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Vanishing electric dipole transition moment

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We have discovered an interference which results in the vanishing of some electric dipole transition amplitudes. A transition-matrix element T between certain S and P sublevels with the same value of M_F vanishes when an applied magnetic field satisfies $X_s + X_p = -2M_F$, where $X = \mu_B g_J B/A$, and A is the hfs constant. There must be an avoided crossing between the two states of the same M_F which can only occur when one of the hyperfine manifolds has an inverted Zeeman effect. These conditions imply $M_F < 0$ and $I > J$. We observed the vanishing of T in ^{23}Na for one of the $\Delta M_F = 0$ optical transitions of the $3S_{1/2}$ $(F = 1) \rightarrow 3P_{1/2}(F = 1)$ manifold. Absorption of cw laser light was monitored by observing total fluorescence perpendicular to both the atomic and laser beams, and absorption vanished near an applied field of 155 G. T vanishes for a large number of cases including rf and microwave frequencies and is a rather general consequence of angular momentum selection rules and perturbation theory. This phenomenon may have application in optical pumping, Lamb-shift measurements, and atomic weak neutral-current experiments.

The vanishing of certain radiative transition-matrix elements in a static applied external field is familiar to rf and microwave spectroscopists.[1] We have discovered an interference which results in the vanishing of a transition amplitude between levels which may be separated by optical frequencies. These forbidden transitions differ from others which have been reported in that they do not require energy degeneracy (level crossing). In fact, the phenomenon occurs because of the coupling between the angular momenta of the electron and the nucleus.

As an example, consider the case of electric dipole transitions between states with total electronic angular momentum $J = \frac{1}{2}$ and nuclear spin I, coupled by the hyperfine structure (hfs) interaction to form $\vec{F} = \vec{I} + \vec{J}$ $(F = I \pm \frac{1}{2})$. In an applied magnetic field, two states of a given hyperfine manifold with the same value of $M_F = M_J + M_I$ will be mixed and the relative $M_J = +\frac{1}{2}$ and $M_J = -\frac{1}{2}$ character of each is field dependent. Optical radiation polarized along the magnetic field (z-polarized light) will induce $\Delta M_F = 0$ transitions between optically separated hyperfine manifolds. Since the optical field acts only on the electron, the transition amplitude results only from couplings where M_J (and thus M_I) does not change. More specifically, there is a transition amplitude which couples the $M_J = +\frac{1}{2}$ component of the ground state and excited state and another which couples the $M_J = -\frac{1}{2}$ components. As discussed below, the two amplitudes have opposite signs, resulting in destructive interference which can be complete if there is sufficient mixing of the two M_J states.

We now calculate specifically the conditions for the vanishing transition moment. The transition-matrix element T for a resonantly oscillating field with the electric vector polarized along the quantization axis z is[2]

$$T = \langle \gamma' IJF' M_F | z | \gamma IJFM_F \rangle , \tag{1}$$

where γ contains all other quantum numbers. The wave function is expanded in the uncoupled basis as

$$|\gamma IJFM_F\rangle = \sum_{M_I + M_J = M_F} (IM_I JM_J | IJFM_F) | \gamma JM_J \rangle | IM_I \rangle . \tag{2}$$

Since $J = \frac{1}{2}$, each coupled state described in Eq. (2) has only two terms in the sum (except for the stretched states where $|M_F| = I + \frac{1}{2}$), and for reference these are expressed explicitly as

$$|\gamma, I, J, I + \tfrac{1}{2}, M_F\rangle = a|\gamma, \tfrac{1}{2}, \tfrac{1}{2}\rangle | I, M_F - \tfrac{1}{2}\rangle + b|\gamma, \tfrac{1}{2}, -\tfrac{1}{2}\rangle | I, M_F + \tfrac{1}{2}\rangle , \tag{3a}$$

$$|\gamma, I, J, I - \tfrac{1}{2}, M_F\rangle = -b|\gamma, \tfrac{1}{2}, \tfrac{1}{2}\rangle | I, M_F - \tfrac{1}{2}\rangle + a|\gamma, \tfrac{1}{2}, -\tfrac{1}{2}\rangle | I, M_F + \tfrac{1}{2}\rangle , \tag{3b}$$

$$a^2 + b^2 = 1 , \tag{3c}$$

where a and b are non-negative real numbers and the relations between them have come from orthogonality, normalization, and the choice of a phase convention.

Forming the Hamiltonian matrix for a one-electron atom ($g_S = 2$, $g_l = 1$) in a magnetic field B with $H_{\text{hfs}} = A\vec{I}\cdot\vec{J}$, we find that for the case $J=\frac{1}{2}$ only the two sublevels with the same value of M_F are mixed by the field. Diagonalizing a 2×2 matrix yields eigenvalues $E_\pm \equiv \frac{1}{2}A\epsilon_\pm$:

$$\epsilon_\pm = -\tfrac{1}{2} \pm [(X+M_F)^2 + p^2]^{1/2} \equiv -\tfrac{1}{2} \pm r\,; \tag{4}$$

and eigenfunctions

$$a^2 = \frac{p^2/2}{p^2 + (X+M_F)^2 - r(X+M_F)}\,, \tag{5}$$

where $X = \mu_B\, g_J B/A$, μ_B is the Bohr magneton, and

$$p^2 = (I+\tfrac{1}{2})^2 - M_F^2\,. \tag{6}$$

We have neglected the nuclear Zeeman energy in all these expressions. Note that the states of smaller F [lower sign in Eq. (4)] have an inverted Zeeman effect at low fields. That is, the energy of states with negative M_F increases with increasing fields, while the energy of states with positive M_F decreases.[3]

The matrix element of Eq. (1) may be calculated for the case $F=F'$ by using the decoupled basis of Eqs. (3a)–(3c). We consider an electric dipole transition between an S and a P state, and use subscripts on the coefficients a and b to refer to these states. Of the four terms in the resulting expression, two vanish since z does not operate on M_I. Finally, for states described by Eq. (3a),

$$T = (a_s a_p - b_s b_p)\langle \gamma'\tfrac{1}{2}\,\|\,\mathrm{r}\,\|\,\gamma\tfrac{1}{2}\rangle/\sqrt{6}\,, \tag{7}$$

where the reduced matrix element contains various selection rules and the radial integral. The negative sign arises because $\langle J,M|z|J,M\rangle = -\langle J,-M|z|J,-M\rangle$. Note that T vanishes when

$$a_s a_p = b_s b_p\,, \tag{8a}$$

which means

$$a_s^2 + a_p^2 = 1\,. \tag{8b}$$

For states described by Eq. (3b), the condition will be identical.

At zero field, $a_s^2 = a_p^2 < \frac{1}{2}$ for states with $M_F<0$ [see Eq. (5)] but to satisfy Eq. (8), at least one of the a^2 terms must become $\geqslant\frac{1}{2}$. The condition $a^2=\frac{1}{2}$ implies complete mixing of the states of the decoupled basis which requires an avoided crossing between two states of the same M_F. This avoided crossing can only occur between Zeeman sublevels with $M_F<0$ in atomic systems with $I>J$. Therefore, Eq. (8) is satisfied only for the proper value of magnetic field applied to an atomic system with $I>J$ and $M_F<0$.

The extremum in the energy eigenvalue implied by an avoided crossing occurs at $X=-M_F$ [see Eq. (4)], for which Eq. (5) gives $a^2=\frac{1}{2}$. If the energy extrema for both the S and P states occur at the same field, that is, $X_s = X_p$, then Eq. (8) is satisfied and T vanishes at this field. Hydrogen-like atoms provide one example of $X_s = X_p$. The g_J values are $g_J(^2S_{1/2}) = 2 = 3g_J(^2P_{1/2})$, and for states of the same J and n the hfs scales as $1/(2l+1)$, yielding $A(^2S_{1/2}) = 3A(^2P_{1/2})$. Hydrogen itself does not satisfy $I>J$, but deuterium and many hydrogen-like ions satisfy this condition. All the alkalis have large enough I to exhibit vanishing transition moments as a result of the mixing described here, but do not necessarily have $X_s = X_p$. T will still vanish but at a field between the anticrossing in the P state and that in the S state.

There are a number of interesting properties of the atomic system at the field where T vanishes. First, note that $a_s^2 + a_p^2 = 1$ implies that $\langle J_z\rangle_s = -\langle J_z\rangle_p$. Since

$$\mu g\langle J_z\rangle = dE/dB \tag{9}$$

by the Hellmann-Feynman theorem,[4] $d\epsilon/dX$ of the S and P energy levels are equal and opposite at the field where T vanishes. This leads to

$$X_s + X_p = -2M_F \tag{10}$$

for the transition amplitude to vanish. When $X_s = X_p$, Eq. (10) reduces to $X=-M_F$ which was discussed earlier. Furthermore, the other P state of the same M_F [as described by Eq. (3b), for example] will have $\langle J_z\rangle_s = \langle J_z\rangle_p$, at this field.

Second, at an arbitrary field, the S state will radiatively couple to both P states so that a short pulse of z-polarized light will result in quantum beats at the hfs frequency in the flourescence from the P states. At this field, however, the amplitude of the quantum beats vanishes with the transition amplitude even though their frequency remains finite.

Third, consider more carefully the action of a short pulse (or spectrally broad light) connecting an S state with a pair of P states.[5] For $\Delta M_F = 0$ transitions, $\langle I_z\rangle$ averaged over the P states just after the pulse will be the same as $\langle I_z\rangle$ in the S state, since the light does not affect I and the pulse is too short for the hyperfine coupling to occur. If the transition to one of the P states vanishes, we have $\langle I_z\rangle_S = \langle I_z\rangle_P$, which leads to $a_s^2 + a_p^2 = 1$, and an alternative way of deriving the condition where T vanishes.[6]

We have observed the vanishing of T for the case of $\Delta M_F = 0$ optical transitions in ^{23}Na ($I=\frac{3}{2}$) between certain Zeeman components of the $3S_{1/2}(F=1) \rightarrow 3P_{1/2}(F=1)$ transition. The strength of each transition was determined by measuring the fluorescence from Na as a function of an applied magnetic field. It can be readily shown that the absorption and fluorescence curves are the same if decays to all ground-state sublevels are observed.

A beam of thermal sodium atoms was directed through holes bored in the pole pieces of a small electromagnet so that the atoms moved parallel to the field. The 1.8-mm diam. holes in the 19-mm diam. pole pieces were enough to provide a well collimated beam (half angle $\cong 0.01$ rad) without causing too much distortion of the field in the 5-mm gap between magnet poles.

Light from a Coherent Radiation Model 599 single-frequency, scannable dye laser using Rhodamine 6G was directed perpendicular to the atomic beam. The laser, which has a spectral width of 2 MHz, was polarized linearly in the direction of the magnetic field so that only $\Delta M_F = 0$ transitions were driven (M_F is a good quantum number at all fields, even though F is not). Absorption of the intensity stabilized laser light was monitored by observing total fluorescence perpendicular to both the atomic and laser beams with a phototube.

Spectra were obtained by recording the photocurrent as a function of laser frequency at a fixed magnetic field. Frequency scans covered about 2 GHz and were calibrated with respect to a 300-MHz free spectral range étalon. Subsequent scans were taken at various values of the magnetic field which was determined from the splitting between the $3S(F=1, M_F=1) \rightarrow 3P(F=2, M_F=1)$ and $3S(F=1, M_F=0) \rightarrow 3P(F=2, M_F=0)$ transitions (highest-frequency transitions).

The observed fluorescence lines had a width of about 30 MHz of which 10 MHz can be ascribed to the natural width, and 15 MHz to residual Doppler width. We attribute the remaining width to magnetic field inhomogeniety caused by the geometry of the electromagnet and the holes in the pole faces.

We determined the heights of each peak in the spectrum from the X-Y recording of the phototube output. The results are plotted against the applied field in Fig. 1. Also in Fig. 1 (solid curve) is the value of T^2 calculated by substituting Eqs. (5) and (3) into Eq. (1) and using $A_s = 886$ MHz and $A_p = 96$ MHz. This is the expected fluorescence curve since our detector could not distinguish between transitions to the $F=2$ and the $F=1$ ground-state levels. Apart from the choice of intensity scale, there were no free parameters in plotting Fig. 1.

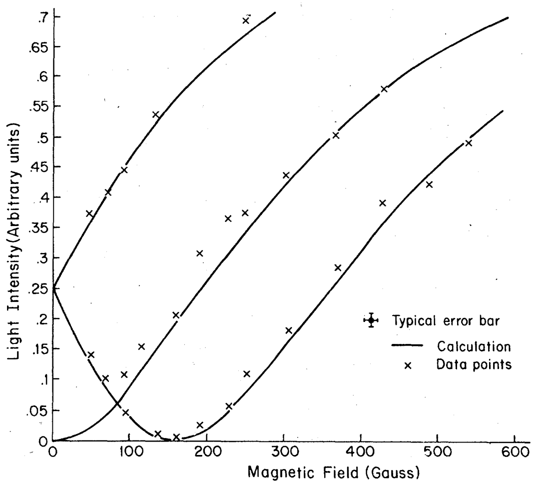

FIG. 1. Data points are the fluorescence peak heights for the three $\Delta M_F = 0$, $\Delta F = 0$ transitions ($F=1$). (Transitions to the $F=2$ excited state were also observed, but their amplitudes have been omitted for clarity.) The solid line is the absorption calculated from Eqs. (5), (3), and (1). The absorption and fluorescence curves are expected to be the same because our detector is equally sensitive to both hyperfine transitions.

The vanishing matrix element is a simple and rather general consequence of angular momentum selection rules and perturbation theory. The phenomenon occurs in a large number of transitions independent of changes in the principle quantum number n. For example, the phenomenon occurs in $n=2S$ to $2P$ transitions in hydrogenlike ions (Lamb-shift transitions) when I is large enough. It may have applications in Lamb shift or other microwave experiments for state selection, and may also be useful for controlling transition amplitudes in atomic weak neutral-current experiments.

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[1] C. Kocher, Phys. Rev. A 6, 35 (1972).

[2] We have used the conventions and notation of A. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton University, Princeton, N.J., 1957). Note that we have treated F as a good quantum number. Since this is not the case in general, the coupling coefficients will be Clebsch-Gordan coefficients only in zero-field. When we refer to F at finite field we mean

a state which connects adiabatically to that F at zero field.

[3] Zeeman diagrams of this case are shown by G. Harnwell and W. Stephens, *Atomic Physics* (Dover, New York, 1966), p. 193 and N. Ramsey, *Molecular Beams* (Oxford University, New York, 1963), p. 79.

[4] E. Merzbacher, *Quantum Mechanics* (Wiley, New York, 1961), p. 442.

[5] Spectrally broad light can be characterized as having a short correlation time which produces transitions similar to those produced by comparably short pulses.

[6] We are indebted to Franck Laloë for this viewpoint.