Anisotropy and field dependence of the electron-paramagnetic-resonance linewidth of *Ag*:Dy

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Here we report measurements of the EPR linewidths for the Γ_7 ground state of *Ag*:Dy at 9 and 35 GHz and 1.45 K. Our data show linewidth anisotropies that depend on the magnetic field. To analyze the experimental data, two models are discussed. In the first, we consider modifications of the Korringa relaxation due to the Zeeman interaction; in the second, the effect on the linewidth of random stresses in the sample using a model that preserves the full cubic local symmetry at the impurity sites. The second model is able to account for the experimental data and allows us to obtain information about the stress distribution in the sample.

I. INTRODUCTION

Earlier,[1] we reported measurements of the crystal-field and exchange parameters in *Ag*: ^{164}Dy. In that work, we concentrated on the g anisotropy at 35 GHz,[1] which was explained as being due to the mixing of excited states with the ground Γ_7 doublet due to the Zeeman interaction. An anisotropy of the linewidth was also observed at that frequency.[1]

In this paper, in an effort to understand the source of the anisotropic broadening, we have studied this phenomenon at two well-separated frequencies, 9 and 35 GHz, where we found that the linewidth anisotropy is magnetic field dependent.

We discuss two mechanisms which produce anisotropic broadening for Kramer's doublets in cubic crystals. One is the modification of the Korringa relaxation due to the Zeeman mixtures of excited levels with the ground state. The other is the presence of random stresses in the sample. This is similar to the mechanism proposed by Feher[2] and McMahon[3] for magnetic impurities in insulators and by Davidov *et al.*[4] for Dy in the metallic compound LaSb.

Comparison of our experimental data at both frequencies with the predictions of the models indicates that the residual linewidth extrapolated to 0 K is a consequence of random internal stresses in the sample. Using a simple model for the orbit-lattice interaction, we are able to characterize the distribution of these stresses.

II. EXPERIMENTAL TECHNIQUES AND RESULTS

The single crystal (the same as used previously[1]) was grown *in vacuo* by the Bridgeman technique. The silver, 99.9999%-purity Cominco, was doped with ^{164}Dy obtained via reduction of the oxide. The concentration was 220 ± 20 ppm as measured by atomic absorption and dc susceptibility.

The reflection EPR data were obtained both at 9 and 35 GHz with superheterodyne spectrometers equipped with magnets which could be rotated 180°. The temperature of the sample was determined from the vapor pressure of the surrounding liquid. Sample alignment, magnetic field, frequency measurements, and other experimental details were carried out as previously discussed.[1]

The EPR spectrum of *Ag*:Dy, both at 9 and 35 GHz, consists of a single line that is an admixture of the pure, real, and imaginary responses due to the skin effect. Excellent signal-to-noise ratios and repeated measurements allowed an accuracy in the half-linewidth ($\Delta H_{1/2}$), defined as the half-power halfwidth of the absorption part of the resonance line, of 1 G at 9 GHz and 3 G at 35 GHz. Assuming an intrinsic Lorentzian line shape, we extract the linewidth as before.[1] These linewidths are presented as a function of field orientation in the (110) crystal plane in Figs. 1 and 2. The theoretical curves shown in Figs. 1 and 2 are calcu-

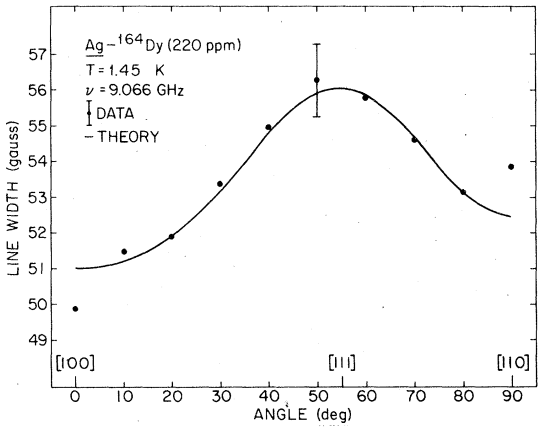

FIG. 1. Angular variation of the EPR linewidth of 220-ppm *Ag*:Dy measured at 9.06 GHz in the (110) crystal plane. The solid line is the best fitting with Eq. (1).

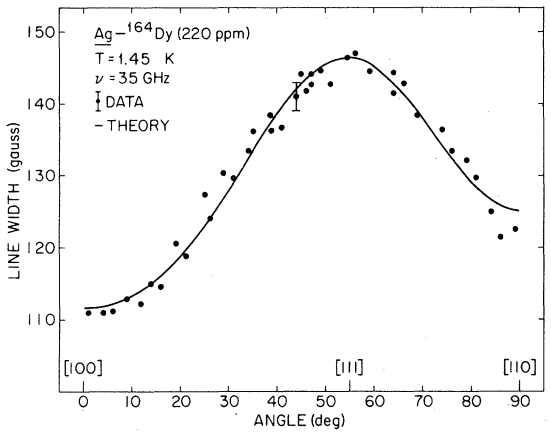

FIG. 2. Angular variation of the EPR linewidth of 220-ppm Ag:Dy measured at 35.0 GHz in the (110) crystal plane. The solid line is the best fitting with Eq. (1).

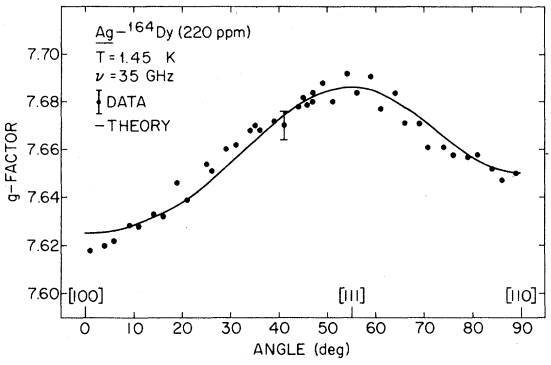

FIG. 3. Angular variation of the gyromagnetic factor g of 220-ppm Ag:Dy measured at 35.0 GHz in the (110) crystal plane. The solid line is the best fitting with Eq. (3).

lated using the best fit to the expression

$$\Delta H^2(\theta,\varphi)=A+Bf_4(\theta,\varphi)+Cf_6(\theta,\varphi), \tag{1}$$

where $f_4(\theta,\varphi)$ and $f_6(\theta,\varphi)$ are the linear combinations of spherical harmonics of fourth and sixth order having cubic symmetry

$$f_4(\theta,\varphi)=35\cos^4\theta-30\cos^2\theta+3+5\sin^4\theta\cos4\varphi, \tag{2a}$$

$$f_6(\theta,\varphi)=231\cos^6\theta-315\cos^4\theta+105\cos^2\theta-5-21(11\cos^2\theta-1)\sin^4\theta\cos4\varphi, \tag{2b}$$

In Fig. 3 we show data on the angular variation of the g factor measured at 35 GHz with the magnetic field in the (110) plane. We added new points to those previously published[1] where the curve shown is the best fit to the equation

$$g(\theta,\varphi)=A'+B'f_4(\theta,\varphi)+C'f_6(\theta,\varphi). \tag{3}$$

The fitting of the experimental values with Eqs. (1) and (3) was done using a least-squares calculation, and the results for the parameters are given in Table I. Additional, but less accurate, data on the linewidth at 35 GHz in the (001) crystal plane fit Eq. (1) well using the values given in Table I.

III. THEORY

Reflection and transmission EPR and magnetic-susceptibility measurements allow one to obtain the parameters of the crystalline field and the exchange interaction acting on Dy ions in metallic Ag.[1] The Hamiltonian for the crystalline field can be written[5] as

$$H_{CF}=B_4(O_4^0+5O_4^4)+B_6(O_6^0-21O_6^4), \tag{4}$$

where $O_n^m=O_n^m(J)$ are Stevens' operators.[5] In the notation of Lea *et al.*,[6] crystalline field parameters W and x are defined by

$$Wx=60B_4 \quad \text{and} \quad W(1-|x|)=13\,860B_6.$$

The values for the crystalline field parameters of Ag:Dy extracted from the data of Oseroff *et al.*[1] are

$$x=0.53, \quad W=0.32\ \text{cm}^{-1}$$

$$B_4=28\times10^{-4}\ \text{cm}^{-1} \quad \text{and} \quad B_6=0.11\times10^{-4}\ \text{cm}^{-1}.$$

The ground state is a Γ_7 doublet with $g=7.555$ (the experimental value is 7.65); the level scheme within the $J=\frac{15}{2}$ multiplet is shown in Fig. 4.

In order to explain the Knight shift and the Korringa rate measured from the broadening of the EPR line, an isotropic exchange interaction between the rare-earth and the conduction electrons has been proposed[7]

TABLE I. Values of A, B, C, and A', B', C', obtained from the best fit of the experimental data with Eqs. (1) and (3), at 9 and 35 GHz.

	A (G^2)	B (G^2)	C (G^2)	A'	B'	C'
9.06 GHz	2482	−36.2	5.0			
35 GHz	16156	−602.6	68.8	7.6524	−0.0042	0.00040

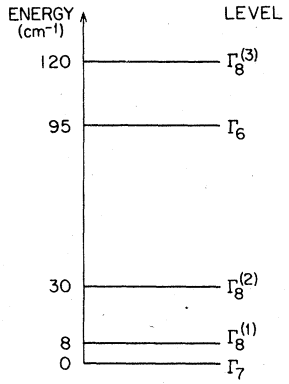

FIG. 4. Scheme of the energy levels of Dy and *Ag*:Dy as obtained from the data in Ref. 1.

$$\mathcal{H}_{\text{exch}} = \sum_i \mathcal{J}\vec{S}\cdot\vec{s}_i\,\delta(\vec{R}-\vec{r}_i) = \sum_i (\Lambda-1)\mathcal{J}\vec{J}\cdot\vec{s}_i\delta(\vec{R}-\vec{r}_i). \quad (5)$$

$\vec{R}$, $\vec{S}$, and $\vec{J}$ correspond to the rare earth and $\vec{r}_i$ and $\vec{s}_i$ to the conduction electrons, $\Lambda=\frac{4}{3}$ for Dy^{3+}, and $\mathcal{J}$ is the exchange interaction.[8] Equation (5) gives $\mathcal{J}=0.47$ eV using Knight-shift data and $\mathcal{J}=0.22$ eV utilizing the Korringa rate.[10] The difference between these two values was attributed[1] to different averages of the exchange interaction over different wave vectors for the electrons.

We now turn to discussion of the two mechanisms mentioned in the introduction that lead to anisotropic, magnetic-field-dependent broadening.

A. Change of the Korringa relaxation induced by the Zeeman interaction

In the low-temperature regime, when kT is much smaller than the crystalline field splittings, the contribution ΔH to the linewidth from the Korringa relaxation is given by[7]

$$\Delta H_0 = \frac{\pi g_0}{\mu_B}\left(\frac{\Lambda-1}{\Lambda}\right)^2 \eta(E_F)^2\mathcal{J}^2 \sum_{q=x,y,z} |\langle\alpha|J_q|\beta\rangle|^2 kT, \quad (6)$$

where g_0 is the gyromagnetic ratio of the ground doublet and μ_B is the Bohr magneton; $\eta(E_F)$ is the conduction-electron density of states at the Fermi surface; $\mathcal{J}$ is the exchange-coupling constant defined in Eq. (5); and $\langle\alpha|J_q|\beta\rangle$ are matrix elements of the angular momentum components J_x, J_y, and J_z within the Γ_7 eigenstates $|\alpha\rangle$ and $|\beta\rangle$.

When the Zeeman interaction

$$\mathcal{H}_z = \Lambda\mu_B\vec{H}\cdot\vec{J} \quad (7)$$

is not negligible compared to the crystalline field splittings, Eq. (6) should be modified in order to consider the admixtures of the ground doublet with excited states via $\mathcal{H}_z$. In this case, the matrix elements which appear in Eq. (6) should be replaced by

$$'\langle\alpha|J_q|\beta\rangle' = \langle\alpha|J_q|\beta\rangle + \sum_{i,j}(1/\Delta_i\Delta_j) \times(\langle\alpha|J_q|i\rangle\langle i|\mathcal{H}_z|j\rangle\langle j|\mathcal{H}_z|\beta\rangle + \langle\alpha|\mathcal{H}_z|i\rangle\langle i|J_q|j\rangle\langle j|\mathcal{H}_z|\beta\rangle + \langle\alpha|\mathcal{H}_z|i\rangle\langle i|\mathcal{H}_z|j\rangle\langle j|J_q|\beta\rangle), \quad (8)$$

where $\Delta_i = E_{\Gamma_8}(i) - E_{\Gamma_7}$ and the sum is over the four states of each of the three Γ_8 quarters.

The correction to the Korringa rate introduced in Eq. (5) was calculated using Eqs. (7) and (8), considering only the first excited $\Gamma_8^{(1)}$ quartet of Dy^{3+} (see Fig. 4). We obtain the result

$$\Delta H(\theta,\varphi) = \Delta H_0\left[1+\left(\frac{h\nu}{\Delta}\right)^2\frac{F^2}{2D^3}\left[\left(\tfrac{9}{10}H-\tfrac{27}{40}K\right)f_4(\theta,\varphi) + \left(\tfrac{9}{5}H+\tfrac{12}{5}K\right)\right]\right], \quad (9)$$

where D, F, H, and K are matrix elements, which can be obtained using the tables of Lea *et al*.,[6] for the wave functions of this quartet as a function of the parameter x ($D=\frac{17}{6}$, $F=-2.37$, $H=0.65$, and $K=-0.30$ for $x=0.53$). ν is the frequency used in the experiment and Δ is the energy difference between the first excited quartet and the ground state. $f_4(\theta,\phi)$ is the cubic function defined in Eq. (2a). The contributions of other quartets to Eq. (9) are negligible.

A similar perturbation calculation was performed in order to obtain the anisotropic contribution to the g factor of the ground state introduced by the Zeeman admixtures with excited states. With the same definitions as in Eq. (9), we obtain

$$g(\theta,\varphi) = g\left\{1+(h\nu/\Delta)^2F^2/2D^3\left[\left(-\tfrac{3}{10}H+\tfrac{9}{40}K\right)f_4(\theta,\varphi) + \left(-\tfrac{3}{5}H-\tfrac{4}{5}K\right)\right]\right\}. \quad (10)$$

It is seen that the anisotropic parts of $\Delta H(\theta,\varphi)$ and $g(\theta,\varphi)$ are proportional and differ by a factor of -3, a fact which will be discussed below

B. Broadening of the EPR line due to random internal stresses

Feher[2] accounted for the EPR linewidths of magnetic impurities in MgO assuming a random distribution of internal stresses in the host sample. A somewhat similar idea was used by Davidov *et al.*[4] to interpret the angular variation of the EPR linewidth of Dy in metallic LaSb. Here we apply the random stress model to *Ag*:Dy. Our analysis is divided into two parts; in the first, a phenomenological spin-Hamiltonian formalism is used to evaluate the predictions of the model in terms of a few constants which can be evaluated from the experiments. In the second, these constants are related to the more fundamental orbit-lattice interaction.

1. Phenomenological theory

The spin-Hamiltonian for an effective spin $S=\frac{1}{2}$ in cubic symmetry and in the presence of small deformations around this symmetry can be written as[4,9]

$$\begin{aligned}\mathcal{H}_{S-L} &= g\mu_B \vec{H}\cdot\vec{S} + g_1\mu_B \epsilon_1 \vec{H}\cdot\vec{S} \\ &+ g_3\mu_B\left[(3H_zS_z - \vec{H}\cdot\vec{S})\epsilon_{3,\theta} + \sqrt{3}\,(H_xS_x - H_yS_y)\,\epsilon_{3,\epsilon}\right] \\ &+ g_5\mu_B\left[\tfrac{1}{2}(H_yS_z + H_zS_y)\epsilon_{5,\xi} + \tfrac{1}{2}(H_xS_y + H_yS_x)\epsilon_{5,\eta} + \tfrac{1}{2}(H_xS_y + H_yS_x)\epsilon_{5,\zeta}\right], \end{aligned} \tag{11}$$

where

$$\begin{aligned}&\epsilon_1 = \epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz}, \quad \epsilon_{5,\xi} = \epsilon_{yz}, \\ &\epsilon_{3,\theta} = 2\epsilon_{zz} - \epsilon_{xx} - \epsilon_{yy}, \quad \epsilon_{5,\eta} = \epsilon_{xz} \\ &\epsilon_{3,\epsilon} = \sqrt{3}\,(\epsilon_{xx} - \epsilon_{yy}), \quad \epsilon_{5,\zeta} = \epsilon_{xy}, \end{aligned} \tag{12}$$

are normal strains with specific transformation rules within the cubic group.

The changes in the gyromagnetic factor g of the spin doublet as a function of the deformation can be written as

$$\delta g(\theta,\varphi,\epsilon) = \delta g_1 + \delta g_3(\theta,\varphi) + \delta g_5(\theta,\varphi), \tag{13}$$

with

$$\delta g_1 = g_1\epsilon_1, \tag{14a}$$

$$\delta g_3 = g_3\left[(3\cos^2\theta - 1)\,\epsilon_{3,\theta} + \sqrt{3}\,\sin^2\theta\cos 2\varphi\,\epsilon_{3,\epsilon}\right], \tag{14b}$$

$$\delta g_5 = g_5\left[\tfrac{1}{2}(\sin 2\theta\sin\varphi)\epsilon_{5,\xi} + \tfrac{1}{2}\sin 2\theta\cos\varphi)\epsilon_{5,\eta} + \tfrac{1}{2}(\sin^2\theta)\sin 2\varphi\,\epsilon_{5,\zeta}\right]. \tag{14c}$$

The shift in magnetic field of the EPR line is

$$\delta H = -\beta H\delta g/g\mu_B. \tag{15}$$

We assume now that the probability of the system, being in a certain state of strain, i.e., a certain set of values for the normal deformations, is given by the product of the probabilities of each of the normal deformations. That is to say, we assume that the probabilities of the normal deformations are uncorrelated. We further assume that each normal deformation is random, leading to a probability function $P(\epsilon_{i,\alpha})$ that is Gaussian.

We wish to consider deformations that conserve cubic symmetry in mean value. This requires that the width of the probability functions $P(\epsilon_{i,\alpha})$ be equal for the different components α within the same irreducible representation.

With these assumptions, we evaluate the second moment ΔH^2 of the observed resonance line as

$$\Delta H^2 = \int\cdots\int (\delta H)^2 \pi_{i,\alpha}\left[P(\epsilon_{i,\alpha})\,d\epsilon_{i,\alpha}\right],$$

where δH is obtained from Eqs. (13)–(15). The result is

$$\begin{aligned}\Delta H^2 = H^2/g^2\big[&(g_1^2\sigma_1^2 + \tfrac{8}{5}g_3^2\sigma_3^2 + \tfrac{1}{5}g_5^2\sigma_5^2) \\ &+ (\tfrac{3}{10}g_3^2\sigma_3^2 - \tfrac{1}{40}g_5^2\sigma_5^2)f_4(\theta,\varphi)\big],\end{aligned} \tag{16}$$

where $f_4(\theta,\varphi)$ is the cubic function defined in Eq. (2a). This result should be compared with Eq. (5) of Davidov *et al.*[4] Their result predicts an angular variation of the linewidth which has axial symmetry, i.e., gives different linewidths for the magnetic field along the [001] and [010] or [100] directions. Our result, Eq. (16), maintains cubic symmetry. This fact will be discussed later.

2. Orbit-lattice interaction

The orbit-lattice interaction, which describes the interaction of the magnetic ion in the $\vec{J}$ representation with the deformations of the lattice, can be written as[9]

$$\mathcal{H}_{O\text{-}L} = \sum_{n,i,\alpha} G_i^{(n)}\,O_{i,\alpha}^{(n)}\,\epsilon_{i,\alpha}, \tag{17}$$

where the $O_{i,\alpha}^{(n)}$ are linear combinations of the Stevens' operators[5] of order n, transforming like the α component of the Γ_i irreducible representation of the cubic group. $G_i^{(n)}$ are the corresponding orbit-lattice coefficients. It is known that in general, the most important contributions to $\mathcal{H}_{OL}$ are those for $n=2$. Thus, we will neglect the fourth- and sixth-order terms of Eq. (17). The change of the gyromagnetic factor is produced by a second-order process involving the orbit-lattice and Zeeman interactions. These second-order g shifts for a Γ_7 Kramer's doublet are given by[9]

$$\delta g = -(4/\mu_B H)$$

$$\times \mathrm{Re} \sum_{i,m} \left(\frac{\langle \alpha | \mathcal{H}_z | \Gamma_{8,m}^{(i)} \rangle \langle \Gamma_{8,m}^{(i)} | \mathcal{H}_{OL} | \alpha \rangle}{E_{\Gamma 8}^{(i)} - E_{\Gamma 7}} \right) ,$$

where Re means "the real part of."

The relations between the parameters g_i of the phenomenological theory with the second-order orbit-lattice parameters $G_i^{(2)}$ can be easily found using the Wigner-Eckart theorem. We chose

$$O_{3,\theta}^{(2)} = 3J_z^2 - J(J+1)$$

and

$$O_{5,\zeta}^{(2)} = \tfrac{1}{2}(J_x J_y + J_y J_x) ,$$

and with this normalization for the orbit-lattice operators, we obtain

$$g_3 = -2\Lambda G_3^{(2)} \sum_i \frac{F_i T_i}{E_{\Gamma_8}^{(i)} - E_{\Gamma_7}} \tag{18a}$$

and

$$g_5 = 4\sqrt{3}\,\Lambda G_5^{(2)} \sum_i \frac{F_i V_i}{E_{\Gamma_8}^{(i)} - E_{\Gamma_7}} , \tag{18b}$$

where the sums in Eqs. (18a) and (18b) are over the three excited quartets. F_i, T_i, and V_i are matrix elements of the Zeeman and orbit-lattice interactions between the doublet and the quartets, and can be calculated as a function of the parameter x using the tables of Lea *et al.*[6]

The energy denominators in Eqs. (18a) and (18b) allow us to consider only the excited quartets $\Gamma_8^{(1)}$ and $\Gamma_8^{(2)}$ (see Fig. 4). In this case and for $x = 0.53$, $F_1 = -2.37$, $T_1 = 0.94$, $V_1 = 5.60$, $F_2 = 2.7$, $T_2 = 53$, and $V_2 = -9.5$. Then

$$g_3 = -12.1 G_3^{(2)} , \tag{19a}$$

$$g_5 = -23.5 G_5^{(2)} , \tag{19b}$$

where $G_3^{(2)}$ and $G_5^{(2)}$ are given in cm^{-1}. On the other hand, using a point-charge model to evaluate the orbit-lattice parameters, we find[9]

$$G_3^{(2)} \simeq -\tfrac{4}{3}\alpha_J e^2 Z_{\mathrm{eff}} \langle r^2 \rangle / R^3 = -8.3\ \mathrm{cm}^{-1} , \tag{20a}$$

$$G_5^{(2)} \simeq \tfrac{32}{3}\alpha_J e^2 Z_{\mathrm{eff}} \langle r^2 \rangle / R^3 = 67\ \mathrm{cm}^{-1} , \tag{20b}$$

where $\alpha_J = -2/315$ is defined in Ref. 5; Z_{eff} is the effective charge of the ligands in units of the electronic charge e, equal to -1 in our case; $\langle r^2 \rangle$ is the mean-square radius for Dy^{3+}, given by Abragam and Bleaney.[5] $R = 2.85$ Å is the paramagnetic ion-ligand distance for the silver lattice.

IV. DISCUSSION

The accuracy of our experimental data allows us to separate the anisotropic contributions to ΔH^2 proportional to $f_4(\theta, \varphi)$ and to $f_6(\theta, \varphi)$ of Eqs. (2a) and (2b), as shown in Eq. (1) and Table I. In order to obtain the anisotropic contribution proportional to $f_6(\theta, \varphi)$ in Eqs. (9) and (16), a higher-order perturbation calculation would be needed. In this section we will compare our experimental results with the predictions of the models discussed in Sec. III.

In order to check the accuracy of the perturbation calculation which led to Eqs. (9) and (10), we use Eq. (10) to evaluate the crystalline field splitting Δ_1 from the experimental data. With the values given for D, F, H, and K, and $\nu = 35$ GHz, we get $\Delta_1 = 9\ \mathrm{cm}^{-1}$ in good agreement with the result 8 cm^{-1} obtained from a full diagonalization of the $J = \frac{15}{2}$ energy matrix.[1] Using the fact that the coefficients of $f_4(\theta, \varphi)$ in Eqs. (9) and (10) differ by a factor of -3, it is seen that this model predicts an angular variation of the linewidth of less than 2% of ΔH_0 at 35 GHz. This contribution is smaller than that observed experimentally, and has the wrong sign.

To compare the data with the prediction of the random-stress model, we extrapolate the linewidths to 0 K using[1] $\delta(\Delta H)/\delta T = 18.5$ G/K. Then, at 0 K

$$(\Delta H_{0\ \mathrm{K}}^2)_{9\mathrm{GHz}} = 751.3 - 36.2 f_4(\theta, \varphi) + 5.0 f_6(\theta, \varphi) , \tag{21}$$

$$(\Delta H_{0\ \mathrm{K}}^2)_{35\mathrm{GHz}} = 10243 - 603 f_4(\theta, \varphi) + 69 f_6(\theta, \varphi) . \tag{22}$$

The ratio between the values of the three coefficients at 9 and 35 GHz are in good agreement with the ratio of the squares of the frequencies (or the magnetic field) as predicted by Eq. (16). In Table II, we show the values obtained when Eqs. (21) and (22) are used together with Eq. (16). The good agreement between the values at 9 and 35 GHz shows the applicability of the random-stress model to Ag:Dy. In the following, we will use the values obtained at 35 GHz, since the experiments are

TABLE II. Values of $|g_3\sigma_3|$ and $|g_5\sigma_5|$ obtained from the values given in Table I, using Eq. (16).

Frequency	$\lvert g_3\sigma_3 \rvert$	$\lvert g_5\sigma_5 \rvert$
9 GHz	0.097	0.48
35 GHz	0.086	0.47

more accurate. In order to estimate the widths of the distributions of stresses in the sample we use the point-charge estimates given in Eqs. (20a) and (20b) together with Eq. (19); we obtain

$$g_3 = 100 \quad \text{and} \quad g_5 = -1574 . \tag{23}$$

There is a large error in the estimation of g_3. The value $T_1 = 0.94$ for $x = 0.53$ used there would be -14 and 8 for $x = 0.4$ and $x = 0.6$, respectively. Thus, a small error in the value of x produces a large error in g_3.

Using Eq. (23) together with Table II, we find

$$\sigma_3 = 8.6 \times 10^{-4} \quad \text{and} \quad \sigma_5 = 3.0 \times 10^{-4} .$$

In spite of the limitations of the point-charge model and the additional uncertainty of g_3 mentioned above, we get values for σ_3 and σ_5 similar to those measured for paramagnetic impurities in insulators.[2,10] In order to avoid the uncertainty in the values of the widths of the distributions of stresses introduced by any model used to estimate the parameters of the orbit-lattice interaction, uniaxial stress experiments[2,9,10] are needed. Recently, Dodds and Sanny[11] observed the magnetic resonance of dilute Dy in thin polycrystalline Ag films deposited in fused quartz substrates. They obtained a lower limit for $G_3^{(2)}$ that, in our notation, is $G_3^{(2)} = -1.2\ \text{cm}^{-1}$. The agreement in sign and order of magnitude of this lower limit and the value we obtain using a simple point-charge model support our idea that uniaxial stress experiments can be performed. These experiments are readily feasible and our estimation for *Ag*:Dy indicates shifts of the EPR line of about one linewidth at any frequency for typical strains of 5×10^8 dyn/cm^2.

We have estimated σ_3 and σ_5 by equating the second moment calculated in Eq. (16) to the halfwidth of the resonance line. This is clearly an approximation because, in general, the *lineshape* will not be Gaussian. Thus a factor of unity order would be needed to pass from the second moment to the linewidth. This same factor would enter in the calculation of σ_3 and σ_5. We observe that the quadratic dependence of the linewidth anisotropy on the field, as predicted by the model, would *not* be affected by the factor because the line shape would be the same at different fields due to the linear approximation in Eq. (11).

We carried out the integration which led to Eq. (16) assuming a Gaussian distribution of strains. If we assume a different distribution with some characteristic width and finite second moment proportional to the square of that width, we would (i) predict a different line shape than in the present calculation and (ii) calculate quantities analogous to σ_3 and σ_5, corresponding to the assumed distribution. The different line shape would produce a factor as discussed in the previous paragraph, but would not change the results in an important way.

In the work of Davidov *et al.*,[4] a random stress model is used to explain the broadening of the EPR line of Dy in the metallic lattice of LaSb. It is not clear to us, however, what assumptions were made by these authors to obtain Eq. (5). The equation applies to a model where the distribution of stresses is not random, but, rather in a preferred direction. Thus there is axial symmetry for the angular dependence of the linewidth. This question will be pursued elsewhere.

Recently, Dahlberg[12] measured the residual linewidth of *Ag*:Er as a function of concentration, with Er dopings between 125 and 2000 ppm at 1.7 and 9.2 GHz. His data is particularly useful to understand the frequency-independent broadening which he attributed to spin-spin interactions between magnetic impurities. Since he used powdered samples, the detailed form of the anisotropy is not readily available. Thus information about random stress broadening contributions is limited. He obtained frequency-dependent contributions of two types; one depends on the concentration of Er and is attributed to Kohn-Vosko oscillations of the charge density. The other does not depend on concentration and is attributed to residual stresses in the sample.

As a concluding remark, we feel that, in order to have a complete view of the sources of broadening of the EPR lines of magnetic impurities in metallic lattices, more measurements of the dependence of the residual linewidths as a function of concentration and frequency in single crystals are needed, together with uniaxial stress experiments.[9]

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