Ionization and Charge Transfer in Proton-Hydrogen Atom Collisions*

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The cross sections for charge transfer and for ionization in collisions between protons and hydrogen atoms have been determined over the energy range from 400 to 40 000 ev. The experiment used modulated crossed-beam techniques. Experimental results are compared with several theoretical predictions.

I. INTRODUCTION

In an earlier paper,[1] experimental values of the cross section for production of slow protons in collisions between fast protons and slow hydrogen atoms were presented for the energy range from 100 to 14 000 ev. At that time it was not possible to distinguish experimentally between the two processes producing the slow protons—namely, charge transfer,

$$p+\mathrm{H} \rightarrow \mathrm{H}+p,$$

and ionization,

$$p+\mathrm{H} \rightarrow 2p+e.$$

However, from theoretical considerations it was known that over the energy range of the experiment, the cross section for the former process must be greatly in excess of that for the latter process, so that to within the experimental error it was justified to identify the cross section for slow-proton production with that for charge transfer.

Since those earlier measurements were made, a number of improvements in experimental technique have allowed experimental distinction between the charge-transfer and ionization processes in collisions between protons and hydrogen atoms, and improvements in signal-to-noise ratio have permitted the extension of the energy range of the experiments up to 40 000 ev. This paper summarizes these later measurements.

II. APPARATUS

The basic experimental method used in the present experiments was the same as that used previously[1] and is shown in Fig. 1. An arbitrarily highly dissociated beam of hydrogen was produced by thermal dissociation in a furnace located in the first of three differentially pumped vacuum chambers. The neutral beam emerged from an aperture in the furnace and passed through a collimating slit in the wall separating the first and second chambers and then through another slit in the wall between the second and third chambers. The beam was chopped at a frequency of 100 cps by a rotating, toothed wheel in the second chamber, so that it was a modulated beam that entered the third chamber, wherein the experiment was carried out. After entering the third chamber, the beam passed between two deflector plates, where an electric field swept out any charged particles that accompanied it, and then passed through an aperture in the mass-spectrometer repeller plate into the interaction region.

A dc proton beam crossed the modulated atom beam in the interaction region, care being taken that all of the ions passed through the neutral beam. The slow ions formed in the collisions of the fast ions with the slow atoms were detected by either of two ion detectors. The first was a simple magnetic-sector mass spectrometer, and the second consisted of two plates located above and below the interaction region and parallel to the plane of the two beams. Using either detector, those signals associated with the interaction of the two beams were separable from those produced by the interaction of the ion beam with the residual gas in the vacuum chamber, because the former occurred at the modulation frequency and in a specified phase, while the latter were dc signals plus noise.

The voltage signals at both ion detectors resulted from the passage of the ion currents through 10^{10}-ohm resistors; at each detector a plate-follower preamplifier

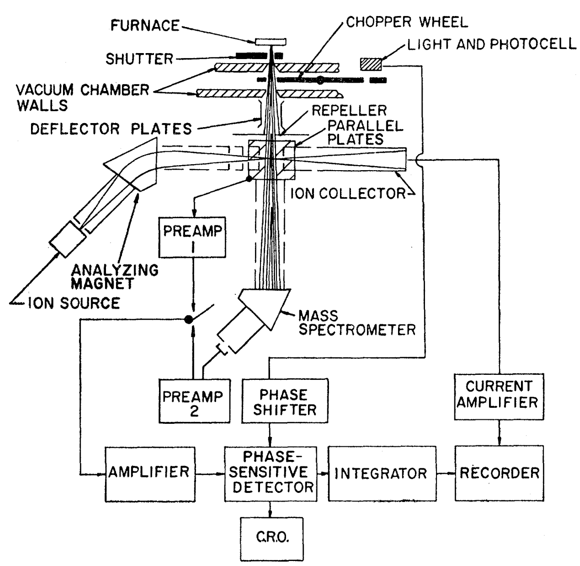

Fig. 1. Schematic diagram of experiment.

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[1] W. L. Fite, R. T. Brackmann, and W. R. Snow, Phys. Rev. **112**, 1161 (1958).

transformed the impedance down to a low value so that the voltage signals could be taken out of the vacuum chamber without loss from distributed capacity. The signals from either preamplifier passed through a tuned amplifier and then to a phase-sensitive detector, whose reference signal was taken from a light and photocell that monitored the chopper wheel. An oscilloscope was used to observe the rectified output of the phase-sensitive detector and to monitor the phasing of the reference signal. The output of the phase-sensitive detector was integrated and displayed by one pen of a two-pen recorder. The other pen gave a simultaneous display of the fast-ion current.

The ion source used in the present experiments was a hot-cathode arc source similar to the duoplasmatron of Von Ardenne,[2] except that the magnetic field was provided by permanent magnets rather than by an electromagnet, and the power dissipated by the arc was kept down to 50 w or less so that forced-air cooling was adequate. For the production of protons, the gas normally used in the source was water vapor.

To separate protons from the other ions produced by the source, magnetic analysis was used. The analyzed proton beam entered the experimental chamber and passed axially through six cylinders, between the fourth and fifth of which the neutral beam crossed the proton beam. The gap between the first and second cylinders was sometimes used as an electrostatic "trimming lens" for final focusing of the beam. The third cylinder contained a rectangular aperture which collimated the ion beam, and the fourth cylinder contained a similar, although larger, aperture. By maintaining the third cylinder somewhat positive with respect to the fourth, secondary electrons formed by ion impact at the edges of the aperture in the third cylinder were restrained from entering the region of intersection of the two beams. The fifth cylinder also contained an aperture, the height of which was less than that of the neutral beam, and the sixth cylinder was the fast-ion collector. The currents to both the fifth and sixth cylinders were read, and satisfactory alignment and focusing were defined as occurring when the fifth-cylinder current was very small compared with the sixth-cylinder current, i.e., of the order of 1%. Under these conditions, it was ensured that the ion current read at the sixth cylinder represented to the required accuracy those protons which had traversed the neutral beam.

The ion collector (sixth cylinder) was biased positive with respect to the fifth cylinder in order to suppress secondary electrons, and because of its being positive, it was necessary to enclose the sixth cylinder in a shield to prevent stray electron currents from reaching the outside of the cylinder and giving erroneous ion-current readings. Such stray electron currents arose from the ionization gauge and from ionization of the residual gas by heavy ions whose trajectories after magnetic analysis took them into the vacuum chamber.

For simplicity of representation, the schematic diagram of Fig. 1 omits four features of the experiment. The first is a filament, located inside the ion collector, which was used as a source of electrons. With the electron beam from this filament running from right to left in the diagram, electron-impact ionization could be used to mass-spectrometrically monitor the neutral beam and determine its degree of dissociation at any time.

The second and third omissions from this schematic are magnetic fields provided by Helmholtz coils. The "horizontal" field was parallel to the neutral beam, and the "vertical" field was perpendicular to the plane formed by the intersection of the two beams. These two fields were used in conjunction with the nondiscriminating parallel-plate detector of slow particles, as will be discussed later.

The fourth omission from Fig. 1 is a guard ring around the collector plate of the parallel-plate detector. It was necessary to maintain this ring at the same potential as the collector plate in order to suppress microphonic noise associated with vibration of the collector plate. It is pertinent to note that while guard rings and plates are customarily used in studies of collisions of ions with chemically stable gases to define the path length of the ions through the gas, in crossed-beam experiments this is unnecessary, since the interaction region is determined from the geometry of the two beams. If the area of the collecting plate is large compared with the geometrical projection of the interaction volume upon it, complete collection efficiency is obtainable.

III. MEASUREMENT PROCEDURE

The two detectors used in these experiments were, as noted above, a mass spectrometer and a parallel-plate detector. The only advantage of the former detector is that in measuring the cross section for the production of slow ions in collisions of protons with a mixture of atoms and molecules, one can distinguish between the processes giving rise to slow protons and to slow molecular ions. In the present experiments, where the neutral beam was highly dissociated ($\gtrsim 95\%$), this advantage was more than offset by the fact that the mass spectrometer cannot discriminate between ions formed by charge transfer and those formed by ionization on ion impact. Consequently, it was used primarily as a check on measurements made using the parallel-plate detector.

When using the mass spectrometer, the procedure was to set the potentials of the fourth and fifth cylinders and of the two parallel plates above and below the interaction region, at 180 v with respect to ground. The potentials of the repeller and the first cylindrical lens leading to the mass spectrometer were set at a slightly different value, as determined from the mass-spectrom-

[2] M. von Ardenne, *Tabellen der Elektronenphysik, Ionenphysik, und Übermikroskopie* (Deutscher Verlag der Wissenschaften, Berlin, 1956), Vol. I, p. 544.

eter focusing requirements. The mass-spectrometer magnet and collector were operated at ground potential, so that the slow ions were accelerated to an energy of 180 ev for passage through the mass-spectrometer magnet. By varying the fast-ion energy and measuring the slow-ion currents, relative cross sections were readily determined.

The use of the parallel-plate detector is a modification of the "condenser method" of measuring charge-transfer and ionization cross sections which has been employed previously[3,4] in studying the interaction between ions and chemically stable gases. In experiments where the purity of the neutral species is sufficiently high, the fact that this detector will collect slow particles of either charge sign with 100% efficiency far outweighs the disadvantage of not being able to discriminate the masses of the ions. In principle, the collection of positive particles yields the sum of the cross sections for ionization and charge transfer, whereas the collection of negative particles (electrons) yields only the cross section for the ionization process.

When the parallel-plate detector was used in the present experiments, the fourth and fifth cylinders, the mass-spectrometer repeller and first lens, and the lower of the two parallel plates were all set at ground potential. For collection of positive ions, the upper plate, which was the actual collector, was biased negatively at a potential of considerably larger magnitude than that at which saturation occurred in a plot of signal versus potential on this plate.

For the collection of negative particles, the polarity of the collector potential was reversed and the vertical magnetic field was employed. This field, being perpendicular to the parallel plates, confined electrons which might otherwise have escaped collection by the weak electrostatic field because of high initial energy and unfavorable initial direction of motion. The vertical field also presented a magnetic barrier against secondary electrons produced by ion impact at the third, fourth, and fifth cylinders which were sufficiently energetic to escape the electrostatic suppression provided. Saturation curves of electron signal as a function of vertical magnetic field were taken, and a field strength of the order of 30 oersteds was found adequate.

When collecting positively charged particles, it was customary to use a horizontal magnetic field of about the same magnitude. Since this field, like the vertical field, was perpendicular to the ion beam's direction, it too served as a magnetic barrier to secondary electrons formed at the ion beam collimation apertures. In addition, the horizontal field, being parallel to the detector, would act as a suppressor of secondary electrons formed at the detector by impact of the slow ions being collected. However, it was straightforward to demonstrate that the number of secondaries formed by these slow ions was quite negligible. This was indicated by the absence of change in the positive-ion signal when the vertical field replaced the horizontal field. With the vertical magnetic field in use and with the electrostatic field set to collect positive ions, any secondary electrons would have increased the signal. The use of the horizontal field was therefore optional.

In collecting negative particles it was essential to demonstrate (1) that the particles were electrons rather than negative ions and (2) that the particles arose from ion-impact ionization rather than from secondary emission caused by ions reaching the parallel plate which was *not* being used as the current detector, i.e., the lower plate. The first point was demonstrated by means of the mass spectrometer, which showed no measurable negative-ion signals. To demonstrate the second point, the incident proton energy was reduced to a few hundred volts, where the p+H charge-transfer cross section becomes large but the ionization cross section becomes negligibly small. Under these circumstances, electrons collected at the upper parallel plate could arise only from secondary emission at the lower plate by ions formed through charge transfer and accelerated in the weak electrostatic collector field between the parallel plates. The signals from such secondary electrons could not be discerned above the noise level.

In measuring relative cross sections with either the mass spectrometer or the parallel-plate detector, signals per unit ion current were compared at different ion energies.

In arriving at absolute cross section values, the basic measurement was that which determined the ratio Q_1/Q_2 of the slow ion production cross section of the hydrogen atom to that of the hydrogen molecule. It was made by running a fixed mass flow of gas in the beam and comparing the slow-ion signals when the gas was purely molecular and when it was arbitrarily highly dissociated. The arguments pertinent to this measurement were presented in reference 1. Measurements of Q_1/Q_2 were made using both detectors.

To separate the cross section for slow-ion production into its separate charge-transfer and ionization components, it is necessary to rely upon the parallel-plate detector. Since for the hydrogen atom this total cross section, Q_1, is simply the sum of the ionization cross section, Q_1^e, and the charge-transfer cross section, Q_1^x, by first noting the slow-ion signal and then the electron signal, using the same atom and ion beams, the ratio Q_1^e/Q_1 is immediately obtained. Subtracting this ratio from unity gives the ratio Q_1^x/Q_1. For the atom, therefore, both the ionization and charge-transfer cross sections are readily measurable in terms of Q_1, which in turn is expressible in terms of Q_2, the cross section for slow-ion production in $p+H_2$ collisions.

The cross section Q_2 was obtained using measurements made by Stier and Barnett[5] of the electron

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

[4] J. B. Hasted and J. B. H. Stedeford, Proc. Roy. Soc. (London) **A227**, 466 (1955).

[5] P. M. Stier and C. F. Barnett, Phys. Rev. **103**, 896 (1956).

capture cross section, σ_{10}, for protons in H_2, which have recently been confirmed by Curran, Donahue, and Kasner.[6] From the list of possible processes involved in collisions between protons and hydrogen molecules,[7] it can be shown that if two processes, $p+H_2 \rightarrow H+2p+e$ and $p+H_2 \rightarrow H^-+2p$ are neglected, then $\sigma_{10}+Q_2^e=Q_2$, where Q_2^e is the cross section for production of electrons when protons strike hydrogen molecules. The neglect of the two processes in question appears justified in view of their both having energy defects of the order of 30 ev, so that the near-adiabatic theory of heavy-particle collisions[8,9] would lead one to expect that the cross sections for these processes would be negligibly small up to ion energies of the order of 200 kev, considerably beyond the energy range of the present experiments. By using the approximation that $Q_2=\sigma_{10}+Q_2^e$, reversal of the collection field in the parallel-plate detector with fixed neutral atom and ion beams gave signals from which the cross-section ratios Q_2^e/Q_2 and σ_{10}/Q_2 were immediately obtained. The latter ratio was used to determine Q_2 from the published values of σ_{10}.

The measurements of the various cross-section ratios indicated above were repeated at a number of proton energies, with the most extensive data being taken at 2, 10, and 30 kev.

IV. RESULTS AND DISCUSSION

In our experiments, which covered the energy range from 400 to 40 000 ev, it was of interest first to examine rather cursorily the cross section Q_2^e for electron production in $p+H_2$ collisions. Values of this cross section previously reported by Keene,[3] Fogel' *et al.*,[10] Gilbody and Hasted,[11] and Afrosimov, Il'in, and Fedorenko,[7] show considerable disagreement in this energy range. In the present experiment, from the determinations of the ratio Q_2^e/σ_{10}, the most probable values of Q_2^e fall between the values of Keene and the two Russian results, with the statistical uncertainties being sufficient to include either set. We regard this as indicative of there being no substantial systematic errors in our measurements of electron-production cross sections in terms of charge-transfer cross sections.

The principal results of this experiment are the cross sections for charge transfer and ionization of the free hydrogen atom in collisions with protons. Figure 2 shows the charge-transfer cross section as compared with several theoretical predictions. The present measurements, like those reported in reference 1, show close agreement at energies of less than 10 kev with the values calculated by Dalgarno and Yadav[12] using the perturbed-stationary-states approximation method.[13]

[6] R. Curran, T. M. Donahue, and W. H. Kasner, Phys. Rev. **114**, 490 (1959).

[7] V. V. Afrosimov, R. N. Il'in, and N. V. Fedorenko, Zhur. Eksp. i Teoret. Fiz. **34**, 1398 (1958) [translation: Soviet Phys.-JETP **7**, 968 (1958)].

[8] H. S. W. Massey, *Reports on Progress in Physics* (The Physical Society, London, 1949), Vol. 12, p. 248.

[9] H. B. Gilbody and J. B. Hasted, Proc. Roy. Soc. (London) **A238**, 334 (1956).

[10] Ya. M. Fogel', L. I. Krupnik, and B. G. Safronov, Zhur. Eksp. i Teoret. Fiz. **28**, 589 (1955) [translation: Soviet Phys.-JETP **1**, 415 (1955)].

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

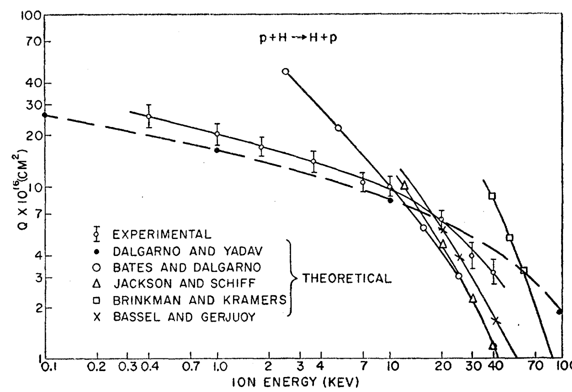

FIG. 2. Cross section for charge transfer in collisions between protons and hydrogen atoms.

Figure 2 also shows a number of high-energy calculations which have been reported. In 1930 Brinkman and Kramers[14] calculated the cross section for charge transfer into only the ground state, using a Born approximation in which the interaction potential was simplified to comprise only the interaction between the atomic electron and the incident proton. Since the experimental values are for charge transfer into all states, rather than just the ground state, it is necessary to modify the Brinkman and Kramers ground-state calculation for comparison with experiment. The modification made here was to multiply the ground-state capture cross section curve by the ratio of total to ground-state capture cross sections calculated by Jackson and Schiff,[15] and it is this modified curve which is ascribed to Brinkman and Kramers in Fig. 2.

As noted above, the only interaction term retained in the calculation of Brinkman and Kramers was that for the interaction between the incident proton and the atomic electron. They omitted the other term which would have been present in a formalistically correct Born approximation calculation, namely, the term describing the interaction between the two protons. This omission is reasonable on the physical basis that in a purely electronic transition such as charge transfer, the interaction between the two nuclei could not exert appreciable influence on the cross section, particularly at high energies.

The principal pragmatic objection to the omission of this proton-proton term arose from the custom of comparing calculated cross sections of *atomic* hydrogen

[12] A. Dalgarno and H. N. Yadav, Proc. Phys. Soc. (London) **A66**, 173 (1953).

[13] D. R. Bates, H. S. W. Massey, and A. L. Stewart, Proc. Roy. Soc. (London) **A216**, 437 (1953).

[14] H. C. Brinkman and H. A. Kramers, Proc. Acad. Sci. Amsterdam **33**, 973 (1930).

[15] J. D. Jackson and H. Schiff, Phys. Rev. **89**, 359 (1953).

with experimental cross sections for *molecular* hydrogen by using the rule that one hydrogen molecule equals two hydrogen atoms. It was found that the Brinkman and Kramers cross section for $p+H$ charge transfer was considerably larger than one-half the cross section for $p+H_2$ charge transfer. The introduction of the proton-proton interaction term in the Born approximation effected a reduction of the cross section to values more nearly in agreement with the molecular data. The curve ascribed to Jackson and Schiff[15] is the total cross section as calculated using the first Born approximation with the interaction potential being included in the proton-proton term. Bates and Dalgarno,[16] using the same approximation as Jackson and Schiff, evaluated the cross section for charge transfer into each state up to and including $n=4$. The curve attributed to Bates and Dalgarno in Fig. 2 is the sum of the cross sections for the states which have been calculated by those authors.

However, in spite of the proton-proton interaction term's leading to better agreement between $p+H$ Born theory and $p+H_2$ experiment, the physical argument in favor of this term's being of little importance is not easily put aside and has led to recent theoretical advances in two directions. The first was an examination by Tuan and Gerjuoy[17] of the validity of treating the hydrogen molecule as equivalent to two free hydrogen atoms for comparison of charge-transfer theory and experiment, a procedure to which they find serious theoretical objections. The second theoretical advance resulted from the recognition that the interaction used in the Born approximation is but one of many equally justifiable interactions which can be used to make a first-order estimate of the cross section. In particular, Bates[18] and Bassel and Gerjuoy,[19] by quite different arguments, have been led to essentially the same interaction, in which the average potential distorting the incoming proton wave is subtracted from the proton-proton and proton-electron interactions.

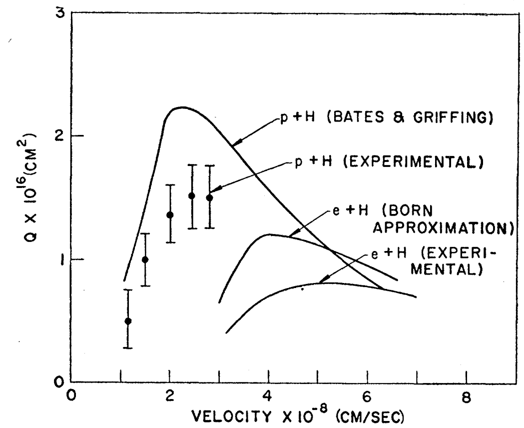

FIG. 4. Comparison of ionization cross sections of the hydrogen atom on proton and electron impact (the electron-impact data are taken from reference 19).

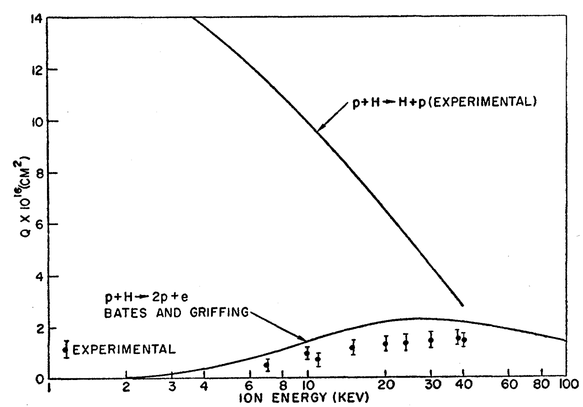

FIG. 3. Cross section for ionization of the hydrogen atom on proton impact, comparing the theoretical prediction of Bates and Griffing and the cross section for charge transfer.

While Bates derives the term largely on physical grounds within the framework of the Born approximation, Bassel and Gerjuoy formulate the charge-transfer problem in the distorted-wave approximation, rigorously derive the scattering amplitudes, and then specialize their solutions to the case of plane waves giving a Born-like approximation. They find that at moderate energies the effect of the proton-proton interaction term is partially offset by another term, and at the higher energies (several hundred kev) these two terms exactly cancel, so as to yield the Brinkman and Kramers result. From Fig. 2 it seems likely that at energies higher than were available in the present measurements, the calculations of Brinkman and Kramers do become correct, as would be expected from Bassel and Gerjuoy's calculations. However, there is still considerable discrepancy between the Bassel and Gerjuoy values and the experimental values in the energy range of these measurements, and it is clear that further theoretical work at moderate energies and experimental work at high energies remain to be done before charge transfer between protons and hydrogen atoms is fully understood.

It is also worth while to note that up to 40 kev, the hydrogen atom cross section for charge transfer with protons appears quite comparable to that of the hydrogen molecule. Comparing theoretical $p+H$ charge-transfer cross sections with one-half the experimental $p+H_2$ charge-transfer cross sections in this energy range is not a valid procedure.

Figure 3 shows the experimental cross section for ionization or electron production (the two being completely identical for $p+H$ collisions) and compares the values calculated by Bates and Griffing[20] using the first

[16] D. R. Bates and A. Dalgarno, Proc. Phys. Soc. (London) **A66**, 972 (1953).
[17] T. F. Tuan and E. Gerjuoy, Phys. Rev. **117**, 756 (1960).
[18] D. R. Bates, Proc. Roy. Soc. (London) **A247**, 294 (1958).
[19] R. H. Bassel and E. Gerjuoy, Phys. Rev. **117**, 749 (1960).
[20] D. R. Bates and G. Griffing, Proc. Phys. Soc. (London) **A66**, 961 (1953).

Born approximation. This figure also shows, for comparison, the $p+H$ charge-transfer cross section.

It is of some interest to compare the experimental cross sections for ionization of the hydrogen atom with Born approximation calculations for both electron and proton impact. Figure 4 shows this comparison plotted on a relative velocity scale. The values for the electron-impact ionization of atomic hydrogen are taken from the paper by Fite and Brackmann.[21] The first point of interest is that for the two processes the relative discrepancies between experiment and the Born approximation calculations are similar, irrespective of the ionizing particle. Second, it is to be noted that the maximum of the theoretical electron-impact ionization curve appears at a higher velocity than that of the proton-impact ionization curve, and the experimental evidence seems to confirm this velocity shift. Similar velocity shifts have been observed in the comparison of the cross sections for ion- and electron-impact ionization in other gases.[22]

[21] W. L. Fite and R. T. Brackmann, Phys. Rev. **112**, 1141 (1958).

ACKNOWLEDGMENTS

We are greatly indebted to F. C. Burke for his aid in the construction of the components of the apparatus, to J. A. Rutherford for his assistance in carrying out the present measurements, and to Dr. Edward Gerjuoy for numerous discussions of the theory of heavy-particle collisions.

[22] N. V. Fedorenko, Uspekhi Fiz. Nauk **68**, 481 (1959).

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Charge Transfer and Electron Production in H^-+H Collisions*

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The cross sections for charge transfer and electron production in collisions between hydrogen atoms and hydrogen negative ions (H^-) have been measured over the energy range 100 to 40 000 ev using modulated atomic-beam techniques in a crossed-beam experiment. Agreement of the experimental results with the perturbed-stationary-states calculation for charge transfer of Dalgarno and McDowell is quite satisfactory.

I. INTRODUCTION

IN the preceding paper,[1] the rather good agreement of experiment with the predictions of the method of perturbed stationary states for low-energy charge transfer in collisions between protons and atomic hydrogen[2] was noted. It is natural to question whether this good agreement is indicative of the merit of the method of perturbed stationary states, or whether the agreement is fortuitously good for this one process. Clearly, an experimental investigation of some other collision process to which the method of perturbed stationary states has been applied would be desirable.

This approximation method has been applied to two other processes—elastic scattering of hydrogen atoms by hydrogen atoms,[3] and charge transfer between negative hydrogen ions (H^-) and atomic hydrogen.[4] Since the examination of the latter process could be carried out with the apparatus described in the preceding paper, it was deemed desirable to carry out the measurement of the H^-+H charge-transfer cross section simultaneously with the measurements of the cross sections for collisions between protons and hydrogen atoms.

During the course of these measurements on the cross sections for collisions between H^- and H, McDowell communicated to us a calculation of the cross section for electron detachment in such collisions, which has since been published.[5] The experimental testing of this calculation constituted a second reason for carrying out the measurements which are the subject of the present paper.

II. EXPERIMENTAL APPROACH

The apparatus which was used in the measurements of the cross sections for charge transfer and for free-electron production in collisions between hydrogen nega-

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[1] W. L. Fite, R. F. Stebbings, David G. Hummer, and R. T. Brackmann, preceding paper [Phys. Rev. **119**, 663 (1960)].

[2] A. Dalgarno and H. N. Yadav, Proc. Phys. Soc. (London) **A66**, 173 (1953).

[3] V. S. Kudriavtsev, Zhur. Eksp. Teoret. Fiz. **33**, 243 (1957), [translation: Soviet Phys. JETP **6**, 188 (1958)].

[4] A. Dalgarno and M. R. C. McDowell, Proc. Phys. Soc. (London) **A69**, 615 (1956).

[5] M. R. C. McDowell and G. Peach, Proc. Phys. Soc. (London) **A74**, 463 (1959).