section of  $1.4\pi a_0^2$  in the cone of observation of Brackmann et al., which is within their experimental spread of points. The destructive interference between the s and d waves at right angles to the incident beam is such as to reduce the scattering there relative to the forward and backward directions by a factor of twenty. These *d*-wave phase shifts, however, are at least an order of magnitude larger than what is expected on the basis of the Born approximation.<sup>5</sup> A calculation of the phase shifts using the method of polarized orbitals is in progress here.

The importance of complete experimental results can be appreciated when one realizes that the assumptions regarding the structure of the wave function that have thus far been made in the scattering problem are still extremely crude compared to what has been done in the bound-state problem.

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#### PHYSICAL REVIEW

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## Lifetime of the 2S State of Atomic Hydrogen\*

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A sensitive test for any mixing of the  $2S_{\frac{1}{2}}$  and  $2P_{\frac{1}{2}}$  state of atomic hydrogen is the measurement of the rate for single-quantum decay of the 2S atom to the ground state. A new upper limit of this decay rate has been determined. A section along a beam of 2S atoms, produced by electron excitation of a ground-state atom beam, was viewed by an iodine-vapor-filled ultraviolet photon counter, which responds to the Lymanalpha radiation of the single-quantum decay process. From the counts observed when an electrostatic quenching field was superposed on the counter's field of view, the necessary experimental parameter (product of 2S atom current and counter efficiency) was determined. With the field removed, a portion of the remaining counts could be ascribed to quenching on collision of the 2S atoms with residual gases in the vacuum chamber, the quenching cross sections for which were measured. The decay rate not ascribable to known quenching effects was 420 sec<sup>-1</sup>. Since unknown quenching effects may have been operative, this figure must be considered only as an upper limit for the natural single-quantum decay rate.

# I. INTRODUCTION

 $\mathbf{R}$  ECENTLY Salpeter pointed out that if a permanent electronic electric dipole moment were to exist, one manifestation of it would be a shortening of the lifetime of the  $2S_{\frac{1}{2}}$  metastable state of atomic hydrogen.<sup>1</sup> Its presence would mix the metastable state with the  $2P_{\frac{1}{2}}$  state, and the lifetime would be intermediate between the  $\frac{1}{8}$  sec associated with the twophoton decay<sup>2</sup> of the 2S state and the  $1.6 \times 10^{-9}$ -sec lifetime of the P state. Clearly, by measuring a lower limit of the natural lifetime of the  $2S_{k}$  atom for singlequantum decay, an upper limit for the strength of any perturbation of a fundamental kind, such as that produced by an electronic electric dipole moment, becomes, in principle, determinable. The present paper describes an experiment which yielded a lower limit of 2.4 msec for the lifetime of the 2S atom and in which the cross sections for quenching the metastable atoms

in collisions with several common gases were determined.

#### **II. EXPERIMENTAL APPROACH**

A schematic diagram of the experiment is shown in Fig. 1. A beam of ground-state hydrogen atoms was produced from a furnace source and a fraction of these were excited by electron impact to the 2S state, in a manner similar to that used by Lamb and Retherford.<sup>3</sup> In the present case, however, the initial ground-state hydrogen atom beam was modulated at 100 cps by a chopper wheel so that ac as well as dc measuring techniques could be used. The 2S atom beam was then passed through two successive electrostatic-field quenching regions. In the second of these regions, the field was produced by a pair of parallel plates, a known portion of the region between them being viewed by an iodinevapor-filled ultraviolet photon counter.<sup>4</sup> Since the range of this counter is from 1050 A to 1270 A, the detected

<sup>\*</sup> This research was supported by the Advanced Research Projects Agency through the Office of Naval Research. <sup>1</sup> E. E. Salpeter, Phys. Rev. **112**, 1642 (1958). <sup>2</sup> J. Shapiro and G. Breit, Phys. Rev. **113**, 179 (1959).

<sup>&</sup>lt;sup>8</sup> W. E. Lamb, Jr., and R. C. Retherford, Phys. Rev. 79, 549 (1950).

<sup>&</sup>lt;sup>4</sup> Brackmann, Fite, and Hagen, Rev. Sci. Instr. 29, 125 (1958).



FIG. 1. A schematic diagram of the experiment.

photons from decay of metastable atoms in view of the counter could be only Lyman-alpha photons, i.e., associated with a single-quantum decay process. Both the ac and dc outputs of the counter were used, the former employing the circuitry and phase-sensitive detection techniques described by Fite and Brackmann.<sup>5</sup>

The counts registered by the photon counter come from two sources. The major source is the decay of metastable atoms within view of the counter, from which the counts are ac at the atom-beam modulation frequency. The second, and smaller, source is the dc background produced by cosmic rays and the countable ultraviolet photons, produced in collisions of the dc electron beam with the gun electrodes and residual gas in the vacuum chamber, which are multiply reflected into the photon counter from the surfaces inside the vacuum chamber. The first electrostatic quenching region was used to measure this background in the dc measurements. By purposely quenching all metastables before they reached the photon counter's field of view, only background was registered. With this quenching field present, a zero value of the ac output of the counter confirmed that this background was indeed dc, and therefore not associated with the modulated hydrogen atom beam. Generally, primary reliance was placed on dc measurements, since a given statistical accuracy could be obtained in somewhat less time than by use of ac techniques, and the ac measurements were used to check the dc results.

Certain extra precautions had to be taken in these measurements in regard to extraneous electric fields in the second quenching region. The rate of decay of the 2S atom in an electric field is given by

$$W_F = \alpha F^2 = 2780 F^2 \text{ sec}^{-1}, \tag{1}$$

where F is given in volts per centimeter.<sup>6</sup> Thus, stray fields of the order of 0.7 v/cm would produce apparent lifetimes of the order of 1 msec. It was mandatory to minimize the possibility of stray fields existing in the field of view of the photon counter, if anything other than field-quenching were to be observed. To this end, special care was exercised in the construction of the second quenching region. All metal surfaces were made of copper and all were gold-plated in the same plating bath in order to minimize contact potential fields. The lithium fluoride window of the photon counter was placed back of two gold-plated copper meshes. All surfaces were heated to deter the formation of surface layers which might become charged. The entire second quenching region was enclosed in a gold-plated copper box with entry and exit slits for the atom beam. These slits were made sufficiently narrow to preclude the possibility that metastable atoms could reach surfaces and be quenched by surface collisions, thus giving off photons which could be counted.

Since the electrons crossed the atom beam at  $90^{\circ}$ , the excited atoms were deflected from the original direction of the ground-state atom beam. For the use of electrons of 12.5-ev energy, detection was made at 8.5°, which is the angle for maximum current of 2S atoms.

## III. ANALYSIS OF THE EXPERIMENT

Defining  $I_v$  as the current of 2S atoms per unit velocity range, the loss of current with distance traversed in the vacuum system is given by

$$dI_v = -w(x)I_v dx/v, \qquad (2)$$

where x is the distance along the beam, and w(x) is the total transition probability per unit time at x. It is assumed that this transition probability may be broken down into

$$w(x) = w_0 + \sum_i w_i + W_F, \qquad (3)$$

where  $w_0$  represents the natural single-quantum decay rate,  $W_F$  is the transition probability induced by electric fields [see Eq. (1)], and  $w_i$  is the transition probability induced by collisions of the 2S atoms with the *i*th chemical species in the residual gas in the vacuum chamber. It is given by

$$w_i = n_i Q_i(v) v, \tag{4}$$

where  $n_i$  is the number density and  $Q_i$  is the collision quenching cross section of the *i*th species. For other metastable atoms than hydrogen, such gas collision quenching terms are generally negligible because the energy spacing between metastable states and states from which radiative transitions can occur are larger than gas kinetic energies. However, for the H(2S) atom, the energy spacing between the metastable level and the  $2P_{\frac{1}{2}}$  state is only the Lamb shift which is small compared to thermal energies. Not only is collision quenching energetically allowed, but, as will be seen in Sec. IV, it is a quite important consideration in experiments on metastable hydrogen atoms.

In writing Eq. (3), we assume that attenuation of the 2S beam by processes other than those giving a single-photon decay may be neglected. Certainly this

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<sup>&</sup>lt;sup>6</sup> W. L. Fite and R. T. Brackmann, Phys. Rev. **112**, 1151 (1958). <sup>6</sup> H. Bethe and E. E. Salpeter, *Encyclopaedia of Physics* (Springer-Verlag, Berlin, 1957), Vol. XXXV, p. 372.

assumption is appropriate for the natural two-photon decay process because its rate<sup>2</sup> is only 8 per sec. While gas collision processes which do not involve the emission of a Lyman-alpha photon may also attenuate the 2S beam (e.g., in a process wherein the excitation energy is transferred to a gas molecule), it may be shown that unless the cross sections for such processes are larger than  $10^{-12}$  cm<sup>2</sup> the ensuing analysis is but slightly affected. It seems doubtful that such nonradiative de-excitation processes have cross sections of this magnitude.

The counts registered by the photon counter from decay of atoms of speed v, radiating in the length interval dx at x is given by

$$dC_v = E(x)G(x)\{w_0 + \sum_i w_i + \frac{3}{2}W_F(x)\}I_v dx/v. \quad (5)$$

Here, E(x) is the probability that a photon reaching the counter window will register a count. It is dependent on x because the sensitivity of the counter is a function of the angle of incidence. G(x) is the probability that a photon originating at x will reach the counter window, if the angular distribution of photons is isotropic. It is assumed that the photons are distributed isotropically from natural and collision-induced decay. For decay in an electric field, the distribution is not isotropic, since the electric dipole is directed parallel to the field. For this reason, the appropriate figure of  $\frac{3}{2}$  has been introduced as a factor before the field-induced decay rate,  $W_F$ . The field-induced decay rate is, of course, spacedependent since the field, F, is a function of position.

Straightforward integration of Eqs. (2) and (5) gives

$$C_{v} = \frac{I_{v}(0)}{v} \int_{0}^{\infty} E(x)G(x) \{ u + \frac{3}{2}\alpha F^{2}(x) \} \\ \times \exp\left\{ -\frac{ux}{v} - \frac{\alpha}{v} \int_{0}^{x} F^{2}(x')dx' \right\} dx, \quad (6)$$

where x is measured from the position of origin of the 2S atoms (i.e., the point of collision of the electron beam and the ground-state atom beam),  $I_v(0)$  is the current of 2S atoms per unit velocity range at x=0 with direction such as to enter the slits and pass through the quenching regions, and  $u \equiv w_0 + \sum_i w_i$ .

In order to evaluate Eq. (6), knowledge of the functions E(x), F(x), and G(x) is required. Since the quenching fields were established by parallel plates, and the beam ran in the mid-plane between the two plates, F(x) is readily determined from potential theory. Indeed, the major reason for the selection of parallel plates in the second quenching region is ready availability of the solution of the fringe fields associated with them. The function G(x) is a simple geometrical function and E(x) had to be determined from the measured angular response of the photon counter.

In order to obtain the total counter output, it is necessary to integrate Eq. (6) over the distribution of velocities,  $I_{\nu}(0)$ . It is straightforward to show from momentum and energy conservation that when a ground-state atom of speed  $v_0$  is excited by an electron whose direction of motion is perpendicular to that of the atom, and when the atom's direction of motion is changed by an angle  $\Psi$  in the plane formed by the two initial directions of motion, then the excited atom possesses one of two speeds given by

$$\psi_{\pm} = v_0 \cos\Psi + \frac{p_0}{M} \sin\Psi$$
$$\pm \left[\frac{p^2}{M^2} - \left(v_0 \sin\Psi - \frac{p_0}{M} \cos\Psi\right)^2\right]^{\frac{1}{2}}, \quad (7)$$

where M is the mass of the atom, and  $p_0$  and p are the scalar momenta of the exciting electron before and after the excitation process, respectively. First, it may be noticed that for the solution to be mathematically real,  $v_0$  must have a limited range. Second, both solutions, when they exist, are physically real, so that a group of atoms of initial speed  $v_0$  splits into two groups on excitation and detection at a fixed angle. In the cases of isotropic scattering of the electron by the atom in the excitation process, or of the momentum of the initial electron being small compared to the initial ground-state atom momentum (as was the case in these experiments), the two final velocity groups are very nearly equally populated.

The distribution of initial velocities,  $I_v(0)$ , of the 2S atoms which would enter the slits in this experiment, was determined by applying the above considerations to a Maxwellian distribution of initial ground-state velocities,  $v_0$ , at a temperature of 2700°K. Integration of Eq. (6) was then carried out numerically for the cases of interest.

There were two major cases of interest. The first was F=0, in which the counter output is given by

$$C(0) = \bar{u} \int_{v_{\rm min}}^{v_{\rm max}} H(v) \, \exp(-ux_0/v) dv, \qquad (8)$$

where H(v) is the tabulated function encompassing all the various experimental and geometrical parameters discussed above,  $x_0$  is the distance to the center of the quenching region, and  $\bar{u}=w_0+\sum_i n_i \langle Q_i v \rangle_{\text{Av}}$ , the average being based on the distribution function H(v). It may be noted that H(v) is very similar to a Maxwellian density distribution at  $T\sim 2900^{\circ}$ K,  $v_{\min}\sim 5.5\times 10^5$ cm/sec, and  $v_{\max}\sim 1.0\times 10^6$  cm/sec. The second case is where the electric-field quenching dominated all other processes. Experimentally, a field of 10 v/cm was usually used, in which the counter output is given by

$$C(10) = \frac{3}{2} \times 0.267 \int_{v_{\min}}^{v_{\max}} H(v) \\ \times \exp\left\{\frac{0.97}{v} - \frac{0.036}{v^2} - \frac{ux_0}{v}\right\} dv, \quad (9)$$

where v is expressed in units of  $10^6$  cm/sec.

To solve for the transition rate,  $\bar{u}$ , a first approximation is made by setting  $\exp(-ux_0/v)=1$ , solving the integrals and dividing Eqs. (8) and (9). This first approximation,  $\bar{u}$ , is iterated back as u into Eqs. (8) and (9), which are then resolved. For the usual experimental conditions where  $ux_0/v_{\min} \leq 0.1$ , a single iteration is required and the transition probability per unit time is given by

$$\bar{u} = w_0 + \sum_i \langle w_i \rangle_{\text{Av}} = 9.8 \times 10^4 C(0) / C(10) \text{ sec}^{-1},$$
 (10)

with mathematical uncertainties being about 1%. This is not the accuracy of the experiment, however, for cumulative physical uncertainties (e.g., inexact knowledge of counter response and geometrical errors) are estimated to be perhaps as high as 20%.

As remarked earlier, it is clear that in this approach, where a reading with no electrical quenching field is required, care must be taken in regard to residual fields which might be left between the plates because of work-function differences, charged surfaces, and other possible effects. Some assurance can be obtained in this regard by observing counter output as a function of electric field in the limit of small fields. It is clear from Eq. (6) that for F sufficiently small, the exponential term containing F may be set equal to 1, in which case  $C_v(F)$  is parabolic with F. On integration over velocity, the same is true. It was found that the counter output was indeed parabolic and by reversing the direction of F that the minimum of the parabola lay between F=0.2 and -0.2 v/cm. Since the decay rate of the 2S atom in a 0.2 v/cm field is only about 100 sec<sup>-1</sup>, and observed decay rates were an order of magnitude higher than this, we can be sure that primary quenching was not caused by residual electric fields normal to the plates. Symmetry of the counter output with reversal of field direction was observed for all values of F.

## IV. COLLISION QUENCHING OF H(2S)

The measured decay rate, u, given in Eq. (10) differs from the natural decay rate in that it contains contributions from quenching of the atoms in collisions with each of the constituents of the residual gas in the vacuum chamber. Although pressures were normally between 5 and  $10 \times 10^{-7}$  mm Hg, the collision quenching was not negligible, since the cross sections for quenching the delicate metastable atom are quite large.

It is to be recalled that  $\langle w_i \rangle_{Av} = n_i \langle Q_i v \rangle_{Av}$ , where  $n_i$  is the number density of the *i*th species,  $Q_i$  is its cross section for quenching the metastable atom, and v is the velocity of the 2S atom. Rather than attempt to measure  $\sum_i \langle w_i \rangle_{Av}$  in a single measurement, the approach used here was to determine, to the accuracy required, (1) the density in the background gas of each constituent through mass analysis of the residual gas and (2) the cross section of each constituent for collision quenching.

#### 1. Mass Analysis of the Residual Gas

For these measurements, a simple small magnetic sector mass spectrometer was inserted into the vacuum chamber. Its electron gun ionizing the residual gas was run at an electron energy similar to the electron energy used with the ionization gauge (so that the same relative proportions of multiply charged ions of any single neutral species would be measured, using either the mass spectrometer or the nondiscriminating ionization gauge). The mass spectrometer used only electrostatic fields in the ionizing region, so that its collection efficiency would be independent of the ion mass, as is the case with the ionization gauge. An electric lens of the mass spectrometer was modulated at 100 cps, so the same ac circuitry could be used on the ac ion current, when a preamplifier of the type described by Fite and Brackmann<sup>7</sup> was used as an impedance changer before the external circuitry.

When the atom beam was being run, the mass analysis of the residual gas in the vacuum chamber (at  $\sim 1 \times 10^{-6}$  mm Hg) showed that the predominant species were H<sub>2</sub>, H<sub>2</sub>O, N<sub>2</sub>, and O<sub>2</sub>. Small traces of Hg (presumably from the diffusion pumps), A, and CO<sub>2</sub> were always found. Several other small peaks, whose identification is not certain, were observed on occasion. The relative magnitude of the mass spectrometer signals for masses 2, 18, 28, and 32 were 3, 1, 3.5, and 1, respectively.

#### 2. Cross Sections for Collision Quenching

In these measurements, the transition probability was measured as a function of density of a particular species. Rewriting Eq. (10) the decay rate may be given by

$$\bar{u} = w_0 + \sum_{i \neq j} \langle w_i \rangle_{\text{Av}} + n_j \langle Q_j v \rangle_{\text{Av}} = 9.8 \times 10^4 \frac{C(10)}{C(0)}.$$
 (11)

By adding to the vacuum chamber additional amounts of the *j*th species of the gas, and plotting *u* as a function of  $n_j$ , a linear plot is found. The slope of this plot is, clearly, the cross section  $Q_j$  times the atom speed ( $\sim 8 \times 10^5$ ). The density,  $n_j$ , was determined from the ionization gauge readings, after suitable correction of the gauge readings for the ionization cross section of the species under study.

Table I summarizes approximate values of these cross sections for the four predominant species in the vacuum. In this table the slopes of the  $\bar{u}$  versus  $n_j$  plots

TABLE I. Approximate values of the cross sections for collision quenching.

Species	$H_2$	$N_2$	$O_2$	$H_2O$
$O \times 10^{14} (\text{cm}^2)$	$0.\tilde{7}$	1.0	0.6	10 <sup>a</sup>

\* To arrive at this figure, the ionization cross section of water was assumed equal to that of molecular nitrogen.

<sup>7</sup> W. L. Fite and R. T. Brackman, Phys. Rev. 112, 1141 (1958).

have been uniformly divided by a speed of  $8 \times 10^5$ cm/sec; the actual velocity range of the accepted 2Satoms is from 5.5 to  $10 \times 10^5$  cm/sec.

The effectiveness of water in quenching the metastable 2S hydrogen atom is quite striking. Its large cross section may be related to the strong electric dipole moment of the water molecule.

### V. RESULTS AND DISCUSSION

For residual gas pressures from 7 to  $9 \times 10^{-7}$  mm Hg, the average rate of single-quantum decay of the 2Shydrogen atom was found to be  $930 \text{ sec}^{-1}$ . Of this, 510sec<sup>-1</sup> is ascribable to collision quenching by the four most abundant gases present in the vacuum chamber. Thus, the final result of this measurement is that the apparent natural lifetime of the metastable hydrogen atom is 2.4 msec, with an estimated over-all probable error of 50%.

This figure can be regarded only as a lower limit on the true natural lifetime for two reasons. First, stray electric fields within the quenching region directed other than normal to the quench plates may have been present. Although a field of only 0.4 v/cm would completely account for the apparent natural decay, in view of the geometry of the quench region and the care

taken to avoid contact potentials and surface charging it is very doubtful that the observed decay arose solely from stray field quenching. Second, gas collision quenching corrections were made for only the four most prominent background gases; gas collision quenching by both trace gases and ground-state hydrogen atoms accompanying the metastable atoms may have contributed substantially to the decay rate.

From the measured lower limit of the metastable atom lifetime, it follows that an upper limit on the admixture (amplitudes squared) of the  $2P_{\frac{1}{2}}$  state with the  $2S_{\frac{1}{2}}$  state is  $7 \times 10^{-7}$ . By applying the result of this measurement in the formulas of Salpeter relating the lifetime of the H(2S) atom and the strength of an electronic electric dipole moment, it follows that the dipole moment strength cannot exceed  $0.0045e\hbar/mc$ .

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# Mathematical Analysis of a Simple Model Related to the Stripping Reaction

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A scattering process in which an incoming bound particle can split up into its component parts when its total energy is above a threshold is of considerable interest physically (deuteron stripping, etc.) and mathematically (analytic properties of the S-matrix, etc.). For such complicated problems it is obviously convenient to have a simple, analytically soluble model for reference, but although many models have been suggested in the past none have proved analytically tractable. In this paper we propose and completely solve a one-dimensional model which, although it is not very physical, has all the desired characteristics. The problem is not mathematically trivial, however, and leads to a Wiener-Hopf integral equation.

**X** E have to begin by apologizing for the presumptuous title of the paper, our excuse being that a similar title has already been used for the same purpose.<sup>1</sup> The mathematical model treated in this paper has only one feature in common with the stripping reaction: It describes a scattering process in which an incoming complex particle might re-emerge as a complex particle or might be split into its components. Problems of this type lead to difficulties since they cannot be treated in the convenient interaction rep-

resentation. Therefore, there has been some desire for a simple mathematical model of such a reaction, and as such the problem has a minor history.

Heisenberg proposed an investigation of the Hamiltoniant

$$-\frac{1}{2}\frac{\partial^2\psi}{\partial u^2} - \frac{1}{2}\frac{\partial^2\psi}{\partial v^2} - \{A\delta(u) + A\delta(v) + 2C\delta(u-v)\}\psi = E\psi.$$

This can be interpreted as describing two particles moving in one dimension and interacting with each other as well as with a fixed scattering potential at the origin. (They can be bound together with an

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 $<sup>\</sup>ddagger$  Throughout we use units in which  $\hbar = m = 1$ .