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COHERENT ANTICROSSING SIGNALS

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Anticrossing signals have been observed in resonance fluorescence in which the two repelling levels are both excited from the same level in the ground state. The two anticrossed levels both decay to the same ground-state level as well. This extension of the anticrossing technique is successfully applied in a state of the hfs of the ytterbium atom.

The technique of level-crossing spectroscopy requires coherence in both the excitation and detection channels. Coherence in this sense means that the two crossing levels must both be excited from the same one or more levels, and must both decay to the same one or more levels. By contrast, the techniques of optical double resonance and of anticrossing[1] spectroscopy achieve their greatest utility when only one of the excited levels is populated, or when the decay of only one of the excited levels is observed. We report here the first observation of coherent anticrossing signals. In this case, the repulsion of two Zeeman levels is observed with either coherent excitation, or coherent fluorescence, or both. The anticrossing signal was identified by its position, intensity, linewidth, and lineshape, and most important, by its dependence on the polarization of the incident and scattered light. Coherent anticrossings may prove useful in obtaining information not easily provided by level crossings or by pure anticrossings.

The following sequence of experiments was performed in the $6s\,^1P_1$ level of the ytterbium atom. Resonance radiation, corresponding to the transition $(6s)^2\,^1S_0 \rightarrow 6s6p\,^1P_1$ at 3987 Å, from a hollow-cathode lamp of natural Yb, was incident on a dense atomic beam of natural Yb. Light scattered at 90° to both the incident light beam and the magnetic field passed through a narrow-band interference filter peaked at 4000 Å and into a photomultiplier. As the magnetic field was increased, weak level-crossing signals were observed in the vicinity of 40 G and a single strong signal appeared at approximately 147 G. Natural Yb contains 14% Yb^{171} ($I=\frac{1}{2}$) and 16% Yb^{173} ($I=\frac{5}{2}$). The strong signal vanished when a sample enriched to 85% in Yb^{173} and containing only 0.7% of Yb^{171} was substituted to scatter the resonance radiation. This identified the strong signal as belonging to Yb^{171}. In addition, the trace shown in Fig. 1 was obtained. Besides the Hanle ef-

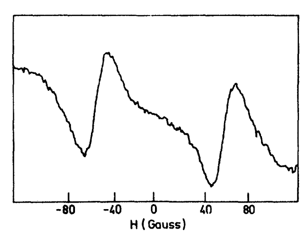

FIG. 1. Level-crossing signals obtained with an enriched sample of Yb^{173}. The incident (or detected) light was linearly polarized at an angle of 55° to the magnetic field direction.

fect or zero-field level crossing, one easily observes a second strong crossing at approximately ±35 G and, with somewhat greater difficulty, a third crossing at approximately ±60 G. Fig. 1 was obtained with the incident (or detected) light linearly polarized at an angle of 55° with respect to the direction of the magnetic field for reasons to be explained below. The detected (or incident) light was unpolarized. It differs from a similar sweep obtained with unpolarized light in both channels only in that the third level crossing is rendered even less distinct with polarized light. The hfs constants of Yb^{173} can now be deduced in the following unambiguous way. Neglecting any hfs anomaly, we may use the ratio A/g_J for Yb^{171} inferred from the signal at 147 G to yield a value for the similar ratio for Yb^{173}. The strong signal at approximately 35 G, identified as the main crossing between the $F=\frac{7}{2}$, $M_F=-\frac{7}{2}$ and $F=\frac{3}{2}$, $M_F=-\frac{3}{2}$ levels, and corrected for overlap, may now be used to extract a value for B/A, the ratio of quadrupole to dipole hfs constants.[2] We use the formula

$$\frac{3A}{g_J}\left(1+\frac{9}{40}b-\frac{9}{400}b^2\right)=\mu_0H_c\left(1+\frac{9}{40}b\right), \tag{1}$$

where $b=B/A$ and H_c is the crossing field.[3] Higher-order terms are negligible. We find $A=59.7(12)$ Mc/sec and $B=600(15)$ Mc/sec. g_J has been taken as 1.05(2).[4] With these constants an energy-level diagram was constructed and is shown in Fig. 2. The third hfs level, $F=\frac{5}{2}$, has an energy of −540 Mc/sec and cannot be shown conveniently. In addition to the two observed level crossings (circled in the figure), one notices the striking anticrossing between the levels $F=\frac{7}{2}$, $M_F=-\frac{3}{2}$ and $F=\frac{3}{2}$, $M_F=-\frac{3}{2}$, as well as a somewhat weaker repulsion between the $M_F=-\frac{1}{2}$ levels.

The resonance fluorescence signal, expressed as a function of Δ, the energy separation of the two repelling levels, has been given by Eck[1]:

$$S=\frac{1}{\gamma}\sum[|f_a|^2|g_a|^2]+\frac{1}{\gamma}\sum[|f_b|^2|g_b|^2]+\frac{2\gamma}{D}\sum[f_af_bg_ag_b]-\frac{2|V|^2\gamma}{D}\sum[fg]$$
$$+\frac{2V^2}{\gamma D}\sum[(2f_af_b)(2g_ag_b)]+\frac{2V\Delta}{D}\sum[f(g_ag_b)+g(f_af_b)], \tag{2}$$

where $\gamma=1/2\pi\tau$ (τ is the lifetime of the excited level), $D=\gamma^2+|2V|^2+\Delta^2$ is the resoance denominator, $f=(1/\gamma)(|f_a|^2-|f_b|^2)$, $g=(1/\gamma)\times(|g_a|^2-|g_b|^2)$, and V is the matrix element of the perturbation which couples states a and b. The symbol f_a is used to represent the dipole matrix element $f_{am}=\langle a|\vec{f}\cdot\vec{r}|m\rangle$ that connects state a to the ground-state Zeeman level m. Similarly, $g_a\equiv g_{m'a}=\langle m'|\vec{g}\cdot\vec{r}|a\rangle$ is the matrix element for the decay. The summation is carried out over all initial levels m and final levels m' involved in the excitation and decay. We have simplified Eck's original expression to obtain Eq. (2) by noting that both anticrossing levels have the same lifetime and that the perturbation V is real. In addition, with the geometry described above, the matrix elements f_a, f_b, g_a, and g_b may all be chosen as real.

Eck has discussed the significance of each of the terms in Eq. (2). The first and second terms represent the nonresonant background from the states a and b. The third term resembles the normal level-crossing signal by requiring coherence in both excitation and decay. The fourth term is the pure anticrossing sig-

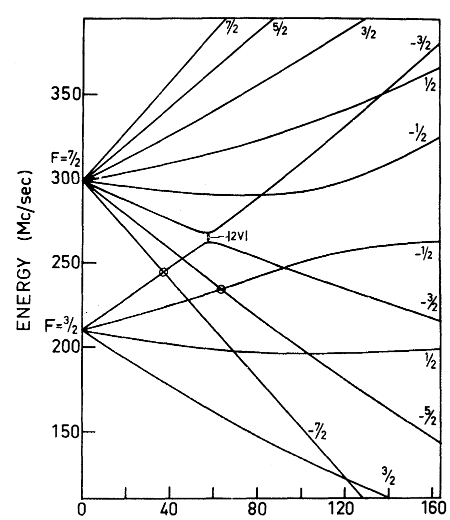

FIG. 2. Zeeman levels of the $^1P_1$ state of Yb^{173}. The $F=\frac{5}{2}$ state has an energy of −540 Mc/sec, and is not shown. Observed level crossings are circled.

nal observed by Eck. The fifth and sixth terms are mixed crossing and anticrossing terms that require coherence in at least one of the two steps of the resonance fluorescence process.

Neither the hfs interaction nor the magnetic field couples states with $\Delta F = 2$ in first order. We may therefore take states a and b in an F, M_F representation. The perturbation V is, in fact, provided by a second-order magnetic field perturbation via the $F=\frac{5}{2}$, $M_F=-\frac{3}{2}$ level. This explains the practically straight-line behavior of the anticrossing levels before and after the point of closest approach.

Assuming the light to be propagating in the y direction with the magnetic field in the z direction, we may evaluate the last four terms in Eq. (2), at the position of the anticrossing. The third, fourth, fifth, and sixth terms have respective intensities of 1.84, −0.01, 0.08, and 0.23 in units of $(\gamma^2+|2V|^2)^{-1}$. All terms have the same angular dependence, $(3\cos^2\theta_1 -1)(3\cos^2\theta_2-1)$, where θ_1 and θ_2 are the angles between the direction of polarization and the z direction for the incident and scattered light, respectively. This suggests that these terms may be distinguished from the more intense $\Delta m = 2$ and $\Delta m = 1$ level-crossing signals shown in Fig. 1, whose dependence on angle is $\sin^2\theta_1 \times \sin^2\theta_2$ and $\sin 2\theta_1 \sin 2\theta_2$, respectively. With Polaroids in both the incident and detection channels rotated to $\theta_1 = \theta_2 = 0$, the curve shown in Fig. 3 was observed. The sweep shows two anticrossing signals at $H = \pm 56$ G, as predicted by Fig. 2. Moreover, the signals diminished, vanished, and then reversed sign as the polarizers were turned through an angle of 55° with respect to the z axis. This explains the choice of 55° in obtaining Fig. 1 mentioned above, since this allows observation of a pure crossing signal.

The third term is by far the largest, although it requires coherence in both parts of the resonance fluorescence process. The sixth term contributes a slight asymmetry to the signal due to its dispersion line shape, but the overall line shape is predicted to be Lorentzian. The dispersion-type line shapes shown result from lock-in detection used in our experiment. This is in good agreement with observation, although perturbations from the nonresonant background, from the anticrossing signal at negative (positive) magnetic field, and possibly from the anticrossing of the $M=-\frac{1}{2}$ levels

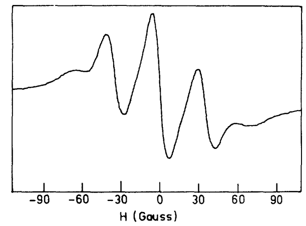

FIG. 3. Coherent anticrossing signals observed with π light in both steps of the resonance fluorescence process. The phase reversal of the two signals is due to the sign reversal of the magnetic field.

rule out any precise determination of the signal asymmetry.

Assuming that the signal is perfectly symmetric (the predicted asymmetry is of the same order as that produced by a slight misalignment of our optics), we find that the peak-to-peak separation is given by[1]

$$\delta_p = (2/\sqrt{3})(\gamma^2+|2V|^2)^{1/2}. \qquad (3)$$

A value of $\gamma = 29.0$ Mc/sec can be inferred from the measured lifetime of the $^1P_1$ level.[5] For $2V$ we may take the distance of closest approach shown in Fig. 2 of 6.3 Mc/sec. This leads to a prediction of 22.9 G for δ_p, where the slope 1.51 Mc/G sec taken from the figure has been used as a conversion factor. The value found from a succession of curves, in which the density of scattering atoms was reduced to eliminate coherence narrowing,[5] is 22.5 ± 1.0 G.

Finally, the intensity of the anticrossing signal at 56 G is predicted to be one-eighth that of the crossing signal at 35 G. A ratio of between five and ten to one was observed in favor of the crossing signal.

In view of the evidence presented above, we feel justified in concluding that we have, in fact, observed signals from the coherent excitation and decay of anticrossing levels. The measured position and linewidth of the anticrossing can now be used to extract an improved value for the quadrupole hfs constant. We find $B = 604(7)$ Mc/sec (assuming $g_J = 1.05$; B is proportional to g_J).

The utility of coherent anticrossing signals in our experiments is apparent from a comparison of Fig. 1 with Fig. 3. More information is obtainable from the single, well-resolved anticrossing than from the superposition of level-crossing signals. This suggests that coherent anticrossings may prove generally useful for the particularly difficult class of atoms with small hfs and short lifetime. A limitation of the technique is that an inversion of the hfs levels may be necessary to avoid a first-order repulsion of levels with the same magnetic quantum number.

[1]T. G. Eck, L. L. Foldy, and H. Wieder, Phys. Rev. Letters 10, 239 (1963); H. Wieder and T. G. Eck, Phys. Rev. 153, 103 (1967).

[2]This identification is based on preliminary optical spectroscopy results kindly supplied to us by S. Gerstenkorn: $A = 3 \pm 1$ mK and $B = 19 \pm 3$ mK. Our experiments cannot rule out the possibility of an inversion of the $F = \frac{7}{2}$ and $F = \frac{3}{2}$ levels. In that case, our work implies $A = 59.7$ Mc/sec and $B = 983$ Mc/sec. Such an inversion has been reported by J. S. Ross and K. Murakawa [J. Phys. Soc. Japan 19, 249 (1964)], who found $A = 1.3$ mK and $B = 23.2$ mK. Our alternative value for B given below is in much better agreement with Gerstenkorn's value and we have therefore taken the level order to be as in Fig. 2.

[3]A. Landman and R. Novick, Phys. Rev. 134, A56 (1964).

[4]W. Meggers, to be published.

[5]M. Baumann and G. Wandel, Phys. Letters 22, 283 (1966).

APPROACH TO HIGH-RESOLUTION nmr IN SOLIDS*

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Consider a spin system having an internal static Hamiltonian $\mathcal{H}$, initially prepared in the state $|0\rangle$ and then subjected to a train of brief, intense, magnetic field pulses whose effects can be represented by the rotations

$$P_k = \exp(-i\theta_k \vec{n}_k \cdot \vec{I}), \tag{1}$$

where $\vec{n}_k$ is a unit vector in the direction of the pulse field in the rotating frame. The state of the system following the nth pulse at time

$$T = \sum_{k=1}^{n} \tau_k$$

is

$$|T\rangle = \left\{ \prod_{k=1}^{n} [P_k \exp(-i\mathcal{H}\tau_k)] \right\} |0\rangle. \tag{2}$$

As usual, it is to be understood that the operators in a product act successively in order of increasing index. Equation (2) can be rewritten as

$$|T\rangle = \left\{ \prod_{k=1}^{n} \exp(-i\tau_k \mathcal{H}_{nk}) \right\} \left\{ \prod_{m=1}^{n} P_m \right\} |0\rangle, \tag{3}$$

where

$$\mathcal{H}_{nk} = \left\{ \prod_{l=k}^{n} P_l \right\} \mathcal{H} \left\{ \prod_{l=k}^{n} P_l \right\}^{-1}. \tag{4}$$

Imagine that the n pulses are so chosen that

$$\prod_{m=1}^{n} P_m = 1.$$

A sequence having this property will be called a cycle. The pulse trains used by Carr and Purcell,[1] Ostroff and Waugh,[2] and Waugh and Huber[3] are cyclic for $n = 2$, 4, and 2, respectively. After N cycles (time $t = NT$) the state of the system is

$$|t\rangle = \left\{ \prod_{k=1}^{n} \exp(-i\tau_k \mathcal{H}_{nk}) \right\}^{N} |0\rangle. \tag{5}$$

It can be shown[4] that if $T \ll T_2$, where T_2 is a natural time of the system associated with $\mathcal{H}$, and often under less restrictive conditions, $|t\rangle$ becomes describable in terms of an average Hamiltonian $\bar{\mathcal{H}}$:

$$|t\rangle \approx \exp(-i\bar{\mathcal{H}}t)|0\rangle, \tag{6}$$

$$\bar{\mathcal{H}} = \frac{1}{T} \sum_{k=1}^{n} \mathcal{H}_{nk} \tau_k. \tag{7}$$

This result is in some ways reminiscent of the phenomenon of motional averaging,[5] but here it is the spin operators rather than the lattice operator which are time dependent, and the "fluctuations" of $\mathcal{H}$ are periodic rather than random. More importantly, the average Hamiltonian obtained is under the control of the experimenter through choice of the pulses P_k and weighting factors τ_k/T.

An interesting case arises when $\mathcal{H}$ consists of static, homonuclear, dipolar interactions