces to air, solvents, and the dynamic vacuum system.

Commercial tank hydrogen of purity 99.5% or better was introduced into the scattering chamber through a "Deoxo" filter, two successive cold traps cooled with dry ice and acetone, and a sensitive needle valve. A steady flow through a "bubbler" was maintained to prevent buildup of impurities. The diffusion pump on the scattering chamber was throttled with a butterfly valve before letting in the hydrogen gas, causing the residual pressure to rise to no more than $1.0\times10^{-5}$ Torr as indicated by a dry ice trapped VG-1A ionization gauge. The scattering chamber was separated from the accelerator vacuum system by a 0.89-mm-diam aperture, and from the analyzer and electron-multiplier vacuum system by a 0.5-mm by 5.0-mm slit. The latter system was pumped with a VMF-10 diffusion pump and had a base pressure of $5\times10^{-6}$ Torr as read on a dry ice trapped VG-1A gauge. This pressure increased to about $3\times10^{-5}$ Torr when the scattering chamber contained hydrogen at a pressure of $10^{-3}$ Torr. The effect on the accelerator pressure was negligible. Measurement of the pressure of hydrogen gas in the scattering chamber was made with a VG-1A ionization gauge which was calibrated to within 10% by a McLeod gauge.

To ensure negligible disturbance to low-energy electrons, the ambient magnetic field was reduced to $10^{-3}$ G or less with three pairs of coils. Field measurements were made with an electronic magnetometer[16] with a sensitivity of 0.07 mG. It was found that 60 cps magnetic fields with sufficient amplitude to disturb low-energy electrons were present in the measurement region. These fields were reduced to a negligible level by introducing compensating currents into each of the coil pairs.

## III. MEASUREMENTS

With a proton beam of about 0.1 to 0.3 $\mu$A, and hydrogen in the scattering chamber at a pressure of about $10^{-3}$ Torr, the number of electron counts corresponding to 4 or 8 $\mu$C of protons was recorded for about 30 settings of the analyzer voltage, covering an electron range of 0.2 to 100 eV. At small angles measurements were made to 300 eV. The hydrogen gas supply was

[15] J. A. Cowan, Can. J. Phys. **32**, 101 (1954); W. P. Alford and D. R. Hamilton, Phys. Rev. **105**, 673 (1957).

[16] C. E. Kuyatt, Ph.D. Thesis, University of Nebraska, 1960 [(unpublished) available from University Microfilms, Ann Arbor, Michigan].

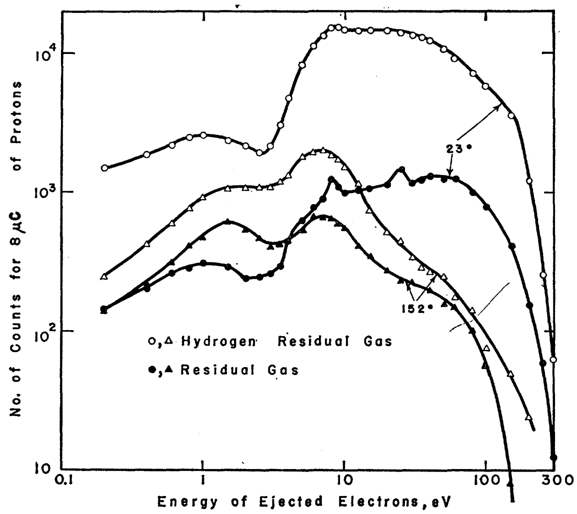


FIG. 3. Typical experimental data for 100-keV protons in hydrogen gas at about $10^{-3}$ Torr.

then shut off, the pressure allowed to come to an equilibrium value, and the series of measurements repeated on the residual gas in the scattering chamber. Figure 3 shows some typical results for the extreme angles 23° and 152°. At 23° the background counting rate was about 10% of that with hydrogen in the scattering chamber. At 152° the background was 50% of the counting rate for hydrogen, showing that production of secondary electrons from the residual gas falls off much slower with angle than from hydrogen. The background curve has a similar shape to that for hydrogen and was quite reproducible showing only small changes from day to day. The structure below 3 eV is probably associated with preaccelerator distortion and by absorption in the residual gas.

Several experimental tests of the apparatus were made. One such test was to repeat a given measurement several times to see if the variations were within normal counting statistics. In every case the mean deviations were close to the square root of the number of counts which is the expected statistical error.

Several plots were made of the number of counts as a function of the gas pressure, resulting in straight lines to within the counting statistics. This result showed that the increase in number of counts is directly proportional to the increase in pressure of hydrogen gas, and demonstrated the absence of multiple collisions of the incident protons and the absence of absorption of electrons in the hydrogen gas before reaching the detector.

Early attempts to test for possible dependence of results on beam intensity gave a change in counting rate of about 15% for a beam intensity ratio of ten to one. This change was demonstrated to be caused by two other effects: The proton beam was striking the edge of the Faraday cup and producing extraneous electrons and the analyzer slits and plates were charging due to insulating layers. The first effect was eliminated by reducing the second proton collimating aperture from 1.40-mm to 0.89-mm diameter. The second effect was eliminated by making the analyzer slits out of gold sheet and having the plates gold plated. After these changes there was no observable dependence on beam intensity. To further verify that the Faraday cup was collecting all of the proton beam, the number of counts per 8 μC collected in the cup was measured for positions of the cup 5, 10, and 15 mils above its usual position. Each time the number of counts was the same to within the statistical error. Another test, in which the Faraday cup bias was increased from $67\frac{1}{2}$ to 135 V, showed that no appreciable quantity of secondary electrons was escaping from the cup.

The differential cross section for production of secondary electrons is defined by the relation[6]:

$$N_e = N_p \sigma(E,\theta) \tau n l \csc\theta d\Omega dE, \qquad (2)$$

where $N_e$ is the number of secondary electrons; $N_p$ is the number of incident protons; $\sigma(E,\theta)$ is the cross section, per unit solid angle and unit energy range, for production of secondary electrons of energy $E$ at an angle $\theta$ from the incident proton beam (cm²/eV-sr); $\tau$ is the effective transmission of the detector; $d\Omega$ is the solid angle intercepted by the detector (sr); $dE$ is the effective electron energy range accepted by the detector (eV); $n$ is the number of gas molecules per cm³; and $l$ is the effective thickness of the gas target when the detector slit system is set at $\theta = 90°$ (cm).

The method of calculating $l$ and $d\Omega$ from the geometry of the detector slits is given by Herb *et al.*[6] Using carefully measured dimensions, the values obtained were $l = 0.480 \pm 0.004$ mm and $d\Omega = (2.48 \pm 0.02) \times 10^{-4}$ sr.

In calculating the number of gas molecules per cm³ from the measured hydrogen gas pressure, it was assumed that the temperature of the gas was equal to the temperature of the scattering chamber.

The number of scattered electrons is determined by subtracting the number of counts $N_2$ from the residual gas from the number of counts $N_1$ with hydrogen in the scattering chamber, and dividing by the efficiency of the electron multiplier. The efficiency was assumed to be 65% for the 50-keV data and the differential cross section calculated. Extrapolation and integration over all angles and energies of ejection should give the total ionization cross section $\sigma_i$:

$$\sigma_i = \int_0^\infty \int_0^\pi \sigma(E,\theta) 2\pi \sin\theta d\theta dE. \qquad (3)$$

Using the measured value of Schwirzke[4] at 50 keV of $2.6 \times 10^{-16}$ cm²/molecule, the efficiency of the multiplier was calculated to be 27%. This value was then used to calculate differential cross sections at 50, 75, and 100 keV.

The error due to counting statistics when two independent counting measurements are subtracted is the square root of the sum of the squares of the individual statistical errors.[17] In this case the individual statistical errors are $N_1^{1/2}$ and $N_2^{1/2}$. Hence, the statistical error of the difference of $N_1$ and $N_2$ is given by $(N_1+N_2)^{1/2}$.

## IV. RESULTS AND DISCUSSION

Measurements were made for incident protons of 50, 75, and 100 keV, the electron energy distribution being taken at angles of 23°, 45°, 67.5°, 90°, 112.5°, 135°, and 152° from the incident proton direction. Differential cross sections were computed from Eq. (2) as discussed above and are shown in Figs. 4, 5, and 6. The cross sections at 112.5°, 135°, and 152° are nearly the same as at 90° and are omitted from the figures for clarity. The error bars shown when possible on the data points at 4, 10, 20, 40, 60, and 80 eV represent the estimated effects of counting statistics and errors in measuring the relative pressure of the scattering gas (3%) and the number of incident protons (2%). These errors are combined in rms fashion.

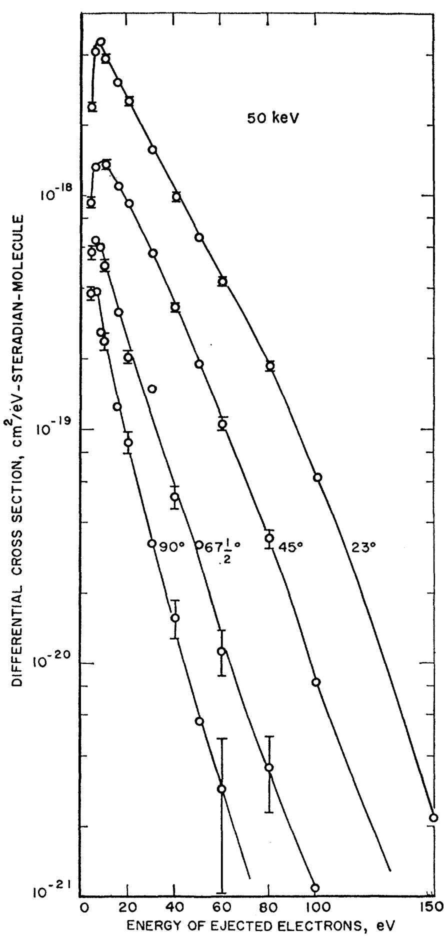


FIG. 4. Differential cross section for production of secondary electrons by 50-keV protons in hydrogen gas. Error bars shown where possible at 4, 10, 20, 40, 60, and 80 eV represent the estimated error due to counting statistics, pressure measurements, and measurement of the number of incident protons.

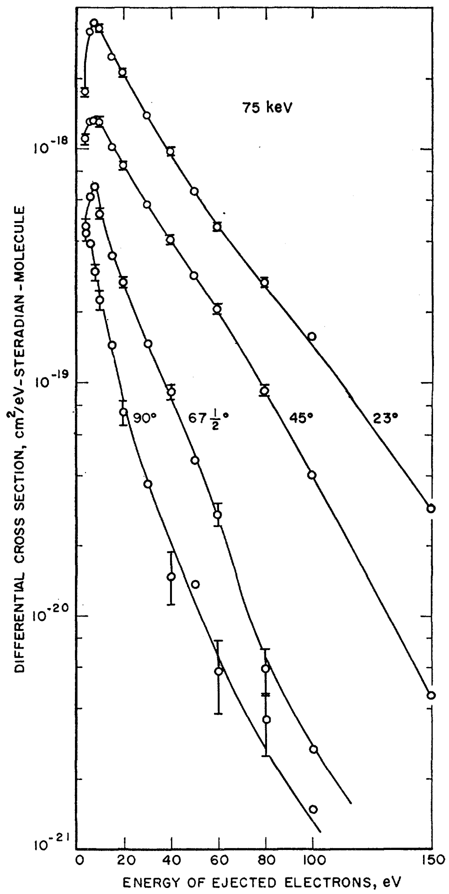


FIG. 5. Differential cross section for production of secondary electrons by 75-keV protons in hydrogen gas. Error bars shown where possible at 4, 10, 20, 40, 60, and 80 eV represent the estimated error due to counting statistics, pressure measurement, and measurement of the number of incident protons.

All of the differential cross section curves show a maximum for electron energies between 4 and 8 eV. At higher electron energies the differential cross section decreases monotonically for all angles investigated. Very recent measurements made in this laboratory using an improved scattering chamber show a maximum at 1 to 2 eV. The difference is thought to be due to the much better vacuum obtained in the new apparatus, the peak in the present results being caused by absorption of low-energy electrons in the background gas. The present results below about 8 eV are low but are presented because the angular distribution at a fixed electron energy should be little affected by absorption.

A fairly direct comparison of the present results with the work of Blauth[1] is possible. He measured the energy distribution of secondary electrons ejected at 54.5° for protons of 8.8, 11.8, and 49 keV in hydrogen gas. A

[17] E. B. Mode, *Elements of Statistics* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1958), 2nd ed., p. 195.

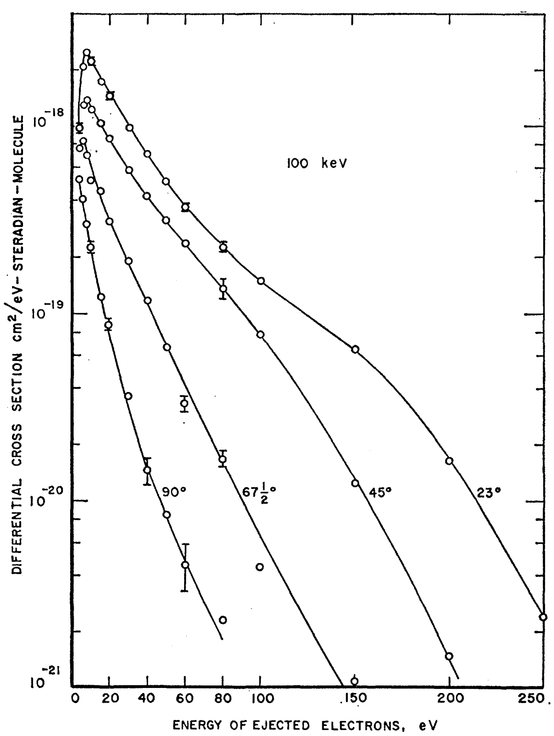


FIG. 6. Differential cross section for production of secondary electrons by 100-keV protons in hydrogen gas. Error bars shown where possible at 4, 10, 20, 40, 60, and 80 eV represent the estimated error due to counting statistics, pressure measurement, and measurement of the number of incident protons.

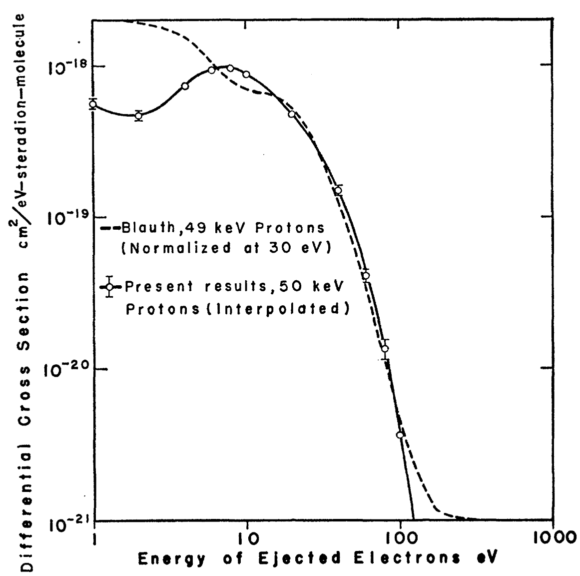


FIG. 7. Comparison of the present differential cross sections with the relative measurements of Blauth. The present results for 50-keV protons were interpolated for 54.5° ejection of electrons. Blauth's results for ejection of electrons at 54.5° by 49-keV protons were normalized to the present results at 30 eV.

comparison of his results for 49-keV protons and the present results for 50-keV protons interpolated for 54.5° is shown in Fig. 7. Since Blauth does not give cross sections, his results are normalized to give agreement at 30 eV. With this normalization the agreement is within 20% from 6 to 100 eV. Below 6 eV and above 100 eV, Blauth's results are much higher. Particularly surprising is the nearly constant cross section measured by Blauth from 200–800 eV. The present results show a cross section which drops smoothly toward zero as the electron energy increases, in accord with the Born approximation treatment of ionization of hydrogen atoms by protons.[18,19] Blauth's somewhat anomalous results may be caused by the production of extraneous electrons in his analyzer by secondary emission or reflection. Absorption of electrons in the background gas in the present experiment may account for the disagreement below 6 eV.

By plotting the angular variation of the differential cross section for various energies of the ejected electrons the following characteristics are observed: The differential cross section drops off rapidly with increasing angle to about 100°, remains nearly constant at larger angles, and usually shows a moderate rise at the largest angle.

By integrating the differential cross sections over the energy of the ejected electrons at various angles of ejection, the cross section $\sigma(\theta)$ for ejection of electrons of all energies, per unit solid angle, as a function of angle is obtained:

$$\sigma(\theta)=\int_0^\infty \sigma(E,\theta)dE. \qquad (4)$$

The results are shown in Fig. 8, plotted as a function of $\cos\theta$. The curves for 75- and 100-keV incident protons are displaced upward one and two decades, respectively, to eliminate confusion from overlapping. Although the curves are similar in shape, it is interesting to note that the 50-keV protons produce the most electrons at small and large angles, while the 100-keV protons produce the most electrons at the intermediate angles. The production of electrons by 75-keV protons is intermediate except in the transition regions at 25°–37° and at 89°–97°.

The cross section for emission of electrons into a unit energy interval, integrated over all angles of emission, has also been obtained. Let this cross section be denoted by $\sigma(E)$. Then

$$\sigma(E)=\int\sigma(E,\theta)d\Omega=2\pi\int_0^\pi \sigma(E,\theta)\sin\theta d\theta. \qquad (5)$$

The integration is facilitated by using $\cos\theta$ as the variable of integration rather than $\theta$. Using primes to

[18] H. Bethe, Ann. Physik 5, 325 (1930).
[19] D. R. Bates and G. W. Griffing, Proc. Phys. Soc. (London) A66, 961 (1953).

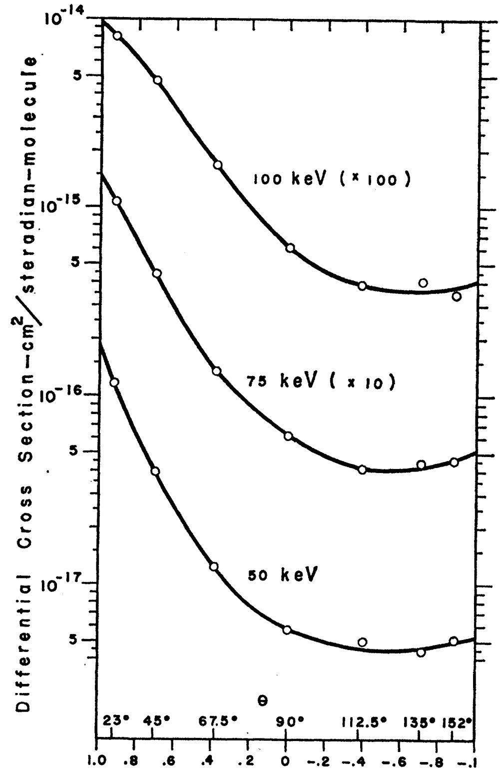


FIG. 8. Differential cross section for ejection of electrons of all energies as a function of the cosine of the angle of ejection. The incident proton energy is noted on the curves. The results for 75- and 100-keV protons are multiplied by 10 and 100, respectively.

indicate cross sections as a function of cos$\theta$, we have

$$\sigma(E)=2\pi\int_{-1}^{+1}\sigma'(E,\cos\theta)d(\cos\theta). \qquad (6)$$

To perform the integration it was necessary to extrapolate the differential cross sections to $\theta=0°$ and 180°. The results are shown in Fig. 9, with the 50- and 75-keV curves displaced upward for clarity. The curves have a pronounced peak at 7–8 eV (presumably due to the previously mentioned background absorption) and drop smoothly toward zero for higher electron energies. At low electron energies, 50-keV protons produce the most electrons, while at large electron energies, 100-keV protons produce the most electrons.

The integral of $\sigma(E)$ over all energies is the total ionization cross section $\sigma_i$. This cross section may also be obtained by integrating the cross section $\sigma(\theta)$ over all angles:

$$\sigma_i=2\pi\int_0^{\pi}\sigma(\theta)\sin\theta d\theta=2\pi\int_{-1}^{+1}\sigma'(\cos\theta)d(\cos\theta). \qquad (7)$$

The integral has been simplified by using cos$\theta$ as a variable and letting $\sigma'$ indicate the cross section as a

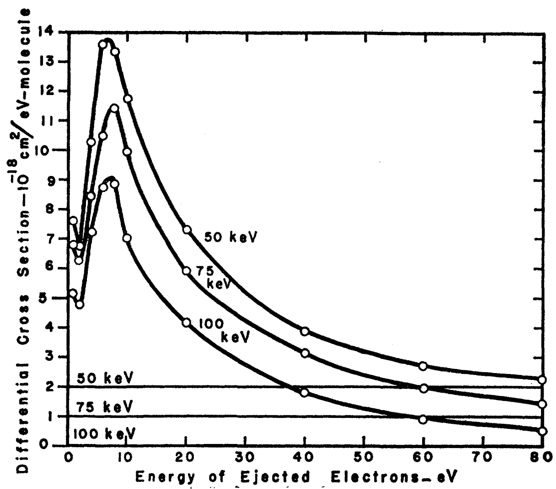


FIG. 9. Differential cross section for ejection of electrons at all angles as a function of the energy of ejection. The base line for each curve is labeled with the appropriate proton energy.

function of cos$\theta$. To evaluate Eq. (7), $\sigma(\theta)$ must be extrapolated to $\theta=0°$ and 180°. This extrapolation is easier when performed on $\sigma'(\cos\theta)$ as shown in Fig. 8. The total ionization cross sections obtained by evaluating the two integrals numerically are shown in Table I. Equation (7) was used for normalization to Schwirzke's measured value at 50 keV. The two methods give values which differ by 3% or less, showing that the extrapolations involved in the two methods are consistent. Values of the total ionization cross section from Eq. (7) are also plotted in Fig. 10, along with the measured values of Schwirzke,[4] Hooper *et al.*,[20] Afrosimov *et al.*,[21] Gilbody and Hasted,[22] Fogel *et al.*,[23] and Keene,[24] and the theoretical values of Bates and

TABLE I. Total ionization cross section for protons in hydrogen gas, obtained by integrating the measured differential cross sections over all angles and energies.

| | Total ionization cross section (units of $10^{-16}$ cm²/molecule) | |
|---|---|---|
| Proton energy (keV) | Integration over energy, then angle | Integration over angle, then energy |
| 50 | 2.60[a] | 2.63 |
| 75 | 2.59 | 2.67 |
| 100 | 2.40 | 2.38 |

[a] This value has been normalized to the measured cross section of Schwirzke, reference 4.

[20] J. W. Hooper, E. W. McDaniel, D. W. Martin, and D. S. Harmer, Phys. Rev. **121**, 1123 (1961).

[21] V. V. Afrosimov, R. N. Il'in, and N. V. Fedorenko, Zh. Eksperim. i Teor. Fiz. **34**, 1398 (1958) [translation: Soviet Phys.—JETP **7**, 968 (1958)].

[22] H. B. Gilbody and J. B. Hasted, Proc. Roy. Soc. (London) **A240**, 382 (1957).

[23] Ia. M. Fogel, L. I. Krupnik, and B. G. Safronov, Zh. Eksperim. i Teor. Fiz. **28**, 589 (1955) [translation: Soviet Phys.—JETP **1**, 415 (1955)].

[24] J. P. Keene, Phil. Mag. **40**, 369 (1949).

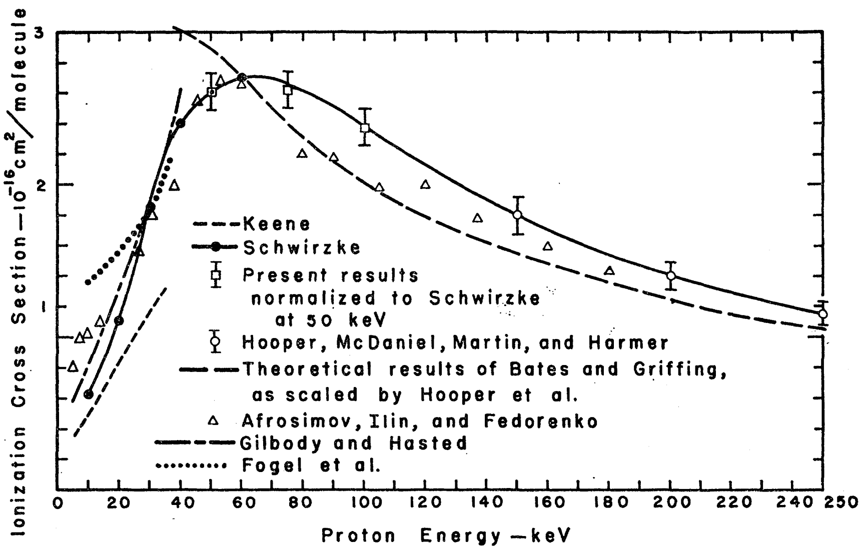


FIG. 10. Total ionization cross section for protons in hydrogen gas. The cross sections obtained in the present work are used to connect the low-energy results of Schwirzke and the high-energy results of Hooper *et al.* Results of other experiments and theory are shown for comparison.

Griffing[19] as scaled for molecular hydrogen by Hooper *et al.* The integrated ionization cross sections from the present measurements, the results of Schwirzke (10–60 keV), and those of Hooper *et al.* (150 keV and up) are connected with a smooth curve which exhibits a maximum of $2.7\times10^{-16}$ cm$^2$/molecule at a proton energy of 65 keV. On the low-energy side of the peak the results of Gilbody and Hasted agree with those of Schwirzke to within the combined experimental error. Keene's values appear to be too low, while the measurements of Fogel *et al.* are too high at low proton energies but are in good agreement for energies above 25 keV. The measurements of Afrosimov *et al.* are below the composite curve but within the combined errors.

In view of the excellent internal consistency of the integrations used to obtain relative total ionization cross sections from the differential cross sections, and considering the good fit to the measurements of Schwirzke and Hooper *et al.*, an estimated error of 5% has been assigned to the *relative* total cross sections. On the basis of Schwirzke's estimate of a maximum error of 10% for his measurements, the 50-keV cross section has been assigned an absolute error of 10% and the 75-keV and 100-keV cross sections have been assigned an absolute error of 15%. An additional error of 5% has been added to obtain an estimate of the normalization error of the differential cross sections. This error is, of course, in addition to the random errors discussed above and indicated in Figs. 4, 5, and 6 by error bars.

Four additional quantities have been calculated from the experimental results: (1) The average energy $E_{\rm av}$ acquired by an electron in an ionizing collision:

$$E_{\rm av}=\int_0^\infty E\sigma(E)dE\Big/\int_0^\infty \sigma(E)dE. \qquad (8)$$

(2) The average energy $\Delta\mathcal{E}_{\rm av}$ lost by a proton in an ionizing collision:

$$\Delta\mathcal{E}_{\rm av}=E_{\rm av}+15.6 \text{ eV}.$$

(3) The average energy lost by a proton in producing an ion pair when account is taken of ion pairs produced by the ejected electrons. Dalgarno and Griffing[25] have calculated the number of ion pairs $N(E)$ produced when an electron of energy $E$ is completely absorbed in a gas of hydrogen atoms. Using their result the average number of ion pairs $N_{\rm av}$ produced by the ejected electrons has been calculated from

$$N_{\rm av}=\int_0^\infty N(E)\sigma(E)dE\Big/\int_0^\infty \sigma(E)dE. \qquad (9)$$

Taking account of the ion pair produced in the primary ionizing collision, the mean specific energy per ion pair $w_p$ for ionizing collisions of protons in hydrogen gas is $w_p=\Delta\mathcal{E}_{\rm av}/(N_{\rm av}+1)$.

(4) The rate of energy loss of a proton due to ionization $(d\mathcal{E}/dx)_i$:

$$(d\mathcal{E}/dx)_i=\int_0^\infty (E+15.6 \text{ eV})\sigma(E)dE. \qquad (10)$$

The calculated quantities $E_{\rm av}$, $\Delta\mathcal{E}_{\rm av}$, $N_{\rm av}$, $w_p$, and $(d\mathcal{E}/dx)_i$ are tabulated in Table II.

An examination of Table II shows that as the proton energy increases there is a corresponding increase in the average energy $\Delta\mathcal{E}_{\rm av}$ lost by a proton in an ionizing collision, the average energy $E_{\rm av}$ of the ejected electrons, and the average number of ion pairs $N_{\rm av}$ produced by the ejected electrons. These quantities combine to give a nearly constant value for the mean energy $w_p$ lost by protons in ionizing collisions in hydrogen for each ion

[25] A. Dalgarno and G. W. Griffing, Proc. Roy. Soc. (London) A248, 415 (1958).

pair formed in primary and secondary processes. Experimental measurements of energy loss per ion pair $W$ for protons in hydrogen gas have been analyzed by Gray[26] who derives a value of $35\pm1.5$ eV per ion pair for impact energies greater than 30 keV. The experiments are performed by completely stopping protons in hydrogen gas. Hence, the experimental quantity is an average over energies from zero to the incident energy. Charge exchange introduces a further complication. Hence no detailed comparison will be made. It will only be pointed out that because of excitation and charge exchange, $W$ is expected to be higher than $w_p$ and this is borne out by the present results.

The experimental results of Dunbar *et al.* from the review article of Allison and Warshaw[27] are also given in Table II, labeled $(d\mathcal{E}/dx)_{\text{exp}}$. Because the measurements of Dunbar *et al.* are made with a beam of protons and hydrogen atoms in charge equilibrium, no special significance should be attached to the fair agreement with the values of $(d\mathcal{E}/dx)_i$ from the present work.

TABLE II. Values of quantities calculated from the measured differential cross sections, together with some experimental and theoretical results for comparison.

| Proton energy | 50 | 75 | 100 | keV |
|---|---|---|---|---|
| $E_{\text{av}}$[a] | 22.4 | 27.9 | 36.2 | eV |
| $\Delta\mathcal{E}_{\text{av}}$[b] | 38.0 | 43.5 | 51.8 | eV |
| $N_{\text{av}}$[c] | 0.35 | 0.53 | 0.77 | ion pair/ejected electron |
| $w_p$[d] | 28 | 28.5 | 29 | eV/ion pair |
| $(d\mathcal{E}/dx)_i$[e] | 9.9 | 11.4 | $12.4\times10^{-15}$ | eV-cm²/molecule |
| $W_{\text{exp}}$[f] | 35 | 35 | 35 | eV/ion pair |
| $(d\mathcal{E}/dx)_{\text{theor}}$[g] | 10.4 | 9.4 | $8.4\times10^{-15}$ | eV-cm²/molecule |
| $(d\mathcal{E}/dx)_{\text{exp}}$[h] | 12.9 | 12.6 | $11.2\times10^{-15}$ | eV-cm²/molecule |

[a] Average energy acquired by an electron in an ionizing collision, calculated from the present data using Eq. (8).
[b] Average energy lost by a proton in an ionizing collision, calculated from the present data: $E_{\text{av}}+15.6$ eV.
[c] Average number of ion pairs produced by the ejected electrons, calculated from Eq. (9) using the present data and the results of Dalgarno and Griffing, reference 25.
[d] Mean specific energy per ion pair for ionizing collisions of protons in hydrogen, calculated from the present data: $\Delta\mathcal{E}_{\text{av}}/(N_{\text{av}}+1)$.
[e] Rate of energy loss of protons due to ionization, calculated from the present data using Eq. (10).
[f] Experimental value of energy per ion pair produced by completely stopping protons in hydrogen gas, from reference 26.
[g] Rate of energy loss of protons in atomic hydrogen, calculated by Dalgarno and Griffing, reference 28.
[h] Experimental rate of energy loss of protons in hydrogen gas, from reference 27.

## V. COMPARISON WITH BORN APPROXIMATION RESULTS

At present there exists no theoretical method which would be expected to give accurate differential cross sections for ionizing collisions of protons in the energy range used in this experiment. The only theoretical method which has been applied to ionizing collisions of protons in this energy range is the Born approximation, even though it is expected to be valid only at somewhat higher energies. Measurements by Fite *et al.*[28] indicate that in the case of ionization of hydrogen atoms by protons the Born approximation is probably valid for energies of 100 keV and above.

Since no calculations have been made for proton collisions with hydrogen molecules, experimentally measured cross sections for hydrogen molecules have, in the past, been compared to twice the calculated cross sections for hydrogen atoms, or for hydrogen atoms in which the binding energy has been altered to agree with the ionization potential of the hydrogen molecule. The procedure of equating a hydrogen molecule to two hydrogen atoms for the purpose of comparison with experiment has been questioned by Dalgarno and Griffing,[29] and has been examined by Tuan and Gerjuoy[30] in the case of charge transfer and found to have serious theoretical objections. The validity of this procedure for ionizing collisions has not been examined theoretically. A study of Fig. 10 shows that the measured total ionization cross section for energies of 50 keV and above is within 15% of that predicted by the Born approximation[19] on the assumption that a hydrogen molecule is equivalent to two hydrogen atoms. The agreement is within the experimental uncertainties, although the shapes of the curves differ for energies below about 80 keV. As in the case of charge exchange[31] the agreement may be fortuitous, but until this has been shown to be the case, the range of validity can only be tested by comparison with experimental results.

Since the Born approximation procedure gives reasonably good agreement with experimental results for the total ionization cross section in the energy range of the present experiment, it seems reasonable to make similar comparisons of differential cross sections. Such comparisons may give further information on the range of validity of the Born approximation and how it breaks down.

Bates and Griffing[19] have calculated the cross section for ejection of electrons into a unit energy interval, integrated over all angles of emission, for protons of 3.2, 32, 320, and 3200 keV incident on hydrogen atoms. Figure 11 compares the theoretical results for 32-keV protons and the experimental results for 50-keV protons. The agreement is fair. It would be very desirable to have more calculated values so that a detailed comparison could be made.

There are no published theoretical results which can be directly compared with the measured differential cross sections. Massey and Mohr[32] have performed the corresponding calculation for electron impact on hydrogen like atoms, and Dalgarno and Griffing[25] have

[26] L. H. Gray, Proc. Cambridge Phil. Soc. **40**, 72 (1944).
[27] S. K. Allison and S. D. Warshaw, Rev. Mod. Phys. **25**, 779 (1953).
[28] W. L. Fite, R. F. Stebbings, D. G. Hummer, and R. T. Brackmann, Phys. Rev. **119**, 663 (1960).
[29] A. Dalgarno and G. W. Griffing, Proc. Roy. Soc. (London) **A232**, 423 (1955).
[30] T. F. Tuan and E. Gerjuoy, Phys. Rev. **117**, 756 (1960).
[31] R. H. Bassel and E. Gerjuoy, Phys. Rev. **117**, 749 (1960).
[32] H. S. W. Massey and C. B. O. Mohr, Proc. Roy. Soc. (London) **A140**, 613 (1933).

calculated the cross section differential in both energy and angle, for ejection of electrons in hydrogen atom–hydrogen atom collisions. Although the Massey and Mohr calculation can be extended to proton impact on hydrogen-like atoms, no numerical results have been reported. Therefore, such a calculation has been carried out.

The starting point for the calculation is the cross section, calculated in Born approximation, for the ejection of electrons from hydrogen atoms by incident electrons. This cross section has been given by Massey and Mohr,[32] Mott and Massey,[33] Massey,[34] and Landau and Lifschitz.[35] Only the result given by Landau and Lifschitz is free from misprints. The extension of the cross section to proton collisions is straightforward[36] and results in multiplication of the electron cross section by $(M/m)^2$, and suitable modification of the conservation of energy equation. The resulting cross section is differential in the direction of scattering of the incident proton, and in the direction of the ejected electron. To enable comparison with experiment the cross section must be integrated over all directions of scattering of the incident proton, a two-dimensional integration. The integral can be reduced to a one-dimensional integral by integrating over the angle $\gamma$ between the momentum change vector $q$ of the incident proton and the momentum vector $\kappa$ of the ejected electron. The result, in atomic units,[37] is

$$d\sigma'=\frac{2^8M^2\kappa}{qk^2}\frac{\exp\{-(2/\kappa)\tan^{-1}[2\kappa/(q^2-\kappa^2+1)]\}}{[(q+\kappa)^2+1][(q-\kappa)^2+1][1-\exp(-2\pi/\kappa)]}\frac{CD^3+4CDE^2-4BD^2E-BE^3+2AD^3+3ADE^2}{(D^2-E^2)^{7/2}}dqd\Omega d\kappa, \quad (11)$$

where $k$ = momentum of incident proton, and

$$A=q^2-2q_m\kappa\cos\theta+(\kappa^2+1)(q_m/q)^2\cos^2\theta,$$
$$B=2(q^2-q_m^2)^{1/2}\kappa\sin\theta-(\kappa^2+1)(2q_m/q^2)(q^2-q_m^2)^{1/2}\sin\theta\cos\theta,$$
$$C=(\kappa^2+1)[(q^2-q_m^2)/q^2]\sin^2\theta,$$
$$D=q^2-2q_m\kappa\cos\theta+\kappa^2+1,$$
$$E=2\kappa(q^2-q_m^2)^{1/2}\sin\theta;$$

$\theta$ = angle of ejection of the electron, and $q_m$ = minimum value of $q\approx\frac{1}{2}M(\kappa^2+1)/k$.

TABLE III. Comparison of measured and calculated values of the differential cross section for production of secondary electrons. The measured values are for 100-keV protons in hydrogen gas. The calculated values were obtained from the Born approximation and are twice the values for 100-keV protons in a gas of hydrogen atoms.

| Energy of ejected electron (eV) | Angle of ejection (degrees) | Differential cross section (Units of $10^{-20}$ cm²/eV-sr-molecule) Experimental | Calculated |
|---|---|---|---|
| 3.4 | 23 | 58 | 187.2 |
| | 45 | 38 | 194.8 |
| | 90 | 47 | 118.8 |
| | 135 | 27 | 68.8 |
| 8.6 | 23 | 248 | 96.2 |
| | 45 | 77 | 114.6 |
| | 90 | 27 | 57.8 |
| | 135 | 29 | 20.7 |
| 13.56 | 23 | 160 | 58.6 |
| | 45 | 96 | 77.0 |
| | 90 | 10 | 33.4 |
| | 135 | 5 | 8.7 |

To obtain the desired cross section, the expression must be numerically integrated over $q$. This was done for several values of $\kappa$ and $\theta$, and the results, multiplied

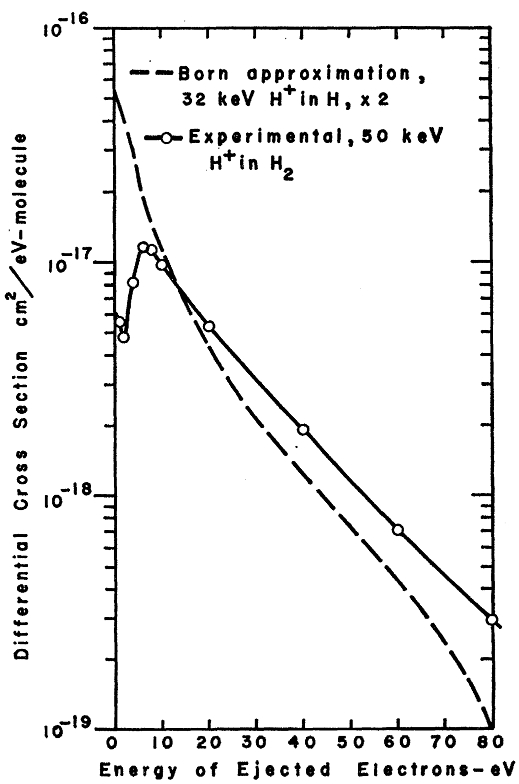


FIG. 11. Differential cross section for ejection of electrons at all angles as a function of electron energy. The experimental results are for 50-keV protons. The Born approximation results for 32-keV protons are from reference 19.

[33] N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions* (Oxford University Press, New York, 1949), 2nd ed., p. 234.

[34] H. S. W. Massey, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1956), Vol. 36, p. 356.

[35] L. D. Landau and E. M. Lifschitz, *Quantum Mechanics, Non-Relativistic Theory* (Addison-Wesley Publishing Company, Inc., Reading, Massachusetts, 1958), p. 459.

[36] See reference 35, p. 464.

[37] See reference 35, p. 122.

by 2, are presented in Table III, together with the corresponding experimental values. The agreement is poor. It is seen that as one proceeds from the total ionization cross section to the doubly differential cross section, the agreement with experiment becomes poorer.

Because of the meagerness of Born approximation results the comparison with experimental results is very sketchy. A need is clearly shown for more extensive calculations, as well as a need for experimental measurements at higher proton energies where the Born approximation is expected to be valid and deviations could be attributed to the treatment of the hydrogen molecule as equivalent to two hydrogen atoms.

**ACKNOWLEDGMENTS**

The authors wish to thank Dr. Charles J. Cook for designing and testing some of the apparatus used in this experiment and for his continued interest in the problem. We wish to thank W. Lang for expert accelerator operation, F. J. Sazama for assisting with the reduction of data, Eugene Rudd and D. E. McArthur for the computation of the Born approximation calculation, L. H. Sohl for construction and testing of the magnetometer, and A. Maschke for stimulating discussions. The expert assistance of J. S. Heiser and D. J. Fuehring in construction of the apparatus is gratefully acknowledged.

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PHYSICAL REVIEW VOLUME 130, NUMBER 4 15 MAY 1963

# Positron Annihilation in Liquid and Solid Mercury*

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Results are presented of a study of the angular correlation of photons from the annihilation of positrons with electrons in solid and liquid mercury. The angular distribution of photon coincidences can be separated into contributions arising from annihilations with the ionic-core electrons and with the conduction electrons. The angular variation of the former does not appear to change at the solid-liquid transition, but the distribution for the conduction electrons is considerably modified. The plot of the number of coincidences against angle for the conduction electrons in the solid can be fitted very well by a parabola corresponding to two free electrons per atom, which indicates that the Fermi surface in extended $k$ space does not depart very significantly from a sphere. The relative number of annihilations from the conduction electrons in the liquid is considerably greater and the distribution departs from the free electron parabola at large angles. These effects are interpreted in terms of the distortion of the wave functions and the broadening of the electronic energy levels by the disorder in the liquid. It is concluded that the uncertainty in the wave vector of an electron at the Fermi surface in the liquid is about 20% of the Fermi wave vector.

## INTRODUCTION

THE electronic band structure of liquid metals has recently been the subject of considerable theoretical and experimental investigation. On the one hand, an attempt has been made to calculate the electronic eigenfunctions and energy levels,[1,2] and on the other a number of measurements of the optical[3] and transport properties of liquid metals[4] have been made to determine experimentally some features of the electronic band structure and scattering mechanisms. Because of their inherent nature as disordered structures, the electronic free path in liquid metals is short and it is not therefore feasible to carry out such experiments as the de Haas–van Alphen or magnetoacoustic effects, which have been successfully applied to the determination of the Fermi surface in solid metals. This limitation does not apply to the study of the angular correlation of the photons created when positrons annihilate with the electrons in a metal however, and valuable information can be obtained from such measurements.[5]

The purpose of the experiments described in this paper was to make a comparison of the electronic structures of mercury in the solid and liquid states by comparing the photon distribution from the two phases. A study of positron annihilation in liquid mercury has previously been made by Stewart,[6] but he did not compare the angular distribution with that for the solid and so was unable to draw any explicit conclusions about the electronic structure of the liquid. In this work the distribution of angular correlations was obtained both for the liquid and the solid phases and it has proved possible to deduce from the results a number of con-

* Contribution No. 1257. Work was performed in the Ames Laboratory of the U. S. Atomic Energy Commission.

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[5] A. R. Mackintosh, U. S. Atomic Energy Commission Report IS-299 (1962).
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