PHYSICAL REVIEW LETTERS

VOLUME 23 | 11 AUGUST 1969 | NUMBER 6

IDENTIFICATION OF A JAHN-TELLER TUNNELING LEVEL

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(Received 9 June 1969)

The effects of a nearby tunneling level are observed in the ESR of the lowest optically excited Γ_8 level of Eu^{2+} in CaF_2. These results are substantiated by the previous observation of this level by other workers using piezospectroscopic techniques. The ESR evidence for such a level can be found in other systems and may allow its approximate position and symmetry to be determined in these cases.

The apparent effects of dynamical Jahn-Teller distortions have been observed in ESR spectra in several situations recently. In particular, Cu^{2+} in MgO[1] or CaO, Sc^{2+} in CaF_2 or SrF_2,[2] and La^{2+} in $SrCl_2$[3] all have d^1 or d^9 ground-state configurations in which the cubic crystalline field leaves an e_g orbital lowest. The first two examples involving Cu^{2+} and Sc^{2+} exhibit coexisting isotropic and anisotropic spectra at temperatures in the region 1.5-4.2°K. The isotropic spectrum was interpreted as arising from transitions within a nearby vibronic singlet tunneling level a few cm^{-1} above the ^2E orbital doublet ground state. This interpretation has been disputed,[3,4] at least for Sc^{2+} and La^{2+}, and an alternative explanation proposed involving an averaging of the anisotropic ^2E spectrum at some lattice sites by rapid spin-lattice relaxation between the random strain-split[4] orbital components of the ^2E ground state. The purpose of this Letter is to show, from a specific example, how the presence of such a tunneling level can be further tested through observation of an asymmetric random strain broadening of the resonance transitions in the orbital components of the e_g level.

An e_g orbital in sixfold or eightfold cubic coordination is split by interaction with E_g distortions of the ligands, and the energy of the entire complex can be lowered by any linear combination of the degenerate E_g mode distortions, Q_ϵ and Q_θ.[5] If the electron-lattice coupling is linear in the Q_i, the resulting energy change depends only on the modulus $(|Q_\epsilon|^2+|Q_\theta|^2)^{1/2}$ of the distortion, and the solutions of the coupled electronic-vibrational Hamiltonian are rotations in the space of Q_ϵ and Q_θ. Interactions nonlinear in the Q_i (due to anharmonicity or nonlinear electron-lattice coupling) tend to stabilize a distortion, elongation or compression, along the cube axes. This leads to hindered rotations in Q_i space which are sometimes discussed in terms of tunneling between the equivalent cube-axis distortions. A doublet E and a singlet (A_1 or A_2) levels are the lowest vibronic levels of the complex, the E level being the ground state. We adopt the tunneling description of these levels which then have wave functions[2]

$$\psi(A) = (3-6\gamma)^{-1/2}[\chi_x\psi_x+\chi_y\psi_y+\chi_z\psi_z], \quad (1a)$$

$$\psi_\epsilon(E) = (2+2\gamma)^{-1/2}[\chi_x\psi_x-\chi_y\psi_y], \quad (1b)$$

$$\psi_\theta(E) = (6+6\gamma)^{-1/2}[2\chi_z\psi_z-\chi_x\psi_x-\chi_y\psi_y], \quad (1c)$$

where χ_x is a vibrational wave function for the complex localized in a distortion along the x axis,

ψ_x is the associated d orbital, and $\gamma = \langle \chi_x | \chi_y \rangle$. $\psi_x \simeq 2x^2 - r^2$ (or $y^2 - z^2$) if A_1 (or A_2) lies lower corresponding to stabilization by the nonlinear coupling of an elongation (or a compression) along cube axes. The separation between E and the lower A level is denoted by 3Γ and decreases as either the linear or nonlinear coupling increases. The spin degeneracy of each level is omitted in (1).

The usual situation encountered in the E level is that of a random strain splitting of the vibronic components in (1b) and (1c) that is much larger than their stabilization by the magnetic field. In the absence of any coupling to the A level the magnetic splitting of the spin degeneracy of the resulting vibronic components is given by[4,6]

$$g = g_1 \pm \tfrac{1}{2} g_2 [\cos\varphi (3n^2 - 1) + \sqrt{3} \sin\varphi (l^2 - m^2)], \quad (2)$$

where $\tan\varphi = e_\epsilon / e_\theta$, the e_i are the E-symmetry random strain components at a site, and (l, m, n) give the orientation of the applied field relative to cube axes (x, y, z). The resulting ESR spectrum is a "powder pattern" with sharp edges at the positions of extrema in the magnetic splitting[4,6] as shown in Fig. 1(a).

There exist matrix elements of strain and anisotropic terms in the spin Hamiltonian which

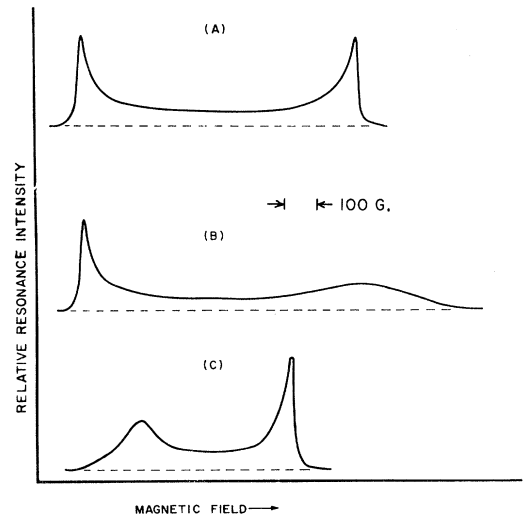

FIG. 1. Line shapes for ESR transitions within the strain-split orbital states of the E level of $CaF_2:Eu^{2+}$. (a) Expected shape for $H \parallel [001]$ assuming no coupling to the nearby tunneling level; (b), (c) form of observed spectra at 24 GHz for $H \parallel [001]$ and $H \parallel [110]$, respectively. The actual resonances are complicated by the six-line hyperfine structure for each of the Eu^{151} and Eu^{153} isotopes. The observed resonances are fitted by $g_1 = 3.68$, $g_2 = 0.40$ in Eq. (2). These parameters are identical with those found for $SrF_2:Eu^{2+}$.

selectively couple the components of E and A. We have observed the effects of such interactions in the optically detected[7] resonance spectrum of the lowest $4f^6 5d$ level[8] of Eu^{2+} at a Ca^{2+} site in CaF_2. The magnetic and strain splittings of this level are qualitatively similar to those of a $5d^1$ (e_g) level, although the magnitude (but not the symmetry) of the magnetic splitting is affected by exchange polarization of the $4f^6$ core which has a nonmagnetic singlet ground state. In Figs. 1(b) and 1(c) are shown the shapes of the resonances observed in this level for applied-field orientations near the [110] and [001] axes in a $(1\bar{1}0)$ plane. In this plane the two vibronic functions (1b) and (1c) are eigenfunctions of the Zeeman interaction and have resonance transitions at the sharp extremes in Fig. 1(a). They are also eigenfunctions of a random strain distortion e_θ along [001] corresponding to $\varphi = 0$ in (2). Other strain configurations stabilize a linear combination of these wave functions and their resonances are nearer the center of the broadened doublet. This one-to-one relationship between resonance position and local strain configuration allows, in effect, the performance of a uniaxial strain experiment using the random strains in the sample.

It can be shown that only the ψ_θ (or ψ_ϵ) E level in (1) is coupled to A_1 (or A_2) by an e_θ strain, and we assume that its resonance transition is at the high-field side for $H \parallel [001]$ and at the low-field side for $H \parallel [110]$. In most Jahn-Teller systems, the position of this resonance could be calculated directly, but in this case the coupling to the $4f^6$ levels may reverse the splittings. The broadening of this transition results from perturbations of the type, assuming A_1 is lowest,

$$h\Delta\nu = \langle E_\theta | \mathcal{H}_z | A_1 \rangle \langle A_1 | V(E) e_\theta | E_\theta \rangle / 3\Gamma, \quad (3)$$

where $\mathcal{H}_z$ is the Zeeman interaction and $V(E)$ is the reduced matrix element for strain coupling by E-symmetry strains in the e_g orbital. The observed broadening is linear in $|\Delta g \beta H + \Delta A m_I|$ as expected from (3) where Δg and ΔA are the anisotropic components of g and the hyperfine constant A.

Using the width of the 4130-Å zero-phonon optical transition to the E level measured for this sample and corrected for coupling to A_1 strains,[9] to obtain an average $V(E)e_\theta$, the measured broadening gives $3\Gamma = 18 \pm 4$ cm^{-1} for $CaF_2:Eu^{2+}$. Our ESR measurements on this Eu^{2+} level in SrF_2 show similar and more severe broadening effects, but the signals are much weaker due to rapid relaxation, prohibiting meaningful esti-

mates of 3Γ, which must be considerably smaller than for CaF_2.

The zero-phonon transition from the orbital singlet ground state of Eu^{2+} to the A level is expected to be forbidden in absence of strain due to vanishing vibrational overlap with the singlet ground state.[5] However, this tunneling level has been observed optically by Kaplyanskii and Przevuskii[9] who measured the stress splitting of the E level of Eu^{2+} in CaF_2 and SrF_2 and found a weak transition, with strain-dependent intensity, to an unidentified level at 15.3 and 6.5 cm^{-1}, respectively, above the E level. In SrF_2 this level is close enough to anticross with the split components of E and borrow transition moment from them when stress is applied along [001] and [110]. The magnitude of the stress coupling of this level to E and the diagonal matrix elements of the stress within the E level can be related by the wave functions in (1). The result is, for $\gamma \ll 1$, $\langle A_1|e_\theta|E_\theta\rangle/\langle E_\theta|e_\theta|E_\theta\rangle = \sqrt{2}$. This ratio was used in Eq. (3) to obtain 3Γ from the ESR data. In the opposite limit where the second-order coupling is negligibly small so that the levels are almost purely rotational, this ratio can be shown to be unity. The measured strain data[9] give a ratio of 1.49 for which we estimate a possible error of up to 10% from the spread in the published data. It thus appears that the optical data for SrF_2:Eu^{2+} are better fitted by the tunneling model of Eq. (1). On the basis of the evidence we propose that the work of Ref. 9 is the first conclusive optical observation of such a tunneling level.

Selective broadening similar to that of Fig. 1 is also found in the resonances of Cu^{2+} in MgO and CaO[10] and possibly in Sc^{2+} in CaF_2[2] and can be used to estimate the position and symmetry of the tunneling level in these cases.

The author is grateful to Dr. T. A. Fulton, Dr. S. Geschwind, Dr. F. S. Ham, and Dr. D. C. Krupka for helpful comments and to J. L. Davis for technical assistance.

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DIRECT OBSERVATION OF THE LORENTZIAN LINE SHAPE AS LIMITED BY QUANTUM PHASE NOISE IN A LASER ABOVE THRESHOLD*

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The quantum-phase-noise–limited Lorentzian power spectral densities of $Pb_{0.88}Sn_{0.12}Te$ diode lasers were directly measured. Linewidths as narrow as 54 kHz were observed and quantitatively analyzed. The predicted inverse dependence of linewidth on laser power was also demonstrated. These results were obtained from the beat-note spectra produced by heterodyning the diode laser, operating cw at 10.6 μm, with a stable, single-frequency CO_2 gas laser.

This paper describes measurements of the output power spectra of $Pb_{0.88}Sn_{0.12}Te$ diode lasers above the threshold of oscillation.[1] The observed spectral widths as narrow as 54 kHz were dominated by fluctuations of the oscillation phase due to spontaneous emission (quantum phase noise), rather than extraneous modulation caused by environmental disturbances.

Theoretical results[2] for amplitude and phase noise in lasers have been generally derived from solutions of equations for a van der Pol oscillator with appropriate noise sources. Thus it was predicted that the output power spectrum of a single-frequency laser may be approximated by a Lorentzian profile whose width is given by

$$\Delta\nu = A\alpha\pi h\nu_0(\Delta\nu_c)^2/P, \quad (1)$$

where h, ν_0, $\Delta\nu$, and $\Delta\nu_c$ denote Planck's constant, the laser center frequency, and the full widths between half-power points of the laser out-