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Stark-Perturbed L_α Decay in Flight*

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The L_α decay in flight of a 150-keV beam of single-foil-excited hydrogen atoms was tracked with a Geiger-Muller counter transversely viewing successive cylindrical segments of the beam as the counter was moved downstream. An adjustable transverse magnetic field encompassing both the foil and viewing regions produced Stark-induced L_α intensity oscillations. The experimental fluctuation frequency disagrees with a previously published measurement of Bickel but agrees well with the usual Bethe-Lamb theory of Stark quenching. No frequency doubling effect is found. New information about foil collision dynamics emerges from studying the phase of the fluctuation patterns and the decay rates of the observed radiation. Our conclusion concerning relative $s_{1/2}$- and $p_{1/2}$-state populations is also in disagreement with the earlier work.

INTRODUCTION

Our recent work[1] on periodic intensity fluctuations of Balmer lines from Stark-perturbed single-foil-excited fast hydrogen atoms showed that an entension of the Bethe-Lamb theory[2] of Stark quenching provided good agreement between a small number of theoretically dominant frequencies and the measured fluctuation frequencies. We were also able to account in fairly quantitative fashion for other measurements,[3] made at the University of Arizona, on various transitions in H and He^+. The sole exception was a single measurement of the fluctuation frequency of the L_α line made by Bickel[4] with which our calculated frequency was in serious disagreement. The experimental frequency he reported was nearly twice the one we calculated at an assumed field strength of 120 V/cm. Because the fine structure for $n=2$ is simple and since Lamb's classic experiments[2] has been the most intensively studied atomic fine structure (fs), it seemed particularly important to reconcile the discrepancy for this case. A frequency doubling effect was unexpected. We have repeated this L_α experiment at a field strength of 118 V/cm (provided by a motional electric field $\vec{v}\times\vec{B}$ rather than by condenser plates). An additional frequency measurement was made at 197 V/cm.

A more detailed discussion of these fs interference phenomena and of our experimental method is found in our earlier publication.[1] We repeat here a few of the essential facts regarding these phenomena. The intensity fluctuations can be attributed to Stark mixing of fs levels decaying with very different line intensities, with the result that weakly radiating levels can periodically "feed" a strongly radiating level, thus giving rise to short bursts of light. The atoms are created in times no greater than the transit time of the particle through the foil, times of order 10^{-14} sec. Distance along the flight path from the foil can be translated into time decay following creation of the atom. Whatever mechanisms cause the atom to alternately emit and not emit light as it proceeds along its path must not only affect all atoms created in the foil uniformly but must always generate the same relative phase. Otherwise one atom would emit light at places where another atom would be quiescent and all evidence of light intensity fluctuations would be obliterated. The wave function describing a suitable superposition of Stark-mixed fs levels must at time zero (at the foil) have definite average values for the population probabilities of the various constituent states. Within experimental error, the first maximum in the fluctuation pattern occurs at the foil, indicating a high initial population of the strongly radiating fs states relative to others which are subsequently "fed" by them and also indicating random relative phases between unperturbed eigenfunctions which are superposed to describe the initial state (as will be discussed). Why this should be true seems to involve model-dependent assumptions about the collision process. Such physical constraints certainly would not be contained in the general set of Stark-coupled wave equations which would be constructed for an otherwise isolated atom in an arbitrary excited state, since any set of initial relative amplitudes and phases would be mathematically acceptable. Specific dynamical boundary conditions would have to be applied to establish the initial populations of the states in the mixture. Much information can be obtained by studying the frequencies alone. When application of boundary conditions becomes necessary, the assumed conditions turn out to be reasonable and not unduly restrictive.

Further information concerning the amplitudes of the various states making up the wave function can in principle be deduced from the exponential attenuation of the envelope of the oscillations. This is particularly true for the L_α decay, since only the perturbed s- and p-state mean lives are involved. Unfortunately, cascade effects from higher levels largely spoil the precision with which these perturbed lifetimes can be taken into account, particularly for the long-lived s state, which is repopulated at a somewhat indeterminate rate from above. Since the fluctuation periods are fairly short compared to the characteristic mean lives of these decay processes, their frequencies can be determined with adequate accuracy.

EXPERIMENTAL ARRANGEMENT

As shown in Fig. 1, protons from a 0–200-keV accelerator were collimated into a 3.2-mm-diam horizontal beam and passed through a 10-$\mu g/cm^2$ carbon foil, which was located on the axis of a scanning chamber clamped between the pole pieces of a 4-in. electromagnet. The symmetrically located pole pieces provided a vertical magnetic field whose associated motional electric field $\vec{v}\times\vec{B} \sim 100$ V/cm was parallel to the direction of view. A L_α-sensitive Geiger-Muller counter filled with a helium-iodine mixture viewed successive cylindrical segments of the beam as it tracked the beam longitudinally at intervals of about 0.6 mm. A stationary counter viewing a segment of beam near the pole-piece axis provided a normalization for the moving counter. Operating characteristics of these particular counters have been discussed in more detail by Sellin.[5] The basic design is that of Brackmann *et al.*[6] The field of view of the moving counter was defined by a pair of parallel knife-edged slits 0.8 mm wide and 6 mm in length. Allowance for the angular acceptance of the counter gave an effective beam-segment length of 1.6 mm. The finite slit width reduces the apparent amplitude modulation of the detected light and causes an apparent shift by about one-half slit width of the maximum at the foil, since the upstream half of this first peak is missing. Radiative background from collisional excitation of the residual gas (pressure $\sim 10^{-6}$ Torr) in the observation region accounted for less than 10% of the total counting rate in the worst case. Allowance for a mean energy loss of about 7 keV in the 10-$\mu g/cm^2$ foil was made in calculating the frequencies from the distance between intensity maxima and the motional field $\vec{v}\times\vec{B}$. The magnetic field probe was calibrated against the known field of a magnetic-resonance spectrometer.[7]

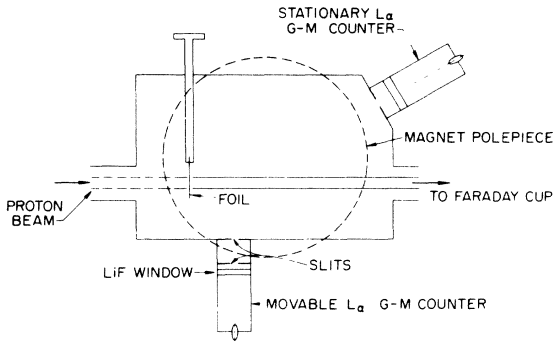

FIG. 1. Schematic diagram of the experimental apparatus.

DISCUSSION AND RESULTS

We assume that the wave function for a hydrogen atom emerging with principal quantum number n from a foil located within a weak electric field F can be described in terms of a coherent mixture of the fs levels. Sudden excitation conditions are appropriate since the particle transit time through the foil is $\lesssim 10^{-14}$ sec, whereas the inverse fs splittings are $\gtrsim 10^{-9}$ sec. The Stark matrix elements couple the levels pairwise, according to the rules $\Delta L = 1$ and $\Delta m_j = 0$. It is well known that in the simplest case of a weakly damped two-level system, oscillations of the amplitudes (a_1, a_2) of the two coupled states occur. This leads to terms in the occupation probability of each state which oscillate at a frequency corresponding to the perturbed energy separation of the two levels. In the three-level case there are terms in the occupation probability of each state which oscillate at each of the three possible level-separation^2 frequencies. At our field strengths, the Stark matrix elements are somewhat smaller than the Lamb shift, so that the $s_{1/2} - p_{1/2}$ coupling is fairly weak and the $s_{1/2} - p_{3/2}$ coupling is negligible. To an accuracy sufficient for the present experiment, then, the $p_{3/2}$ level is regarded as unperturbed and decays in exponential fashion with the usual p-state lifetime of 1.6 nsec. The $p_{1/2}$ lifetime is slightly lengthened and the $s_{1/2}$ lifetime appreciably shortened in a field-dependent manner. The oscillatory component of the $p_{1/2}$- and $s_{1/2}$-state occupation probabilities modulates the downward transition probability for L_α from these two levels. The frequency is just the perturbed energy separation of these two levels. This separation is calculated using the usual time-independent perturbation theory of the weak-field Stark effect. We thus neglect quadratic or higher terms in F and the effects of radiative widths on the level energies, as in the treatment of Bethe and Salpeter.[8] Once the level terms are established, the harmonic content of the Lyman radiation from specific super-

positions of the levels can be studied. As in Lamb's treatment,[2] we consider each state amplitude a_i to be expressible as a sum over appropriate constants times the corresponding time dependences

$$a_i = \sum_k A_k \exp[(-\mu_k + i\omega_i)t] , \quad (1)$$

where $$\psi = \sum_i a_i u_i \exp(-i\omega_i t). \quad (2)$$

In this expression, a_i is the amplitude of one of the unperturbed atomic eigenstates such as $p_{1/2}{}^{1/2}$ or $s_{1/2}{}^{1/2}$ (the superscript denotes m_j); A_k is an arbitrary constant which depends implicitly upon the relative importance of the kth perturbed eigenstate in constructing a_i as well as explicitly upon the initial conditions; ω_i is the unperturbed energy of level i in frequency units; μ_k is the complex number corresponding to the energy of the kth perturbed eignestate; and m_j is a good quantum number. The imaginary part of μ_k is just 2π times the perturbed-term value of the kth perturbed state. The real part of μ_k is half the corresponding radiative width of mode k. Its value is close to half the unperturbed decay probability of the corresponding pure state at small electric fields and approaches an average characteristic of all three levels at sufficiently large fields.

The wave function may be calculated directly from Eq. (2). The sum over i goes over all j and m_j. When the electric-dipole matrix element to the ground state is squared, cross terms of the type

$$A_k^* A_j \exp[(-\mu_k^* - \mu_j)t] + \text{c. c.} \quad (3)$$

occur, one pair for each possible level-separation frequency $i\omega_{jk}' = \mu_k^* + \mu_j$.

The dependence on initial conditions is contained in the constants A_k. Many choices could, of course, be made. A choice that matches our experimental observations well is as follows: The occupation probability $|a_i|^2$ at $t = 0$ is assumed to have a certain mean value when averaged over all collisions, but the phase between a_i and a_j is assumed to be random from collision to collision.

The possible phase relations between the $s_{1/2}$- and $p_{1/2}$-state amplitudes deserves some discussion. First of all, if the amplitudes of the $p_{1/2}$ and $s_{1/2}$ states corresponded to the mixture of states required to form a pure perturbed eigenstate, no intensity oscillations would occur. That fluctuations do occur shows that the average eigenstate of an atom emerging from the foil is appreciably different from that of either pure perturbed eigenstate.

Inspection of the cross terms in Eq. (3) corresponding to $s_{1/2}$-$p_{1/2}$ coupling shows that the damped oscillatory terms will contain additive terms proportional to $\mathrm{Re}(A_k^* A_j)\cos\omega' t$ and to $\mathrm{Im}(A_k^* A_j)\sin\omega' t$, respectively ($t = 0$ corresponds to the foil location and ω' is the perturbed $s_{1/2}$-$p_{1/2}$ splitting). Our experiments show that, within about 0.5 mm, the first oscillatory maximum is located at the foil, corresponding to a $\cos\omega' t$ dependence. If all radiative widths are small compared to ω', then there are three conditions under which $\mathrm{Im}(A_k^* A_j)$ goes to zero.[9] If one fixes the initial $s_{1/2}$-state amplitude as $[a\exp(i\phi)]$ and the initial $p_{1/2}$-state amplitude as $[b\exp(i\theta)]$, then the $\sin\omega' t$ terms are proportional to $b(1-b^2)^{1/2} \times \sin(\theta - \phi)$. If $b = 0$ or 1, corresponding to pure s-state or pure p-state formation, then only $\cos\omega' t$ terms would occur. If, however, one averages over a random $(\theta - \phi)$ from collision to collision, then once again only $\cos\omega' t$ terms survive.[9] Of these possibilities, the random-phase condition seems least arbitrary and most reasonable.

Each of the direct and cross terms in the absolute square of Eq. (1) for the $p_{1/2}$ state is exponentially damped. The damping of the direct terms will go as the perturbed decay rate of the $p_{1/2}$ and $s_{1/2}$ states, respectively. The cross terms are damped at a rate equal to the mean of these two perturbed rates. The total light intensity, which is approximately proportional to the instantaneous summed population probabilities of the $p_{1/2}$ and $p_{3/2}$ states, should thus be resolvable into a sum of three exponentials plus an exponentially damped sinusoidal term. The three exponentials should be damped with the decay rate of the essentially unperturbed $p_{3/2}$ state, the slightly perturbed decay rate of the $p_{1/2}$ state, and the significantly perturbed decay rate of the $s_{1/2}$ state.

Figure 2 shows our experimental data. A least-squares adjusted sum of two exponentials, one of which is chosen to match the calculated p-state decay rate and the other the calculated s-state rate,[10] provides a good smooth fit to the data. What remains resembles a damped sinusoidal curve, which is plotted for the two cases in Fig. 3. In principle, one could work backwards and extract the decay rates and s- and p-state population probabilities from Fig. 2, but the precision is very poor, expecially in the determination of relative populations. Cascades severely alter the magnitude and slope of the fit to the weaker s-state decay, which in turn induces error in the fit to the p-state decay slopes and magnitudes.

Some information concerning relative populations of s and p states is inherent in the phase at $t = 0$. The fact that the $\cos\omega' t$ contribution is positive rather than negative at $t = 0$ implies that the initial-state mixture is rich in $p_{1/2}$ compared to $s_{1/2}$ initially. If, for example, one had a pure $s_{1/2}$ state at $t = 0$, initially no radiation would emerge and would thereafter oscillate in time as

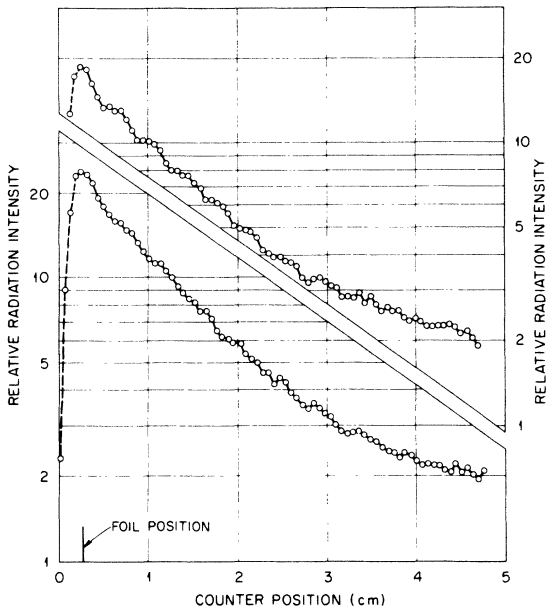

FIG. 2. Plots of the beam intensity versus detector position for motional electric fields of 118 (lower curve) and 197 V/cm.

$(1-\cos\omega' t)$. A $(|c_1|+|c_2|\cos\omega' t)$ dependence would be found for an initially pure $p_{1/2}$ state. For $a=b$, the positive and negative cosines cancel, leaving no net oscillation. From the positive sign of the $\cos\omega' t$ term, then, one concludes that the state mixture is rich in $p_{1/2}$ compared to $s_{1/2}$.

This dominance of $p_{1/2}$ over $s_{1/2}$ population is inconsistent with the results of Bickel,[4] which indicated that the hydrogen $2s$-state population was greater than the sum of all $2p$-state populations under similar collision conditions. Our results are free of cascade effects, and we suggest that further work be done to determine if these effects are responsible for the discrepancy.

The tendency of the base line for the damped sinusoid in Fig. 3 to wander merely reflects an imperfect fit to the purely exponential components of Fig. 2. The experimental sinusoidal frequency can be read directly from Fig. 3 by dividing the velocity by the mean distance between successive intensity maxima.

Our results are 1.34 ± 0.07 GHz and 1.22 ± 0.06 GHz at 197 and 118 V/cm, respectively. They are to be compared with Bickel's result[4] of 2.20 ± 0.20 GHz at 120 V/cm. Our corresponding theoretical values are 1.37 and 1.18 GHz, respectively.

We conclude that the match between our experiments and theory is entirely adequate, and that we find no frequency doubling effect as suggested by the far different result of Bickel.[4,11]

Note added in proof. Bickel has subsequently informed us that the frequency discrepancy very likely resulted from a scale reading error, and that new measurements on this and other L lines are now in progress. Use of an electrostatic rather than a motional field will not further elucidate the phase question, since the precise field value at the moment of birth is important. Assuming random initial phases of unperturbed states yields a successful but not necessarily unique collision model.

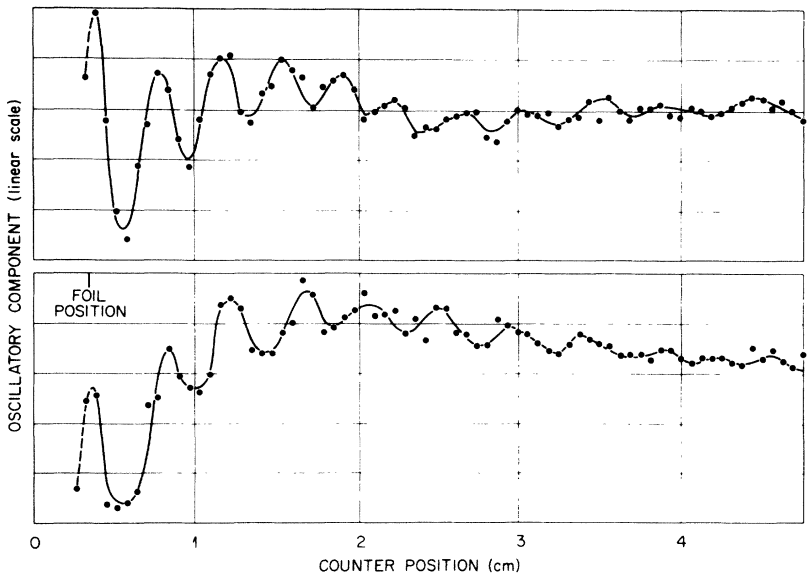

FIG. 3. Damped oscillatory components of the decay of L_α as a function of distance downstream from the foil, at fields of 118 (lower curve) and 197 V/cm.

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[1]Sellin *et al.*, Phys. Rev. (to be published).

[2]A particularly pertinent discussion which bears on the present work is given by W. E. Lamb, Jr., Phys. Rev. 85, 259 (1952).

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[4]W. S. Bickel, J. Opt. Soc. Am. 58, 219 (1968).

[5]I. A. Sellin, Phys. Rev. 136, A1246 (1964).

[6]R. T. Brackmann, W. L. Fite, and K. E. Hagen, Rev. Sci. Instr. 29, 125 (1958).

[7]P. D. Miller of this laboratory kindly provided this calibration.

[8]H. A. Bethe and E. E. Salpeter, Quantum Mechanics of One- and Two-Electron Atoms (Academic Press Inc., New York, 1957), p. 290. See also Ref. 9.

[9]Inclusion of the p-state radiative widths gives a small $\sin\omega' t$ term which, when added to the $\cos\omega' t$ term, produces a small phase shift. The shift is less than 5 deg for the present case and is beyond our measurement accuracy. It should be stressed that for cases when $\gamma_i \sim \omega_i$ this phase shift may be substantial, and that the energy splitting calculated with neglect of γ_i (as in time-independent perturbation theory) will be seriously in error.

[10]The $p_{1/2}$-state decay rate is sufficiently close to the $p_{3/2}$ rate that their difference can be neglected in the fitting procedure.

[11]It should be noted that Bickel used an electrostatic field rather than a motional $\vec{v}\times\vec{B}$ field. No consequential difference should occur except that a foil may be placed within a magnetic field without perturbing the resulting motional electric field.

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Accurate Low-Energy Atom-Atom Scattering

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Measurements of low-temperature thermodynamic properties of gases yield information concerning the pairwise scattering parameters of the atoms of the gas. For recent helium-gas experiments, the He-He scattering potential has been determined with a high degree of accuracy. This necessitates a reanalysis of low-energy He-He scattering theory with inclusion of previously neglected effects of order m/M. The formulation requires a nonlinear transformation of coordinates to ensure the correct form of the incident wave, and a novel definition of the adiabatic Hamiltonian to ensure the dissociation of the "molecular" states into correct atomic states. In addition to the usual nonadiabatic terms, we find additional terms of the same order not usually encountered. They are necessary to make the potentials vanish at infinity. It is shown that the procedure of determining a potential interaction from the second virial coefficient (scattering data) and then predicting bound-state properties (if any) is a correct one.

I. INTRODUCTION

Measurement of thermodynamic functions as a function of temperature can yield extremely accurate information concerning the low-energy scattering parameters of the atoms of the gas. For example, the second virial coefficient can be related to the bound states and phase shift of the pairwise scattering of the atoms.[1] This analysis is based on a treatment of the atoms as structureless particles interacting through a local-energy-independent potential. Various potentials have been used to calculate the second virial coefficient in helium with the result that only potentials lying in a very narrow range will reproduce the experimental data. Typically, the uncertainty in the potential (near the minimum) is quoted at about 10^{-3}–10^{-4} eV[2]. This accuracy is well beyond the usual situation encountered in atomic scattering phenomena so that a reanalysis of the atom-atom scattering event is necessary to take into account small effects previously neglected.

We shall pursue the scattering aspect of the problem here. It is clear that at the energies in question (10–300°K or 0.001–0.025 eV), the adiabatic [Born-Oppenheimer(BO)] representation of