made by them in collaboration with one of the authors (W.L.F.), but they have continually provided us with results of their subsequent experiments. Their generosity has substantially advanced the progress of the measurements described here.

The first attempts at this experiment were made while one of the authors (W.L.F.), was a National Science Foundation Post-doctoral Fellow at University College London, in 1955.

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Collisions of Electrons with Hydrogen Atoms. II. Excitation of Lyman-Alpha Radiation

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The cross section for excitation of Lyman-alpha radiation in electron-hydrogen atom collisions has been measured as a function of electron energy. The measurement is made by observing Lyman-alpha photons with an iodine-vapor-filled photon counter. A relative-cross-section curve is normalized to fit the Born approximation for high electron energies. The experiment utilizes a 100-cps-modulated atomic beam whose purity in hydrogen-atom content is determined by ionization and mass analysis. A dc electron beam crosses the modulated ground-state atomic beam. The photon counter, which looks at the interaction region, has its output passed through a tuned amplifier and phase-sensitive detection system. Although this treatment of the counter output as an ac current (in which the quantum of charge is that of a Geiger-Müller pulse) introduces some unique noise problems, it satisfactorily separates the photons which arise from the interaction under study from those which arise from other processes (e.g., electron collisions with the residual gas in the high-vacuum chamber).

Some implications of the measured excitation cross section upon scattering theory are discussed.

I. INTRODUCTION

IN the first paper of this series, the measurement of the total cross section for ionization of the hydrogen atom on electron impact was described.[1] The present paper discusses the excitation of the hydrogen atom to certain discrete states upon electron impact. The states of interest are those which lead to the emission of Lyman-alpha radiation, which occurs at 1216 A and corresponds to a transition from the $n=2$ to the $n=1$ state. Because of the differences in experimental procedures, this second experiment in General Atomic's program of research on the free hydrogen atom is presented in a separate paper, although objectives of the experiment are essentially the same as those of the ionization-cross-section measurements, namely, to ascertain the validity of scattering approximations and to measure atomic cross sections of interest in controlled thermonuclear research.

In the present measurements a relative-cross-section curve was taken directly. Absolute values were obtained by normalizing the relative measurements to Born approximation values at high energies.

[1] W. L. Fite and R. T. Brackmann, preceding paper [Phys. Rev. 112, 1141 (1958)].

II. EXPERIMENTAL APPROACH

The experimental arrangement was as shown in Fig. 1. The atomic beam flowed from a tungsten furnace in the first of three differentially pumped vacuum chambers. It was modulated at 100 cps by a mechanical chopper wheel located in the second vacuum chamber. The modulated atomic beam then entered the third vacuum chamber, in which the experiment was carried out.[2]

In the third vacuum chamber, a single electron gun, constructed of cathode-ray-tube gun components, provided electrons for both ionization monitoring of

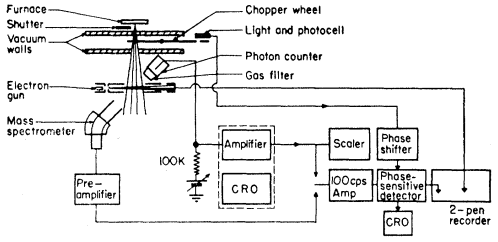

FIG. 1. Schematic diagram of experiment.

[2] For further details of the beam apparatus and general experimental procedures, see reference 1 and W. L. Fite, General Atomic Report GA-267, December 20, 1957 (unpublished).

the neutral beam and excitation of ultraviolet radiation. In the excitation experiments the elements of the gun near the collision region were grounded, and energy was determined by the cathode potential. The mass spectrometer used for ionization monitoring was the second instrument described in reference 1. Knowing the ionization cross sections of both the hydrogen atom and the molecule, we could ascertain the degree of dissociation of the neutral-beam particles and also know that the neutral beam contained only hydrogen atoms and molecules. The beam was normally from 92% to 96% dissociated.

For the detection of Lyman-alpha radiation an iodine-vapor-filled ultraviolet photon counter,[3] a Geiger-Müller counter in which ion pairs were made by photoionization of the I_2 molecule, was used. In front of this counter was a gas filter, consisting of a small cell with lithium fluoride windows at each end, through which a stream of liquid-oxygen-trapped molecular oxygen flowed. The absorption properties of oxygen are such that the filter was black to radiation in the wavelength range detectable by the counters, except at seven "windows," one of which occurs at about 1216 A.[4] Thus, the filter strongly attenuated ultraviolet radiation excited by electron collisions with the residual gas in the vacuum chamber, and also countable bremsstrahlung, but was transparent to Lyman alpha. The counter looked directly at the interaction region.

The most unorthodox procedure in these experiments was the treatment of the counter output. The background gas, consisting of air and molecular hydrogen at a pressure of $\sim 10^{-6}$ mm Hg, produced enough ultraviolet radiation, at electron energies in excess of 15 ev, to obscure completely the counts arising from electrons interacting with the atoms in the beam. Consequently, to identify the desired signal it was necessary to treat the counter output as an ac signal and pass the pulses through the electronics system used with the mass spectrometer. This procedure introduced remarkably large shot noise, because the quantum of charge was the Geiger pulse, about 10^{10} electrons, rather than the usual single electronic charge. In spite of this, however, when a 40-sec time constant was used in the final integration of the signal, the signal-to-noise ratio was usually about 15:1.

The output of the photon counter was developed over the 100-kilo-ohm resistor shown in Fig. 1, and the pulses themselves were observed on a Tektronix oscilloscope at this point, to ensure that the counter tube was operating properly. The amplifier on the oscilloscope served as a preamplifier for the ac signal, which was taken simultaneously to a scaler, to register dc count, and to the tuned amplifier. From this point, the signal proceeded exactly as described previously.[1] A second oscilloscope, which determined if the reference signal was phased correctly, also showed that each counter pulse was Fourier-analyzed to a 100-cps wave train of about 20 oscillations.

In the present experiments the dc shutter immediately in front of the furnace was used, as in the case of the ionization experiment, to distinguish the effects of the beam from those of the bulk gas flowing between the differentially pumped vacuum chambers. Its major use, however, was in the determination of the cross section for excitation of countable ultraviolet radiation in collisions between electrons and hydrogen molecules. When the beam is admitted to the third chamber, an increase in dc pressure in that chamber occurs, and since this increase is almost pure molecular hydrogen, formed by the reassociation of the atoms in the beam as they arrive at the vacuum-chamber walls, differences in dc counts on the scaler with the shutter open and closed could be used to determine this relative cross section, which was checked by making ac measurements using a pure molecular-hydrogen beam. Although the intensities of the various wavelengths were not directly ascertainable in this measurement, the wavelengths are defined by the seven windows in the oxygen absorption spectrum.

III. COMPARISON OF ATOMIC AND MOLECULAR EXCITATION CROSS SECTIONS

It has already been remarked that the beam in this set of experiments was kept only 92% to 96% dissociated. While purer atomic beams may be produced with furnaces, they generally require reduced pressure in the furnace and thus a lower actual density of atoms in the beam as it crosses the electron beam. To obtain a high actual atomic signal, it was advantageous to take greater beam intensity and less purity of the atomic beam. However, such a choice necessitates a correction for the signal derived from the molecules in the beam. It becomes necessary to determine the ratio of atomic to molecular cross sections for excitation of countable ultraviolet radiation. Of course, the only countable ultraviolet radiation emanating from the atom is Lyman-alpha radiation.

In determining this cross-section ratio, advantage was taken of the facts that (1) complete thermal equilibrium apparently is achieved in the furnaces and (2) the amount of mass flow per unit time in the beam is constant and independent of both the furnace temperature and the degree of dissociation in the furnace, under the experimental conditions. (The reader is again referred to reference 1 in regard to these statements, as well as for further details of the present arguments.) Under these circumstances, it is convenient to define the signal $S_0(T_1)$ as that which would be observed with the furnace at absolute temperature, T_1, if molecules of hydrogen did not dissociate, for a given electron current. It can be related to the signal observed, with the same gas flow in the beam, for the furnace at

[3] Brackmann, Fite, and Hagen, Rev. Sci. Instr. **29**, 125 (1958).
[4] Watanabe, Inn, and Zelikoff, J. Chem. Phys. **21**, 1021 (1953).

absolute room temperature, T_r, by

$$S_0(T_1)=(T_r/T_1)^{\frac{1}{2}}S_r. \quad (1)$$

This equation occurs because the signal is proportional to the density of particles in the beam, which for fixed mass flow decreases as the square root of temperature. Actually, the signal at temperature T_1 arises from both molecules and atoms, and their contributions are, respectively,

$$S_2(T_1)=(1-D)S_0(T_1)=(1-D)(T_r/T_1)^{\frac{1}{2}}S_r, \quad (2)$$

$$S_1(T_1)=(Q_1/Q_2)(2D/\sqrt{2})S_0(T_1)$$
$$=(Q_1/Q_2)(2D/\sqrt{2})(T_r/T_1)^{\frac{1}{2}}S_r, \quad (3)$$

where D is the dissociation fraction and Q_1/Q_2 is the ratio of atomic to molecular cross sections for excitation of countable ultraviolet radiation. In Eq. (3), the 2 in the numerator occurs because the molecule dissociates into two atoms, and $\sqrt{2}$ in the denominator occurs because the atom moves faster than the molecule at the same temperature by this amount, and hence spends less time in the region of interaction between the neutral beam and the electron beam.

The total signal, $S_{\text{total}}(T_1)=S_1(T_1)+S_2(T_1)$, obtained with the furnace at temperature T_1, can be expressed alternatively as

$$S_{\text{total}}(T_1)=S_r(T_r/T_1)^{\frac{1}{2}}[\sqrt{2}D(Q_1/Q_2)+1-D], \quad (4)$$

or

$$S_{\text{total}}(T_1)=S_1(T_1)\{1+(Q_2/Q_1)[(1-D)/\sqrt{2}D]\}. \quad (5)$$

The dissociation fraction, D, is determined from comparing peak intensities with the mass spectrometer and using the formula

$$D=1/[1+\sqrt{2}(Q_1{}^i/Q_2{}^i)(S_2{}^i/S_1{}^i)], \quad (6)$$

where $S_1{}^i$ and $S_2{}^i$ are atomic and molecular peak strengths on the mass spectrometer and $Q_1{}^i/Q_2{}^i$ is the ratio of cross sections for ionization of the atom and molecule, as determined in the measurement described in reference 1.

To illustrate how the ratios of cross sections for the atom and molecule to excite and radiate countable photons were determined, we take an experimental case at 60-ev energy, where with a dissociation fraction $D=0.92$ and with $T_1=2700°\text{K}$ (T_r having been 300°K), S_{total} was observed to be twice S_r. Substituting into Eq. (4) and rearranging, we have

$$\frac{Q_1}{Q_2}=\frac{1}{\sqrt{2}D}\left[\left(\frac{2700}{300}\right)^{\frac{1}{2}}\frac{S_{\text{total}}}{S_r}-(1-D)\right]=4.5. \quad (7)$$

At other energies, similar measurements indicated that the atomic cross section exceeds the molecular cross section by about the same amount.

The important result, for present purposes, is seen immediately from Eq. (5). It is evident that for D greater than 90% the contribution to the total signal arising from excitation of the remaining molecules in the beam, i.e., the second term in Eq. (5), is less than 2% of the total observed signal.

IV. ATOMIC CROSS SECTION

A. Total Cross Section

As was remarked previously, the approach adopted for the present experiments was to take directly relative cross sections for excitation as a function of electron energy and then obtain absolute values by assuming Born approximation calculations at high energies to be correct. Since the published theoretical work on excitation of the hydrogen atom to discrete states above the ground state presents total cross sections, it was of primary interest to obtain the total cross section.

Experimentally, the most satisfactory situation, from signal-to-noise considerations, was to observe photons emitted perpendicular to the direction of the electron beam. Such relative-cross-section measurements would, however, represent the relative total cross section only if the angular distribution of photons were independent of electron energy. Generally, it would be necessary to apply corrections for photon angular anisotropy to these 90° relative measurements, and this would entail measuring the angular distribution of the photons as a function of electron energy.

Since Lyman-alpha radiation is electric dipole radiation, the angular distribution must be expressible as proportional to $(1-P\cos^2\theta)$, where P is a constant and θ is the angle between the direction of photon emission and the direction of the incident electron beam. Alternatively, one can define the cross section per unit solid angle for photon emission, and relate it to the total cross section by

$$q(\theta)=\{3Q/[4\pi(3-P)]\}(1-P\cos^2\theta). \quad (8)$$

Since observations made at two angles θ will give relative signals proportional to relative q's for the two angles, P may be determined at each electron energy by measurements at the two angles, and the entire angular distribution will be known.

To make the measurement experimentally, the electron gun was mounted on a table which rotated on an axis coincident with the neutral beam. The photon counter was located so that it looked at the interaction region at an angle of 45° with respect to the neutral beam's direction of travel. Thus, rotating the table made it possible to continuously vary the angle of observation from 45° to 135° with respect to the electron beam's direction. The actual correction term for the relative-cross-section data taken by observing the photons at 90° was obtained by making observations at 90° and 45°. Defining S_{45} and S_{90} as the ac photon signals at 45° and 90°, respectively, it may be shown that

$$R\equiv S_{45}/S_{90}=1-(P/2). \quad (9)$$

Thus, the total relative cross section of the atom for excitation of Lyman-alpha is obtained from

$$Q_1 \propto S_{90}(2R+1). \tag{10}$$

This approach of using the data taken under the convenient experimental conditions of making 90° observations and then correcting for angle seemed entirely satisfactory for electron energies above 25 ev. In this range there was no significant difference between signals at 45° and 135°, and the average of readings at these angles was taken for S_{45}. Below 25 ev, however, the signals at 45° exceeded those at 135° by significant amounts. We therefore regard this procedure for correcting the 90° data as satisfactory only above 25 ev.

(While the cause of the 45°–135° discrepancy has not yet been completely investigated, we believe it may arise from a Doppler-shift effect and an imperfect coincidence of the wavelengths of Lyman-alpha radiation and the 1216-A oxygen absorption window. The experimental arrangement is such that if momentum is transferred to the atom in the collision with the electron, radiation observed at 45° will be Doppler-shifted toward shorter wavelengths, and that observed at 135° will be shifted toward longer wavelengths. The observed discrepancies below 25 ev, where considerable momentum transfer to the atom is to be expected, suggest that the wavelength of Lyman-alpha radiation lies on the long-wavelength side of the oxygen window. Further experiments on this matter will be made in the near future.*)

A second method for obtaining the relative total cross section was used to check the method described above. It will be noted from Eq. (8) that there is a "magic angle" at which signal strengths [which are proportional to $q(\theta)$] are proportional to the total cross section. This magic angle is defined by $\cos^2\theta = 1/3$, i.e., $\theta = 54.5°$. Relative-cross-section measurements were made by observing photons emitted at 54.5° from the direction of the electron beam; and these agreed with the relative measurements for the total cross section obtained from correction of the 90° data to within experimental uncertainty.

Having obtained a curve of the relative total cross section for excitation of Lyman-alpha radiation, we found that this curve would fit the Born approximation calculations for the $1s$-$2p$ excitation over the energy range 200 to 700 ev (the highest electron energy used in these experiments). Below 200 ev, substantial deviations from the first Born approximation values occurred. Figure 2 compares experimental results with three theoretical predictions—the first[5] and second[6] Born approximation calculations and a recent calculation by Massey and Khashaba[7] using the distorted wave approximation. Two features are of interest in this curve when it is compared with the cross section for ionization of the hydrogen atom, presented in the first paper in this series. The first is that the deviations between experimental values and first Born approximation values at lower energies are more pronounced in the case of excitation of Lyman-alpha radiation. The second is the suggestion that for electron scattering problems both the second Born approximation and the distorted wave approximation are hardly worth the computational effort—their values are very little better than the much simpler first Born approximation values, if indeed the observed radiation does arise from the simple $1s$-$2p$ excitation.

While this comparison of experimental data with calculations for the $1s$-$2p$ excitation is the most reasonable one to make, there are other excitations which might lead to the emission of the observed Lyman-alpha radiation. Three types of processes suggest themselves particularly strongly.

The first is the excitation of the hydrogen atom from the ground state to the $2s$ metastable state, with quenching of this state by very weak stray fields in the collision region. However, the smallness of the computed values[8] and of the experimental values[9,10] for the cross section for the $1s$-$2s$ excitation strongly suggest that this process is insignificant compared with the $1s$-$2p$

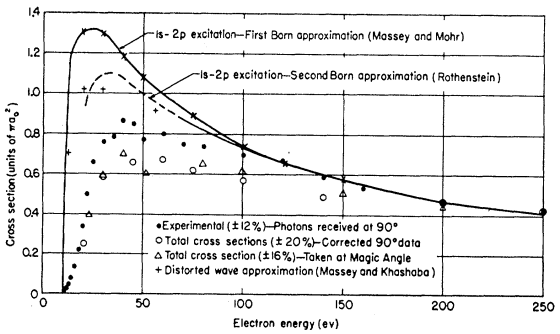

FIG. 2. Comparison of theoretical and experimental cross sections. The experimental cross sections are normalized to fit the first Born approximation over the range 200 to 700 ev.

* *Note added in proof.*—Since submission of this paper, an experiment has been performed on the absorption of molecular oxygen in the immediate neighborhood of Lyman alpha radiation, where the source of light was the collision-excited atomic beam and where Doppler shift due to the beam's motion provided the variation of wavelength. This experiment showed that Lyman alpha from atomic hydrogen does not lie on a steep side of the oxygen absorption window, and thus makes the proposed explanation of the 45°–135° discrepancy untenable. This experiment will be described in full in a future paper.

[5] H. S. W. Massey, *Encyclopedia of Physics* (Springer-Verlag, Berlin, 1956), Vol. 36, p. 354.
[6] W. Rothenstein, Proc. Phys. Soc. (London) **A67**, 673 (1954).
[7] H. S. W. Massey and S. Khashaba, Proc. Phys. Soc. (London) **71**, 574 (1958).
[8] G. A. Erskine and H. S. W. Massey, Proc. Roy. Soc. (London) **A212**, 521 (1952); H. S. W. Massey and B. L. Moiseiwitsch, Proc. Phys. Soc. (London) **A66**, 406 (1953); and B. H. Bransden and J. S. C. McKee, Proc. Phys. Soc. (London) **A70**, 398 (1957).
[9] W. E. Lamb and R. C. Retherford, Phys. Rev. **79**, 549 (1950).
[10] W. Lichten and S. Schultz, paper read at New York University Conference on Physics of Electronic and Atomic Collisions, January, 1958 (unpublished), and private communication.

excitation, except possibly in the immediate vicinity of threshold.

The second is a double collision process in which the ground-state hydrogen atom coming from the furnace is excited to the $2s$ state by an electron and then is induced to radiate by a collision with a second electron. The experimental evidence against any significant contribution by this process to the results of the present experiment consists of the observed linearity of an ac counter signal with electron current. A double process would indicate itself by a signal which varied quadratically with electron current. Unfortunately, in the present experiments it was not possible to increase electron current to the point where such a quadratic effect would be expected on the basis of presently estimated cross sections for the double process. Since the photon counter was a Geiger counter, currents could be increased only to where the dc count rate was such that the mean time interval between counts was approximately equal to the recovery time of the counter. For lesser current the signal appeared to vary linearly with the electron current to within an experimental uncertainty of about 8%. It may be noted that for the geometry of the apparatus and with Born approximation values for the $1s$-$2s$ excitation cross section, a cross section for the $2s$-$2p$ excitation[11] of the order of 10^{-8} cm^2 would contribute a singnal of less than the 8% experimental uncertainty. We therefore believe that this double process did not contribute significantly to the final signal.

The third possible type of process is the cascade, in which the atom is excited to states of quantum number $n>2$, from which it radiates back to the $2p$ state and thence emits Lyman-alpha radiation. We believe that the contribution of such cascades to the final signal is quite negligible. This opinion is based on consideration of the cross section for excitation of the atom to higher states, from which it could radiate to the $2p$ state, and the probability that, given the excitation, it would do so. Using Born approximation calculations[5] for the excitation cross sections, all such possible single cascades give contributions to well within the experimental error. Since the Born approximation values for the cross sections must be high, the actual contributions must be even less significant than is indicated by the calculations. More complicated cascading processes appear to be even less important.

Since these most obvious processes competing with $1s$-$2p$ excitation appear to be insignificant, considering the experimental uncertainty, we shall continue, in this paper, to compare experiment results with predictions for only the $1s$-$2p$ excitation process.

B. Angular Distribution of Radiation

To correct the 90° data to obtain the total cross section, it was necessary to learn the angular distribution of the radiation. This was done by measuring the ratio of signals with the photon counter observing at 90° and at 45° with respect to the electron beam, as indicated in Eqs. (8) through (10). From this ratio the constant P was determined as a function of electron energy. The constant P is the customary polarization fraction, i.e.,

$$P=(I_{\parallel}-I_{\perp})/(I_{\parallel}+I_{\perp}). \tag{11}$$

where $I_{\parallel}$ and $I_{\perp}$ are radiation intensities observed at an angle of 90° to the direction of the electron beam which are polarized with electric vector parallel and perpendicular, respectively, to the direction of the electron beam.[12] Figure 3 shows a plot of the polarization fraction as a function of electron energy. The experimental points shown are those taken directly from measurements of R [see Eq. (9)], and the experimental uncertainty at each point is indicated. These results indicate that above about 200 ev the radiation appeared isotropic in angular distribution, while at lower energies a very clear preference for radiation perpendicular to the direction of the electron beam was evident, both from measurements of R down to 25 ev and from comparison of relative cross sections taken at 90° and 54.5°.

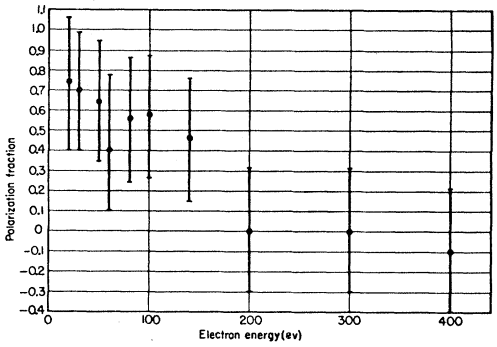

FIG. 3. Polarization fraction for Lyman-alpha radiation from electron-hydrogen atom collisions.

C. Behavior Near Threshold

The behavior of a cross-section curve near threshold gives information on the type of incoming electron wave responsible for the excitation in this energy region. Wigner[13] has shown that the cross-section contributions from electrons having an angular-momentum quantum number l after the excitation process go as $E^{l+\frac{1}{2}}$, where E is the kinetic energy of the electron after the excitation process is completed. While the range in which this energy dependence should hold is rather ill-defined, it is interesting to note that in the range from 11 to 16 ev, the cross-section values best fit a curve of the form E^n, with $n\sim 1.3$. The uncertainty in this exponent arises from the uncertainty in the signals rather than

[11] M. J. Seaton, Proc. Phys. Soc. (London) A68, 457 (1955).

[12] J. A. Smit, Physica 2, 104 (1935).

[13] E. P. Wigner, Phys. Rev. 73, 1002 (1948).

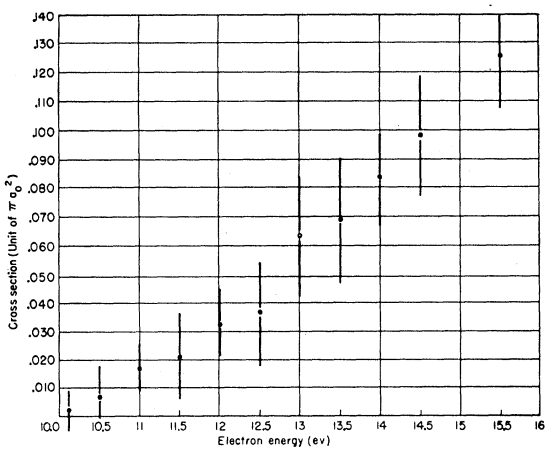

FIG. 4. Cross section for excitation of Lyman-alpha radiation near threshold.

from scattering of the mean-value signals about the assumed mathematical form. This clearly suggests that the primary excitation involves an incoming *s*-wave electron and an outgoing *p*-wave electron. In the first electron volt above threshold the signal strength became so small that it was not possible to verify the general theoretical prediction that in the ill-defined *immediate* vicinity of threshold all outgoing electrons in an electron-excitation process must be *s*-wave electrons. Figure 4 shows low-energy cross-section data taken with observations at 90°. The absolute values for the cross section are appropriate to the assumption of zero polarization of the radiation.

V. EXCITATION OF THE HYDROGEN MOLECULE

Reference has already been made to information which had to be gathered on excitation of the ultraviolet radiation accepted by the photon counter which arose from electron collisions with the hydrogen molecule. As miscellany to this paper, this information is summarized in Fig. 5, which compares excitation cross sections of the atom and the molecule for which the photons were observed at 90° with respect to the direction of the electron beam. In this graph, the values of the cross section for the molecule must be considered as only approximate; no correction was made for angular distribution of the photons.

One feature of interest is not indicated on the graph. This is the behavior of the molecule's excitation cross section at low energies. From data on a dc scaler count with the shutter open and closed, it was evident that no excitation appeared until the electron energy was about 11.5 ev, when a very slight background count from H_2 appeared. It is presumed that this radiation arose from excitation of the B state of the molecule, with subsequent radiation. A more substantial increase in background count from H_2 appeared at about 14.5 ev. This energy coincides quite well with the energy required to dissociate the molecule and excite one of the dissociated atoms. However, a mild violation of the Franck-Condon principle as applied to the hydrogen atom is involved in this interpretation; according to this principle, this process should not be observed.[14] A similar violation was observed by Lamb and Retherford[15] in the formation of $2s$ atoms in electron-molecule collisions, although their threshold energy was somewhat higher.

VI. DISCUSSION OF ERROR

The uncertainty of the measurements reported here has already been presented. It seems unlikely that the approach used here can be made to give appreciably more precise results. The basic difficulty is that the usual method of increasing a signal-to-noise ratio by increasing both signal and noise, the former at a faster rate than the latter, is inapplicable. Such a program can continue in these experiments only to the point where the photon counter begins to operate on its recovery time and the separate counts seriously overlap each other. In the present experiments, count rates were normally kept at about the maximum that this limitation would allow.

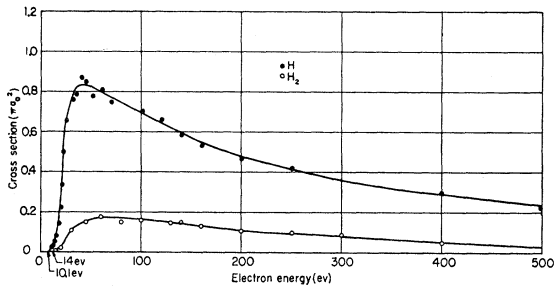

FIG. 5. Comparison of cross sections for excitation of Lyman-alpha radiation from the hydrogen atom and countable ultraviolet radiation from the molecule.

VII. ACKNOWLEDGMENTS

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The assistance of Miss Eugenia Rossell in this experiment is deeply appreciated.

[14] H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Impact Phenomena* (Clarendon Press, Oxford, 1952), pp. 229–232.
[15] W. E. Lamb and R. C. Retherford, Phys. Rev. **75**, 1332 (1949); **81**, 222 (1950).