The $S_{N_\tau,N_{\tau'}}$ are the asymmetric rotor line strengths and have been tabulated.[12]

The $\Delta\nu(F_U-F_L)$ of $3_{03}-3_{12}$ was used along with the $1_{01}$ and $1_{10}$ parameters to obtain $(0)_S$, $(aa)_S$, $(bb)_S$, $(cc)_S$. For $Cl^{35}$, $\Delta\nu(F_U-F_L)$ predicted for $2_{12}\rightarrow3_{03}=410$ Mc, $\Delta\nu_{obs}=380$ Mc; $\Delta\nu(F_U-F_L)$ predicted for $5_{14}\rightarrow4_{23}=-612$ Mc, $\Delta\nu_{obs}=-563$ Mc. For $Cl^{37}$, $\Delta\nu(F_U-F_L)$ predicted for $2_{12}-3_{03}=368$ Mc, $\Delta\nu_{obs}=355$ Mc; $\Delta\nu(F_U-F_L)$ predicted for $5_{14}-4_{23}=-556$ Mc, $\Delta\nu_{obs}=-533$ Mc. The spacings inside the quartet groups are predicted in error by $\sim$10 Mc. All the parameters obtained are listed in Table III.

[12] R. H. Schwendeman and V. W. Laurie, *Line Strengths of Rotational Transitions* (Pergamon Press, New York, 1958).

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# Ionization Cross Sections for Protons on Hydrogen Gas in the Energy Range 0.15 to 1.10 Mev*

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Measurements have been made of the ionization cross section for protons incident on hydrogen gas in the energy range 0.15–1.10 Mev. The experimental cross section in this region can be represented by $\sigma_i=(3.45\pm0.20)E^{-(0.874\pm0.010)}\times10^{-17}$ cm$^2$/molecule, where $E$ is the incident proton energy in Mev. The experimental results are in excellent agreement with a Born approximation calculation, which is discussed.

## I. INTRODUCTION

THE gross ionization cross section for protons incident on hydrogen gas has been measured for incident particle energies over the range from 0.15 to 1.10 Mev. Previous measurements in this area have been confined to incident-particle energies below 0.18 Mev.[1,2] The work reported here represents an extension into a region that is largely unexplored.

The atomic and molecular reactions that can occur when fast atoms or atomic ions collide with the molecules of a target gas may be conveniently classed as either "ionization" or "charge-transfer" events. There is no general agreement on the exact definition of these terms—we choose to define them as follows: In an "ionization" event, the fast particle ionizes the struck molecule but emerges with no change in its own charge state, while in a "charge-transfer" event the fast particle either gains one or more electrons from, or loses one or more electrons to, the target particle. For a given projectile on a given target, each class of events in general includes several distinct kinds of reactions differing in the array of slow residual particles that are produced. The energies of the latter are usually low, although a small fraction of them may have energies as high as a few hundred electron volts. In either ionization or charge transfer, the incident particle almost always suffers only a small loss of energy and emerges with only a slight deviation from its original direction of motion.

In charge-transfer studies, the sum of the cross sections for all types of events that produce a given change in the charge state of the fast particle may be measured by observing the distribution of charge states in the emerging fast beam. Such measurements have been made previously for hydrogen atoms and ions incident on hydrogen gas with energies up to 1.0 Mev.[3] The observed cross sections indicate that in our energy range charge transfer events should not make a significant contribution to the gross ion production. (Our experimental results bear out this expectation.)

To study ionization events one must collect and observe the slow charged particles produced by the collision, since the emerging fast beam contains no information about the occurrence of these events. To avoid confusion due to multiple reactions by a single incident particle, the target must be "thin" in the sense that most of the incident particles will traverse the target with no collisions at all. One previous study of protons on hydrogen has been made for energies up to 0.18 Mev,[1] but the bulk of other previous work has been confined to energies of less than 0.04 Mev.[2]

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[1] V. V. Afrosimov, R. N. Il'in, and N. V. Fedorenko, Soviet Phys.—JETP **34**, 968 (1958).

[2] J. P. Keene, Phil. Mag. **40**, 369 (1949); Ia. M. Fogel, L. I. Krupnik, and B. G. Safronov, Soviet Phys.—JETP **1**, 415 (1955); H. B. Gilbody and J. B. Hasted, Proc. Roy. Soc. (London) **A240**, 382 (1957).

[3] C. F. Barnett and H. K. Reynolds, Phys. Rev. **109**, 355 (1958); P. M. Stier and C. F. Barnett, Phys. Rev. **103**, 896 (1956).

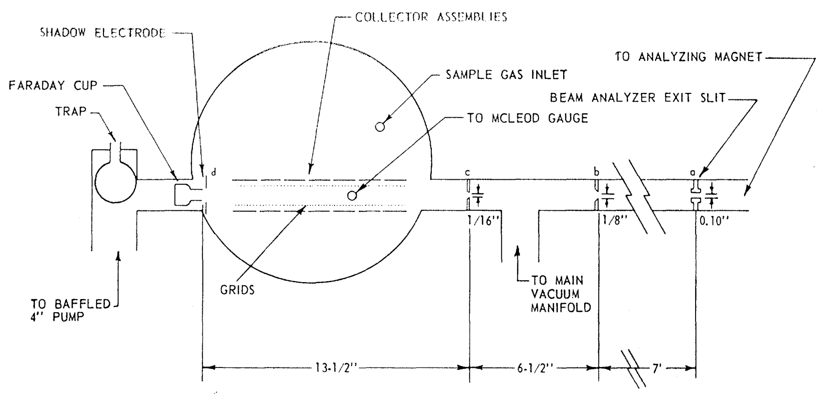


FIG. 1. Schematic view of apparatus for gross ionization measurement. The analyzed proton beam enters from the right through collimating slits, *a*, *b*, *c*, traverses the active volume between the collector assemblies, and is collected in the Faraday cup at the left. One set of collector electrodes is shown in detail in Fig. 4.

## II. EXPERIMENTAL METHOD

The source of incident protons was a 1-Mev Van de Graaff positive-ion accelerator, equipped with a beam analyzing and stabilizing system. The incident proton energy was determined by a 90° deflection in a regulated magnetic field, whose value was measured with a precision gauss-meter. The nominal proton energy spread was $\pm 2$ kev at 1 Mev.

The beam was passed through collimating apertures and into a collision chamber containing the target gas. (See Fig. 1.) The target thickness (5.5 inch) and gas pressure ($10^{-4}-10^{-3}$ mm Hg) was such that the target was "thin." A transverse electric field was maintained between two sets of electrodes parallel to the beam axis in the collision chamber, the "collector assemblies" in Fig. 1. The slow charged particles produced in ionizing collisions were collected, while the original incident particles passed through the collision volume and into a Faraday cup. Detection of both the slow and the fast particles was accomplished by electrometer measurements of the electron and ion currents to the collectors. The chamber pressure was measured with a carefully cleaned McLeod gauge. While this experiment is very simple in principle, completely spurious results can be obtained unless a number of factors are considered with care. A complete discussion of the design considerations and the detailed testing of the apparatus has been presented elsewhere,[4] and we mention the more important factors only briefly here: 1. Beam apertures should have knife edges to minimize scattering from surfaces, Fig. 1 (b and c). 2. The Faraday cup detector of the fast axial beam should be large enough to intercept essentially all of the beam, allowing for scattering from aperture edges and target gas molecules. 3. The Faraday cup should be designed in such a way as to trap secondary electrons ejected by the impinging beam particles, see Fig. 2. 4. The level of background contaminant gas should be held to a minimum and held constant, so that the ionization of the background constituents will be small and capable of being assessed, Fig. 3. 5. Pressure equilibrium should be established before each run, and appreciable pressure gradients in the gas avoided. 6. The collection electrodes for the slow residual particles must be covered with high-transparency grids, adequately biased to eliminate the effects of secondary electron emission from the collectors. 7. Guard electrodes must be provided to define accurately the collection volume from which slow residual particles are collected, Fig. 4. 8. The transverse sweeping electric field must be strong enough to allow collection of essentially all of the slow particles, Fig. 5. 9. Leakage currents must be minimized and accurately assessed. 10. The contributions from charge transfer, if appreciable, must be assessed and taken into account.

A drawing of one of the two collector assemblies is shown in Fig. 4. It is seen to consist of nine segments. Although all nine are always held at the same potential, we measure the ion currents to only one or more of the five interior segments. The inactive segments serve as guards to assure that the collection field in front of the active segments is parallel and uniform. Then the "effective thickness" of the slab of target gas from which all slow ions produced are drawn is just the combined

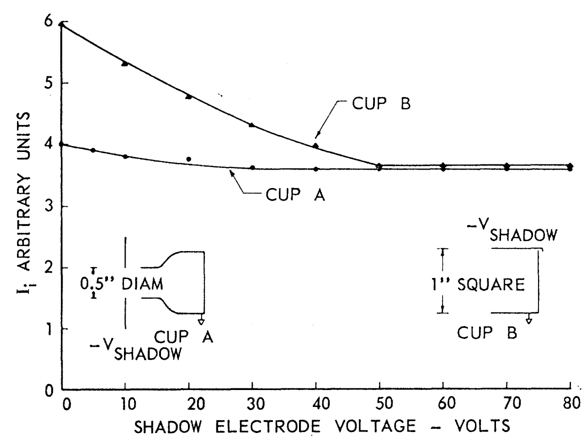


FIG. 2. Incident beam collected in Faraday cup vs suppression voltage. Constancy of the indicated current above 30 volts for cup A and above 50 volts for cup B assures that secondary electron suppression is complete.

[4] E. W. McDaniel, D. W. Martin, J. W. Hooper, and D. S. Harmer, Technical Report, September 1, 1960 (unpublished).

length of the active collector segments. In the present apparatus, it has been verified that the measured ion currents are precisely proportional to the number of segments included in the electrometer circuit. An electron suppressor grid is located $\frac{1}{4}$ inch in front of each collector assembly. The grids consist of parallel, 0.004-inch diameter stainless steel wires and have a geometrical transparency of 96%.

Leakage currents in the electrometer circuits were measured frequently and subtracted from all current measurements. The correction was usually less than 5%. A constant pumping arrangement (described in reference 4) was used to provide a residual background gas density that is independent of the sample gas density insofar as possible. The hydrogen target gas was admitted through a palladium leak in order to achieve purity of the entering gas.

The pressure of the residual gas averaged about $6\times10^{-6}$ mm Hg as indicated by ionization gauges, using the nitrogen calibration. The actual value was uncertain since the composition was unknown. A typical run of the ionization currents produced in the residual gas is shown in Fig. 3. The slope of the line is almost the same as that obtained with hydrogen in the chamber, and was not found to vary from day to day. The impurity currents at several energies were read daily before the hydrogen was admitted, and again at the end of a day's run. The impurity ionization current for each energy inferred from these data was subtracted directly from each hydrogen ionization current reading. Except for the lowest hydrogen pressures, this amounted to a correction of less than 5%.

In a given run the incident particle energy was varied over the entire range while the hydrogen gas pressure

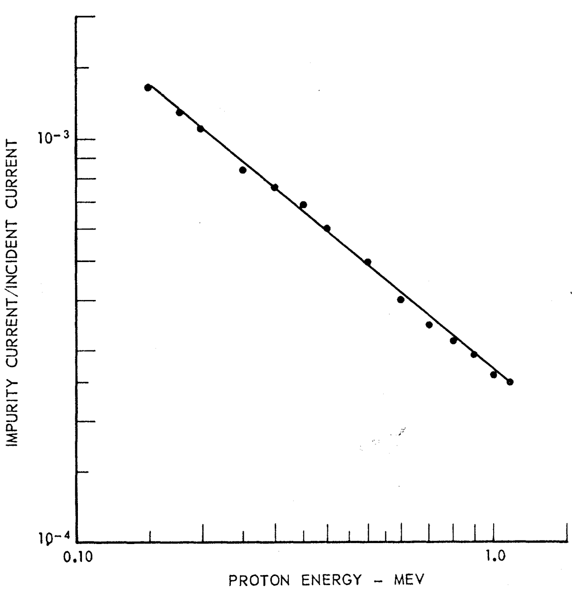


FIG. 3. Ion current contribution from protons incident on the residual background gas. The energy shape of this curve is approximately the same as for hydrogen gas.

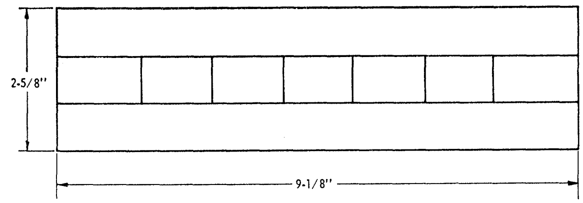


FIG. 4. Collector electrode assembly showing guard electrodes (outer plates). The "effective collision volume" is defined by such of the five central segments as are connected to the electrometer circuit.

was held nominally constant. Usually the setting of the palladium-leak-heater power was left fixed for at least one hour before readings were begun, to allow pressure equilibrium to be reached. Even so, the McLeod gauge was read frequently during the run.

Complete runs were made for hydrogen pressures throughout the range from 0.1 to $12.0\times10^{-4}$ mm Hg. The residual gas pressure was not subtracted from the indicated total pressure since it was not really well known; however, if it was really of the order of $6\times10^{-6}$ mm Hg as the ionization gauges indicated, it would represent a correction of less than 5% for all hydrogen pressures above $1.2\times10^{-4}$ mm Hg. In computing the molecular density of the target gas, its temperature was taken to be that of the room.

A set of values obtained for the gross ionization cross section at one energy from a series of runs at different pressures is shown in Fig. 6, plotted to a relative scale. The falloff at pressures below $2.5\times10^{-4}$ mm Hg can be identified with the above failure to take account of the residual gas in computing the target gas density. Similarly, the indication of rising values for pressures above $10\times10^{-4}$ mm Hg can be identified with multiple collisions and failure of the "thin target" assumptions. The existence of a definite plateau between these regions lends confidence that all the important assumptions are valid there. All of the data used in compiling the final results have been taken from runs lying within this plateau.

## III. EXPERIMENTAL RESULTS

Our final values for the absolute gross ionization cross section for protons incident on hydrogen gas with energies from 0.15 to 1.10 Mev are plotted in Fig. 7. The data give an excellent fit to a straight line in this log-log plot throughout the energy range.

The uncertainties in the ratios of the corrected ionization currents to the incident beam current should not exceed about $\pm2\%$. The target gas temperature is not directly measured and may be uncertain by perhaps $\pm1\%$. By far the largest uncertainty is in the measurement of the target gas pressure. Our McLeod gauge scale extends only to $10^{-5}$ mm Hg, and the instrument has not been absolutely calibrated. We believe we can read its scale to less than 5% in the range around $5\times10^{-4}$ mm Hg, but must admit a probable error of

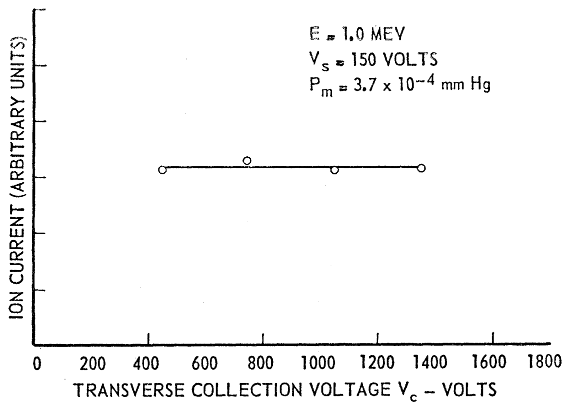


FIG. 5. Data showing the lack of dependence of the transverse ion current on the collection voltage.

about $\pm 5\%$ in the absolute reading. Combining these errors leads to an estimated probable error of about $\pm 6\%$ in the absolute normalization of the data, and this is the vertical error indicated by the brackets on the points in Fig. 7.

The slope of the line is less uncertain, however. The proton energy has a nominal uncertainty of only $\pm 0.2\%$ at 1 Mev, and we believe the uncertainty is not over $\pm 0.5\%$ at 0.15 Mev. Since individual runs were made at constant nominal pressure, the slopes obtained depend only on the relative scale-reading accuracy rather than on the absolute accuracy of the McLeod gauge. Further, self-consistency of the slopes from many individual runs at different pressures gives confidence that the ratios of the cross sections at the extreme energies are known to $\pm 2\%$ or better. The straight line drawn through the data in Fig. 7 corresponds to the expression:

$$\sigma_i = (3.45 \pm 0.20) E^{-(0.874 \pm 0.010)} \times 10^{-17} \text{ cm}^2/\text{molecule},$$

where $E$ is the proton energy in Mev.

## IV. COMPARISON WITH THEORY

In the present case of protons incident on molecular hydrogen, the gross ionization measurements described here include contributions from the following four distinct kinds of ionization events:

$$H^+ + H_2 \rightarrow H^+ + H_2^+ + e, \quad (1)$$

$$H^+ + H_2 \rightarrow H^+ + H^+ + H^0 + e, \quad (2)$$

$$H^+ + H_2 \rightarrow H^+ + H^+ + H^+ + 2e, \quad (3)$$

$$H^+ + H_2 \rightarrow H^+ + H^+ + H^-, \quad (4)$$

plus the four kinds of charge-transfer events:

$$H^+ + H_2 \rightarrow H^0 + H_2^+, \quad (5)$$

$$H^+ + H_2 \rightarrow H^0 + H^+ + H^0, \quad (6)$$

$$H^+ + H_2 \rightarrow H^0 + H^+ + H^+ + e, \quad (7)$$

$$H^+ + H_2 \rightarrow H^- + H^+ + H^+. \quad (8)$$

Among the first four, reactions (3) and (4) represent more complex events than do (1) and (2), and it seems quite likely that they will be correspondingly improbable and contribute in a minor fashion to the total ionization. The sum of the cross sections for (5), (6), and (7) is the gross charge transfer cross section $\sigma_{10}$ which has been measured previously.[3] This cross section is found to be of such magnitude that charge transfer should make a barely significant contribution of about 2% at 0.15 Mev, but be negligible above 0.2 Mev. In verification of this assertion is the fact that the collected electron currents observed were always equal to the positive-ion currents within our reading accuracy of $\pm 2\%$. Any significant amount of charge transfer would lead to an excess of positive-ion current over electron current.[5] Reaction (8) has a completely negligible cross section at our energies.[3] Therefore, the present gross ionization measurements yield essentially the sum of the cross sections for processes (1) and (2).

Theoretical cross-section calculations using the Born approximation have been made[6] for the atomic ionization process

$$H^+ + H^0 \rightarrow H^+ + H^+ + e. \quad (9)$$

A method of obtaining an approximate theoretical treatment for the present molecular processes has been indicated in reference 6. Although the results calculated for reaction (9) were not given in explicit analytic form, the following generalization was made:

If a fast proton collides with a nucleus of atomic number $Z_b$, to which one electron is bound in the 1$s$ state, then the cross section for removal of that electron takes the general form [Eq. (21) of reference 6]

$$\sigma^+ = (Z_b/\Delta E)^2 f(M\Delta E/E),$$

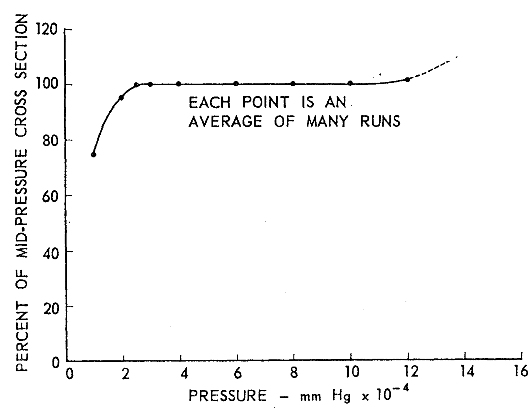


FIG. 6. Computed ionization cross section at various target gas pressures showing plateau from $2.5 \times 10^{-4}$ to $10.0 \times 10^{-4}$ mm Hg.

[5] Secondary electron emission produced by positive-ion impact on the grid shielding the slow-ion collector has the opposite effect. Data presented in reference 4 indicate that this mechanism increases the electron current by less than 2%.

[6] D. R. Bates and G. Griffing, Proc. Phys. Soc. (London) **A66**, 961 (1953).

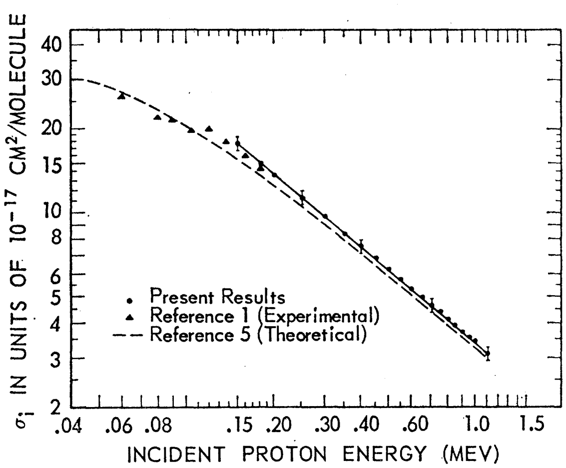


FIG. 7. Gross ionization cross section for protons incident on molecular hydrogen.

in which $\Delta E$ is the ionization energy for removal of the electron, $M$ is the reduced mass of the colliding system, $E$ is the kinetic energy of the relative motion, and $f$ is a function of unspecified analytic form. This formula permits scaling of the graphical results given for reaction (9) to any other reaction that meets the above description.

It has been often assumed that a hydrogen molecule is simply equivalent, in an energetic collision process, to two independent hydrogen atoms, so that the molecular cross section would be expected to be simply twice the atomic cross section. However, in the formula above there is an explicit dependence on the ionization energy $\Delta E$ of the electron to be removed. The *vertical* ionization energy of one electron in the hydrogen molecule is appreciably different from the atomic ionization energy, being in fact greater by the factor 1.2.

The procedure followed is this: The molecule is considered to be equivalent to two free neutral atoms in every respect except that account is taken of the fact that the ionization energy is 1.2 times the normal atomic value. Ignored are the effects of the second atom on the reduced mass of the system consisting of the projectile and the first atom, on the ratio of the incident particle to the relative motion energy, and of course on the form of the electronic wave function that was used in the calculation of the atomic cross section. To this approximation, a theoretical cross section for the removal of one electron from the molecule by the impact of an incident proton of energy $E$ will be twice the given atomic cross section for the incident proton energy $E/1.2$, divided by $(1.2)^2$. This cross section should actually correspond to the sum of the cross sections for all of the several kinds of molecular ionization events, since the theoretical assumptions made no assertion as to the final state of the molecule. Therefore, this theoretical cross section should correspond to our measured gross ionization cross section.

The dashed line in Fig. 7 is the described extrapolation from the theory of Bates and Griffing. Knowledge of the proper location of the line is limited by our ability to read the rather small graphs in the published paper. There is excellent agreement within our stated experimental uncertainties.

The triangles in Fig. 7 indicate the results of Afrosimov *et al.*,[1] which extend upward in energy only to 0.18 Mev. The agreement in the overlap region is quite satisfactory.

## ACKNOWLEDGMENTS

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