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LEVEL-CROSSING SIGNAL LINE SHAPES AND ORDERING OF ENERGY LEVELS*

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Theory and experiments are presented to show that, when correctly interpreted, the sign of the dispersion-shaped term which describes a part of the change in resonance scattering at level crossings can be used to determine the energy ordering of the crossing levels, and hence, in some cases, to obtain the sign of the nuclear magnetic moment. Because of a discrepancy in the literature in the theoretical prediction for the sign of the dispersion-shaped term, we have made a recalculation. The result is borne out by our experiments.

Colegrove, Franken, Lewis, and Sands[1] introduced the use of the "level-crossing" effect in atomic resonance fluorescence as a very fruitful technique for the measurement of fine and hyperfine structures.[2] The experiments have relied largely on the study of the Lorentz-shaped term that describes a part or, for certain geometries, all of the variation of the resonance scattering at level crossings. We wish to show that if the dispersion-shaped term is used, in addition to obtaining the level-crossing magnetic fields, we can determine the energy ordering of the crossing levels which may be of importance in the analysis of the level structure. In particular instances this can serve to obtain the sign of the hfs interaction constant, and hence that of the nuclear magnetic moment. Since the sign of the dispersion-shaped term is crucial we call attention to the fact that our calculations show that this sign is opposite to that obtained by Franken,[3] whose result is used in much of recent level-crossing literature. In contrast, our result —which was corroborated by our experiments—is in agreement with early work of Weisskopf, whose result also appears in the review article by Breit, and with that of Rose and Carovillano and of Lassila.[4]

We have calculated in a manner analogous to that of Franken[3] the rate R at which photons of polarization $\vec{f}$ are absorbed from ground-state levels with magnetic quantum numbers m, m', and are re-emitted with polarization $\vec{g}$ from excited-state levels μ, μ'. We find

$$R \propto \sum_{\substack{m\,m' \\ \mu\,\mu'}} f_{\mu m} f_{\mu' m}{}^* g_{m'\mu'}{}^* g_{m'\mu} \times \frac{1}{1+i(E_\mu - E_{\mu'})/\Gamma\hbar}, \qquad (1)$$

where $f_{\mu m}$, $g_{\mu' m'}$, etc. are the appropriate electric dipole matrix elements, Γ is the excited-state decay constant, and E_μ and $E_{\mu'}$ are the energies of the excited-state sublevels. In this expression we differ in the sign of the factor that multiplies $E_\mu - E_{\mu'}$ from that obtained in Ref. 3. The consequence is a change in the sign of the dispersion-shaped term in (2b). The variation in the rate R produced when levels cross is given by the "signal" term S, which is obtained from (1) as

$$S \propto \sum_{\mu \neq \mu'} A_{\mu\mu'} \frac{1}{1 + i(E_\mu - E_{\mu'})/\Gamma\hbar}, \tag{2a}$$

where we define

$$A_{\mu\mu'} \equiv \sum_{m, m'} f_{\mu m} f_{\mu' m}^* g_{m'\mu'}^* g_{m'\mu}.$$

For a given pair of crossing levels this can also be written as

$$S \propto \frac{A_{\mu\mu'} + A_{\mu\mu'}^*}{1 + [(E_\mu - E_{\mu'})/\Gamma\hbar]^2} - i \frac{[(E_\mu - E_{\mu'})/\Gamma\hbar](A_{\mu\mu'} - A_{\mu\mu'}^*)}{1 + [(E_\mu - E_{\mu'})/\Gamma\hbar]^2}. \tag{2b}$$

$A_{\mu\mu'}$ can be expressed in terms of the matrix elements of the spherical tensor components of $\vec{r}$ and geometrical factors that depend on the directions of the incident and scattered photons $\vec{k}_i$ and $\vec{k}_S$, of the applied magnetic field $\vec{H}$, and of the polarization vectors $\vec{f}$ and $\vec{g}$. We give here only the result for the level crossings of the type $|\mu - \mu'| = 2$, which are the ones observed in the experiments that we shall describe. The direction of the magnetic field $\vec{H}$ defines the positive z axis and $\vec{k}_i$ is taken, by definition, to lie in the x-z plane. The vector $\vec{k}_i$ makes an angle θ_i with the z axis. We can then resolve the incident polarization vector $\vec{f}$ into $f_A u_y + f_B u_\theta$, where u_y and u_θ are unit vectors in the y and θ directions. The vector $\vec{k}_S$ lies at angles θ_S, φ_S as measured in a right-handed coordinate system. Since we do not use any analyzers, we sum over the two orthogonal components of $\vec{g}$. With these definitions we obtain

$$A_{\mu\mu'}(\mu > \mu') = |\langle m | r_{-1}^{\ 1} | \mu\rangle|^2 |\langle m | r_1^{\ 1} | \mu'\rangle|^2 (2\pi/15)^{1/2} Y_2^{\ 2}(\theta_S, \varphi_S) \times [|f_A|^2 - |f_B|^2 \cos^2\theta_i - i(f_A f_B^* + f_A^* f_B)\cos\theta_i]. \tag{3}$$

We note that if we interchange the roles of μ and μ', i.e., choose $\mu < \mu'$, the expression corresponding to (3) is given by $A_{\mu'\mu} = A_{\mu\mu'}^*$. The result for S, (2b), remains unaffected. Our experiments were performed with mercury using the 2537-Å $^3P_1$-$^1S_0$ intercombination resonance line. Because the electronic angular momentum in the ground state is zero, we have therefore only a single value $m = m'$, and the sum over these quantum numbers does not appear in (3).

The commonly used geometry, which was also that for our experiments, is $\theta_i = \theta_S = 90°$ for which the factors multiplying the spherical harmonic $Y_2^{\ 2}(\theta_S, \varphi_S) = (15/32\pi)^{1/2} \sin^2\theta_S \exp(2i\varphi_S)$ are positive. The remaining angular dependence of $A_{\mu\mu'}$ in (3) is $+\exp(2i\varphi_S)$, with which (2b) becomes

$$S \sim \frac{\cos 2\varphi_S}{1 + [(E_{\mu>} - E_{\mu<})/\Gamma\hbar]^2} + \frac{[(E_{\mu>} - E_{\mu<})/\Gamma\hbar]\sin 2\varphi_S}{1 + [(E_{\mu>} - E_{\mu<})/\Gamma\hbar]^2}. \tag{4}$$

We emphasize the specific positions of $\mu_>$ and $\mu_<$, the larger and smaller magnetic quantum numbers of the crossing levels, in the second term of (4). Clearly the sign of the "dispersion" term in the signal S depends on whether the energy level $\mu_>$ lies above or below the level $\mu_<$. Thus for a zero-field crossing (Hanle effect), the sign of the "dispersion" term is determined by the sign of the g factor.

A knowledge of the energy ordering can be of value in the identification of the crossing levels. As an example where this may be important, we consider an atom with electronic angular momentum $J = 1$ and nuclear spin $I = \frac{3}{2}$. A $|\mu - \mu'| = 2$ level crossing observed at a magnetic field that corresponds to approximately $3A/\mu_0 g_J$ (A, magnetic dipole hfs interaction constant; μ_0, Bohr magneton; and g_J, electron gyromagnetic ratio) could correspond to a B/A ratio of either −0.11 or 2.7. B is the electric quadrupole hfs interaction constant. However the dispersion-shaped term has opposite sign and hence distinguishes the two cases.

Though actually not used at the time, as an example where the application of this technique would have served to determine the sign of the nuclear magnetic moment μ_I, we cite our experiments[5] on 47-d Hg^{203} ($I=\frac{5}{2}$) in which we were able to observe spectroscopically only a single component in the hfs of the 2537-Å resonance line, the others being blended in the spectral region of natural mercury. Thus, with a normal ordering of the hfs levels, we were observing either the component of maximum total angular momentum F for a negative nuclear magnetic moment or of minimum F for the opposite sign of μ_I. Since the energy ordering of the m_F levels is opposite in the two cases, a study of the line shape of the

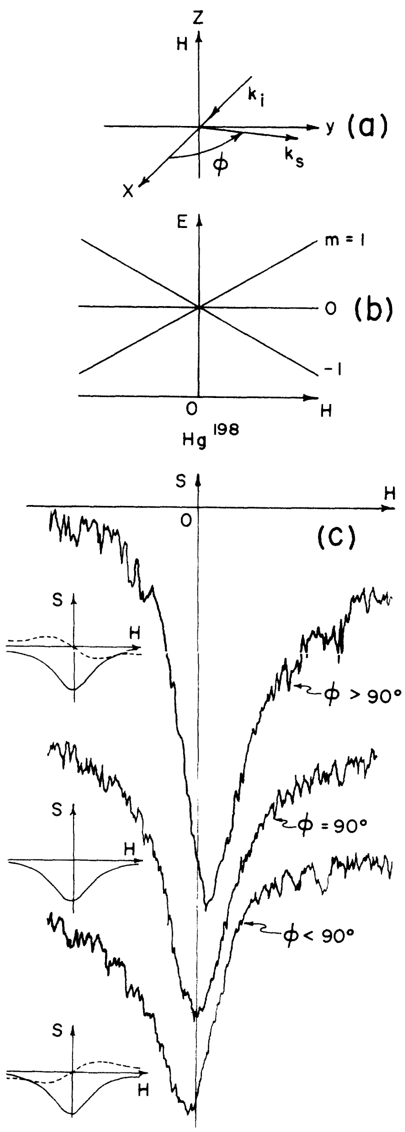

FIG. 1. (a) Geometry for zero-field level-crossing experiment. (b) Energy levels of Hg^{198} in $^3P_1$ state near zero-field crossing. (c) Level-crossing signals obtained for rectangular geometry $\varphi = 90°$, and for $\varphi = 90° \pm 10°$. Contributing Lorentz and "dispersion" terms are indicated.

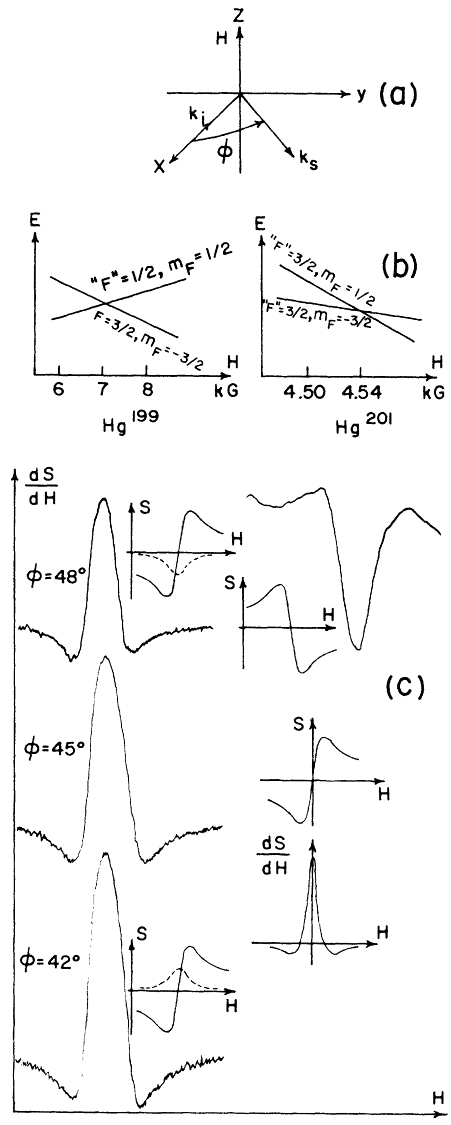

FIG. 2. (a) Geometry for high-field experiments. (b) Crossing energy levels for Hg^{199} and Hg^{201}. (c) Derivatives with respect to H of level-crossing signals. For Hg^{199}, results are shown for $\varphi = 45°$ and for $\varphi = 45° \pm 3°$. Contributing Lorentz and "dispersion" terms are indicated. For $\varphi = 45°$ the expected derivative shape is also shown. Note the inversion of the curves for the two crossings.

zero-field crossing would discriminate between these two possibilities.

Because the sign of the dispersion-shaped term is essential in these studies, we have carried out several experiments which confirm the above results. Both high-field and near-zero-field level crossings were studied. The sense of the conventional positive magnetic field, a knowledge of which is required for the correct definition of the angles in $A_{\mu\mu'}$, was ascertained relative to the field of a current loop. The detector response to a positive signal was also established. In the case of the zero-field level crossings the observations were made without lock-in detection. In the high-field work results were obtained both with and without lock-in detection. For the former case only the recorded detector output, which is proportional to the derivative of S with respect to H, is presented in Fig. 2.

Hg^{198}, which has $I=0$, was used in the zero-field experiments. With rectangular geometry ($\varphi_S=90°$), $\sin 2\varphi_S=0$, and the second term in (4) is zero. Eq. (4) therefore gives a pure, negative, Lorentz-shaped signal. If, as was done, we make a small change δ in the angle φ, measured in the right-handed coordinate system referred to positive H, the first term remains the same in first approximation. The second term, however, now contributes with the angular factor equal to $-\sin 2\delta$. This introduces an asymmetry in the line shape from which the sign of the dispersion-shaped term can be determined. The experimental geometry and the results are shown in Fig. 1.

High-field level-crossing experiments were used to show the reversal in sign of the "dispersion" term with ordering of the energy levels in (4). Near the level crossing, on the lower magnetic field side, $E_{\mu>}-E_{\mu<}$ is negative for Hg^{199} while for Hg^{201}, in the "foldover" crossing (same low-field F value for both states), $E_{\mu>}-E_{\mu<}$ is positive. The sign of the dispersion-shaped term is thus expected to be opposite in the two cases.

In the high-field experiments we observed the scattered light at $\varphi_S=45°$, so that the signal is given entirely by the second term in (4) with the angular factor equal to +1. The results are shown in Fig. 2. As an additional verification of (4) we studied the influence on the line shape produced by a small variation δ in the angle $\varphi=45°$. The dispersion-shaped term remains approximately unchanged, but the Lorentz-shaped term now contributes with an angular factor equal to $-\sin 2\delta$. These experimental results are also shown in Fig. 2.

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