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Theory of Atomic Lifetime Measurements*

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The theory of atomic lifetime measurements is developed on the basis of the quantum theory of radiation. The effect of coherent emission on the shape of the decay curves is studied in detail. It is shown that the light intensity will oscillate in time when two closely spaced levels decay coherently. Oscillations due to interference between fine-structure levels of the hydrogen atom in particular are discussed.

INTRODUCTION

Recently lifetimes of excited states of atomic hydrogen have been measured using beam-foil techniques.[1] Excited hydrogen atoms are formed by passing H^+, H_2^+, or H_3^+ through a thin foil (usually carbon). The beam on the downstream side of the foil consists of a mixture of many components, some of which are excited hydrogen atoms. The excited states decay by emission of line radiation, and the intensity of this radiation is measured as a function of the distance of the excited atom from the foil. Knowing the velocity of the atoms in the beam enables one to relate the distance of the detector from the foil to the time interval between formation of the excited atom and its subsequent decay. The intensity of the emitted light can, therefore, be measured as a function of time. The intensity usually decreases exponentially with time so the decay curve can be fitted to $e^{-t/\tau}$ and the lifetime τ of the state determined.

The exponential decay curve is expected on the basis of the usual theory,[2] but the usual theory applies only to incoherent decay. If two excited states decay to the same lower state, they decay coherently.[3] Such decays are common, especially for the hydrogen atom. When the decay is coherent, the population of a lower state can oscillate as a function of time. The oscillation frequency is just the difference in energy of the two upper levels divided by Planck's constant. For most decays, this frequency is so large that measurements average over many cycles of oscillation and the oscillatory part of the decay curve averages to zero. When time intervals as short as a nanosecond are measured, oscillations with frequencies of the order of 1000 MHz can be resolved. Since the separation in energy between many of the levels of the hydrogen atom is of the order of 100 MHz, one may expect the oscillations to appear in the experimental data. A brief discussion of the conditions for the observation of the oscillations, and the relevance of these conditions to interpretation of experiments, has been given earlier.[4] The purpose of this paper is to extend the earlier discussion.

The number of photons emitted from an atom excited by a beam-foil collision varies as a function of the angle θ between the emitted ray and the beam according to[5]

$$I(\theta) = I(1 - P\cos^2\theta)/4\pi(1 - \tfrac{1}{3}P), \tag{1}$$

where P is the polarization of light emitted at $\theta = 90°$, and I is the total number of photons. Letting $I_{\parallel}$ denote the number of photons of light polarized parallel to the beam, and letting $I_{\perp}$ denote the number of photons of light polarized perpendicular to the beam, we have

$$P = (I_{\parallel} - I_{\perp})/(I_{\parallel} + I_{\perp}) = (3I_{\parallel} - I)/(I_{\parallel} + I), \tag{2}$$

where $$I = I_{\parallel} + 2I_{\perp}. \tag{3}$$

From (1) and (2) we see that the number of photons of light emitted at any angle is determined by $I_{\parallel}$ and I. The relationship of $I_{\parallel}$ and I to excitation cross sections, decay widths, and the time interval after collision is the subject of this paper. Reference to specific experiments is made via Eqs. (1)–(3).

In the earlier publication,[4] we emphasized that the total number of photons I is unmodulated. If measurements are made at 54.8°, then $\cos\theta$ equals $1/\sqrt{3}$ and the observed intensity is proportional to I. The decay curve will not oscillate and will be better suited to the determination of atomic lifetimes. On the other hand, one may emphasize the oscillations by measuring light polarized parallel to the beam axis or perpendicular to the beam axis. The frequency of oscillation provides a direct measurement of the splitting of the decaying levels. Further, the amplitudes of the oscillations are determined by the relative population of excited states. Thus, an extension of the previous discussion to include some general conditions for the occurrence of the oscillation and the relationship between the amplitudes of the oscillations and excitation cross sections is desirable.

Section I reviews the theory of Ref. (4) and Sec. II treats cascading. Since the cascade portion of decay curves contains contributions from an infinite number of states, this portion is inherently difficult to analyze in terms of properties of individual states. For this reason Sec. II presents only an outline of the general theory of cascading, the main objective being to determine when the interference of coherent decays is likely to lead to noticeable oscillations. Section III discusses general conditions for the vanishing of the oscillations. We show that the total intensity is unmodulated, and that light from the decay of fine or hyperfine multiplets is unmodulated if the cross section for an orbital angular momentum substate LM_L is independent of M_L. Section IV presents a detailed application of the theory to hydrogenlike ions.

In Secs. I and II, atomic states are denoted by ajm, where j is the total angular momentum of the atom, and a denotes all other quantum numbers. If only fine-structure splitting is important, j and m denote the total angular momentum quantum numbers of the electron J and M_J. If hyperfine splitting is significant, j and m refer to the total internal angular momentum of the atom F and M_F. The axis of quantization is the beam axis.

I. DECAY TO A SHARP LOWER LEVEL

Consider a group of upper levels with quantum numbers a_1, j_1, and m_1 which are degenerate in the magnetic quantum number m_1, but are nondegenerate in the j_1. The decay is shown in Fig. 1. The upper levels decay to a group of lower levels ajm. We are interested in the probability amplitude that the atom is in state ajm at time t, if it was in state $a_1j_1m_1$ at $t=0$.

If the atom is in state $a_1j_1m_1$ at $t=0$, the amplitude that it will be, in the state ajm and that a photon of angular frequency ω_S is in the surrounding field, is[6,7]

$$B(a_1j_1m_1, a_0j_0m_0) = H(a_1j_1m_1, a_0j_0m_0) \times \frac{\exp[i\omega_S - i\omega(a_1j_1, a_0j_0) - \gamma_{a_1}]t}{\omega_S - \omega(a_1j_1, a_0j_0) - i\gamma_{a_1}}, \quad (4)$$

where

$$H(a_1j_1m_1, a_0j_0m_0) = i(2\pi e^2/k_S)^{1/2} \times \omega(a_1j_1, a_0j_0)(a_1j_1m_1|\vec{\epsilon}_S \cdot \vec{\mathbf{X}}|a_0j_0m_0), \quad (5)$$

$$\omega(a_1j_1, a_0j_0) = (E_{a_1j_1} - E_{a_0j_0})/\hbar, \quad (6)$$

and $k_S = \hbar\omega_S$.

In (5) $\vec{\epsilon}_S$ is the polarization vector of the emitted photon, $\vec{\mathbf{X}}$ is the dipole length operator, k_S is the magnitude of its wave vector, and $1/\gamma_{a_1}$ is the mean lifetime of all states $a_1j_1m_1$. Equation (1) is a good approximation to the decay amplitude for $t \gg 1/\omega(a_1j_1, a_0j_0)$. For visible light $1/\omega$ is of the order of 10^{-6} nsec and the restriction $t > 1/\omega$ is of no consequence.

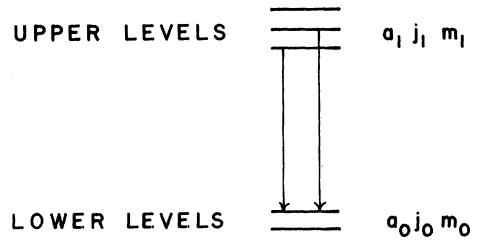

FIG. 1. Schematic level diagram showing a coherent decay.

In a collision, the excited states are populated in a time of the order of 10^{-15} sec, which is much shorter than typical decay times ($\sim 10^{-8}$ sec). Then at $t=0$ the excited states are populated according to the excitation amplitudes $A(a_1j_1m_1)$, and the amplitude for finding the atom in state ajm and a photon in the surrounding field is the sum

$$B = \sum_1 A(a_1j_1m_1)B(a_1j_1m_1, a_0j_0m_0), \quad (7)$$

where $\sum_1$ is short for $\sum_{a_1j_1m_1}$.

It should be emphasized that (7) is an approximation because (4) is an approximation. Neglected in (4) is the direct coupling between states of the same multiplet. This neglect is justified because the coupling matrix element (5) is proportional to the fine-structure or hyperfine-structure splitting which is small compared to the difference $E_{a_1j_1} - E_{a_0j_0}$. Indirect coupling via the lower states $a_0j_0m_0$ is proportional to the average of

$$\sum_0 (a_1j_1m_1|\vec{\epsilon}_S \cdot \vec{\mathbf{X}}|a_0j_0m_0) \times (a_0j_0m_0|\vec{\epsilon}_S \cdot \vec{\mathbf{X}}|a_1j_1m_1) \quad (8)$$

over all polarizations. Breit shows[3] that this average is zero, thus (7) is a good approximation. When the atom is in an external field, the eigenstates of the atom are no longer eigenstates of angular momentum. Then the sum (8) does not vanish and (7) is a poor approximation. The theory in this paper assumes (8), therefore, it does not apply to atoms in external fields; for example, it does not apply to the Stark effect.

The probability for finding the atom in state $a_0j_0m_0$ is obtained by integrating $|B|^2$ over all normal modes of the field

$$\rho_S dk_S = k_S^2 dk_S d\Omega/(2\pi hc)^3, \tag{9}$$

where $d\Omega$ is the element of solid angle into which the photon is emitted. The squared modulus of B is a sum of terms of the form

$$A(a_1 j_1 m_1)B(a_1 j_1 m_1,\ a_0 j_0 m_0)A(a_1' j_1' m_1')^* \times B(a_1' j_1' m_1',\ a_0 j_0 m_0)^* . \tag{10}$$

The integral of each term over k_S may be performed by noting that the main contribution to the integral comes from the region $\omega_S \approx \omega(a_1 j_1, a_0 j_0)$, therefore, the integral over k_S can be extended to $-\infty$ with negligible error. Then all integrals over k_S can be converted to contour integrals.

Integrating (10) over the density of states (9) gives

$$KA(a_1 j_1 m_1)(a_1 j_1 m_1|X_q|a_0 j_0 m_0) \times (a_1' j_1' m_1')^*(a_1' j_1' m_1'|X_q|a_0 j_0 m_0)^* \times \frac{1-\exp[-i\omega(a_1 j_1, a_1' j_1') - \gamma_{a_1} - \gamma_{a_1'}]t}{\omega(a_1 j_1, a_1' j_1') - i(\gamma_{a_1}+\gamma_{a_1'})}, \tag{11}$$

where $K = 4e^2\omega^3/3c^3$, (12)

and we have set $\omega(a_1 j_1,\ a_0 j_0) \approx \omega \simeq$ const since the splitting of the upper levels is much smaller than the energy difference between the upper and lower levels. The subscript q on x refers to the polarization of the emitted light referred to a space-fixed coordinate system in which the beam axis is taken to be the z axis.

The probability $P'(t)$ (not to be confused with the polarization P which is also a function of time) for finding the atom in one of the lower levels and a photon of polarization q in surrounding space is the sum of (11) over $a_1 k_1 m_1$, $a_1' j_1' m_1'$, and $a_0 j_0 m_0$:

$$P'(t) = K \sum_{011'} A(a_1 j_1 m_1)(a_1 j_1 m_1|X_q|a_0 j_0 m_0) \times A(a_1' j_1' m_1')^*(a_1' j_1' m_1'|X_q|a_0 j_0 m_0)^* \times \frac{1-\exp[-i\omega(a_1 j_1, a_1' j_1') - \gamma_{a_1} - \gamma_{a_1'}]t}{\omega(a_1 j_1, a_1' j_1') - i(\gamma_{a_1}+\gamma_{a_1'})}. \tag{13}$$

Because $P'(t)$ is the probability for finding the atom in one of the lower levels at time t if it was in an upper level at $t=0$, $P'(t)$ is proportional to the number of photons emitted between $t=0$ and $t=t$. In some experiments this integrated number of photons is measured, and (11) describes the measured quantity. Because the upper states decay coherently, the integrated intensity oscillates as a function of time.

Experiments may also measure the number of photons emitted in a short time interval. Then the number of photons of a specific polarization q is proportional to the time rate of change of $P'(t)$. This rate of change is just $\dot{P}'(t)$, obtained by differentiating (13),

$$\dot{P}'(t) = K \sum_{011'} A(a_1 j_1 m_1)(a_1 j_1 m_1|X_q|a_0 j_0 m_0) \times A(a_1' j_1' m_1')^*(a_1' j_1' m_1'|X_q|a_0 j_0 m_0)^* \times \exp[-i\omega(a_1 j_1,\ a_1' j_1') - \gamma_{a_1} - \gamma_{a_1'}]t. \tag{14}$$

Equations (13) and (14) are the fundamental equations of our theory of lifetime measurements. They are implicit in much earlier work,[3,8] but this derivation is presented here for completeness and because it is easily extended to include cascading. Equations (13) and (14) contain the sum of oscillatory terms as well as the usual exponential terms. When the period of the oscillations is very short compared to the time interval over which the photon flux is averaged by the measuring apparatus, the interference terms average to zero and only the exponential decay terms remain in both (13) and (14). In (13) the oscillatory terms are small compared to the nonoscillatory terms when the ratio

$$(\gamma_{a_1}+\gamma_{a_1'})/\omega(a_1 j_1,\ a_1' j_1') \tag{15}$$

is small. Consequently, when the decay widths are much smaller than the level splitting the time-integrated photon flux will be unmodulated, even though a specific polarization is detected. When the number of photons emitted in a short time interval is measured, the intensity is given by (14). The oscillatory terms are as large as the nonoscillatory terms, and therefore must be included to obtain the correct value of $P'(t)$. However, when $\omega(a_1 j_1,\ a_1' j_1')$ is small compared to the average decay width $\gamma_{a_1}+\gamma_{a_1'}$, the light intensity will decrease rapidly during one period of oscillation, and the oscillation will not be noted. An example of this behavior is given in Sec. IV.

II. CASCADING

Lifetimes of excited states can also be measured indirectly by detecting light emitted in the transition of the intermediate states $a_0 j_0 m_0$ to a group of lower levels when the intermediate states are populated by cascading from the upper states $a_1 j_1 m_1$. The cascade is shown in Fig. 2. The con-

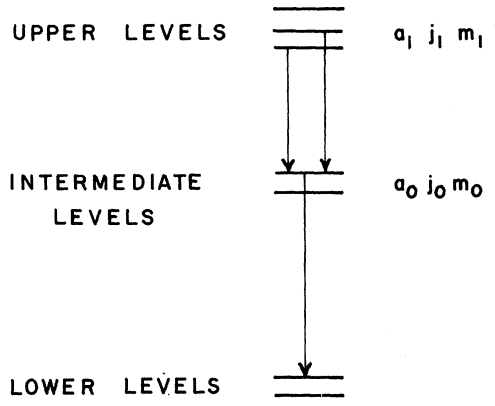

FIG. 2. Schematic level diagram showing a cascade for which an intermediate level is populated by a coherent decay from a group of upper levels. Light emitted in the subsequent transition from the intermediate levels to the lower levels is modulated with a frequency corresponding to the splitting of the upper levels.

ventional theory of these measurements has been discussed by Bickel.[2] On the basis of the discussion in Sec. I, the light intensity can be expected to oscillate if the group of upper levels is closely spaced in energy. The conventional theory does not apply, and a treatment based on the quantum theory of radiation is necessary.

We may qualitatively discuss the decay using Eq. (13). Here the number of photons emitted in the decay of state $a_0j_0m_0$ is proportional to the probability of finding the atom in state $a_0j_0m_0$. This probability is given approximately by (13) (when summed over q but not over $a_0j_0m_0$), and we see that the population of a given level $a_0j_0m_0$ will oscillate in time. This probability multiplied by the square of the dipole matrix element for the decay to a lower level is proportional to the number of photons emitted in the decay, provided the levels $a_0j_0m_0$ are degenerate, or have splittings which are much larger than the decay widths so that one need not consider interference due to the splittings of the intermediate states. Since m_0 refers to a specific magnetic sublevel, we expect the number of photons of a specific polarization to oscillate in time.

Our discussion of cascading on the basis of (13) is correct only when the decay width of $a_0j_0m_0$ is small compared to the decay width of $a_1j_1m_1$. In this case $a_0j_0m_0$ is a fairly sharp level and (13) applies. Right here, however, we are interested in precisely the opposite limit, namely when the lower level decays much faster than the upper level. To investigate this case, we can follow our previous approach except that (4) must be replaced by the amplitude appropriate for the case when $a_0j_0m_0$ can itself decay. This amplitude has been obtained by Weisskopf and Wigner.[7] They find

$$B(a_1j_1m_1, a_0j_0m_0) = H(a_1j_1m_1, a_0j_0m_0)\exp(-\gamma_{a_0}t)\frac{\exp[i\omega_s - i\omega(a_1j_1, a_0j_0) - (\gamma_{a_1}-\gamma_{a_0})]t}{\omega_s - \omega(a_1j_1, a_0j_0) - (\gamma_{a_1}-\gamma_{a_0})}, \tag{16}$$

where the amplitude has been rewritten in the notation of Ref. (6). We see that (16) differs from (4) in that γ_{a_1} is replaced by $\gamma_{a_1}-\gamma_{a_0}$ and the amplitude is multiplied by $\exp(-\gamma_{a_0}t)$. With this observation and recalling Eq. (13), we can immediately write down the probability $P'_{a_0j_0m_0}(t)$ for finding the atom in the state $a_0j_0m_0$:

$$P'_{a_0j_0m_0}(t) = K\sum_{q11'} A(a_1j_1m_1)(a_1j_1m_1|X_q|a_0j_0m_0)A(a_1j_1m_1)^*$$

$$\times (a_1'j_1'm_1'|X_q|a_0j_0m_0)^*\frac{\exp(-2\gamma_{a_0}t) - \exp[-i\omega(a_1j_1, a_1'j_1') - \gamma_{a_1} - \gamma_{a_1'}]t}{\omega(a_1j_1, a_1'j_1') - i(\gamma_{a_1}+\gamma_{a_1'}-2\gamma_{a_0})}. \tag{17}$$

The main significance of (17) derives from the observation that the magnitude of oscillating terms to the nonoscillatory terms is governed by the ratio

$$(\gamma_{a_1}+\gamma_{a_1'}-2\gamma_{a_0})/\omega(a_1j_1, a_1'j_1'). \tag{18}$$

Since we are mainly interested in cases where $\gamma_{a_0} \gg \gamma_{a_1}$ or $\gamma_{a_1'}$, we see that the oscillatory terms can be large even if the upper levels decay slowly, consequently, oscillations may even be important in the cascade region.

III. GENERAL CONDITIONS FOR THE VANISHING OF THE OSCILLATIONS

A. Proof that the Total Intensity is Unmodulated

The theory presented in Secs. I and II can be applied to two somewhat different types of experiments. One may wish to determine only the decay

rates of excited states. The additional oscillatory terms merely complicate the analysis of the data since now $\log I$ is no longer a linear function of time. On the other hand, one may wish to measure the excitation amplitudes and excitation cross sections. As mentioned in the Introduction, the coefficients of the oscillatory terms can be extracted from experimental data, and excitation cross section can in turn be extracted from the coefficients. One may also wish to measure the oscillation frequencies and thereby determine the splittings of the upper levels. For purposes of designing and interpreting the results of these various experiments, it is desirable to have some general criteria for the observation or elimination of the oscillations. We first show that the total intensity is unmodulated.

The coefficient of the oscillatory terms in (13), (14), and (17) contains the factor

$$\sum_q \sum_0 (a_1 j_1 m_1 | X_q | a_0 j_0 m_0) \times (a_1' j_1' m_1' | X_q | a_0' j_0' m_0')^*, \tag{19}$$

where the sum over all polarizations is included to obtain the total intensity. Now Breit has shown[3] that (19) vanishes for fine-structure or hyperfine-structure levels with the same orbital angular momenta unless $a_1 j_1 m_1 = a_1' j_1' m_1'$. We need only extend Breit's proof to include the case when the states $a_1 j_1 m_1$ and $a_1' j_1' m_1'$ have different values of orbital angular momenta; for example, S and D states of hydrogenlike ions decaying to a common P state.

In the representation $LM_L SM_S IM_I$, the matrix (19) is diagonal in the electron-spin and the nuclear-spin quantum numbers. The factors depending upon the orbital angular momentum quantum numbers are just

$$\sum_{qM_{L_0}} (L_1 M_1 | X_q | L_0 M_{L_0}) (L_1' M_{L_1'} | X_q | L_0 M_{L0})^*. \tag{20}$$

Using the Wigner-Eckart theorem[9] to factor out the geometrical factors in (20) gives

$$(L_1 \| X \| L_0)(L_1' \| X \| L_0)^* \sum_{qM_{L_0}} (L_0 1 L_1 M_{L_1} | L_0 M_{L_0} 1q)(L_0 M_{L0} 1q | L_0 1 L_1' M_{L_1'}). \tag{21}$$

By the orthogonality properties of the Clebsch-Gordan coefficients, (21) vanishes unless $L_1 = L_1'$ and $M_{L_1} = M_{L_1'}$. Thus, the matrix (19) is diagonal in the representation $LM_L SM_S IM_I$. Furthermore, each submatrix corresponding to states with the same L is a constant times the unit matrix. Since the transformation to the physical states ajm does not mix states with different L, the matrix (19) is diagonal in the representation ajm, and the total intensity does not oscillate. Note that this proof does not hold for atoms in an external field, for which the transformation to the physical states does mix states with different L.

B. Vanishing of the Oscillations When States With Different M_L are Equally Populated

The amplitudes $A(a_1 j_1 m_1)$ contain all the information about the excitation of the upper states in the atomic collision. A detailed theory of these collisions is not available at present, but some results can be obtained using only very general properties of the scattering amplitudes. Of greatest importance here are the symmetry properties of the amplitudes. The electrostatic interaction is primarily responsible for the excitation of the atoms in atomic collisions. For atomic states which obey LS coupling rules, the amplitudes are conveniently written in the representation $LM_L SM_S IM_I$. To facilitate the discussion of the symmetry properties of the amplitudes, we write the amplitudes more explicitly as transition matrix elements:

$$A(a_1 j_1 m_1) = (a_1 j_1 m_1, fkk' \cdots | T | i) = \sum_{M_L M_S M_I} (a_1 j_1 m_1 | LM_L SM_S IM_I) \times (LM_L SM_S IM_I, fkk' \cdots | T | i), \tag{22}$$

where i denotes the initial state of the beam-foil system, f denotes the internal quantum numbers of all undetected particles in the final state of the beam-foil system, $k, k' \cdots$ are the momentum vectors of the outgoing particles, and T is the transition operator. To obtain the intensity of light emitted in a decay we must average (13), (14), or (17) over the initial state i and the sum over the final states. The required sums are obtained using a generalization of a result due to Percival and Seaton.[5] Consider the integral

$$\int dk\, dk' \cdots (LM_L SM_S IM_I, fkk' \cdots | T | i) \times (i | T | L' M_L' S' M_S' I' M_I, fkk' \cdots). \tag{23}$$

We will show that the integral is zero unless $M_L + M_S = M_L' + M_S'$ and $M_I = M_I'$.

The transition amplitude and, consequently (23), is a complex number whose value is independent of the choice of coordinate systems. This is true on very general grounds, but in our case, it is easily seen to hold because all spatial coordinates in the definition of the T matrix element are integrated over. Expression (23) must be the same number if we choose a new system of coordinates to write the T operator and the wave functions. We choose a new system related to the old system by a rotation through an angle γ about the axis of the incoming particles. The states $|LM_L SM_S IM_I)$ transform as

$$|LM_L SM_S IM_I) \to \exp[-i(M_L + M_S + M_I)\gamma] \times |LM_L SM_S IM_I) . \quad (24)$$

The transformation of the plane wave functions is obtained from the partial-wave expansion

$$e^{ik\cdot r} = \sum_{lm} j_l(kr) Y_{lm}^*(\hat{k}) Y_{lm}(\hat{r}) \to \sum_{lm} j_l \times (kr) Y_{lm}^*(\hat{k}) e^{-im\gamma} Y_{lm}(\hat{r}) . \quad (25)$$

Since all azimuthal directions of the outgoing particles $kk'\cdots$ are integrated over and since

$$\int d\phi_k \, Y_{lm}^*(\hat{k}) Y_{l'm'}(\hat{k}) = 0, \quad \text{unless } m = m', \quad (26)$$

all exponential factors $e^{-im\gamma}$ coming from the plane-wave terms in Eq. (23) cancel.

The states $|i)$ and $|f)$ transform as

$$|i) \to \exp(-iM_i\gamma)|i), \qquad |f) \to \exp(-iM_f\gamma)|f) . \quad (27)$$

Here M_i is the projection of the total angular momentum of the initial state of the beam-foil system onto the axis of the incoming beam, and M_f is the projection of the total angular momentum of the final state of all particles, except the decaying atom, along the axis of the incoming beam. Included in the total angular momentum of $|i)$ and $|f)$ are the orbital angular momentum of the electrons, their spin angular momentum, the motional angular momentum of all foil nuclei, and the intrinsic angular momentum of all nuclei. Since $(f|T|i)$ is the complex conjugate of $(i|T|f)$ the factors $e^{-iM_f\gamma}$ and $e^{-iM_i\gamma}$ also cancel. The transformation only multiplies Eq. (22) by

$$\exp[i(M_L + M_S + M_I - M_L' - M_S' - M_I')\gamma].$$

But Eq. (22) is just a complex number whose value is independent of the choice of coordinate systems. This can only be so if (22) is zero or if $M_L + M_S + M_I = M_L' + M_S' + M_I'$. Since T does not depend upon the nuclear spin, Eq. (22) is zero unless both $M_L + M_S = M_L' + M_S'$ and $M_I = M_I'$. When LS coupling holds, we further have $M_L = M_L'$ and $M_S = M_S'$. When the wave function for the final state depends upon J, or when T depends upon the spin variables, (22) vanishes only when $M_J \neq M_J'$ where $M_J = M_L + M_S$.

This proof is a rather standard application of group theory,[10] but it seems worthwhile to dwell on it in some detail to show the essential conditions under which it is valid. The most important requirement is that all azimuthal directions are integrated over. Results which use this theorem are not directly applicable to experiments in which the direction of some outgoing particles is detected. Our proof also assumes that the target is cylindrically symmetric so that T is invariant to rotations about the beam axis. This means that the foil must be isotropic. More precisely, it means that if the incoming particle makes a collision, the probability that it will make a second collision must be independent of the azimuthal direction of the particle momentum after the first collision. This is not the case for a crystal with well-defined channels along which the fragments can travel.

The intensity of light detected in an experiment is proportional to the sum of (13) or (14) over all undetected final states and an average over all initial states of the beam-foil system. We denote the sum and average by $\mathbf{S}$. Included in the sum and average is an integration over all direction and energies of the outgoing particles. The integrations are carried out using (23). When $L' = L, S' = S$, and $I' = I$ the summation of (23) can be written

$$\mathbf{S}(LM_L SM_S IM_I, fkk'\cdots|T|i)(i|T|LM_L' SM_S' \times IM_I', fkk'\cdots) = \delta_{M_L M_L'} \delta_{M_S M_S'} \delta_{M_I M_I'} \times V_i \sigma_{LM_L} / (2S+1)(2I+1), \quad (28)$$

where σ_{LM_L} is the cross section for forming the atom in state LM_L' and V_i is the initial velocity. Because the cross section is independent of the electron and nuclear spin quantum numbers, they have been dropped on the subscripts of σ. This formula will be useful when the excited states are fine-structure or hyperfine-structure states. In these cases all states have the same L, S, and I.

The coefficients of oscillatory terms in (13), (14), and (17) contain the factor

$$\mathbf{S} A(a_1 j_1 m_1) A(a_1' j_1' m_1')^* . \quad (29)$$

Using (22) and (28) to evaluate the sum S when $L = L'$, we have

$$S_{A(a_1 j_1 m_1)A(a_1, j_1, m_1,)} = \sum_{M_L M_S M_L,} (a_1 j_1 m_1 | LM_L SM_S IM_I)(LM_L SM_S IM_I a_1, j_1, m_1,) \times V_i \sigma_{LM_L} / (2S+1)(1I+1). \tag{30}$$

When σ_{LM_L} is independent of M_L the right-hand side of (30) becomes

$$V_i \sigma_L \sum_{M_L M_S M_I} (a_1 j_1 m_1 | LM_L SM_S IM_I)(LM_L SM_S IM_I a_1, j_1, m_1,)/(2S+1)(2I+1)$$

$$= V_i \sigma_L (a_1 j_1 m_1 | a_1, j_1, m_1,)/(2S+1)(2I+1)$$

$$= V_i \sigma_L \delta_{a_1 a_1'} \delta_{j_1 j_1'} \delta_{m_1 m_1'} / (2S+1)(2I+1), \tag{31}$$

and we see that the light intensity is unmodulated even when light of a specific polarization is detected. We emphasize that, in contrast to our result that the total intensity is unmodulated, this result applies only to modulation from states with the same orbital angular momentum. It does not apply for example to interference in the decay of $S_{1/2}$ and $D_{3/2}$ or $D_{5/2}$ states of hydrogen to a common $P_{1/2}$ or $P_{3/2}$ state.

One significant corollary to (31) is that hyperfine splitting of S states does not produce oscillations in decay curves, since $\sigma_{L=0}$ is clearly independent of M_L. Thus hyperfine splittings of S states cannot be measured by observing oscillation frequencies in decay curves as long as the experimental arrangement maintains cylindrical symmetry. Cylindrical symmetry could be destroyed by detecting light from atoms which are scattered at a definite azimuthal angle for example.

Equation (31) holds as long as LS coupling holds. When LS coupling no longer holds, one can show by the techniques used to obtain Eqs. (30) and (31) that the equation

$$S_{A(a_1 j_1 m_1)A(a_1, j_1, m_1)} = \sum_{M_J M_I} (a_1 j_1 m_1 | JM_J IM_I)(JM_J IM | a_1, j_1, m_1,) V_i \sigma_{JM_J} / (2I+1) \tag{32}$$

holds, where σ_{JM_J} is the cross section for exciting a particular state of total electron angular momentum. Analogously to (31) one has that the light is unmodulated if σ_{JM_J} is independent of M_J. An analog of the corollary to Eq. (31) also holds. States with $J=0$ have no hyperfine splitting but states with $J=\frac{1}{2}$ do. However, because the beam-foil system is invariant to reflections in any plane passing through the beam, one has that σ_{JM_J} (and σ_{LM_L}) is independent of the sign of M_J (or M_L). It then follows that σ_{JM_J} is independent of M_J if $J=\frac{1}{2}$, therefore, states with $J=\frac{1}{2}$ will show no oscillations resulting from interference of decays of different hyperfine levels. For example, $P_{1/2}$, $F=0$, 1 states of hydrogen, although split by the hyperfine interaction, do not give rise to oscillations in decay curves.

IV. INTERFERENCE IN THE DECAY OF EXCITED HYDROGENLIKE IONS

In this section, we discuss the decay of excited states of the hydrogen atom ($I=\frac{1}{2}$) in some detail. For reasons stated in the Introduction we will neglect cascading. In addition, we will omit interference of decays from states with different orbital angular momenta. The omission of interference terms from states with different orbital angular momenta cannot be rigorously justified,[11] however, and our treatment is incomplete in this regard.

A. Influence of Hyperfine Structure

Only S and P states have hyperfine splittings comparable to their decay widths. In light of the results of Sec. III B, only the decay of $P_{3/2}$ states will show modulations due to hyperfine structure. Since the hyperfine splitting decreases rapidly as the principal quantum number increases, and since the hyperfine splitting of the $2P_{3/2}$ state is already a factor of 4 smaller than the decay width, we will only consider the effect of hyperfine splitting on the Lyman-α decay curve. The relevant coefficients have already been obtained by Percival

and Seaton.[5] Using the results of Sec. (3.6) of their paper, we find

$$I_{q=0} = 2\gamma 36^{-1}[19\sigma_1 + 17\sigma_0 + 13(\sigma_0 - \sigma_1)\cos\Omega t]\exp(-2\gamma t), \quad (33)$$

where $\Omega = 3.71 \times 10^8$ sec^{-1} and $2\gamma = 6.25 \times 10^8$ sec^{-1}. A graph of $I_{q=0}$ versus time is shown in Fig. 3 for three choices of σ_0 and σ_1. Curve A is for $\sigma_1 = 0$, curve B for $\sigma_0 = \sigma_1$, and curve C for $\sigma_0 = 0$. The oscillations are too small to be noticed as oscillations, but the presence of the $\cos\Omega t$ term changes the apparent slope of the decay curve. Curve B represents a purely exponential decay and the slope of the line gives the correct lifetime. This example illustrates the necessity of either measuring the total intensity, which is unmodulated, or measuring the polarization of the emitted radiation to determine the extent of the modulation, in order to obtain accurate lifetimes. Measurements of the lifetime of the $2P$ state of hydrogen by Chupp *et al.*[1] are unaffected by interference of hyperfine levels since they determined that the Lyman-α light was unpolarized, indicating that $\sigma_0 = \sigma_1$. Their experimental curve corresponds to the pure exponential decay curve B in Fig. 3.

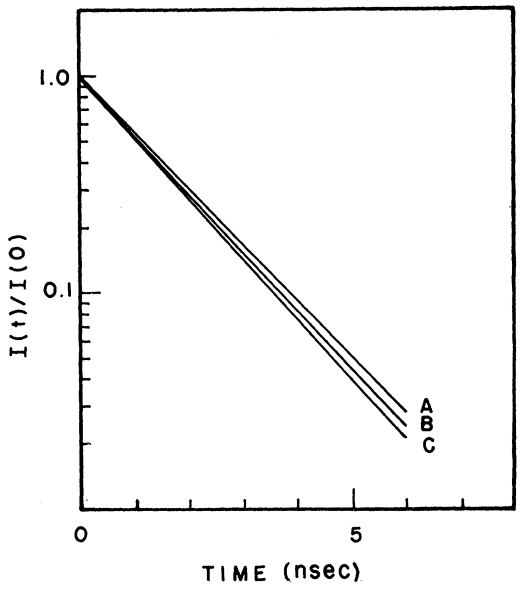

FIG. 3. Effect of hyperfine structure upon the decay curve for the intensity $I(t)$ of Lyman-α radiation polarized parallel to the beam for three magnetic-substate populations. Curve A is for $\sigma_1 = 0.$, curve B is for $\sigma_0 = \sigma_1$, and curve C is for $\sigma_0 = 0$.

B. Influence of Fine Structure

Many fine-structure splittings of excited states of atomic hydrogen are in the 100–1000 MHz range. They can therefore give rise to measurable oscillations in decay curves. A complete analysis of the decay curves, to obtain cross sections σ_{M_L} as well as mean lifetimes, requires expressions analogous to (33). The total intensity I is given by the product of the total cross section, times the transition probability for the decay, times the exponential factor. Denoting the transition probability by $X(L, L_0)$ and the decay width of the excited state by $2\gamma_L'$, we have for the total intensity I,

$$I = X(L, L_0)\sigma_T \exp(-2\gamma_L t). \quad (34)$$

In (34) the dependence of $X(L, L_0)$ upon the principle quantum numbers of the initial and final states has been suppressed. Since σ_{M_L} is independent of the sign of M_L, σ_T is given by

$$\sigma_T = \sigma_0 + \sum_{M_L=1}^{L} 2\sigma_{M_L}. \quad (35)$$

We seek analogous expressions for $I_{q=0} = I_{\|}$. Since only fine-structure splitting is important, the quantum numbers $a_1 j_1 m_1$ in (14) will be explicitly denoted by $LSJM_J$, the quantum numbers $a_0 j_0 m_0$ by $L_0 S_0 J_0 M_{J_0}$, and the quantum numbers $a_1' j_1' m_1'$ by $LSJ'M_J'$. The decay widths γ_{a_1} and $\gamma_{a_1'}$ are equal and will be denoted by γ_L. Furthermore, since $S = \frac{1}{2}$, each fine-structure multiplet consists of two levels and there is only one oscillation frequency Ω. Then Eq. (14) becomes

$$dP/dt = \sum_{J'M_{J'}JM_J J_0 M_{J_0}} A(LSJM_J)(LSJM_J|X_q|L_0S_0J_0M_{J_0}) \times A(LSJ'M_{J'})^*(LSJ'M_{J'}|X_q|L_0S_0J_0M_{J_0})^* \exp[-i\omega(J,J') - \gamma_L]t. \quad (36)$$

Equation (26) is summed over final states and averaged over initial states using (28) and(22). To perform the sums we may use a method of Percival and Seaton.[5] We note that when the sum S has been carried out (36) has the form

$$I_q = (a + b\cos\Omega t)\exp(-2\gamma t). \quad (37)$$

We will calculate a and b by first calculating $a+b$ and a. The sum $a+b$ is most easily calculated. It is equal to the right-hand side of Eq. (36) at time $t=0$, and is simply the product of four matrices in the $LSJM_J$ representation. Since the product must have the same value if expressed in the LM_LSM_S representation, the sum S can then be carried out using Eq. (28) giving

$$I_q = K \sum_{M_L M_S M_{L_0} M_{S_0}} V_i \sigma_{LM_L} (LM_L SM_S | X_q | L_0 M_{L_0} S_0 M_{S_0})^2/(2S+1),$$
$$I_q = K \sum_{M_L M_S M_{L_0}} V_i \sigma_{LM_L} (LM_L | X_q | L_0 M_{L_0})^2, \tag{38}$$

where the last equality holds because X_q does not operate on the spin variables. Using the Wigner-Eckart theorem to factor out the geometrical factors from the dipole matrix element gives finally

$$a+b = X(L, L_0) \sum_{M_L M_{L_0}} V_i \sigma_{LM_L} (L_0 1 LM_L | L_0 M_{L_0} 1q)^2, \tag{39}$$

where the decay probability $X(L, L_0)$ is given by

$$X(L, L_0) = K(L \| X \| L_0)^2/(2L+1). \tag{40}$$

Summation over terms on the right-hand side of (36) with $J = J'$ gives the constant,

$$a = S \sum_{JM_J M_{J'} J_0 M_{J_0}} A(LSJM_J)(LSJM_J | X_q | L_0 S_0 J_0 M_{J_0}) A(LSJM_{J'})^* \times (LSJM_{J'} | X_q | L_0 S_0 J_0 M_{J_0})^*. \tag{41}$$

The sum over J_0 and M_J may be replaced by a sum over M_{L_0} and M_{S_0} since the projection operators

$$\sum_{J_0 M_{J_0}} |L_0 S_0 J_0 M_{J_0})(L_0 S_0 J_0 M_{J_0}|$$

and

$$\sum_{M_{L_0} M_{S_0}} |L_0 M_{L_0} S_0 M_{S_0})(L_0 M_{L_0} S_0 M_{S_0}|$$

project into the same space. All vectors of other states in the $LSJM_J$ representation are written in the LM_LSM_S representation using the transformation (22) and the relation

$$|LSJM_J) = \sum_{M_L M_S} |LM_L SM_S)(LM_L SM_S | LSJM_J). \tag{42}$$

With these transformations, and using (30) and the Wigner-Eckart theorem to evaluate the sum S, we find

$$a = X(L, L_0) \sum_{M_{L_0} M_{S_0} JM_L M_S} V_i \sigma_{LM_L} [(LM_L SM_S | LSJM_{L_0} + q + M_{S_0})(LSJM_{L_0} + q + M_{S_0} | LM_{L_0} + q SM_{S_0}) \times (L_0 1 LM_{L_0} + q | L_0 M_{L_0} 1q)]^2/(2S+1). \tag{43}$$

A FORTRAN program was written to evaluate the coefficients of σ_{LM_L} in (39) and (43) for the case when $q=0$. The results for $L=0$ to 4 and $L_0=L\pm1$ are presented in Table I. They suffice to analyze decay from states with principle quantum numbers up to five. Some qualitative remarks concerning the magnitude of the oscillations can be made on the basis of Table I. First, we note that the modulation is 100% only when $\sigma_{M_L}=0$ for $M_L\neq L$, and the decay takes the atom from a state with orbital angular momentum L to a state with orbital angular momentum $L-1$. This follows directly from the observation that the coefficient $(L_0 1 L M_L | L_0 M_{L_0} 1 q)$ in (39) is zero when $q=0$, $L>L_0$ and $M_L=M_{L_0}=L$. Preferential population of the state with $M_L=L$ seems unlikely, however, and one does not expect the modulation to be as high as 100%. In this connection, we note that the a_{M_L}'s increase relative to the b_{M_L}'s as L increases. For most reasonable types of populations this implies that light coming from the decay of states with high $L>2$, will show only a small modulation, if any.

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TABLE I. Formula for the number of photons polarized parallel to the beam axis for the transitions $L\rightarrow L_0$ in hydrogen. $I_{\parallel}=V_i X(L,L_0)\ (a+b\cos\omega t)\exp(-2\gamma_L t)$, where $a=\sum_{M_L} a_{M_L}\sigma_{M_L}/D$, $b=\sum_{M_L} b_{M_L}\sigma_{M_L}/D$, $M_L=0,\ldots,L$, $X(L,L_0)$ is the transition probability, V_i is the initial velocity of the beam, and $2\gamma_L$ is the decay width.

L	L_0	a_0	a_1	a_2	a_3	a_4	b_0	b_1	b_2	b_3	b_4	D
1	0	5	4				4	−4				9
1	2	16	29				2	−2				45
2	1	44	69	12			6	6	−12			75
2	3	213	388	274			12	12	−24			525
3	2	417	748	490	60		24	36	0	−60		735
3	4	380	717	588	373		12	18	0	−30		882
4	3	628	1181	956	581	56	20	34	16	−14	−56	1134

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