electrons have almost no effect on the inner electrons. This result is not unexpected. Less expected is the fact that the $4s$ perturbation on the $3d$ and inner electrons is less than that suffered by an iron series ion when it is inserted into a crystalline environment.[14,15]

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Collisions of Electrons with Hydrogen Atoms. V. Excitation of Metastable 2S Hydrogen Atoms*

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Ground-state hydrogen atoms produced by thermal dissociation in a tungsten furnace were excited by collision with electrons having energies up to 600 ev. Those atoms which were excited to the metastable $2S$ state were subsequently quenched in an electrostatic field, and the resulting Lyman-alpha radiation was detected with an iodine-vapor-filled photon counter. In order to assign absolute cross-section values to the excitation function obtained in this way, the ratio of the $2S$ to the $2P$ excitation cross sections was determined. From previously obtained knowledge of the cross section for excitation to the $2P$ state, the absolute $2S$ cross section was evaluated. Agreement with the Born approximation was observed at high energies. The angular distribution of the scattered $2S$ atoms was also investigated for electron energies up to 600 ev.

I. INTRODUCTION

THE cross section for excitation of ground-state hydrogen atoms to the metastable $2S$ state on electron impact has been the subject of extensive theoretical investigation.[1–7] The fact that both the initial and final states have zero angular momentum, so that the wave functions are extremely simple in addition to being precisely known, makes this inelastic collision process particularly amenable to a variety of scattering-theory approximations.

However, the predictions of the various scattering-theory approximations differ quite widely, and it is difficult, on theoretical grounds, to select with any confidence the approximation that most closely describes the transition. It seems possible, therefore, that insight into the problem may be gained more directly from a laboratory study of this process and that preference for a particular approximation may be indicated by the results of an accurate experiment. Prior to the present work, experimental evidence on the form of this cross section near threshold was provided by the relative measurements of Lamb and Retherford[8] and by the more recent work of Lichten and Schultz,[9] who used energies up to 40 ev. In the present measurements, the use of modulated-atomic-beam techniques enabled the range of measurements to be extended to considerably higher values of the electron energy.

II. APPARATUS AND PROCEDURE

A schematic view of the apparatus which was employed in the present work is shown in Fig. 1. Atomic hydrogen flowing from a tungsten furnace in the first of three differentially pumped vacuum chambers was modulated at 100 cps by a motor-driven, toothed chopper wheel located in the second chamber. The hydrogen atoms were then excited by a beam of electrons from a gun that was placed in the third chamber with its axis normal to the direction of the atomic beam. After passing through the gun, the atoms entered a "quench" region throughout which an electrostatic field could be established. An iodine-vapor-filled ultraviolet photon counter[10] was mounted on a movable

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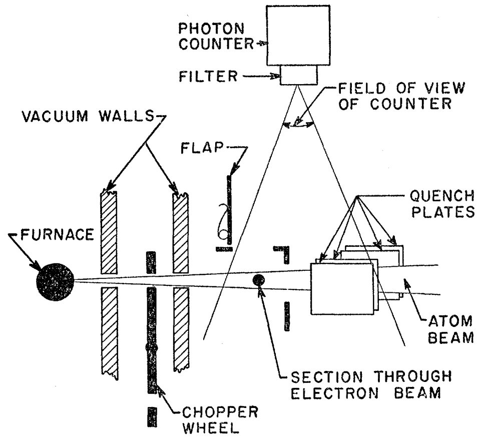

FIG. 1. Schematic diagram of the experimental region in total cross-section measurements.

trolley, so that it could be positioned to view either the region of interaction of the two beams or the quench region.

In these measurements, advantage was taken of the fact that the lifetime of a hydrogen atom in the $2S$ state (which in the absence of perturbations is greater than 2.4 msec[11]) is considerably shortened in the presence of an electric field, where mixing of the $2S$ and $2P$ states results in the accelerated decay of the $2S$ atoms, with accompanying emission of Lyman-alpha radiation. Measurement of these photons provides a convenient mechanism whereby $2S$ metastable atoms may be detected.

The procedure was as follows. Ground-state hydrogen atoms were excited by electron impact. Those atoms which were excited to the $2P$ state, with an excited lifetime of the order of 10^{-9} sec, decayed with emission of Lyman-alpha radiation while still in the collision region. Meanwhile, the $2S$ atoms passed from the collision region and entered the quench region. Two measurements were then made.

For the first measurement, the photon counter was positioned directly above the collision region, and the flap (shown in Fig. 1) was raised to allow the counter to view the region of interaction of the two beams. The electron gun was operated dc, and the signal arising from the atoms could be distinguished from that resulting from the impact of electrons with the gun electrodes or with the background gas, since it occurred at the modulation frequency and in specific phase. The background radiation gave rise to a dc counter signal, plus some noise at the modulation frequency, and was discriminated against by the use of ac counting techniques.

For the second measurement, the counter was moved to a position immediately above the quench region, and the flap was lowered to shield the collision region from the counter's direct view. The ac counter output was then observed for two values of the quench field:

(1) At a value of the field, determined from saturation measurements, sufficient to ensure total quenching of the $2S$ atoms.

(2) At zero field, when the ac counter signal arose from Lyman-alpha radiation which issued from the collision region through the atom beam exit aperture and was subsequently reflected into the detector. This signal was augmented to some extent through natural and collision-induced decay of the $2S$ atoms, while in transit through the field of view of the counter.

This subject will be further discussed in Sec. III. The difference between the two ac counter signals was taken as a measure of the $2S$-atom flux.

If the furnace, electron-gun, and photon-counter conditions were maintained constant throughout these two measurements, the ratio of the cross sections for the $1S$-$2P$ and $1S$-$2S$ excitation processes was calculable. Determination of the absolute response of the photon counter was unnecessary.

Despite the use of ac measuring techniques, the $2P$-atom signal that was registered by direct observation of the collision region was found to be quite noisy, because of the abundance of background ultraviolet radiation. Consequently, for some measurements, a gas filter comprised of a small cell with lithium fluoride windows at each end was mounted on the front of the counter. Molecular oxygen was circulated through this filter after being passed through a trap immersed in an acetone—dry-ice mixture to remove water vapor. Although this oxygen filter strongly absorbed the ultraviolet radiation produced by collisions of the electrons with the gun electrodes and with the background gas, it was nearly transparent to Lyman-alpha radiation, whose wavelength corresponds exactly with one of seven windows in the oxygen absorption spectrum. Use of the filter gave improved signal-to-noise, although some attenuation of the signal was introduced by the lithium fluoride windows.

In order to minimize the possibility of accidental quenching of the $2S$ atoms due to stray electrostatic fields in the vicinity of the collision region, all of the gun elements were gold-plated and heated to deter the formation of insulated layers which could become charged. Those elements in the vicinity of the atom beam were grounded, and the electron energy was determined from the cathode potential. An axial magnetic field of about 50 gauss—sufficiently small to have negligible effect on the lifetime of the $2S$ atoms—was used (1) to reduce lateral spread of the electron beam while passing through the atom beam and (2) to prevent the escape of scattered electrons to the quench-region electrodes, where the generation of soft x rays would give rise to an unacceptable noise level. The final electron collector was biased to prevent the escape of secondary electrons ejected from it by the primary

[11] W. L. Fite, R. T. Brackmann, D. G. Hummer, and R. F. Stebbings, Phys Rev. **116**, 363 (1959).

beam; these electrons would otherwise have been trapped by the magnetic field and would have been constrained to move back across the collision region, thus giving rise to a spurious signal and, at the same time, leading to error in the measured electron current.

The electron gun was enclosed in a grounded box in which apertures were cut to allow passage of the atom beam and to allow the photon counter to view the interaction region of the two beams. The size and position of the exit aperture for the atom beam was dictated by the results of previous measurements of the angular distribution of the 2*S* atoms; these measurements are discussed in Sec. IV.

The requirements for the quench field were quite stringent, and the behavior of several configurations was examined. Basically, it was necessary to ensure the complete quenching of the 2*S* atoms within the rather limited field of view of the counter, while ensuring only negligible premature quenching of the 2*S* atoms by the field penetrating into the collision region. Thus, the field needed to be intense and localized.

As a first step toward this end, the field was established between two plane grids that were placed perpendicular to the axis of the neutral atom beam. The first of these grids was grounded so that the electrostatic quenching would occur only between the grids. With this arrangement, however, the "zero field" 2*S*-atom signal was not negligible, apparently being caused by the reflection of Lyman-alpha radiation from the grid wires into the counter or from accidental quenching of some 2*S* atoms in passage through residual electrostatic fields in the neighborhood of the closely spaced grid wires. This led to uncertainty in the true 2*S*-atom signal and to the abandonment of this arrangement in favor of that shown in Fig. 1. Here, the quench field was established between two plates biased symmetrically about ground and positioned to intercept only a negligibly small fraction of the emerging 2*P* radiation. Tests with an electrolytic tank showed that penetration of the fringe field of this arrangement into the collision region was substantially reduced by the addition of a further pair of "guard" plates. This penetration field was less than 3% of the quench field, and since the lifetime of the 2*S* atoms is inversely proportional to the square of the field strength, accidental quenching in the collision region accounted for considerably less than 1% of the total 2*S*-atom flux. With this arrangement, the 2*S*-atom signal observed was no longer contaminated with reflected 2*P* radiation, as was evidenced by the absence of an appreciable "zero field" signal. Another quench configuration, in which the four plates were replaced by four heated vertical rods, was examined. This configuration was found to give results similar to those for the four-plate arrangement. The results which are presented here are derived from measurements made with both of these systems.

To relate the signals arising from 2*S* and 2*P* atoms to the appropriate excitation cross sections, consideration of the anisotropic angular distribution of the emitted radiation was necessary. Since photon detection was made at an angle of 90° with respect to the electron beam's direction in the case of 2*P* excitation, and at an angle of 90° with respect to the direction of the quench field in detecting 2*S* atoms, it is convenient to introduce two slightly nonphysical cross sections, $Q_{90}(2S)$ and $Q_{90}(2P)$, which may be directly related to the observed signals by

$$Q_{90}(2S)/Q_{90}(2P) = (2S \text{ signal})/(2P \text{ signal}).$$

These cross sections, Q_{90}, correspond physically to the apparent total cross sections deduced from measurements using radiation detection at 90° only and the assumption of isotropic angular distribution of the radiation.

Since in quenching the 2*S* atom, the radiation is electric dipole radiation with the dipole oriented parallel to the quench field, it is necessary to relate the true total 2*S*-production cross section, $Q(2S)$, to $Q_{90}(2S)$ by introducing the factor $\frac{2}{3}$, so that

$$Q(2S) = \tfrac{2}{3}Q_{90}(2S) = \tfrac{2}{3}(2S \text{ signal}/2P \text{ signal}) \times Q_{90}(2P).$$

Thus, the ratio $Q(2S)/Q_{90}(2P)$ is immediately obtained from comparison of the observed signals. Further, since values of $Q_{90}(2P)$ have been determined in a separate experiment,[12] absolute values for the total 2*S*-excitation cross section are immediately deducible. In that experiment, normalization of data was made to the Born approximation over the energy range 200 to 700 ev. Hence, the ultimate precision of the present determination of $Q(2S)$ is limited by the accuracy of the Born-approximation calculations for 2*P* excitation at high energies.

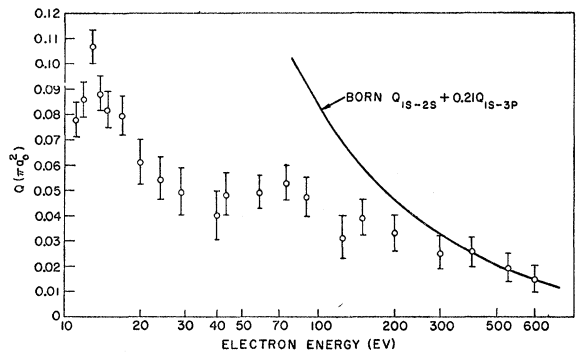

FIG. 2. Total cross section for production of 2*S* hydrogen atoms.

III. RESULTS OF EXCITATION CROSS-SECTION MEASUREMENTS

Observations of the cross section for production of 2*S* metastable atoms are plotted in Fig. 2, with the experimental uncertainties shown. These results include

[12] W. L. Fite, R. F. Stebbings, and R. T. Brackmann, Phys. Rev. **113**, 356 (1959).

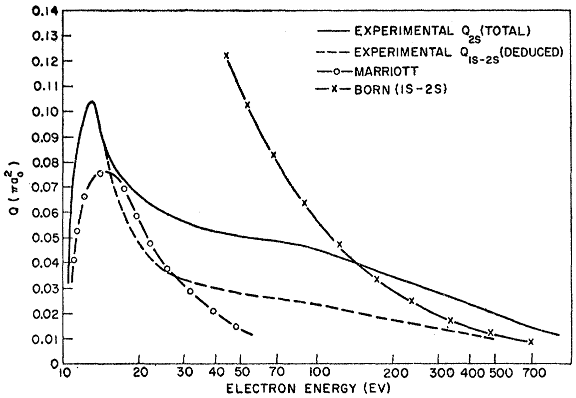

FIG. 3. Comparison of theoretical and experimental cross sections.

not only the direct $1S$-$2S$ excitation cross section, but also that contribution to the $2S$-atom production which is due to cascade from higher levels excited from the ground state by electron impact. Lichten and Schultz[9] have shown that by using the transition probabilities given by Bethe and Salpeter,[13] the measured total $2S$-production cross section is equivalent to the sum of the direct $1S$-$2S$ cross section, plus 21% of the $1S$-$3P$ excitation cross section. This composite cross section, as calculated using the Born approximation, is compared with the experimental results in Fig. 2.

It is seen from this figure that at energies greater than about 300 ev, the measurements agree satisfactorily with this composite cross section, although at low energies a marked divergence from the Born approximation is apparent. The agreement of the high-energy data with theory is taken as strong confirmation of the correctness of the experimental procedure. Near threshold, 10.15 ev, the cross section exhibits a sharp maximum, characteristic of forbidden transitions, at about 13 ev and then falls monotonically with increase in electron energy. In view of the very severe discrepancy between these low-energy measurements and the predictions of the Born approximation, considerable attention was paid to possible mechanisms which would lead to erroneously low values for the measured cross section. These may be listed as follows:

(1) Quenching of $2S$ atoms in the collision region due to residual electrostatic fields.

(2) Quenching due to the space-charge field of the electron beam.

(3) Loss of $2S$ atoms by collision-induced transitions to the $2P$ state in further encounters with electrons.

(4) Natural and collision-induced decay of the $2S$ atoms in the collision region.

(5) Loss of $2S$ atoms due to their being scattered at sufficiently large angles to escape passage through the quench field.

(6) Inefficient quenching and detection of the $2S$ atoms.

With regard to the first of these mechanisms, the apparent correctness of the results at high energies, where the Born approximation should be valid, is held to be sufficient evidence for the absence of any residual electrostatic field capable of inducing significant quenching in the collision region.

The possibility of quenching due to the space-charge field of the electron beam was examined by observing whether or not the $2S$-atom signal varied linearly with electron current. This effect is, of course, of more concern at the lower energies, since for a fixed electron current the space-charge field increases as the electron velocity decreases. It was found that even at the lowest energies, linearity was maintained up to well above 10 μa. Since, in general, a current of 2 μa was used, attenuation of the $2S$-atom beam from this cause should be considerably less than 1%. Estimation of the space-charge field using the known geometry of the beam gave confirmation to this conclusion.

The cross section for production of $2P$ atoms in collisions of electrons with $2S$ atoms has been investigated, and the measurements are discussed in Sec. V. These show that less than one $2S$ atom in ten thousand was de-excited in this way.

The cross sections for quenching of $2S$ atoms in collision with several of the more common gases have been measured and previously reported.[12] Under the present experimental conditions, it may be estimated that loss of atoms from such processes and from natural decay was appreciably less than 1%.

The angular distribution of the scattered $2S$ atoms was examined in some detail, and the results of these measurements are presented in Sec. IV. It may be said here that over the entire range of energies from threshold to 600 ev, the metastable atoms were scattered at angles, in the horizontal plane, of less than about 20°. For any value of the electron energy, the maximum possible vertical scattering angle corresponding to a given horizontal scattering angle may be readily determined. Therefore, dimensions of the apparatus were chosen such that after excitation, all $2S$ atoms, regardless of their angle of scatter, would pass to the quench region without hindrance.

Of final concern was the efficiency of the quench system. As noted above, the requirements of this system were that it should provide a sufficiently intense field to quench all the $2S$ atoms within the field of view of the counter, while at the same time being sufficiently localized so that the fringe field penetrating to the collision region could induce negligibly few premature transitions to the ground state. The agreement between the measured curve and the Born approximation at high energy may again be taken as confirmation that saturation of the detector was achieved and that no significant quenching of the $2S$ atoms took place within the colli-

[13] H. Bethe and E. E. Salpeter, *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 35, p. 352.

sion region. To further examine this question, the field distribution around a scale model of the quench system was plotted with the aid of an electrolytic tank. From the expression for the lifetime of a 2S atom in an electrostatic field[14] of E volts per centimeter,

$$1/t = 2780E^2 \text{ sec}^{-1},$$

it was ascertained that only trivial error resulted from premature quenching in this fringe field. Field plots were made for both the guarded-plate and guarded-rod configurations, and the voltages necessary for saturation, calculated from the expression above, were very closely approximated by those determined experimentally.

These foregoing considerations lend strong argument to the conclusion that the observed low-energy departure from the Born approximation is real, and not an instrumental effect.

It is difficult to make detailed comparison of experimental results with theory, since the experimental results include contributions from cascade processes as well as direct 1S-2S excitation. The many theoretical calculations available have been concentrated on only the direct excitation process. Unfortunately, the contributions from cascade processes constitute a large fraction of the total 2S-production cross section, over most of the energy range. The cross-section contributions caused by cascade processes may be taken as 21% of the 1S-3P excitation cross section as indicated at the beginning of this section. The 1S-3P cross section is estimated here (1) by assuming that the shape of this curve is similar to that of the 1S-2P cross section[12] and (2) by normalizing this relative cross-section curve to the Born-approximation values at energies above 250 ev. When cascading contributions obtained in this way are subtracted from the total 2S-production cross section, the deduced cross-section curve for the direct 1S-2S excitation shown in Fig. 3 is obtained. The total 2S-production cross-section curve serves to indicate the importance of cascade processes.

Because of the large contribution from cascade processes and the experimental uncertainties, it is not very meaningful to make a detailed comparison of the deduced Q_{1S-2S} with theory over most of the energy range. However, at energies near threshold, the cascade contributions become quite small and comparison with theory becomes significant. Figure 3 also shows the theoretical predictions of Marriott, which are expected to be valid only at energies near threshold but show rather good agreement with experiment up to about 20 ev, together with the Born-approximation predictions for the 1S-2S process only.

Additional experimental evidence on the 1S-2S cross section near threshold is provided by the measurements of Lichten and Schultz. They used crossed-beam techniques to determine the excitation function, which they then normalized to the Born approximation at 40 ev, the highest energy used. In addition, by estimation of certain experimental parameters, they were able to make a further absolute estimate of this cross section which was in moderate agreement with that obtained through normalization of the relative measurements. This was taken as evidence in favor of the correctness of the normalization procedure. Disagreement in absolute magnitude between the present results and those of Lichten and Schultz is, therefore, very severe. However, the shapes of the two curves are closely similar. Renormalization of their measurements to the present value of 0.045 πa_0^2 at 40 ev results in excellent overlapping of the two sets of data.

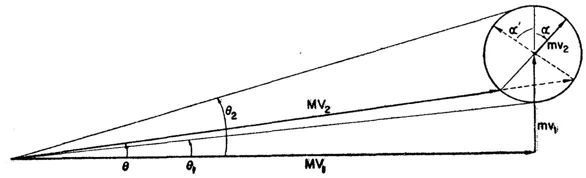

FIG. 4. Momentum diagram showing the atom scattering sphere in a crossed-beam excitation experiment.

The authors incline to the view that the *relative* measurements of Lichten and Schultz are more accurate than those now being reported, particularly in the immediate vicinity of threshold. However, the proximity of the present measurements to the Born approximation within the energy range 300 to 700 ev—where this approximation should certainly be valid—is thought to be undeniable evidence for their correctness.

IV. ANGULAR DISTRIBUTION OF THE 2S ATOMS

A prime requirement of the measurements discussed in Sec. III was that all atoms excited to the 2S state should pass through the quench region within the field of view of the counter. In order for this condition to be satisfied, it was obligatory to know the angular distribution of the scattered 2S atoms.

In collision with electrons, hydrogen atoms that are excited to the 2S state suffer a recoil, measured by the angle θ, which determines the change in their direction of motion. The motion of the colliding particles before and after the impact is represented in Fig. 4, where MV_1 and MV_2 are, respectively, the initial and final momenta of the atom and where mv_1 and mv_2 are the corresponding momenta of the electron. The circle represents the spherical locus of the final electron momentum vector. It is seen that for given values of V_1 and v_1, θ may have a range of values from θ_1 to θ_2. Near threshold, where v_2 is small, the outgoing wave function has zero orbital angular momentum, and the distribution of the outgoing electrons will be spherically symmetrical. Figure 5 shows the loci of the electron momentum vectors for electron energies up to 150 ev and shows the recoil angles for an atom with an initial velocity of 7.5×10^6 cm/sec.

[14] H. A. Bethe and E. E. Salpeter, *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 35, p. 373.

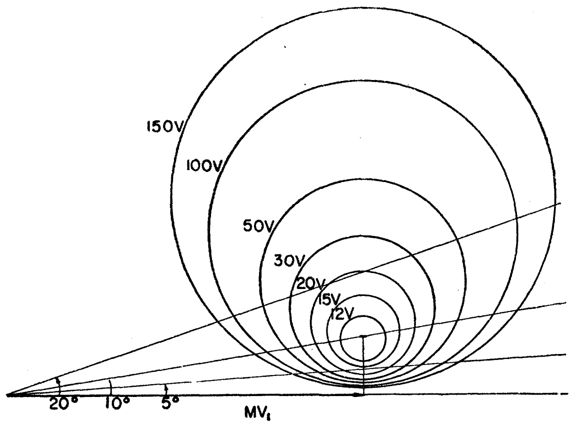

FIG. 5. Scale diagram of atom scattering spheres for ground-state atoms with an initial speed of 7.5×10^5 cm/sec and at a number of electron energies.

The angular distribution of the scattered $2S$ atoms was measured for electron energies up to 600 ev, using the apparatus shown schematically in Fig. 6. In this experiment, those metastable atoms scattered within a narrow angular range traversed two slits. In the region immediately behind the second slit, an electrostatic quenching field transverse to the axis of the beam could be established between two plates. The photon counter was placed to observe part of this quenching region. The signal registered by the counter had two main components, the predominant one being the field-induced decay of metastable atoms within view of the counter, from which the counts were ac at the beam modulation frequency. The second, and smaller, contribution to the count rate came from cosmic rays and from countable ultraviolet photons produced in collisions of the electrons with the background gas or with gun electrodes and then multiply reflected into the counter. The latter signal was dc and was discriminated against by the use of ac measuring techniques. Application of an external electrostatic field—sufficient to quench all metastable atoms before they reached the field of view of the counter—gave confirmation that all

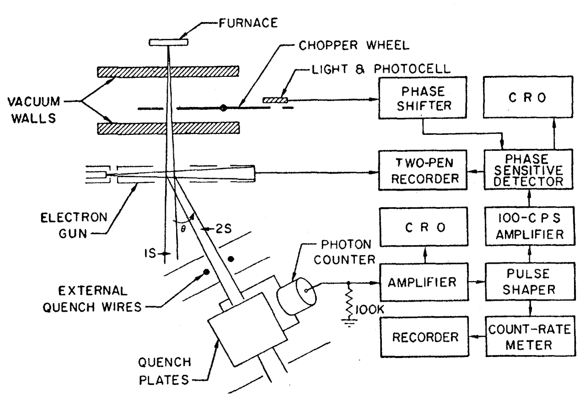

FIG. 6. Schematic diagram of the experimental arrangement for determination of the angular distribution of excited $2S$ atoms.

background effects were indeed dc. The entire quench and detector assembly was mounted on a trolley which could be rotated around a vertical axis passing through the center of the collision region. Measurements of the scattered intensity were made for recoil angles of up to 20° in the horizontal plane; beyond this the signal strength was inadequate to give measurable data (see Fig. 7).

In principle, observation of the angular distribution of the scattered atoms in a crossed-beam experiment can provide information on the differential cross section for the electron scattering. This atom-recoil method has, in fact, been used by Bederson[15] and his co-workers to determine the differential cross section for scattering of low-energy electrons by potassium. However, to determine the equivalent electron scattering angle from the observed atom scattering angle, it is necessary to know both the electron and atom velocities. Consequently, in the present work, where the ground-state atoms had a near Maxwellian distribution of velocities, information on the differential cross section for $2S$ excitation was not readily obtainable.

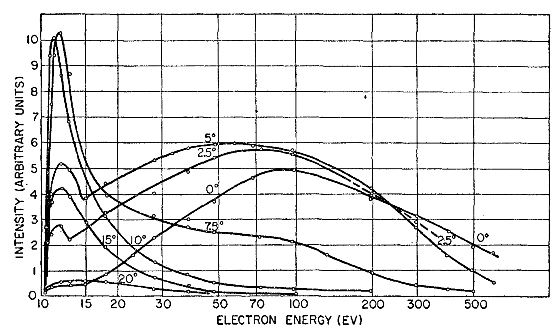

FIG. 7. Angular distribution of $2S$ atoms, showing dependence on electron energy at fixed angles.

No attempt was made to investigate the scattered intensity as a function of vertical scattering angle, and the geometry was such that, in these measurements, only those atoms scattered through angles of less than 5° in the vertical plane contributed to the signal. It may be seen from Fig. 5, however, that the maximum vertical scattering angle corresponding to any scattering angle θ in the horizontal plane is readily determinable for any value of the incident electron energy. Therefore, the geometry in the total cross-section measurements was chosen so that atoms which were scattered through angles up to 45°, in both the vertical and horizontal planes, would enter the quench region within view of the counter.

It is of interest to note that for the production of well-defined beams of $2S$ atoms, there is considerable merit in using electrons of about 70 ev, rather than working near threshold where the cross section is a maximum; larger exciting electron currents may be used at the

[15] K. Rubin, J. Perel, and B. Bederson, Phys. Rev. **117**, 151 (1960).

higher energy without the difficulty of quenching of the 2*S* atoms by space-charge fields. A further benefit arises because the electron scattering is strongly in the forward direction; the angular spread of the resulting 2*S*-atom beam is consequently quite small.

V. QUENCHING OF 2S ATOMS IN COLLISIONS WITH ELECTRONS

To complete this investigation, it was appropriate to examine the extent to which collisions with electrons would transfer hydrogen atoms from the 2*S* state to the 2*P* state, since this effect, if significant, would result in premature quenching inside the electron gun and, thus, in error in the measured 2*S*-production cross section. For these measurements, the instrumentation used for the accumulation of angular-distribution data was modified by the insertion of grids in the two field-quenching plates, so that an electron beam could be made to cross the 2*S*-atom beam within the quench region. The photon counter again observed the Lyman-alpha radiation produced by decaying 2*S* atoms. The essence of the measurement was to compare the quenching of 2*S* atoms by the electrostatic quench field to the quenching by an electron flux. The electron-impact cross section for easing the atoms from the 2*S* to the 2*P* state could then be readily deduced. The electron energy was restricted to less than 10 ev, for above this energy Lyman-alpha radiation would be produced by direct excitation of ground-state atoms accompanying the 2*S* beam.

A second restriction in the experiment was that the electron current density be sufficiently small that only negligible quenching by the macroscopic electric fields from electron space charge could occur.

Under these restrictions, no signals arising from electron collisions with the 2*S* atoms could be detected This permits placing upper limits on the 2*S*-2*P* cross section below 10 ev of something less than an order of magnitude higher than has been calculated by Seaton.[16] If we assume that the cross section at all electron energies is less than our upper limit figure at 6 ev (6×10^{-12} cm^2), which appears reasonable in view of the cross section's probable decrease with increasing electron energy, then it may be shown that less than one atom in 10^4 of those excited to the 2*S* state could have been de-excited in the collision region by impact of electrons.

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