Stark-Induced Temporal Intensity Variations in Spectral Lines*

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The effect of externally produced Stark splitting upon fine-structure-level probabilities is investigated in order to describe periodic intensity variations previously found in hydrogen lines. Coupled differential equations for the probability amplitudes of mixed levels are solved exactly in terms of initial amplitudes; effects due to spontaneous transition probabilities and Lamb shift are included. In general, the time-dependent line intensity is a combination of exponential decays and sinusoidal oscillations. The frequencies of oscillations are discussed in detail, and the effects of different initial conditions upon the intensities are considered for several special cases.

R ECENT observations[1] of the spectra emitted by a beam of hydrogen atoms which had been excited by passing through a thin foil showed a time-periodic variation in line intensity when the beam particles were in the presence of an electric or magnetic field. This effect was ascribed to a quantum-mechanical resonance which resulted in time-dependent populations of emitting states of quite different transition probabilities; the resonance arose from a Stark mixing of the states produced either by the actual electric field or by a motional electric field $(\mathbf{v}/c)\times\mathbf{B}$. The frequencies of the intensity variation were calculated from the usual resonance factor $\sin^2(\Delta E/2\hbar)t$, where ΔE is the energy difference in the states which would be produced by the field; reasonable agreement was found between the calculated and measured frequencies.[1] In this paper, we shall give a more detailed treatment of this phenomenon and obtain more exact expressions for the frequency factors and decay lifetimes which are involved.

For simplicity, we shall consider only the case of two levels whose energies in the absence of the field are E_1 and $E_2 = E_1 + \hbar\delta$. In the case of hydrogen levels related by $l = j \pm \frac{1}{2}$, the energy difference $\hbar\delta$ is simply the Lamb shift. Both of these levels can decay to a lower level with spontaneous transition probabilities per unit time A_1 and A_2; we assume that $\hbar\delta$ is so small that the separate spectral lines cannot be resolved. The observed intensity I of the line will then be proportional to the sum of products of the A's and the average probabilities of occupations of the levels; thus, if c_1 and c_2 are the probability amplitudes of the two levels, we can write

$$I = A_1\langle|c_1|^2\rangle_{\mathrm{av}} + A_2\langle|c_2|^2\rangle_{\mathrm{av}} \qquad (1)$$

with the omission of a constant multiplicative factor.

We assume that in the presence of a small external electric field F, the perturbation potential $V = eFz$ ($e > 0$) has a nonvanishing matrix element $V_{12} = V_{21} = \hbar v$ connecting these two levels. For example, in the case of the hydrogen atom and for the levels related by $l = j \pm \frac{1}{2}$ (such as $s_{1/2}$ and $p_{1/2}$), this matrix element con-

nects states of the same m and is[2,3]

$$\hbar v = -3eFan\{m^2[n^2-(j+\tfrac{1}{2})^2]\}^{1/2}/4j(j+1), \qquad (2)$$

where a is the Bohr radius. If the states are like $(l=j-\tfrac{1}{2})_j$ and $(l+1)_{j+1}$ (such as $s_{1/2}$ and $p_{3/2}$ or $p_{3/2}$ and $d_{5/2}$), the matrix element can be calculated from the Pauli state functions as given by Bethe and Salpeter[3] with the result that, in this case, $\hbar v$ is obtained by multiplying the result given in (2) by the additional factor $j[(j+1)^2 - m^2]^{1/2}$.

In the presence of the field, the original states are no longer eigenstates of the complete atomic Hamiltonian and the coupled differential equations of motion for the probability amplitudes become

$$\dot{c}_1 = -ivc_2e^{-i\delta t} + \dot{c}_{1\text{ spon}} = -ivc_2e^{-i\delta t} - \tfrac{1}{2}A_1c_1, \qquad (3)$$

$$\dot{c}_2 = -ivc_1e^{i\delta t} + \dot{c}_{2\text{ spon}} = -ivc_1e^{i\delta t} - \tfrac{1}{2}A_2c_2, \qquad (4)$$

where we have accounted for the existence of the spontaneous transition probabilities by adding the terms $-\tfrac{1}{2}A_ic_i$.[4]

The exact solutions of (3) and (4) in terms of the initial values c_{10} and c_{20} at $t=0$ are found to be

$$c_1(t) = \omega^{-1}\exp[-\tfrac{1}{4}(A_1+A_2)t - \tfrac{1}{2}i\delta t]$$
$$\times[c_{10}(\omega\cos\omega t - \alpha\sin\omega t) - ivc_{20}\sin\omega t], \qquad (5)$$

$$c_2(t) = \omega^{-1}\exp[-\tfrac{1}{4}(A_1+A_2)t + \tfrac{1}{2}i\delta t]$$
$$\times[-ivc_{10}\sin\omega t + c_{20}(\omega\cos\omega t + \alpha\sin\omega t)], \qquad (6)$$

where

$$\omega = (v^2 - \alpha^2)^{1/2}, \qquad (7)$$

$$\alpha = \tfrac{1}{4}(A_1 - A_2) - \tfrac{1}{2}i\delta. \qquad (8)$$

The results (5) and (6) are more complicated than they appear at first sight because ω is complex when $\delta \neq 0$. If we write $\omega = \omega_r + i\omega_i$, then we find that (5) and

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[1] S. Bashkin, W. S. Bickel, D. Fink, and R. K. Wangsness, Phys. Rev. Letters **15**, 284 (1965).

[2] R. Schlapp, Proc. Roy. Soc. (London) **A119**, 313 (1928); V. Rojansky, Phys. Rev. **33**, 1 (1929).
[3] H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Springer-Verlag, Berlin, 1957), p. 239.
[4] E. Wigner and V. Weisskopf, Z. Physik **63**, 54 (1930); Ref. 3 p. 288.

(6) become

$$c_1(t) = \omega^{-1} \exp[-\tfrac{1}{4}(A_1+A_2)t - \tfrac{1}{2}i\delta t]$$
$$\times \{c_{10}[(\omega \cosh\omega_i t - i\alpha \sinh\omega_i t)\cos\omega_r t$$
$$- (\alpha \cosh\omega_i t + i\omega \sinh\omega_i t)\sin\omega_r t]$$
$$+ vc_{20}(\sinh\omega_i t \cos\omega_r t - i \cosh\omega_i t \sin\omega_r t)\}, \quad (9)$$

$$c_2(t) = \omega^{-1} \exp[-\tfrac{1}{4}(A_1+A_2) + \tfrac{1}{2}i\delta t]$$
$$\times \{vc_{10}(\sinh\omega_i t \cos\omega_r t - i \cosh\omega_i t \sin\omega_r t)$$
$$+ c_{20}[(\omega \cosh\omega_i t + i\alpha \sinh\omega_i t)\cos\omega_r t$$
$$+ (\alpha \cosh\omega_i t - i\omega \sinh\omega_i t) \sin\omega_r t]\}, \quad (10)$$

where

$$\omega_r = 2^{-1/2}\{(\omega_0{}^2 + \tfrac{1}{4}\delta^2) + [(\omega_0{}^2 + \tfrac{1}{4}\delta^2)^2$$
$$+ \tfrac{1}{4}\delta^2(A_1-A_2)^2]^{1/2}\}^{1/2}, \quad (11)$$

$$\omega_i = \delta(A_1-A_2)/8\omega_r, \quad (12)$$

$$\omega_0 = v\{1 - [(A_1-A_2)^2/16v^2]\}^{1/2}. \quad (13)$$

What remains now is to use the results (9) and (10) in order to calculate the intensity from (1). Although this will result in a complicated expression, it is evident that the intensity will in general be some superposition of exponentially decaying terms which will be modulated by factors containing oscillating terms whose circular frequency will be the ω_r of (11). In evaluating the beam averages required in (1), we should take into account the fact that the atoms are excited by independent collisions in the foil, so that the initial probability amplitudes of different states will have incoherent phases and we will be able to set cross terms like $\langle c_{10}{}^* c_{20}\rangle_{av}$ equal to zero. In addition, one must sum over the possible values of m in order to obtain the total level populations needed for (1).

In order to illustrate the effect of the mixing produced by the field, let us consider the extreme case in which one of the initial c's is zero; because of the symmetry in (9) and (10), any choice will do, so let us take $c_{20}=0$ to be specific. We then find from (10) that

$$|c_2(t)|^2 = |c_{10}|^2 [v^2/(\omega_r{}^2 + \omega_i{}^2)](\sinh^2\omega_i t + \sin^2\omega_r t)$$
$$\times \exp[-\tfrac{1}{2}(A_1+A_2)t], \quad (14)$$

which vanishes in the absence of the field so that $v=0$. We note that the oscillating term has an exponential decay factor equal to the average of those associated with the two states, while the $\sinh^2\omega_i t$ term gives rise to three terms with different exponential decay factors of $\tfrac{1}{2}(A_1+A_2)$ and $\tfrac{1}{2}(A_1+A_2)\pm2\omega_i$, although the last two terms will not appear if $A_1=A_2$ so that $\omega_i=0$.

We also see from (11) and (13) that if $\delta=0$ and $A_1=A_2=A$, then the frequency is simply $v=\Delta E/2\hbar$ where ΔE would be the total energy difference produced in the two levels by the Stark effect, the formula which was used previously.[1] In this exceptional case, however, we find from (1), (5)–(7), that $I = Ae^{-At}[\langle|c_{10}|^2\rangle_{av}$

$+\langle|c_{20}|^2\rangle_{av}]$, which is simply an exponential decay with no trace of oscillation. If $\delta=0$, but $A_1\neq A_2$, then the frequency becomes ω_0 as given by (13) and shows the correction necessitated by the difference in spontaneous transition probabilities; for the cases considered in Ref. 1, this correction amounts to only about 1 part in 500. On the other hand, if $\delta\neq0$, but $A_1=A_2$, then ω_r becomes just $(v^2+\tfrac{1}{4}\delta^2)^{1/2}$; this is, however, an unlikely case. In these latter cases, there are oscillatory terms present.

If one is dealing with sufficiently large values of the principal quantum number n, it may not be too unreasonable an approximation to simply get $\delta=0$. If this is the case, then ω in (5)–(7) becomes simply ω_0. We see from (13), however, that if the applied field is sufficiently small, ω_0 will be a pure imaginary and there will be no oscillations at all but only exponential decays. The condition for oscillation is seen to be $|v|>\tfrac{1}{4}|A_1-A_2|$. For the value $n=6$ in hydrogen, the critical value of the applied field turns out to be about 0.08 V/cm; all of the fields used in Ref. 1 were greater than this value. We can see from (5)–(7) that for fields so small that there are no oscillations, the probabilities will decay with three different lifetime factors $\tfrac{1}{2}(A_1+A_2)$, $\tfrac{1}{2}(A_1+A_2)\pm2(\alpha^2-v^2)^{1/2}$ and with corresponding amplitudes which depend upon the initial relative populations. In the special case in which one initial probability is zero, we can easily find from (14) that these terms with different relaxation factors will have relative amplitudes of -2, 1, 1, respectively.

In summary, then, if δ can be neglected and the external field is large enough, then the frequency will be half the Stark shift corrected for different spontaneous transition probabilities, whereas, if $\delta\neq0$, then measurement of the frequencies of oscillation could provide a means of measuring the Lamb shift. Furthermore, it may well be possible to determine the relative populations of the levels as produced in the foil by measuring the relative amplitudes of the oscillating terms as given by (5) and (6).

Note added in proof. The desirability of indicating the existence of other formally analogous phenomena has been pointed out to us. For example, it has been found that the neutral K meson decays with two lifetimes, and Feinberg[6] has shown that as a result of the difference between the K^0 and its antiparticle there can be possible transitions between the long- and short-lived K^0's in the presence of matter.

[5] G. W. Series, Phys. Rev. **136**, A684 (1964).
[6] G. Feinberg, Phys. Rev. **109**, 1381 (1958).