Computing $g(M_I)$ to third order (neglecting all but the three largest terms) one obtains Eq. (B5):

$$g(M_I) = -\frac{A_1^2}{4g_1\mu_B H} M_I + \left(\frac{3}{2}\frac{qA_2qQ}{A_1} + \frac{3}{8}\frac{A_1^3}{(g_1\mu_B H)^2}\right) \times [I(I+1) - M_I^2] \ . \qquad \text{(B5)}$$

Substituting Eq. (B5) into Eq. (B4) and simplifying, one obtains Eq. (9).

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Integrated-Magnetic-Resonance-Absorption Dependence on the Resonance Frequency and Spin-Hamiltonian Parameters for a Non-Kramers Doublet

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A model is derived for the frequency dependence of the magnetic-resonance integrated absorption for non-Kramers doublets. The integrated electron-paramagnetic-resonance absorption is shown to be independent of the resonance frequency through second order in the crystal field perturbation for transitions within a non-Kramers doublet. This in in large contrast with the frequency-squared dependence in first order for the acoustical-paramagnetic-resonance and the paraelectric-resonance integrated intensities. For the cases of the acoustical-paramagnetic-resonance and paraelectric-resonance integrated absorptions as functions of the resonance frequency squared, the intercept is shown to depend explicitly upon the zero-field splitting of the states of the doublet. The slopes determine the relative strengths of the acoustical-paramagnetic-resonance and paraelectric-resonance absorptions. A transformation is derived from the spin Hamiltonian which transforms the absorption line shape from that in field variation (H) to that in frequency variation (ν). The transformation shows that the line shapes in the two cases are basically different when the absorption line is broadened by crystal field perturbations. The model predicts that the integrated intensity in frequency variation is greater than that in field variation by a term proportional to the average value of the square of the crystal field perturbations.

I. INTRODUCTION

Non-Kramers paramagnetic-ion impurities in diamagnetic-crystal host lattices have been studied using electron-spin-resonance[1] (EPR) techniques. The applications of acoustical-paramagnetic resonance (APR) also include studies on the non-Kramers impurities.[2] In fact, the APR techniques are particularly adaptive to the study of transitions within the non-Kramers doublets since the quadrupole transitions are allowed[3] for APR but forbidden for EPR. In some cases both EPR and APR have been readily observed. The transition probability for the EPR in such cases is significant because of mixing of the states of the doublet by local crystal field perturbations.[4]

The nature of the mechanism for the phonon-induced-APR and the photon-induced-EPR transition probability is quite different.[3] If the absorption line shapes are broadened by local crystal field perturbations, as is the case for non-Kramers ions,[1,2] the shapes are expected to be quite different for EPR and APR in the same sample.[4] Line-shape calculations have recently been carried out and applied for several cases.[5] It is shown in this paper that the integrated intensity of the absorption as a function of resonance frequency also gives a meaningful comparison between the APR- and EPR-resonance absorption, and a model is presented for the case of a non-Kramers ion of effective spin one with an axial crystalline-field environment and simple hyperfine coupling.

In addition, the integrated absorption as a function of frequency is derived for the case of paraelectric-resonance (PER) absorption. This kind of absorption is possible when the impurity site lacks inversion symmetry.[6] Electric-field-induced transitions of this type for a non-Kramers doublet have been reported previously.[6-9]

In the development which is to follow, we assume that the resonance absorption line is inhomogeneously broadened by local crystal field perturbations caused by crystal imperfections. In addition, we consider contributions to the broadening from isotropic-hyperfine or transferred-hyperfine interactions. In Sec. II A expressions are developed for the integrated intensity for APR and PER in terms of the resonance-frequency and spin-Hamiltonian parameters. The slope and intercept are obtained from the model for the integrated intensity vs the square of the resonance frequency and can be compared with the results of an experiment. Such a comparison can give pertinent information concerning the spin-lattice coupling and the transition probabilities. It is shown that the integrated intensity for APR and PER varies as the square of the resonance frequency, and this is compared with the frequency invariance, through second order in the crystal field perturbations, of the EPR integrated intensity. In Sec. II B a transformation is derived from the spin Hamiltonian, which transforms the absorption line shape from magnetic field variation (H) with the frequency ν held constant to that for frequency variation (ν) with the magnetic field H held constant. It will be shown from the transformation that the line shapes in the two cases are basically different. It is also shown that the integrated intensity in (ν) is different from that in (H) because of the difference in the line shape in the two cases. This is shown to arise explicitly from the crystal field perturbations and the hyperfine splitting.

II. THEORY

A. Integrated Intensity

We consider the condition that the resonance absorption is inhomogenously broadened by crystal field perturbations and hyperfine or transferred-hyperfine interaction. The paramagnetic absorption of acoustic energy (APR) for a homogeneous component of the resonance absorption is given by the relation

$$Ph\nu n = \alpha \epsilon v \quad , \tag{1}$$

where α is the paramagnetic-absorption coefficient at the Larmor frequency ν, ϵ is the energy density of the phonon field of frequency ν, v is the velocity of sound in the material, n is the population difference between the levels, and, using Fermi's golden rule,

$$P = \frac{1}{\hbar^2} \left| \langle i | \mathcal{H}_{\mathrm{SL}} | f \rangle \right|^2 g(\nu) \tag{2}$$

is the probability per unit time that a transition occurs. Here, $g(\nu)$ is the density of final states for the homogeneous component and $\mathcal{H}_{\mathrm{SL}}$ is the appropriate spin-lattice Hamiltonian[3] for the phonon field at the resonance frequency which couples the initial state i with the final state f.

For the high-temperature approximation, i.e., for $h\nu / KT \ll 1$, and assuming that the frequency ν does not change significantly over the range in which the density of final states for a homogeneous component, $g(\nu)$, is significantly different from zero,[10] the paramagnetic absorption of acoustic energy is

$$\alpha(\nu' - \nu) = K(N) \nu'^2 g(\nu' - \nu) \quad . \tag{3}$$

Here ν' is the "center" frequency of the homogeneous absorption and

$$K(N) = \frac{4\pi^2 N}{\rho v^3 KT} \Big(\sum_{ijkl} G'_{ijkl} \hat{e}_{kl} \Big)^2 \quad , \tag{4}$$

where N is the number of paramagnetic absorbers per unit volume for the homogeneous component, ρ is the density of the material, G'_{ijkl} are the spin-lattice coupling parameters which are related to the spin-lattice coupling coefficients[11] by numerical factors which depend upon the particular spin-operator combination involved, and $\hat{e}_{kl}$ are the strains per unit strain.

The resonance frequency ν' is identified with a homogeneous ensemble of paramagnetic absorbers and can be evaluated in terms of the interaction parameters from the eigenvalues of the appropriate spin Hamiltonian for the given ensemble. The total integrated intensity J for the inhomogenously broadened absorption line is obtained by summing the integrated absorption over all the ensembles of the system,[12]

$$J=\sum_i \int_0^\infty \alpha_i(\nu_i-\nu)\,d\nu$$

$$=\sum_i K_i(N_i)\int_0^\infty \nu_i^2 g_i(\nu_i-\nu)\,d\nu \quad , \tag{5}$$

where ν_i and N_i are the resonance frequency and number of absorbers per unit volume in the ith ensemble, respectively, and $K_i(N_i)$ is given for each ensemble by Eq. (4). We have the normalization condition for the density of final states for each ensemble, i.e.,

$$\int_0^\infty g_i(\nu_i-\nu)\,d\nu=1 \quad . \tag{6}$$

Then we get

$$J=\sum_i K_i(N_i)\nu_i^2 \quad . \tag{7}$$

To evaluate the terms in Eq. (7), in terms of the spin-Hamiltonian parameters, we must solve the eigenvalue problem for each ensemble of the system. As an example we consider the effective spin-one Hamiltonian for a non-Kramers doublet with axial crystal field symmetry,[4,13]

$$\mathcal{H}=\tfrac{1}{2}\Gamma S_z+D[S_z^2-\tfrac{1}{3}S(S+1)]$$

$$+\Delta S_+^2+\Delta^* S_-^2+(AS_z-\mu_N H_z)I_z \quad , \tag{8}$$

where

$$\Gamma=2g\mu_B H_z \tag{9}$$

and H_z is the axial component of the magnetic field, A the isotropic hyperfine coupling parameter, and Δ can be written explicitly in terms of the local-strain components and spin-lattice coupling coefficients.[4,5] Thus, Δ is a distribution function in the local crystal field perturbations,[4] and therefore each value of $|\Delta|^2$ characterizes an ensemble of absorbers of the system. If the axial distortion is large compared to the Zeeman splitting, i.e.,

$$|D|/\Gamma \gg 1 \quad ,$$

then, neglecting any coupling of the singlet into the states of the doublet defined by Eq. (8),[4] the characteristic determinant of Eq. (8) takes the form[5]

$$\begin{vmatrix} a_{11}-\lambda & 0 & a_{13} \\ 0 & a_{22}-\lambda & 0 \\ a_{31} & 0 & a_{33}-\lambda \end{vmatrix}=0 \quad . \tag{10}$$

The resonance condition for an ensemble for transitions within the non-Kramers doublet defined by Eq. (8) is therefore[14]

$$\mathcal{E}=[(\Gamma+Am_I)^2+\Omega^2]^{1/2} \quad , \tag{11}$$

where

$$\mathcal{E}=h\nu, \quad \Omega^2=16|\Delta|^2 \quad . \tag{12}$$

If we use Eq. (11) in Eq. (7), the resulting expression for the integrated APR intensity for a given hyperfine component of the system, characterized by the nuclear-spin quantum number m_I, is

$$J_{m_I}=p_{m_I}\sum_i \frac{K(N_i)}{h^2}[\mathcal{E}_0^2+2m_I A\mathcal{E}_0+m_I^2A^2+\Omega_i^2] \quad . \tag{13}$$

Here p_{m_I} is the normalized hyperfine probability, i.e.,

$$p_{m_I}=\frac{n!}{x_{m_I}!(n-x_{m_I})!}\bigg/\sum_{m_I=-I}^{m_I=I}\frac{n!}{x_{m_I}!(n-x_{m_I})!} \quad . \tag{14}$$

In this expression x is the number of ways in which n objects can be arranged within the spin manifold m_I. Also,

$$\mathcal{E}_0=h\nu_0 \tag{15}$$

is the unperturbed Zeeman splitting. We use Eq. (4) to perform the indicated summation in Eq. (13) to give the resulting expression

$$J_{m_I}=p_{m_I}K(N_T)\left(\nu_0^2+\frac{2m_I A}{h}\nu_0+\frac{1}{h^2}(m_I^2A^2+\langle\Omega^2\rangle)\right) \cdot \tag{16}$$

Here

$$\langle\Omega^2\rangle=\sum_i N_i|\Delta_i|^2/N_T \tag{17}$$

is the ensemble average of the crystal field perturbations and

$$N_T=\sum_i N_i \tag{18}$$

is the total number of paramagnetic absorbers in the system. The total APR integrated intensity for the system is obtained by summing Eq. (16) over all the hyperfine components to give the total integrated intensity J for the inhomogeneously broadened line, i.e.,

$$J=\sum_{m_I=-I}^{m_I=I} J_{m_I} \quad ,$$

$$J=K(N_T)\left(\nu_0^2+\tfrac{1}{3}I(I+1)(2I+1)\frac{A^2}{h^2}p_I+\frac{\langle\Omega^2\rangle}{h^2}\right) \quad (\text{APR}) \quad , \tag{19}$$

where

$$p_I=\sum_{m_I=-I}^{m_I=I} m_I^2 p_{m_I}\bigg/\sum_{m_I=-I}^{m_I=I} m_I^2 \tag{20}$$

and

$$\sum_{m_I=-I}^{m_I=I} m_I^2=\tfrac{1}{3}I(I+1)(2I+1) \quad . \tag{21}$$

In the case of electric-field-induced transitions in PER, the absorption a per unit time per unit volume[15] is

$$a = \tfrac{1}{3}\pi^2 (e^2 |x_{ab}|^2) \nu E_{\mathrm{rf}}^2 \quad , \tag{22}$$

where the term in parenthesis is the electric-dipole moment of the ion and E_{rf} is the applied-rf electric-field intensity in the sample. The expression analogous to Eq. (1), in this case the paraelectric absorption of energy from the radiation field, PER, is

$$a = Ph\nu n \; . \tag{23}$$

If we assume that the undistorted crystal field symmetry is C_{3v}, the PER transition probability has been shown to be given, through second order in $|\Delta|/h\nu_0$, by[5]

$$P = \frac{1}{\hbar^2} R^2 E_{\mathrm{rf}}^2 \left(1 - \frac{8}{(h\nu_0)^2}\left(|\Delta|^2 - \sigma^2 + \rho^2\right)\right) g(\nu) \; . \tag{24}$$

Here R is the spin–electric-field coupling parameter, Δ is defined by Eq. (8), and σ and ρ are the real and complex parts of Δ, respectively.[4,5] The rf-field-normalized integrated intensity J' is

$$J' = \textstyle\sum_i \int a_i'(\nu_i - \nu)\, d\nu \quad , \tag{25}$$

where

$$a' = a / E_{\mathrm{rf}}^2 \quad . \tag{26}$$

If Eqs. (24) and (26) are used in Eq. (23), the indicated integrations and summation can be performed in Eq. (25) using the same assumptions and procedure which led to Eq. (19). The result is, neglecting terms of order $|\Delta|^2/(h\nu_0)^2$ and $A^2/(h\nu_0)^2$ and higher,

$$J' = K'(N_T)\left(\nu_0^2 + \tfrac{1}{3} I(I+1)(2I+1)\frac{A^2}{h^2} p_I + \frac{1}{2}\frac{\langle \Omega^2\rangle}{h^2} - \frac{8}{h^2}\left(\langle \rho^2\rangle - \langle \sigma^2\rangle\right)\right) \quad (\mathrm{PER}) \quad , \tag{27}$$

where

$$K'(N_T) = \frac{2\pi^2}{kT} R^2 N_T \quad , \tag{28}$$

$\langle\Omega^2\rangle$ and p_I are given by Eqs. (17) and (20), respectively, and $\langle\rho^2\rangle$ and $\langle\sigma^2\rangle$ are defined in the same way as $\langle\Omega^2\rangle$, i.e., Eq. (17). It is to be noted that the last term in Eq. (27) is zero in some cases, from symmetry considerations. In particular, it has been shown[5] that for a system such as $\mathrm{CaF_2{:}U^{4+}}$ where the axial distortion is along [111], $\langle\sigma^2\rangle$ and $\langle\rho^2\rangle$ are equal. When this is the case, Eqs. (19) and (27) are quite similar.

We turn attention now to the case for magnetically induced transitions, EPR. The absorption per unit time per unit volume B in this case, the counterpart of Eqs. (1) and (23), is

$$B = Ph\nu n \; . \tag{29}$$

For this case,[16]

$$B = 4\pi H_{\mathrm{rf}}^2 \nu \chi'' \quad , \tag{30}$$

where χ'' is the complex component of the susceptibility and H_{rf} is the magnetic field component of the applied rf field in the sample. The transition probability P is again given by Eq. (2). If we assume that the unperturbed symmetry at the impurity site is either D_{3d} or C_{3v}, the matrix element in Eq. (2) which couples the initial and final states is, through second order in the crystal field perturbations,[5]

$$|\langle i|\mathcal{H}_{\mathrm{rf}}|f\rangle|^2 = 16 g^2 \mu_B^2 H_{\mathrm{rf}}^2 \left[|\Delta|^2/(h\nu_0)^2\right] \; . \tag{31}$$

The crystal field perturbation Δ is again defined by Eq. (8) and is the same distribution function in the local crystalline field which appeared in the previous two cases. Using Eq. (31) in Eq. (29) and under the same assumptions made in the previous two cases, the expression for the absorption B for a given ensemble of the system becomes

$$B(\nu' - \nu) = K''(N)\nu'^2 g(\nu' - \nu)\left[|\Delta|^2/(h\nu_0)^2\right] \; , \tag{32}$$

where

$$K''(N) = (32\pi^2 N/KT) g^2 \mu_B^2 H_{\mathrm{rf}}^2 \quad . \tag{33}$$

The rf-magnetic-field-normalized integrated intensity for the system, J'', is given by

$$J'' = \sum_i \frac{K''(N_i)}{H_{\mathrm{rf}}^2} \nu_i^2 \frac{|\Delta_i|^2}{(h\nu_0)^2} \int g_i(\nu_i - \nu)\, d\nu \quad . \tag{34}$$

Using Eqs. (6), (11), and (17) in Eq. (34), the integrals and summation are performed in the same way as presented in the previous cases. If terms on the order of $|\Delta|^2/(h\nu_0)^2$ and $A^2/(h\nu_0)^2$ and higher are neglected in the result, we obtain the expression for the integrated EPR intensity for the system,

$$J'' = K_n''(N_T)(\langle\Omega^2\rangle/16h^2) \quad (\mathrm{EPR}) \; , \tag{35}$$

where

$$K_n'' = K''/H_{\mathrm{rf}}^2 \tag{36}$$

is independent of the applied radiation density. Equation (35) differs markedly from Eqs. (19) and (27) in that it is independent of the resonance frequency ν_0. This is a direct consequence of the fact that the transition within a non-Kramers doublet is a magnetic-quadrupole transition ($|\Delta M_S| = 2$) and is therefore a forbidden transition in EPR. Transitions occur only when the states of the doublet are mixed by perturbations which lower the symmetry to lower than axial. Thus, there are no first-order contributions to the transition probability as there are for the APR and PER, and the second-order term in effect cancels the frequency dependence of the integrated intensity.

B. Transformation of Line Shape

Magnetic-resonance observations are most often performed in the microwave region, and therefore the applied dc field H is normally varied, while the unperturbed Zeeman frequency ν_0 (klystron frequency in EPR) is constant. The line shape obtained under this condition will be called H variation, and designated as (H). The resulting equations for the integrated intensity, Eqs. (19), (27), and (35), correspond to the condition where the applied dc field H is fixed and the rf frequency ν is varied. This condition will be called ν variation and designated as (ν). The calculations were necessarily done in (ν) because the straightforward counterpart in (H) leads to difficulties in the theory; in particular, the density of final states is not strictly normalizable independent of the unperturbed splitting ν_0.[12]

To compare experimental results with Eqs. (19), (27), and (35), it is necessary to obtain a mapping of the line shape from (H) to (ν). It is the purpose of this section to develop this transformation within the context of the present model. This is done directly from the eigenvalues of the spin Hamiltonian. Consistent with the definitions given by Eqs. (9) and (12), we replace (H) by (Γ) and (ν) by $(\mathcal{E})$. Thus, Eq. (11) in $(\mathcal{E})$ can be written as

$$\Omega^2(\mathcal{E}) = \mathcal{E}^2 - (\mathcal{E}_0 + Am_I)^2 \quad , \tag{37}$$

where $\mathcal{E}_0$ is given by Eq. (15) and is the unperturbed Zeeman splitting. Similarly, in (Γ), Eq. (11) can be written as

$$\Omega^2(\Gamma) = \mathcal{E}_0^2 - (\Gamma + Am_I)^2 \ . \tag{38}$$

Since a particular ensemble of the system is characterized by a particular value of Ω^2, the mapping of the resonance, or "center," absorption of each ensemble between (Γ) and $(\mathcal{E})$ is obtained by equating Eqs. (37) and (38),

$$\mathcal{E}^2 = 2\mathcal{E}_0^2 + 2m_I A(\mathcal{E}_0 - \Gamma) - \Gamma^2 \quad . \tag{39}$$

Thus, if the ith ensemble of absorbers (corresponding to $\Omega = \Omega_i$) is resonant at Γ_i in (Γ) when the unperturbed Zeeman splitting is $\mathcal{E}_0$, then from Eq. (39) we have its corresponding resonance at $\mathcal{E}_i$ in $(\mathcal{E})$ when the unperturbed Zeeman splitting is the same. If the homogeneous components of the resonance absorption are completely unresolved in the line shape, each point on the resonance line can be considered as the resonance position corresponding to some ensemble and hyperfine component. Thus, for each hyperfine component of the absorption line, Eq. (39) is the mapping between Γ in (Γ) and $\mathcal{E}$ in $(\mathcal{E})$.

We proceed now with the derivation of the transformation for the intensity of the absorption. Since this depends explicitly on the nature of the probe, we shall consider only the case for APR. The procedure carries over identically to the other two cases and these will not be considered here.

We begin with the following expressions for the intensity of the absorption in (Γ) and $(\mathcal{E})$, respectively, for each hyperfine component, assumed resolved:

$$J_{(\Gamma)}(\Gamma) = p_{m_I} \sum_i n_i \alpha'(\Gamma_i' - \Gamma) \tag{40}$$

and

$$J_{(\mathcal{E})}(\mathcal{E}) = p_{m_I} \sum_j n_j \alpha''(\mathcal{E}_j' - \mathcal{E}) \quad , \tag{41}$$

where n_i in Eq. (40) is the number of absorbers in the ith ensemble, the ensemble belonging to the resonance splitting Γ_i', and α' in this case is the absorption per ion. The obvious analogy holds for Eq. (41). The terms in the arguments, Γ_i' and $\mathcal{E}_i'$, are connected by the point transformation, Eq. (39), and Γ and $\mathcal{E}$ are corresponding points in (Γ) and $(\mathcal{E})$, respectively, on the resonance-absorption line. Consistent with our assumption that the homogeneous components of the absorption line are unresolved, within each hyperfine component Eqs. (40) and (41) become, in the unresolved limit

$$J_{(\Gamma)}(\Gamma) = p_{m_I} \int_{\mathcal{E}_0}^{0} \alpha'(\Gamma' - \Gamma) q(\Gamma') d\Gamma' \tag{42}$$

and

$$J_{(\mathcal{E})}(\mathcal{E}) = p_{m_I} \int_{\mathcal{E}_0}^{\bar{E}} \alpha''(\mathcal{E}' - \mathcal{E}) p(\mathcal{E}') d\mathcal{E}' \ , \tag{43}$$

where $\bar{E} = (2\mathcal{E}_0^2 + 2m_I A\mathcal{E}_0)^{1/2}$ and $q(\Gamma')$ and $p(\mathcal{E}')$ are the density of absorbers in (Γ) and $(\mathcal{E})$, respectively. We look for the transformation that transforms Eq. (42) into Eq. (43). With this purpose in mind, we transform Eq. (43) from $(\mathcal{E})$ to (Γ).

If $n(\Omega)d\Omega$ is the number of absorbers between Ω and $\Omega + d\Omega$ [$n(\Omega)$ Gaussian for random perturbations], then

$$q(\Gamma) = n(\Omega(\Gamma)) \frac{d\Omega}{d\Gamma} \tag{44}$$

and

$$p(\mathcal{E}) = n(\Omega(\mathcal{E})) \frac{d\Omega}{d\mathcal{E}} \ . \tag{45}$$

Thus we have

$$p(\mathcal{E}) = q(\Gamma(\mathcal{E})) \frac{d\Gamma}{d\mathcal{E}} \ . \tag{46}$$

Consistent with the assumption made in Eq. (3) that $\mathcal{E}$ does not change significantly for an ensemble of the system over the range in which the density of final states $g(\nu' - \nu)$ is appreciably different from zero, the absorption functions α' and α'' in Eqs. (42) and (43) are written in the same form as Eq. (3),

$$\alpha'(\Gamma' - \Gamma) = K'\mathcal{E}_0^2 g'(\Gamma' - \Gamma) \tag{47}$$

and

$$\alpha''(\mathcal{E}' - \mathcal{E}) = K'\mathcal{E}^2 g(\mathcal{E}' - \mathcal{E}) \ , \tag{48}$$

where

$$K' = (K/N) p_{m_I} \tag{49}$$

and K is given by Eq. (4). Consistent with the assumption used in representing the transition probability P according to Fermi's golden rule, we relate the density of final states g' in (Γ) to that in $(\mathcal{E})$, g, by the Jacobian of the transformation,

$$\mathcal{J}\begin{pmatrix}\Gamma \\ \mathcal{E}\end{pmatrix} \quad ,$$

derived from Eq. (39), i.e., in the same way as p and q in Eq. (46). From Eq. (39) we get

$$\mathcal{J}\begin{pmatrix}\Gamma \\ \mathcal{E}\end{pmatrix} = - \frac{[2\mathcal{E}_0^2 + 2m_I A(\mathcal{E}_0 - \Gamma) - \Gamma^2]^{1/2}}{m_I A + \Gamma} \ . \tag{50}$$

If we use Eqs. (39), (46)–(48), and (50) in Eqs. (42) and (43), the result for each hyperfine component, assumed resolved, is

$$J_{(\Gamma)}(\Gamma) = K'\mathcal{E}_0^2 \int g'(\Gamma' - \Gamma) q(\Gamma') d\Gamma' \tag{51}$$

and

$$J_{(\mathcal{E})}(\Gamma) = K' \int \frac{[2\mathcal{E}_0^2 + 2m_I A(\mathcal{E}_0 - \Gamma') - \Gamma'^2]^{3/2}}{m_I A + \Gamma'} g'(\Gamma' - \Gamma) q(\Gamma') d\Gamma' \ . \tag{52}$$

We note that Eqs. (51) and (52) are in the form of convolution products if the density of final states, g', is taken to be zero for $\Gamma < 0$, $\Gamma > \mathcal{E}_0$. Thus, taking the transform of Eq. (51) and making use of the convolution theorem,[17]

$$T[J_{(\Gamma)}(\Gamma)] = K\mathcal{E}_0'^2 T[g'] T[q] \ . \tag{53}$$

Similarly for Eq. (52),

$$T[J_{(\mathcal{E})}(\Gamma)] = K' T[g'] T[F] \ , \tag{54}$$

where

$$F = \frac{[2\mathcal{E}_0^2 + 2m_I A(\mathcal{E}_0 - \Gamma') - \Gamma'^2]^{3/2}}{m_I A + \Gamma'} q(\Gamma') \ . \tag{55}$$

We may combine Eqs. (53) and (54) to get

$$T[J_{(\mathcal{E})}(\Gamma)] = \frac{1}{\mathcal{E}_0^2} \frac{T[F]\, T[J_{(\Gamma)}(\Gamma)]}{T[q]} \ . \tag{56}$$

Since $J_{(\Gamma)}$ is the measured intensity and F/q is determined if the hyperfine splitting A is known, the right-hand side of Eq. (56) can be determined numerically from the observed line shape in (Γ) if some assumption is made about the form for the probability density in the crystal field perturbations[12] q. Thus Eqs. (56) and (39) comprise the transformation of the line shape from (Γ) to $(\mathcal{E})$.

A particularly simple special case arises if the density of final states in Eqs. (51) and (52) is approximated as a δ function. This, of course, will be justifiable in a given situation only if the line broadening due to the crystal field perturbations is much greater than that caused by lifetime broadening. In this special case, Eqs. (51) and (52) become

$$J_{(\Gamma)}(\Gamma) \approx K'\mathcal{E}_0^2 q(\Gamma) \tag{57}$$

and

$$J_{(\mathcal{E})}(\Gamma) \approx K' \frac{[2\mathcal{E}_0^2 + 2m_I A(\mathcal{E}_0 - \Gamma) - \Gamma^2]^{3/2}}{(m_I A + \Gamma)} q(\Gamma) \tag{58}$$

for $g'(\Gamma' - \Gamma) \rightarrow \delta(\Gamma' - \Gamma)$. Thus, combining the last two expressions,

$$J_{(\mathcal{E})}(\Gamma) = \frac{[2\mathcal{E}_0^2 + 2m_I A(\mathcal{E}_0 - \Gamma) - \Gamma^2]^{3/2}}{\mathcal{E}_0^2(m_I + \Gamma)} J_{(\Gamma)}(\Gamma) \ . \tag{59}$$

So, for the δ-function approximation for the density of final states, given the line shape in (Γ), Eqs. (59) and (39) can be used to obtain the corresponding line shape in $(\mathcal{E})$ for each resolved hyperfine component.

III. DISCUSSION AND CONCLUSIONS

The integrated intensity for the APR, Eq. (19), predicts a resonance-frequency-squared dependence for transitions within a non-Kramers doublet. This dependence has been observed and reported[18] for $CaF_2:U^{4+}$. However, the results of the model indicate that the ratio of the intercept to the slope for the integrated intensity versus the square of the resonance frequency can give important information about the effects of the hyperfine coupling and average-crystal-field perturbations which occur in second order in Eq. (19). The integrated intensity given by Eq. (19) is in $(\mathcal{E})$. The corresponding integrated intensity in (Γ) is[12]

$$J_{(\Gamma)} = K(N_T)\nu_0^2 \ . \tag{60}$$

So it is seen that the integrated intensity in $(\mathcal{E})$, Eq. (19), yields more information than that in (Γ), Eq. (60). The fact that the integrated intensity in $(\mathcal{E})$ is greater than that in (Γ) is a direct manifestation of the fact that in $(\mathcal{E})$, the energy-level splittings corresponding to the various ensembles are sampled at different energy-level separations and consequently different population differences, whereas in (Γ), the energy-level splitting for each ensemble is sampled at the same energy-level separation and population difference. The ratio $\mathcal{R}$ of Eq. (19) to Eq. (60),

$$\mathcal{R} = 1 + \tfrac{1}{3} I(I+1)(2I+1) \frac{A^2}{(h\nu_0)^2} p_I + \frac{\langle \Omega^2 \rangle}{(h\nu_0)^2} , \qquad (61)$$

bears out fundamentally the predicted differences in the two types of experiment using the same probe, i.e., one in $(\mathcal{E})$ and the other in (Γ). If the concentration of impurity N_T is known, the spin-lattice coupling can be evaluated in the usual manner[19] from Eq. (60) and thus $K(N_T)$ is evaluated using Eq. (4). Then, using the transformation for the line shape, Eqs. (39) and (56) or (59), the left-hand side of Eq. (19) can be evaluated from experiment. Thus, if the hyperfine splitting A is known, the average crystal field perturbation $\langle \Omega^2 \rangle$ can be evaluated. Since $\langle \Omega^2 \rangle$ is "hidden" in the line shape for a non-Kramers doublet,[5] and therefore must be determined from the line shape itself, Eq. (19) offers a useful means for evaluating this parameter from an experimental line shape.

The line shapes in $(\mathcal{E})$ and (Γ) are fundamentally different, as indicated by the transformation equations (39) and (59). On the basis of the transformation, the shapes are expected to be very nearly the same for regions of the absorption line in the neighborhood of the undisplaced Zeeman component. However, in the low-field tail of the line, the shapes are expected to be quite different. The crystal field perturbations in this region tend to increase the amplitude and narrow the line in $(\mathcal{E})$ with respect to (Γ). This is caused by the combined effect of the increase in population difference as well as splitting in $(\mathcal{E})$ and the increase in the density of absorbers between (Γ) and $(\mathcal{E})$ as indicated by Eq. (50).

The results of the model for the integrated intensity predict a similar dependence between the APR and PER integrated intensity, Eqs. (19) and (27), as expected, since in each case the transition occurs in first order. On the basis of a development similar to that presented for the APR, the left-hand side of Eq. (27) can be evaluated from an experiment provided the coupling R is significantly different from zero and provided the energy density in the rf electric field, E_{rf}, can be evaluated in the experiment, Eq. (26). In such a case, the ratio of R to the spin-phonon coupling can be determined independently of the impurity concentration N_T by a comparison of the APR, Eq. (19), with the PER, Eq. (27), of the same sample.

A marked difference exists on the basis of the model between the EPR integrated intensity, Eq. (35), and the APR and PER integrated intensities, Eqs. (19) and (27), respectively. This is due entirely to the fact that the magnetic-quadrupole transitions occur in second order, whereas the phonon-induced and electric-dipole transitions each give a first-order contribution to the transition probability for a non-Kramers doublet. In the absence of a first-order contribution to the transition probability, the resonance-frequency-squared dependence inherent in the expression for the integrated intensity for APR and PER, Eqs. (19) and (27), in effect is "cancelled out" in the case for EPR, Eq. (35). Thus, to the same order in the crystal field perturbations, the EPR integrated intensity is expected to be independent of the resonance frequency, in large contrast to the APR and PER.

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