Relation of Crossing and Anticrossing Effects in Optical Resonance Fluorescence*

K. E. LASSILA
Case Institute of Technology, Cleveland, Ohio
and
Institute for Atomic Research and Department of Physics, Iowa State University, Ames, Iowa†

(Received 10 April 1964)

An interaction Hamiltonian analogous to that in a coupled Lee model is used to calculate the transition matrix appropriate for the description of the anticrossing effect in optical resonance fluorescence. The transition matrix is found to contain two terms (in addition to those for resonance scattering through each of the coupled states) which exist in the amplitude only because the damping matrix in the coupled representation is not diagonal. This matrix is diagonal when the uncoupled states have equal radiative widths, and the resonance scattering intensity is then given by the Breit formula. This result justifies the application by Eck, Foldy, and Wieder of the Breit equation to their initial discovery of the anticrossing effect even though this formula would not be expected to apply to the general anticrossing situation. The crossing effect can also be discussed as a special case of the approach presented.

THE Breit formula[1] has been used by Franken[2] and by Rose and Carovillano[3] to explain the interference effects observed[4] in the optical resonance fluorescence by crossed atomic levels (referred to in the following as the crossing effect). In the present article, results will be presented which will serve to clarify some points on the use of the Breit formula, and which will, particularly, show when this formula can be applied to the recently discovered anticrossing effect[5] in optical resonance fluorescence.

The experiments in question and calculation to be presented can be best discussed by referring to Fig. 1. The crossing effect can be discussed in terms of an atomic ground state 0 and excited states a, b with energy levels having a magnetic field dependence as shown. Radiation with a continuous energy spectrum is resonantly scattered by the system. This process is described by an operator for absorption $\mathbf{f}$ specifying frequency, direction, and polarization of the annihilated photon and a similar operator $\mathbf{g}$ for the created photon. When the magnetic field is swept through H_0, the field strength at which $E_a=E_b$, a detector at a fixed scattering angle observes a signal due to redistribution of the scattered radiation because of interference effects.

The experimental situation in the anticrossing effect is similar except that a perturbing interaction (V) coupling states a and b exists. The energy levels, labeled by α and β in Fig. 1, are obtained from the secular determinant and the states themselves are given in terms of a and b with coefficients determined by V,

$$\begin{aligned}\alpha&=A_a a+A_b b\,,\\ \beta&=B_a a+B_b b\,,\end{aligned}\tag{1}$$

where orthogonality and normalization of α and β give

$$\begin{aligned}&|A_a|^2+|A_b|^2=|B_a|^2+|B_b|^2=1\,,\\ &A_a{}^*B_a+A_b{}^*B_b=0\,.\end{aligned}\tag{2}$$

The interaction Hamiltonian between the system and the electromagnetic field is written as

$$H_I=\mathbf{f}a_k a_\alpha{}^\dagger a_0+\mathbf{f}a_k a_\beta{}^\dagger a_0+\mathbf{g}a_k{}^\dagger a_0{}^\dagger a_\alpha+\mathbf{g}a_k{}^\dagger a_0{}^\dagger a_\beta\,,\tag{3}$$

where $a_k{}^\dagger$ and a_k create and annihilate, respectively, a photon of momentum k (with polarization label suppressed), and $a_j{}^\dagger$ and a_j create and annihilate the atom in state $j=0$, α, or β. The rest of the total Hamiltonian (neglecting H_I) simply gives the energies of the states 0, α, and β plus the total energy in the radiation field. Thus, one sees that the description given here for resonance fluorescence is formally identical with a coupled Lee model.[6] The physical justification for this approach is that contributions from intermediate states with more than one photon will be negligible because of the smallness of the electromagnetic coupling

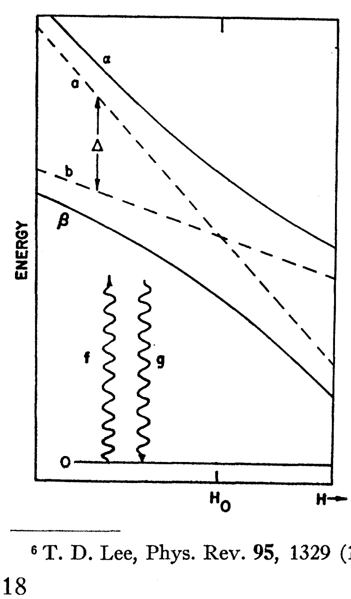

FIG. 1. Energy levels as functions of magnetic field H. The unperturbed levels labeled by a and b are separated by an energy Δ at field H; the solid lines represent the energy levels as a function of H when the coupling perturbation V is included. The ground state (0) energy variation is not of interest; $\mathbf{f}$ is an operator connecting the ground states with the excited states and $\mathbf{g}$ connects the excited states with the ground state.

* Contribution No. 1499. Work was supported by the U. S. Atomic Energy Commission.
† Present address.
[1] G. Breit, Rev. Mod. Phys. **5**, 91 (1933).
[2] P. A. Franken, Phys. Rev. **121**, 508 (1961).
[3] M. E. Rose and R. L. Carovillano, Phys. Rev. **122**, 1185 (1961).
[4] F. D. Colegrove, P. A. Franken, R. R. Lewis, and R. H. Sands, Phys. Rev. Letters **3**, 420 (1959).
[5] T. G. Eck, L. L. Foldy, and H. Wieder, Phys. Rev. Letters **10**, 239 (1963).
[6] T. D. Lee, Phys. Rev. **95**, 1329 (1954).

constant and will not lead to terms with resonant denominators.

Using H_I, one can calculate the T matrix straightforwardly to obtain ($\hbar = c = 1$)

$$T = [(E_0 - E_\alpha + k - D_{\alpha\alpha})(E_0 - E_\beta + k - D_{\beta\beta}) - D_{\alpha\beta}D_{\beta\alpha}]^{-1}[g_{0\alpha}f_{\alpha 0}(E_0 - E_\beta + k - D_{\beta\beta}) + g_{0\beta}f_{\beta 0}(E_0 - E_\alpha + k - D_{\alpha\alpha}) + g_{0\beta}D_{\beta\alpha}f_{\alpha 0} + g_{0\alpha}D_{\alpha\beta}f_{\beta 0}], \quad (4)$$

where k is the energy of the incident (absorbed) photon,

$$f_{\alpha 0} = (\psi_\alpha | \mathbf{f}_\mathbf{k} | \psi_0), \quad g_{0\alpha} = (\psi_0 | \mathbf{g}_\mathbf{k} | \psi_\alpha) \quad \cdots \text{ etc.},$$

and

$$D_{\mu\nu} = \lim_{\epsilon \to 0} \sum \int dk' (\psi_\mu | \mathbf{f}_{\mathbf{k}'}{}^P | \psi_0)(k - k' + i\epsilon)^{-1} \times (\psi_0 | \mathbf{g}_{\mathbf{k}'}{}^P | \psi_\nu), \quad (5)$$

with μ, $\nu = \alpha$ or β. In (5), the sum is over all photon polarizations, directions, and any quantum numbers needed to specify the "intermediate" atomic wave function ψ_0. Since we are not now interested in line shift experiments, the real part of $D_{\mu\nu}$ can be neglected.[7] Thus,

$$D_{\mu\nu} = -i\Gamma_{\mu\nu}/2 = -i\pi \sum \int dk' \delta(k - k') \times (\psi_\mu | \mathbf{f}_{\mathbf{k}'}{}^P | \psi_0)(\psi_0 | \mathbf{g}_{\mathbf{k}'}{}^P | \psi_\nu) \quad (6)$$

gives the damping matrix for the coupled system, which has the following elements in terms of the radiative widths of the uncoupled states,

$$\begin{pmatrix} \Gamma_{\alpha\alpha} = |A_a|^2\Gamma_a + |A_b|^2\Gamma_b & \Gamma_{\alpha\beta} = A_a{}^*B_a\Gamma_a + A_b{}^*B_b\Gamma_b \\ \Gamma_{\beta\alpha} = B_a{}^*A_a\Gamma_a + B_b{}^*A_b\Gamma_b & \Gamma_{\beta\beta} = |B_a|^2\Gamma_a + |B_b|^2\Gamma_b \end{pmatrix}. \quad (7)$$

This matrix is, in general, nondiagonal. It becomes diagonal when (i) $\Gamma_a = \Gamma_b = \Gamma$, by the conditions of Eq. (2) $\Gamma_{\alpha\alpha} = \Gamma_{\beta\beta} = \Gamma$, and (ii) when $V = 0$, as $A_b = B_a = 0$. Therefore, the transition matrix appropriate for describing the anticrossing effect with uncoupled states of equal radiative widths *or* for describing the crossing effect is

$$T' = g_{0\alpha}f_{\alpha 0}(E_0 - E_\alpha + k + i\Gamma_{\alpha\alpha}/2)^{-1} + g_{0\beta}f_{\beta 0}(E_0 - E_\beta + k + i\Gamma_{\beta\beta}/2)^{-1}. \quad (8)$$

For incident photons with a constant energy distribution, the resonance scattering cross section or rate given by (8) will be

$$\sigma \propto \int dk |T'|^2 \propto \Gamma_{\alpha\alpha}{}^{-1} |g_{0\alpha}f_{\alpha 0}|^2 + \Gamma_{\beta\beta}{}^{-1} |g_{0\beta}f_{\beta 0}|^2 + \left[\frac{g_{\alpha 0}f_{0\alpha}g_{0\beta}f_{\beta 0}}{i(E_\beta - E_\alpha) + \frac{1}{2}(\Gamma_{\alpha\alpha} + \Gamma_{\beta\beta})} + \text{c.c.} \right], \quad (9)$$

where constants have been left out. In case (i) with $\Gamma_a = \Gamma_b = \Gamma$, Eq. (9) is the Breit equation in exactly the form discussed by other authors,[2,3] *except* that the matrix elements involve the coupled states. This justifies Eck, Foldy, and Wieder's[5] application of the Breit formula with coupled states to their experimental observations. Incidentally, it also shows that Series'[8] comparison of optical double resonance with the formula of Eck, Foldy, and Wieder was not as general a comparison as possible because of the particular case, $\Gamma_a = \Gamma_b = \Gamma$, assumed.

In case (ii) with $V = 0$ the states are uncoupled ($\alpha = a$, $\beta = b$, $\Gamma_{\alpha\alpha} = \Gamma_a$, and $\Gamma_{\beta\beta} = \Gamma_b$) and Eq. (9) differs from Franken's (where $\Gamma_a = \Gamma_b = \Gamma$ was assumed) and Rose and Carovillanos' result only in the incoherent background term, $\Gamma_a{}^{-1}|g_{0a}f_{a0}|^2 + \Gamma_b{}^{-1}|g_{0b}f_{b0}|^2$, for which these authors get the same Γ^{-1} factor on both terms.

The main result presented here can be summarized as follows: in an optical resonance fluorescence experiment on "tuned" atomic levels, the Breit formula, Eq. (9), may be applied to the anticrossing effect (in addition to the crossing effect) when the damping matrix of Eqs. (6) and (7) is diagonal. This matrix is not diagonal in the general anticrossing situation, and the cross section following from the transition matrix Eq. (4) is correspondingly more complicated[9] than the Breit formula. *A priori*, it would appear therefore that the Breit formula might not be applicable to the anticrossing effect, and its use by Eck, Foldy, and Wieder should be justified. A simply treated situation, for example, occurs when one of the states (in a steady-state experiment of the type under discussion) is nonradiative; the matrix of Eq. (7) will still be nondiagonal as the radiative width of the other state occurs linearly in each matrix element. In this case, the direct application of the Breit formula, as given in Refs. 2 and 3, with coupled levels leads to a result violating the sum rule for total scattering.[10] However, the approach presented here gives a consistent result, the total resonance scattering being independent of the energy level spacing.

ACKNOWLEDGMENTS

It is a pleasure to thank Professor T. G. Eck and Professor L. L. Foldy for suggesting the study of the anticrossing effect and for numerous beneficial discussions.

[7] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, London, 1954), 3rd ed., Sec. 16; W. Heitler and S. T. Ma, Proc. Roy. Irish Acad. **52**, 109 (1949).

[8] G. W. Series, Phys. Rev. Letters **11**, 13 (1963).

[9] K. E. Lassila (to be published). The general cross-section formula, especially in the uncoupled representation, is under study in an effort to make it tractable, as the possibility of an anticrossing experiment with uncoupled states with different lifetimes is being considered by the Columbia group, Columbia Radiation Laboratory Quarterly Progress Report, 15 December 1963 (unpublished).

[10] The author would like to thank Professor T. G. Eck for pointing out this discrepancy and thus stimulating the author's interest in the present problem.