Coherence Effects in Resonance Fluorescence

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The crossed-level method of atomic spectroscopy is discussed and the angular distribution formula for both the incoherent and coherent resonance scattering is derived. The form of this distribution function, as given here, explicitly displays the geometric factors depending on radiation propagation vectors. With the application to hydrogen in mind, the distribution function is expressed explicitly for single electron transitions with external fields possessing axial symmetry. The properties of the distribution function are discussed with emphasis on the case of unpolarized radiation. For the case of hydrogen there are two possible applications of major interest. The first concerns the possibility of a precision measurement of the $2p$ fine structure splitting and, hence, a determination of the fine structure constant. Explicit results for the shape and other properties of the resonance line with a uniform magnetic field obtained. The other application is concerned with the possibility of measuring the $2s$-$2p$ Lamb splitting. This requires an electric field parallel to the magnetic field. Unfortunately, the level crossings which are sensitive to the Lamb splitting cannot radiate sufficiently rapidly while those which do radiate appreciably occur at field strengths which are extremely insensitive to the Lamb splitting.

I. INTRODUCTION

IN a recent paper Colegrove, Franken, Lewis, and Sands[1] have described a novel measurement of the fine structure ($2\,^3P_1-2\,^3P_2$) splitting in atomic helium. The method involves the production of level-crossing of excited atomic states by an external magnetic field. It depends on the fact that the scattering of resonance radiation in the case of degenerate or nearly degenerate excited states is coherent in somewhat the same sense as in the diffraction of light by a pair of slits, the initial and final (sub-) states playing the role of source and detector with which the diffraction pattern is observed. A more direct description of the phenomenon in physical terms will be given in the discussion to follow. An alternative description has also been given by Franken.[2]

It should be emphasized that the process under discussion is not new in that it had been discussed in the literature almost three decades ago. In this connection the work of Weisskopf[3] and of Breit[4] is especially worthy of mention. What is important is the fact that the work of Franken *et al.* constitutes the first application of this method to precision spectroscopy. The measurement of the spin-orbit splitting in helium gives results accurate to better than one part in 2×10^4 and is presumably capable of even greater accuracy. The application to hydrogen is immediately suggested, since the more accurately known wave functions for that case would presumably lead to a highly precise determination of the fine structure constant via a measurement of the $2p_{\frac{3}{2}}-2p_{\frac{1}{2}}$ splitting in a uniform magnetic field. The accuracy of the measurement would then be determined largely by the accuracy with which the magnetic field at the center of the resonance could be determined. In principle the method could be extended to other light atoms, but the application to hydrogen would appear most urgent. Another possible measurement would appear to yield an alternative determination of the Lamb splitting. In this case, as is almost immediately apparent, it is necessary to apply an electric field as well. The question of measuring the Lamb shift in hydrogen by this "crossed-level" method will be taken up in this paper.

Other applications are possible in principle at least. For instance, the investigation of very small splittings in nuclear levels produced by magnetic dipole and electric quadrupole splittings would be amenable to this method. The experiment would now depend on coherent resonance effects in a cascade of two gamma rays, say. Unfortunately, however, in most cases the nuclear spin Hamiltonian contains too many unknown parameters to make the envisaged experiment attractive. For instance, in the simple case that a nucleus (spin $\geqslant 1$) is embedded in a crystal which provides an inhomogeneous electric field with an axis of symmetry and if there is no hyperfine coupling, the ratio of quadrupole to dipole coupling could be determined by placing the sample in an external magnetic field. We shall not pursue these applications in this paper.

Although the process under discussion has been described to some extent in the references cited, we wish to present a few remarks which will clarify the subsequent discussion. The situation considered is one in which an atom (say) undergoes stimulated absorption from an initial state a to a group of excited states

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[1] F. D. Colegrove, P. A. Franken, R. R. Lewis, and R.H. Sands, Phys. Rev. Letters **3**, 420 (1959).

[2] P. A. Franken, Phys. Rev. **121**, 508 (1961).

[3] V. Weisskopf, Ann. Physik **9**, 23 (1931).

[4] G. Breit, Revs. Modern Phys. **5**, 91 (1933).

b, b', $\cdots$. Subsequently, by spontaneous emission the system undergoes a transition to a state c which need not be the same as the initial state. If the states a and c are degenerate or even if this degeneracy is removed, by a magnetic field for example, the labelling of the states includes a description of the sublevels and then a and c will, in general, be different. In our application, however, the initial and final states will be sublevels of the same gross state. The states b, b', $\cdots$ will, in the absence of external fields, be not completely degenerate and we consider a pair of states, b and b', which in the field-free case are split by an amount Δ_0. Moreover, Δ_0 will be assumed to be considerably larger than the average width, $\bar{\gamma}=\frac{1}{2}(\gamma+\gamma')$, of this pair of states. Under such circumstances the scattered intensity is an incoherent superposition of contributions from b and b'. If the $b-b'$ splitting can be "tuned" by application of external fields so that it can be made zero (crossed levels) or of order γ, then an interference contribution to the scattered intensity arises. This will be true for some fixed direction of the outgoing radiation and will be angle dependent. For the total intensity there is, of course, no coherent effect; see Sec. II below. As an incidental point, it is clear that the precise description of the coherent contribution depends also on the polarization states of incident and outgoing radiation.

This discussion shows that as a function of the external field a sharp resonance will appear.[5] The width at half maximum is about $2\bar{\gamma}$.[6] The fact that the width is of the order cited will be evident from the following consideration. If the excited state exists for a time t the nearly degenerate states b and b' undergo random phase changes given by $\exp(-iEt)$ and $\exp(-iE't)$. The net random phase introduced is $\exp(-i\Delta Et)$, where $\Delta E=E-E'$. Since $t\sim 1/\bar{\gamma}$ it follows that coherence is preserved perfectly for $\Delta E/\bar{\gamma}=0$ (level-crossing) and is completely destroyed when $\Delta E/\bar{\gamma}\gg 1$.

In this discussion there are two assumptions: (1) First, the incident radiation is assumed to have a sufficiently broad spectrum so that b and b' can both be excited. Second, the states involved must fulfill the standard selection rules which in the atomic case implies that a and c can be connected to both b and b' by electric dipole radiation. This requirement of the selection rules renders some level crossings in helium ineffective, and in hydrogen it requires that s states be admixed with some p constituent.

Finally, we make the rather obvious remark that the utility of the method, if the objective is to measure Δ_0, depends on an accurate connection between Δ_0 and the values of the field parameters at which the level-crossing occurs. This is most readily established in the hydrogen case.

[5] This is to be distinguished from the resonance as a function of frequency, which has nothing to do with the coherent phenomenon considered here.

[6] This statement is modified when the hyperfine splitting is of the same order as the width γ; see Sec. III.

II. THE ANGULAR DISTRIBUTION FUNCTION

The transition probability for resonance scattering of almost degenerate states has been derived by other authors[3,4] but in this section we give a simplified version of the derivation for the purpose of putting in evidence some physical consideration. In our formalism we follow the procedure used by Abragam and Pound[7] in their discussion of angular correlation. This procedure is not meant to be more rigorous than the perturbation methods adopted in the previous discussions but it has certain pedagogic advantages.

In the spirit of the perturbation theory the states involved in our description are eigenfunctions of a static Hamiltonian, H, which includes the external fields, and are also damped by the coupling to the radiation field. The transition from the initial state a (which actually constitutes a group of substates which we label by m_1) to the intermediate state b occurs at time $t=0$. We consider the subsequent emission process from b to a final state c, substates labelled by m_2, occurring at time t.

The transition probability for this "time-delayed" scattering is proportional to[8]

$$W(t)=\sum_{m_1m_2}|\sum_b\langle m_2|H_r^{(+)}|b_t\rangle\langle b_0|H_r^{(-)}|m_1\rangle|^2. \quad (1)$$

Here $H_r^{(+)}$ is the operator that creates the outgoing photon with specified frequency, direction, and polarization, while $H_r^{(-)}$ is the operator for annihilating the incident photon with specified frequency, direction, and polarization. Clearly the time dependence of the state amplitude b_t is given by

$$|b_t\rangle=|b_0\rangle\exp(-iE_b-\tfrac{1}{2}\Gamma_b)t. \quad (2)$$

In (2) we have used the diagonality of the damping matrix.[3,4] We return to this point in the discussion at the end of this section. Also E_b represents the eigenvalue of H for *one* of the states b, the label b representing a composite of quantum numbers referring to the (in general) nondegenerate states b. We have allowed for the possibility that the radiative widths Γ_b depend on the state b. Also in our units $\hbar=1$.

Using (2), the transition probability will be given by

$$W=\int_0^\infty W(t)dt$$
$$\sum_{m_1m_2}\sum_{bb'}\langle m_2|H_r^{(+)}|b_0\rangle\langle m_2|H_r^{(+)}|b_0'\rangle^*$$
$$\times\langle b_0|H_r^{(-)}|m_1\rangle\langle b_0'|H_r^{(-)}|m_1\rangle^*\frac{1}{\Gamma_{b'b}-iE_{b'b}}, \quad (3)$$

[7] A. Abragam and R. V. Pound, Phys. Rev. **89**, 1306 (1953).

[8] Here and in the following the formalism follows that of angular correlation theory, which is the precise counterpart of the process we discuss. The replacement in the initial transition of an emission process by an absorption does not change this equivalence. See, for example, L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1953).

where we have used the notation

$$\Gamma_{b'b}=\tfrac{1}{2}(\Gamma_b+\Gamma_{b'}), \tag{3a}$$

$$E_{b'b}=E_{b'}-E_b. \tag{3b}$$

Equation (3) agrees exactly with the results given by Weisskopf and Breit. All the states entering in the matrix elements are now true stationary states.

Since the "delay time" t is random, it is clear that the finite lifetime of the intermediate states introduces a random phase in the state amplitudes: $E_{b'b}t$ is the relative phase difference due to the precession caused by H. The interpretation of the resonance as given in the introduction is now clear. If with $b\neq b'$, $E_{b'b}/\Gamma_{b'b}\gg 1$ the only important contributions are those for which $b=b'$ and $E_{b'b}=0$. These are the incoherent terms in (3). When the external field parameters are so adjusted that $E_{b'b}\sim\Gamma_{b'b}$ for some pair b, b' there will be a coherent contribution arising from this pair of states. This will certainly occur at and near a level-crossing. In general, for given external fields only one crossing will occur.

To make the rather formal result (3) more useful we recognize that the $H_r^{(\pm)}$ operators will usually correspond to emission and absorption of pure multipole fields and, in our case, of electric dipole fields. Hence, it is useful to introduce the field-free atomic eigenstates for a single electron. These are pure angular momentum states. These are conveniently described by the quantum numbers κ and μ (aside from the energy quantum number). Here μ is the eigenvalue of j_z, the angular momentum along the quantization axis, and κ gives the total angular momentum j and parity $(-)^l$ according to[9]

$$j=|\kappa|-\tfrac{1}{2},$$
$$l=j+\tfrac{1}{2}\kappa/|\kappa|.$$

The eigenfunctions $|\kappa\mu\rangle$ diagonalize H when the external fields are absent. Then, dropping the subscript 0,

$$|b\rangle=\sum_{\kappa\mu} U^{\mu}(b\kappa)\,|\kappa\mu\rangle \tag{4}$$

and the $U_{b\kappa}{}^{\mu}$ are elements of a unitary matrix. When the external fields have axial symmetry so that j_z and H commute, we write (4) in the form

$$|b\rangle=\sum_{\kappa} U^{\mu}(K\kappa)\,|\kappa\mu\rangle, \tag{4'}$$

where we recognize that b contains μ parametrically as one constituent label and K serves to distinguish the different states with the same μ. Our considerations will be almost entirely confined to the case of axial symmetry and we use (4') in the following.[10] The quantum numbers m_1 and m_2 can now be interpreted as the eigenvalues of j_z for the states a and c. The unitary transformation (4) is obviously unnecessary for a and c.

We introduce the transformation (4') and the notation

$$\mathfrak{M}^{(-)}=\sum_{m_1}\langle\bar{\kappa}\mu|\,H_r^{(-)}\,|\kappa_1 m_1\rangle\langle\bar{\kappa}'\mu'|\,H_r^{(-)}\,|\kappa_1 m_1\rangle^*, \tag{5a}$$

$$\mathfrak{M}^{(+)}=\sum_{m_2}\langle\kappa_2 m_2|\,H_r^{(+)}\,|\kappa\mu\rangle\langle\kappa_2 m_2|\,H_r^{(+)}\,|\kappa'\mu'\rangle^*. \tag{5b}$$

Here we use κ_1 and κ_2 to complete the description of the states a and c. Then the transition probability becomes

$$W=\sum U^{\mu}(K\kappa)U^{\mu*}(K\bar{\kappa})U^{\mu'*}(K'\kappa')U^{\mu'}(K'\bar{\kappa}')\times\frac{\mathfrak{M}^{(+)}\mathfrak{M}^{(-)}}{\Gamma_{b'b}-iE_{b'b}} \tag{6}$$

and the sum in (6) is over K, K', μ, μ', κ, κ', $\bar{\kappa}$, $\bar{\kappa}'$. Of course, b is an abbreviation for the index pair K, μ. The κ and $\bar{\kappa}$ take on the same set of values and κ' and $\bar{\kappa}'$ also take on the same values. The two pairs $(\kappa,\bar{\kappa})$ and $(\kappa',\bar{\kappa}')$ do not necessarily have the same range. It is to be recognized that $\mathfrak{M}^{(\pm)}$ depend on the polarization and direction of the quanta and on μ and μ'. Also $\mathfrak{M}^{(-)}$ depends on $\bar{\kappa}$ and $\bar{\kappa}'$, $\mathfrak{M}^{(+)}$ on κ and κ'. But K and K' do not enter in $\mathfrak{M}^{(\pm)}$. They do occur in the unitary matrix elements. These matrix elements also depend on the external field parameters as do $\Gamma_{b'b}$ and $E_{b'b}$. Because W depends on the direction of the photons it will be henceforth referred to as the angular distribution function. For the case in which the external fields exhibit axial symmetry, the angles which enter are those describing the spherical triangle defined by the symmetry axis and the two propagation vectors $\hat{k}_1$ and $\hat{k}_2$.

To exhibit the properties of the angular distribution function it is necessary to examine the detailed structure of the $\mathfrak{M}^{(\pm)}$. For dipole radiation the absorption operator, omitting irrelevant multiplicative constants, is

$$H_r^{(-)}=\sum_M D_{MP}^{1}(\hat{k})\mathbf{j}\cdot\mathbf{A}_1{}^{M}, \tag{7}$$

where D_{MP}^{1} is an element of the rotation matrix of order three, and the two of the three Euler angles appearing as arguments are the polar and azimuth angles of $\hat{k}$ and the third is zero. $\mathbf{j}$ is the current density operator and $\mathbf{A}_1{}^{M}$ is the vector potential for a dipole (angular momentum 1 and z component M). In (7), $P=\pm 1$ and, in modern usage, $P=+1$ refers to right, $P=-1$ to left circular polarization.[11]

[9] M. E. Rose, *Elementary Theory of Angular Momentum* (John Wiley & Sons, Inc., New York, 1957), Chap. IX.

[10] As will be made more evident in Sec. III it is only in the case of axial symmetry that level crossings (accidental degeneracies) occur.

[11] For linear polarization, (7) is to be replaced by

$$H_r^{(-)}=\Sigma_P e^{-iP\alpha}D_{MP}^{1}(\hat{k})\mathbf{j}\cdot\mathbf{A}_1{}^{M},$$

where α defines the direction of the electric vector with respect to a plane containing the quantization axis.

The result for $\mathfrak{M}^{(-)}$ is then

$$\mathfrak{M}^{(-)}=(-)^n\langle\bar{\kappa}\|1\|\kappa_1\rangle\langle\bar{\kappa}'\|1\|\kappa_1\rangle^* \times[(2\bar{j}'+1)(2\bar{j}+1)]^{\frac{1}{2}}\sum_\nu C(11\nu;P,-P) \times C(\bar{j}\bar{j}'\nu;-\mu\mu')W(1\bar{j}1\bar{j}';j_1\nu)D_{\mu-\mu',0}{}^{\nu}(\hat{k}), \quad (8)$$

where $n=\bar{j}'-\bar{j}+\mu'+j_1$. In (8) the first two factors after the phase are reduced matrix elements for dipole emission, the C factors are Clebsch-Gordan coefficients, and W is a Racah coefficient. We also note that

$$D_{m0}{}^{\nu}(\hat{k})=\left(\frac{4\pi}{2\nu+1}\right)Y_\nu{}^{m*}(\hat{k}) \quad (8')$$

give the angular dependence.

If we sum over polarizations, we employ

$$\tfrac{1}{2}\sum_P C(11\nu;P,-P)=\tfrac{1}{2}[1+(-)^\nu]C(11\nu;1,-1),$$

so that ν is restricted to even values and, explicitly, $\nu=0$ and 2. The $\nu=1$ terms contribute to the polarization-dependent terms. The major part of our special considerations will be restricted to the resonance fluorescence of the Lyman alpha radiation, and for experimental reasons it is sufficient to consider only unpolarized radiation. Recognizing that the extension to polarized radiation can easily be made, we shall henceforth discuss only the case in which the unpolarized radiations are observed.

For $\mathfrak{M}^{(+)}$ we change $\bar{\kappa}$, $\bar{\kappa}'$ to κ, κ', also j_1 to j_2, and finally take the complex conjugate of (8). Obviously, $\hat{k}=\hat{k}_1$ in $\mathfrak{M}^{(-)}$ and $\hat{k}=\hat{k}_2$ in $\mathfrak{M}^{(+)}$.

With these results we can return to the question of the diagonality of the damping matrix which was tacitly assumed in writing (2). For this purpose it is necessary to show that

$$D_{bb'}\equiv\sum_\omega\langle b|H_r^{(-)}|a\rangle\langle b'|H_r^{(-)}|a\rangle^*=0 \quad (9)$$

if the states b and b' are different, where the sum is over all photon frequencies, polarizations, and directions. We need only to integrate (8) over all directions of $\hat{k}$ to see that $D_{bb'}$ is proportional to $\delta_{\mu\mu'}\delta_{\nu0}$. Since

$$D_{bb'}=\sum_\omega\sum_{\bar{\kappa}\bar{\kappa}'}U^{\mu*}(K\bar{\kappa})U^{\mu}(K'\bar{\kappa}')\mathfrak{M}^{(-)},$$

it is seen that $\nu=0$ requires $\bar{\kappa}=\bar{\kappa}'$. For those values of $\bar{\kappa}$ and $\bar{\kappa}'$ for which the product of reduced matrix elements is not zero, this product is independent of $\bar{\kappa}$ (or $\bar{\kappa}'$). Thus the sum over $\bar{\kappa}$ involves only the U matrix elements and gives $\delta_{KK'}$. $D_{bb'}$ is proportional to $\delta_{KK}\delta_{\mu\mu'}$ and is completely diagonal.[12]

A second consequence of the result (8) is that the intermediate states must be linear combinations of states, at least one of which has an angular momentum $j\geqslant\frac{3}{2}$. Otherwise there is no coherent contribution to the scattering. To see this, we first recognize that in the coherent term $\mu-\mu'\neq0$. This follows from a well-known theorem of Wigner and von Neumann[13] which prohibits crossings of levels with the same symmetry. Hence in the product $\mathfrak{M}^{(+)}\mathfrak{M}^{(-)}$ there is no coherent contribution from the isotropic terms $\nu=0$. Incidentally, this confirms the more or less self-evident fact that the coherence does not affect the total intensity of scattering. Since we consider only unpolarized scattered light, $\nu=2$ must occur in both $\mathfrak{M}^{(+)}$ and $\mathfrak{M}^{(-)}$. Hence $\bar{j}$, $\bar{j}'$, and ν as well as j, j', and ν with $\nu=2$ must form a triangle, which is impossible if all angular momenta are equal to $\frac{1}{2}$. This means that for the $n=2$ states in H the role of the $p_{\frac{3}{2}}$ level is vital, so far as producing coherence with unpolarized light is concerned.

The structure of the coherent contribution is now seen to be of the form

$$W_{\text{coh}}=\sum_{b\neq b'}\frac{AY_2{}^{\mu-\mu'}(\hat{k}_1)Y_2{}^{\mu-\mu'*}(\hat{k}_2)}{\Gamma_{b'b}-iE_{b'b}},$$

where A contains the four unitary matrix elements and the remaining factors in $\mathfrak{M}^{(+)}\mathfrak{M}^{(-)}$. Since

$$Y_2{}^m(\hat{k})=(-)^m\left(\frac{5}{4\pi}\right)^{\frac{1}{2}}\left[\frac{(2-m)!}{(2+m)!}\right]^{\frac{1}{2}}P_2{}^m(\vartheta)e^{im\varphi},$$

where $P_2{}^m$ is the associated Legendre function, we can write

$$W_{\text{coh}}\sim|A|P_2{}^{\mu-\mu'}(\vartheta_1)P_2{}^{\mu-\mu'}(\vartheta_2)\frac{\Gamma_{b'b}\cos\beta-E_{b'b}\sin\beta}{\Gamma_{b'b}{}^2+E_{b'b}{}^2}, \quad (10)$$

where

$$\beta=(\mu-\mu')(\varphi_1-\varphi_2)+\arg A.$$

Of course, ϑ_1 and ϑ_2 are the polar angles of $\hat{k}_1$ and $\hat{k}_2$ measured from the axis of symmetry and $\varphi_1-\varphi_2$ is the dihedral angle between the planes formed by this axis with $\hat{k}_1$ and with $\hat{k}_2$. This verifies the statement that for unpolarized radiation only the rotationally invariant spherical triangle is involved. If the energy difference $E_{b'b}$ is varied by changing the field parameters, symbolized for the moment by x, the width of the resonance in x space is

$$\Delta x=2\frac{\partial x}{\partial E_{b'b}}\Gamma_{b'b}, \quad (11)$$

while in energy ($E_{b'b}$) space it is $2\Gamma_{b'b}$. The resonance

[12] An alternative proof is given in reference 4.

[13] J. von Neumann and E. P. Wigner, Physik. Z. **30**, 467 (1929).

line is seen to consist of a Lorentz-shape contribution proportional to

$$\Gamma_{b'b}\cos\beta/(\Gamma_{b'b}^2+E_{b'b}^2)$$

and an asymmetrically shaped contribution proportional to

$$-E_{b'b}\sin\beta/(\Gamma_{b'b}^2+E_{b'b}^2).$$

In the foregoing discussion we have tacitly assumed that over the width of the resonance, A is an extremely slowly varying function of the field parameters. In all our applications this is extremely well fulfilled; see Sec. III.

For the case of external fields for which there is an axis of symmetry, A will be real. A necessary condition for this is the reality of the relevant products of reduced matrix elements. That these products are indeed real can be seen *a priori* by an adaptation of a well-known argument of Lloyd.[14] In this case

$$\beta=(\mu-\mu')(\varphi_1-\varphi_2)=(\mu-\mu')\Phi,$$

and the pure Lorentz shape can be obtained by a suitable choice of the dihedral angle.

An additional selection rule which must be fulfilled for the existence of a coherent term is

$$0<|\mu-\mu'|\leqslant 2,$$

as can be seen from (8). For the $n=2$ levels of hydrogen this is fulfilled for all levels which cross, as will be seen in the following section. For one of the radiations circularly polarized, the restriction is more severe: $|\mu-\mu'|=1$.

Finally, we observe that if either the incident or outgoing radiation is parallel (or antiparallel) to the axis of symmetry, there is no coherent effect. This type of geometry forces the condition $\mu=\mu'$ which precludes the crossing of two levels.

The incoherent part of the radiation is clearly anisotropic if and only if at least one of the intermediate states has an admixture of $j\geqslant\frac{3}{2}$. In any event the incoherent part is independent of the dihedral angle.

As far as optimization of the interference signal is concerned, it is clear that one should make $\vartheta_1=\vartheta_2=\vartheta$, and for $|\mu-\mu'|=2$ the value $\vartheta=\pi/2$ is optimum while for $|\mu-\mu'|=1$ the value $\vartheta=\pi/4$ is optimum. For $|\mu-\mu'|=2$ the pure Lorentz shape results for $|\varphi_1-\varphi_2|=n\pi/2$ with $n=0, 1, 2, 3$; and for $|\mu-\mu'|=1$ the condition is $|\varphi_1-\varphi_2|=0$ or π. In the applications given below, these geometrical conditions will be assumed.

III. APPLICATION TO HYDROGEN

We shall consider the following two applications of the resonance scattering at crossed levels in hydrogen: (1) the scattering in a magnetic field only whereby the $2p_{\frac{3}{2}}-2p_{\frac{1}{2}}$ splitting is ascertainable, and (2) the scattering in combined electric and magnetic fields whereby (*a priori*) one might hope to determine the Lamb shift. Our discussion is intended to be illustrative of the principles of the method and of some of the difficulties one may expect to encounter.

A. Spin-Orbit Splitting

From an experimental point of view resonance scattering is simplest between the ground and the $n=2$ states. However, for reasons having to do with the absorption of Lyman radiation, it is not practical to consider polarized radiation. The results of Sec. II will then apply.

The stationary part of the Hamiltonian of the problem is

$$H=H_0+H', \tag{12}$$

where H_0 contains not only the kinetic and Coulomb energy but also, in a phenomenological way, all necessary radiative corrections. The magnetic coupling is H' which will be expressed in nonrelativistic form,

$$H'=\mu_0\mathfrak{H}(j_z+s_z), \tag{13}$$

where μ_0 is the Bohr magneton and $\mathfrak{H}$, the magnetic field, is in the direction of the quantization axis. The eigenvalues of H_0 for the $n=2$ states are ϵ_1, ϵ_0, and ϵ_2 for $2p_{\frac{1}{2}}$, $2s_{\frac{1}{2}}$, and $2p_{\frac{3}{2}}$, respectively. We introduce

$$\delta=\epsilon_2-\epsilon_1, \tag{13'}$$

the spin-orbit splitting plus radiative corrections. We shall also use

$$\Delta=\epsilon_0-\epsilon_1,$$

the Lamb splitting. In the present case the $2s_{\frac{1}{2}}$ level plays no role since it radiates with a half-life (1/7 sec) which is far too large. The coupled states are $p_{\frac{1}{2}}$ and $p_{\frac{3}{2}}$ with the same eigenvalue μ of j_z. The energies in the field are $E(K\mu)$:

$$E(1\,\tfrac{1}{2})=\tfrac{1}{2}(G-S_+), \tag{14a}$$

$$E(2\,\tfrac{1}{2})=\tfrac{1}{2}(G+S_+), \tag{14b}$$

$$E(1\,-\tfrac{1}{2})=-\tfrac{1}{2}(G+S_-), \tag{14c}$$

$$E(2\,-\tfrac{1}{2})=\tfrac{1}{2}(-G+S_-), \tag{14d}$$

where

$$G=\mu_0\mathfrak{H}/\delta, \tag{15}$$

and

$$S_\pm=(G^2\pm\tfrac{2}{3}G+1)^{\frac{1}{2}}. \tag{16}$$

For zero field the $K=1$, 2 states become $p_{\frac{1}{2}}$, $p_{\frac{3}{2}}$ states, respectively. In addition, for $j=\frac{3}{2}$, $\mu=\pm\frac{3}{2}$ the energy is

$$E_{\frac{3}{2}}(\pm\tfrac{3}{2})=\pm 2G+\tfrac{1}{2},$$

and these states remain pure.

[14] S. P. Lloyd, Phys. Rev. **81**, 161 (1951).

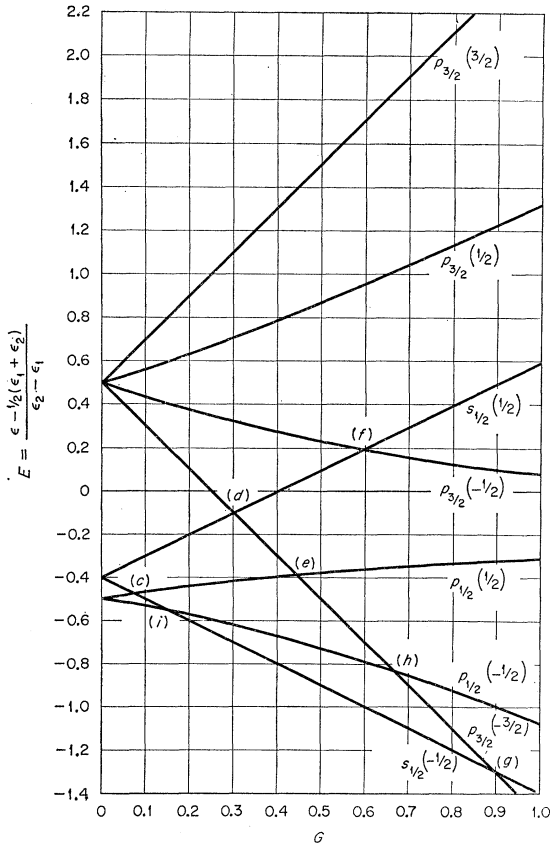

FIG. 1. Energy levels for the $n=2$ state in hydrogen as a function of $G=\mu_0\mathfrak{IC}/\delta$. The symbols are defined in Eqs. (13) and (13′). The s levels are shown with the Lamb shift taken to be 1057 Mc/sec. The letters in parentheses serve to distinguish the various crossings.

For future reference we note that

$$E_{2s}(\pm\tfrac{1}{2})=\pm G-\frac{1}{2}\left(\frac{\sigma-1}{\sigma+1}\right), \tag{17}$$

where

$$\sigma=\frac{\epsilon_2-\epsilon_1}{\epsilon_0-\epsilon_1}=\frac{\delta}{\Delta}-1=9.350. \tag{17′}$$

The expansion coefficients $U^{\mu}(K\kappa)$ are:

$$U^{\pm\frac{1}{2}}(1\ 1)=[\tfrac{1}{2}(1+\rho_{\pm}/S_{\pm})]^{\frac{1}{2}}, \tag{18a}$$

$$U^{\pm\frac{1}{2}}(2\ 1)=[\tfrac{1}{2}(1-\rho_{\pm}/S_{\pm})]^{\frac{1}{2}}, \tag{18b}$$

$$U^{\pm\frac{1}{2}}(1\ -2)=-[\tfrac{1}{2}(1-\rho_{\pm}/S_{\pm})]^{\frac{1}{2}}, \tag{18c}$$

$$U^{\pm\frac{1}{2}}(2\ -2)=[\tfrac{1}{2}(1+\rho_{\pm}/S_{\pm})]^{\frac{1}{2}}, \tag{18d}$$

and

$$\rho_{\pm}=1\pm G/3.$$

Of course, $\kappa=1,\ -2$ refer to $p_{\frac{1}{2}}$ and $p_{\frac{3}{2}}$, respectively.

The energy levels are shown in Fig. 1 where the Lamb-shifted s sublevels are also shown. There are only two crossing between p states and these are marked (e) and (h) in Fig. 1. In the present approximation these occur at

$$G=4/9 \quad (e \text{ crossing}),$$
$$G=2/3 \quad (h \text{ crossing}).$$

Clearly in a precise analysis many small corrections would be considered as well. For instance, relativistic corrections (of order α^2) would increase G by one part in 10^5. Other corrections, arising from finite nuclear mass, hyperfine coupling, and so on, are readily applied as discussed by Lamb.[15] To a good approximation the effect of hyperfine coupling is to broaden the coherent resonance. Thus, each level in Fig. 1 becomes a doublet with m_I, the nuclear magnetic quantum number, equal to $\pm\frac{1}{2}$ so that a given crossing is replaced by four crossings. However, as a consequence of the selection rule $\Delta m_I=0$ only two of these crossings will contribute to the scattering. Hence, a single Lorentz peak is replaced by two peaks which are shifted by an amount equal to hyperfine splitting. Since this splitting is 1.776 Mc/sec for the $n=2$ state and the width Γ (the same for all p states) is 99 Mc/sec, the resonance will appear as two separated peaks.

Crossing (e) corresponds to $|\mu-\mu'|=2$ and for optimum geometry the incoherent scattering is

$$W_{\text{inc}}=(9/8)+\textstyle\sum_{K\mu}\{1+\frac{1}{4}[U^{\mu}(K-2)]^2 -\sqrt{2}\mu U^{\mu}(K1)U^{\mu}(K-2)\}^2. \tag{19}$$

With the same normalization the coherent scattering is

$$W_{\text{coh}}{}^{(e)}=\frac{49}{88}\left(1-\frac{540}{847}g\right)\frac{1}{1+[(24/11)x]^2}, \tag{20}$$

where g is a measure of the magnetic field measured from the center of the resonance; that is,

$$g=G-4/9, \tag{20a}$$

and

$$x=g\delta/\Gamma. \tag{20b}$$

The linear factor in g appearing in (20) arises from the distortion of the wave functions by the magnetic field and, as a consequence, the shape of the resonance is not exactly of the Lorentz type. Since $x=1$ corresponds to $g=0.009$, this deviation from the Lorentz shape is small but may be significant enough to take into account in applications where precision is a consideration. At half maximum, the deviation from the pure Lorentz shape due to this distortion is $\Delta x\cong-6.7\times10^{-4}$.

[15] W. E. Lamb, Jr., Phys. Rev. **85**, 259 (1952).

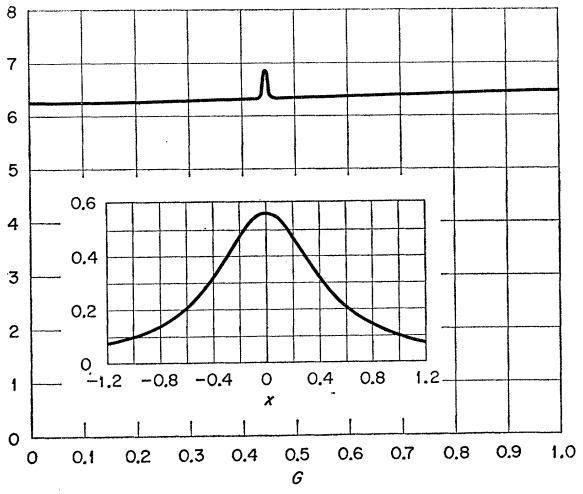

FIG. 2. Coherent scattering for zero electric field as a function of magnetic field. The ordinate scale is arbitrary. The geometry is $\vartheta_1=\vartheta_2=\pi/2$, $\Phi=0$. The resonance arises from crossing (*e*) in Fig. 1. The lower portion of the figure shows an enlarged version of the resonance curve. Splitting due to hyperfine coupling has not been included here. The small deviation from the Lorentz shape is not noticeable.

The ratio of W_{coh} to W_{inc} at the resonance is

$$W_{\text{coh}}/W_{\text{inc}}=0.0895,$$

which should correspond to a coherent signal of ample strength.

The crossing (*h*) corresponds to $|\mu-\mu'|=1$ and the conditions of optimum geometry ($\vartheta_1=\vartheta_2=\pi/4$) are not as convenient as in the $|\mu-\mu'|=2$ case. Nevertheless, we give the results for the incoherent and coherent scattering for this crossing:

$$W_{\text{inc}}=\frac{81}{32}+\sum_{K\mu}\left\{1-\tfrac{1}{8}[U^{\mu}(K-2)]^2+\frac{1}{\sqrt{2}}\mu U^{\mu}(K1)U^{\mu}(K-2)\right\}^2 \quad (21)$$

and

$$W_{\text{coh}}{}^{(h)}=\frac{1}{24}\left(1-\frac{10}{3}g\right)\frac{1}{1+(4x/3)^2}. \quad (22a)$$

Here

$$g=G-\tfrac{2}{3}. \quad (22b)$$

The ratio $W_{\text{coh}}/W_{\text{inc}}$ at $x=0$ is only 0.0068 and, hence, this crossing is not quite as interesting as crossing (*e*).

Figure 2 shows the coherent peak for crossing (*e*) superimposed on the incoherent background. In this figure the coherent resonance is also shown on an enlarged scale. The effect of hyperfine coupling, not shown here, is readily taken into account by superimposing two resonance curves of the type shown. The total width at half maximum, which is 2Γ in energy units, is, in x space, equal to $\Delta x=11/12$ which corresponds to a magnetic field width of 65.2 gauss. The position of the resonance is, in the approximation considered, $\mathfrak{H}_{\text{res}}=3480$ gauss. Since the relation between $\mathfrak{H}_{\text{res}}$ and δ is linear, the fine structure (or better, $2p_{\frac{3}{2}}-2p_{\frac{1}{2}}$) splitting can be determined to the same accuracy as the measurement of the magnetic field at resonance.

B. Lamb Shift

It is immediately clear that the position of the crossings (*e*) and (*h*) of Fig. 1 do not depend on Δ, the $2s_{\frac{1}{2}}-2p_{\frac{1}{2}}$ splitting. On the other hand, the position of the other crossings shown do depend somewhat sensitively on Δ but these crossings cannot be utilized in resonance scattering from the $1s_{\frac{1}{2}}$ ground state. Hence, it is necessary to introduce an electric field $\mathfrak{E}$. For reasons to be made apparent at a later stage of the discussion it is advantageous to make the two fields $\mathbf{H}$ and $\mathfrak{E}$ parallel (or antiparallel). In this case the system considered still exhibits axial symmetry and μ is a good quantum number.

For $\mathfrak{E}\times\mathbf{H}=0$ the eigenvalues are determined from the roots of the secular equation

$$\det(H_{ij}'-E\delta_{ij})=0,$$

where

$$H'=\mu_0\mathfrak{H}(j_z+s_z)+e\mathcal{E}z.$$

The secular determinant splits into two cubics ($\mu=\pm\frac{1}{2}$) and two one-by-one determinants ($\mu=\pm\frac{3}{2}$). With the notation

$$x(\mathfrak{H})=\mu_0\mathfrak{H}, \quad x(\mathcal{E})=-\sqrt{3}ea_0\mathcal{E},$$

where a_0 is the Bohr radius in hydrogen, we have for $\mu=\frac{1}{2}$

$$\det H_{ij}'=\begin{vmatrix} x(\mathfrak{H})+\epsilon_0 & -x(\mathcal{E}) & \sqrt{2}x(\mathcal{E}) \\ -x(\mathcal{E}) & \frac{1}{3}x(\mathfrak{H})+\epsilon_1 & \frac{1}{3}\sqrt{2}x(\mathfrak{H}) \\ \sqrt{2}x(\mathcal{E}) & \frac{1}{3}\sqrt{2}x(\mathfrak{H}) & \frac{2}{3}x(\mathfrak{H})+\epsilon_2 \end{vmatrix}. \quad (23)$$

In (23) the rows and columns refer to $s_{\frac{1}{2}}$, $p_{\frac{1}{2}}$, and $p_{\frac{3}{2}}$ states, respectively. For $\mu=-\frac{1}{2}$ the sign of $x(\mathfrak{H})$ on the diagonal and the sign of $x(\mathcal{E})$ in the $s_{\frac{1}{2}}-p_{\frac{1}{2}}$ matrix elements must be changed. The characteristic equation for $\mu=-\frac{1}{2}$ differs from that pertaining to $\mu=\frac{1}{2}$ by a change of sign of $x(\mathfrak{H})$. For $\mu=\pm\frac{3}{2}$ the energies are

$$E(\pm\tfrac{3}{2})=\pm 2x(\mathfrak{H})+\epsilon_2. \quad (24)$$

Of course, the sum of the eigenvalues for both $\mu=\pm\frac{1}{2}$ is unchanged by the presence of the fields and the center of gravity of the levels remains at $\frac{1}{3}(\epsilon_0+\epsilon_1+\epsilon_2)$.

At any nonzero value of the electric field there are

TABLE I. Information relative to eight level crossings of the $n=2$ states of hydrogen in parallel electric and magnetic fields.

Intersecting states	Energy at intersection	Field strengths at intersection	
(a) $p_{\frac{1}{2}}(\frac{3}{2}):p_{\frac{1}{2}}(-\frac{1}{2})$	$2G+(2-L)/3$	$V^2=3G(G+4/9)(1+3G-L)/[(13/3)G+1]$,	$G\geqslant 0$
(b) $p_{\frac{1}{2}}(\frac{3}{2}):p_{\frac{1}{2}}(\frac{1}{2})$	$2G+(2-L)/3$	$V^2=G(G+\frac{2}{3})(1+G-L)/[(5/3)G+1]$,	$G\geqslant 0$
(c) $s_{\frac{1}{2}}(-\frac{1}{2}):p_{\frac{1}{2}}(\frac{1}{2})$	$-1/12(\lambda+1)$	$G^2=(1/a)(\beta-\omega\lambda)$	
(d) $p_{\frac{1}{2}}(-\frac{3}{2}):s_{\frac{1}{2}}(\frac{1}{2})$	$-2G+(2-L)/3$	$V^2=9G(4/9-G)[(1-L)/3-G]/[(13/3)G-1]$,	$3/13\leqslant G\leqslant(1-L)/3$
(e) $p_{\frac{1}{2}}(-\frac{3}{2}):p_{\frac{1}{2}}(\frac{1}{2})$	$-2G+(2-L)/3$	$V^2=3G(G-4/9)(3G+L-1)/[(13/3)G-1]$,	$G\geqslant 4/9$
(f) $s_{\frac{1}{2}}(\frac{1}{2}):p_{\frac{1}{2}}(-\frac{1}{2})$	$1/12(\lambda-1)$	$G^2=(1/a)(\beta+\omega\lambda)$	
(g) $p_{\frac{1}{2}}(-\frac{3}{2}):s_{\frac{1}{2}}(-\frac{1}{2})$	$-2G+(2-L)/3$	$V^2=G(\frac{2}{3}-G)(1-L-G)/[(5/3)G-1]$,	$\frac{3}{5}\leqslant G\leqslant\frac{2}{3}$
(h) $p_{\frac{1}{2}}(-\frac{3}{2}):p_{\frac{1}{2}}(-\frac{1}{2})$	$-2G+(2-L)/3$	$V^2=G(G-\frac{2}{3})(G+L-1)/[(5/3)G-1]$,	$G\geqslant 1-L$

eight distinct level crossings, not counting the degeneracies at $\mathcal{K}=0$. In general, these occur at different values of $\mathcal{E}$, $\mathcal{K}$ so that the degree of accidental degeneracy does not exceed two. Also, as a general rule, these crossings are sufficiently well separated so that where one crossing occurs, the level separation of other pairs appreciably exceeds $\bar{\Gamma}$. In Table I information relative to the various crossings is given. The table describes the crossings in terms of the spectroscopic designation of the zero-field states from which the states in question arise (first column). The second column gives the energy at which the accidental degeneracy occurs with respect to the center of gravity $\frac{1}{3}(\epsilon_0+\epsilon_1+\epsilon_2)$. The third column gives the equation which determines the relation which must be fulfilled between the field strengths for each crossing. The entries in the table are listed according to increasing $\mathcal{K}$ for given $\mathcal{E}$. The notation employed is

$$V=x(\mathcal{E})/\delta, \quad L=\Delta/\delta, \tag{25}$$

and G defined in Eq. (15). In addition the following abbreviations are used.

$$a=16(12V^2+1), \tag{25a}$$

$$\beta=320V^4+(8/3)V^2(9L^2-24L+26)+(2/9)(45L^2-36L+16), \tag{25b}$$

$$\lambda^2=a+9L^2, \tag{25c}$$

$$\omega=(8/9)+2L(4V^2-1). \tag{25d}$$

In Fig. 3 the energy levels for the eight $n=2$ states are shown as a function of magnetic field with $V=L$. Using the value 1057 Mc/sec for the Lamb shift, this corresponds to an electric field of 477.6 volts/cm. For other values of the electric field similar energy level patterns are obtained.

It will be noted that even for $\mathcal{K}=0$ the separation of the $s_{\frac{1}{2}}$ and $p_{\frac{1}{2}}$ states is slightly more than twice the Lamb shift. Intersection (*c*) between $s_{\frac{1}{2}}$ with $\mu=-\frac{1}{2}$ and $p_{\frac{1}{2}}$ with $\mu'=\frac{1}{2}$ is clearly the most sensitive to the value of L, or the Lamb shift.

Before discussing the intersection (*c*), it is of interest to note that the intersection labelled (*i*) in Fig. 1 referring to zero electric field has disappeared. This intersection between $s_{\frac{1}{2}}$ and $p_{\frac{1}{2}}$, both with $\mu=-\frac{1}{2}$, is not a contradiction to the Wigner von Neumann theorem[13] since it corresponds to a "touching" of the levels. In a 2-by-2 secular determinant,

$$\begin{vmatrix} H_{11}-E & H_{12} \\ H_{21} & H_{22}-E \end{vmatrix}=0,$$

with roots

$$E_{1,2}=\tfrac{1}{2}\{H_{11}+H_{22}\pm[(H_{11}-H_{22})^2+4|H_{12}|^2]^{\frac{1}{2}}\}, \tag{26}$$

this intersection corresponds to $H_{12}=0$ and $H_{11}=H_{22}$. Identifying the levels with these two roots, $E_1\geqslant E_2$ always. Introducing the electric field makes $H_{12}\neq 0$ and hence $E_1=E_2$ is impossible. This "repulsion" of the levels is responsible for the fact that when $\boldsymbol{\mathcal{E}}$ and $\mathbf{H}$ are not aligned all the level crossings of Fig. 3 disappear. Off-axis components of $\boldsymbol{\mathcal{E}}$ introduce nondiagonal matrix elements between all s and p states with $|\mu-\mu'|=1$, as is clear from the fact that

$$\boldsymbol{\mathcal{E}}\cdot\mathbf{r}-\mathcal{E}_z z=\tfrac{1}{2}[(\mathcal{E}_x+i\mathcal{E}_y)(x-iy)+(\mathcal{E}_x-i\mathcal{E}_y)(x+iy)].$$

It is for this reason that we assume aligned fields although it is to be recognized that in an actual experimental arrangement the fields can be aligned accurately over only a limited volume of the scatterer. When the angle θ between fields is small, two levels which cross for $\theta=0$ will approach each other within a minimum separation which is, in general, of order θ, [see Eq. (26)]. The coherent scattering still exhibits a resonance but the maximum coherent scattering intensity is slightly diminished; that is, in the ratio

$$\bar{\Gamma}^2/[\bar{\Gamma}^2+(\Delta E)_{\min}{}^2],$$

as compared to the coherent intensity at a crossing. The geometry for the Lorentz shape is assumed. Hence, small misalignment of the fields is, in itself, not a catastrophe. It is only necessary that $(\Delta E)_{\min}\lesssim\bar{\Gamma}$.

When the mixed states have different radiative widths in the zero-field limit, the states with fields present will

obviously have different widths. It is easy to see that if a state is represented by

$$\Psi_b=\sum_j c_{bj}\psi_j,$$

then the width of this state is, to lowest order,[16]

$$\Gamma_b=\sum_j|c_{bj}|^2\gamma_j, \tag{27}$$

where γ_j are the zero-field widths of the states ψ_j. With this result the angular correlation function is easily obtained for the present case from the results of Sec. II. In the present application there are three terms in the sum (27) and we can take $\gamma_{2s}=0$ with $\gamma_{2p\frac{1}{2}}=\gamma_{2p\frac{3}{2}}$.

If we now consider intersection (*c*) for electric fields which admix appreciable *p* state in the originally pure *s* state, it is seen that the position of this crossing rapidly becomes insensitive to the Lamb shift. Physically, this is an expression of the fact that the Stark effect simply dominates the radiative effects leading to the Lamb splitting at all fields for which $V\gtrsim L$. Since this unfortunate circumstance also influences the crossings which are useful for the determination of the fine structure splitting, it is always advantageous to have no electric field present.

In quantitative terms the lack of sensitivity of *all* the crossings to the Lamb shift can be seen by considering

$$\frac{d\Delta}{\Delta}=\tau_G\frac{dG}{G}+\tau_V\frac{dV}{V}, \tag{28}$$

where

$$\tau_G=\partial\ln L/\partial\ln G,\quad \tau_V=\partial\ln L/\partial\ln V. \tag{28'}$$

For a sensitive determination the relative error $d\Delta/\Delta$ should be small or at least not large compared to dG/G and dV/V. Hence τ_G and τ_V should be of order or less than unity. These quantities τ_G and τ_V can be determined from the third column of Table I. For example, for crossings (*a*), (*d*), and (*e*),

$$\tau_G=\frac{1}{3L}\frac{G}{G-4/9}\left[\frac{V^2}{G^2}-18G+7-3L\right], \tag{29a}$$

$$\tau_V=\frac{2}{L}(L+3G-1). \tag{29b}$$

[16] The simplest proof of the result quoted is to recognize that to the order considered the width is proportional to the square modulus of the matrix element of $H_r^{(+)}$ summed over all quanta. The matrix element is $\Sigma_j c_{bj}(\psi_f|H_r^{(+)}|\psi_j)$; and after squaring and summing, the cross-terms vanish by the same argument used to deduce the diagonality of the damping matrix. An alternative proof from the equations of motion of the probability amplitudes can be devised; see W. E. Lamb, Jr. and R. C. Retherford, Phys. Rev. **79**, 571 (1950). This method involves the solution of the determinantal equation,

$$\det(H_{ij}'-E\delta_{ij}+\tfrac{1}{2}\gamma_j\delta_{ij})=0.$$

Solving to first order in γ_j gives the same result.

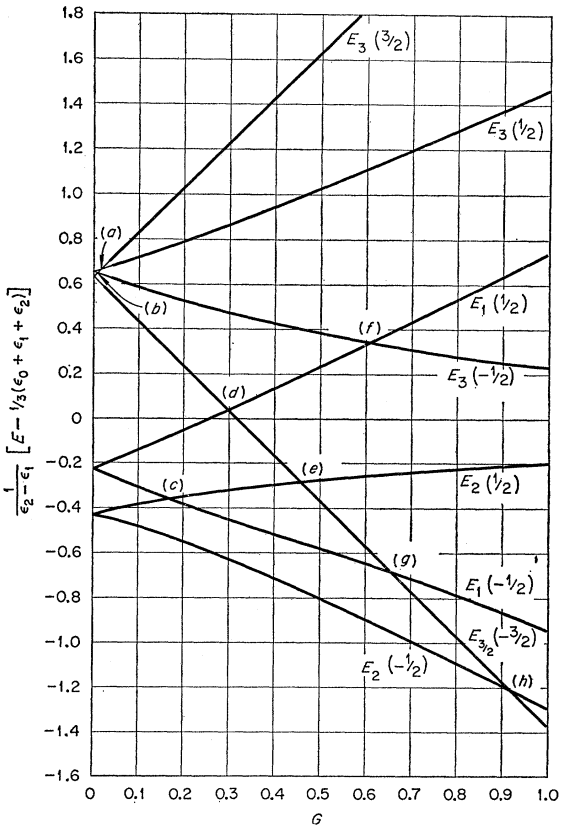

FIG. 3. Energy levels for the $n=2$ state in hydrogen as a function of G (see caption to Fig. 1) for an electric field of 477.6 v/cm [$V=L$, see Eq. (25)]. The electric field introduces new crossings (a) and (b) and removes the "crossing" (i).

For crossings (*b*), (*g*), and (*h*),

$$\tau_G=\frac{1}{L}\frac{G}{G-2/3}\left[\frac{V^2}{G^2}-2G-L+\frac{5}{3}\right], \tag{30a}$$

$$\tau_V=\frac{2}{L}(L+G-1). \tag{30b}$$

For crossings (f) (upper sign) and (c) (lower sign),

$$\tau_G=\frac{2G^2}{L}a\left\{16V^2(3L-4)+4(5L-2)\pm\left[\lambda(8V^2-2)+9L\frac{\omega}{\lambda}\right]\right\}^{-1}, \tag{31a}$$

$$\tau_V=-\frac{8V^2\tau_G}{aG^2}\left\{-24G^2+80V^2+3L^2-8L+\frac{26}{3}\pm(L\lambda+12\omega/\lambda)\right\}. \tag{31b}$$

In Figs. 4 and 5 the quantities τ_G and τ_V are given as functions of V. It is to be remembered that G is a function of V (for given L) as indicated in the third column

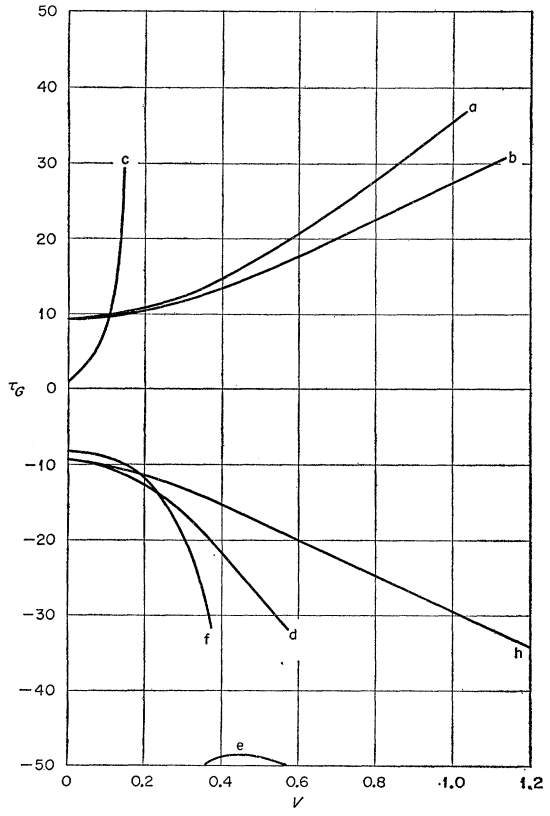

FIG. 4. The parameter τ_G, see Eq. (28′), as a function of electric field. For definition of V see Eq. (25). The letters attached to the curves refer to the various crossings in Fig. 2.

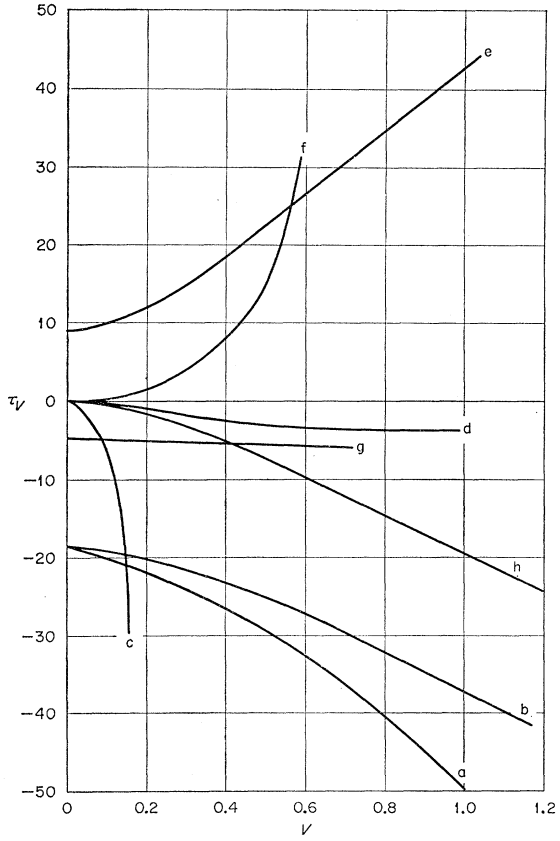

FIG. 5. Same as Fig. 4 except that the ordinate represents τ_V; see Eq. (28′).

of Table I. For τ_G only the (c) crossing can give sensitivity, but at the expense of small p admixture and very low scattering intensity from the initially pure s state. For τ_V the same remark applies to several crossings, including (c). It must be recognized that both τ_G and τ_V must be small for a crossing sensitive to the Lamb shift and this never occurs in a practical case. For small V/L the expansion coefficients $[c_{bj}$ or $U^{\mu}(K\kappa)]$ are such that the p admixture in the initially pure s state is very small (of order V^2/L^2 as is obvious from perturbation theory). The only exception is the case $\mu=-\frac{1}{2}$ where in the absence of an electric field a "touching contact" of two levels was obtained. The rapid repulsion of these two levels in an electric field precludes the possibility of observing any coherent scattering in this case as well. Moreover, the fact that these states are essentially pure $j=\frac{1}{2}$ mixtures at small fields would eliminate any possibility of coherence in that case.