

Crystalline-Field Effects in the Electron-Spin Resonance of Rare Earths in the Noble Metals*

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The effect of a low-lying crystalline-field excited state on the electron-spin-resonance (ESR) linewidth and g shift of the ground state in a dilute magnetic alloy is calculated. Departures from linearity are found for the linewidth at temperature substantially below the excitation energy of the first excited state. Measurements on Ag:Dy¹⁶⁴ and (especially) Au:Er are reported which exhibit this behavior. Upon comparison with Ag:Gd and Ag:Er measurements, it is found possible to estimate the energy separation of the first excited level from the ground state from ESR linewidth and g -shift measurements alone.

I. INTRODUCTION

Static-susceptibility measurements¹ on Ag:Dy and Au:Er (among others) have been interpreted in terms of a crystalline-field ground Γ_7 doublet, with a $\Gamma_8^{(1)}$ first excited state, at an energy of <1 and 19 K, respectively. A close proximity of the excited state should lead to changes in the apparent g shift and thermal broadening of the ground-state resonance. The former arises from Zeeman admixtures of the excited level into the ground level (in these cases, the admixtures are anisotropic because of the Γ_8 character of the excited level); the latter from off-diagonal matrix elements of the localized-conduction-electron exchange coupling between the ground and excited levels. Clearly, the closer the levels, the larger will be these effects.

We report the electron-spin resonance of Ag:Dy¹⁶⁴, Au:Er, and Ag:Gd dilute magnetic alloys. The isotopic purity of the Dy¹⁶⁴ was such as to eliminate complications caused by hyperfine broadening of the Ag:Dy resonance.

Williams and Hirst¹ have set the first excited level in Ag:Dy at 1 K from the ground level. Our calculations indicate that a substantial alteration of the g value (negative shift) should occur as the result of such a small splitting, along with a substantial change in the slope of the temperature dependence of the linewidth. Our measurements indicate no significant negative shift of the g value, and, upon comparison with Ag:Gd and Ag:Er (to establish the weak variation of the exchange coupling across the second half of the rare-earth series) only a slightly larger thermal broadening than expected for an isolated Γ_7 ground state. From both measurements we are able to place the $\Gamma_8^{(1)}$ excited level in the range between 5–10 K, substantially further away from the ground doublet Γ_7 than predicted from susceptibility data.

Williams and Hirst place the first excited level of Au:Er $\Gamma_8^{(1)}$ at 19 K above the ground Γ_7 . No substantial change in the g shift is expected, there-

fore. However, measurements of the linewidth from 1.6 to 25 K exhibit a rather sharp break in slope of the temperature-dependent width at approximately 6 K. Our theoretical analysis demonstrates that this break is caused by off-diagonal transitions of the localized-conduction-electron exchange coupling between the ground and excited states. From our fit, we verify that the susceptibility estimate of the splitting is accurate, finding a value of 16 ± 6 K from our experiments.

Magnetic-resonance measurements therefore can give an independent method for establishment of the ground-excited state splitting in dilute magnetic alloys, and complement magnetic-susceptibility data. In the case of a close proximity of two levels, where it is difficult to use the latter method to sort out the splitting, magnetic resonance can provide more accurate estimates.

The experimental results for Ag:Dy¹⁶⁴, Au:Er, and Ag:Gd are presented in Sec. II. Our theoretical work is displayed in Sec. III, and an analysis of our results given in Sec. IV.

II. EXPERIMENTAL RESULTS

A. Ag:Dy¹⁶⁴

The observation of the magnetic-resonance spectrum of Ag:Dy has been reported previously,² using powdered samples and naturally abundant Dy isotopes. In the present work, measurements were performed on single crystals (as well as powdered samples) using the even Dy¹⁶⁴ isotope. This reduces the residual width and enables one to measure more accurately both the g value and the thermal broadening. The Ag:Dy¹⁶⁴ single-crystal spectrum exhibits a single isotropic line with a field for resonance appropriate to a g value close to the $g = 7.55$ expected for an isolated Γ_7 doublet. The linewidth can be fitted to the formula $a + bT$. Our experimental results are given in Table I. The observed thermal broadening is found to be smaller than that reported previously.² No resonance lines

TABLE I. g value, the residual width a , and the thermal broadening b , in the temperature range $1.4 < T < 4.2$ K for the various Ag:Dy samples measured.

Sample	g value ($T=1.4$ K)	a (G)	b (G/K)
450 ppm (single crystal)	7.66 ± 0.05	28	23 ± 5
500 ppm (single crystal)	7.65 ± 0.05	26	24 ± 5
500 ppm (powder)	7.63 ± 0.08	80	20 ± 8
1000 ppm (powder)	7.63 ± 0.08	95	27 ± 8
1400 ppm (powder)	7.61 ± 0.1	110	24 ± 8

associated with the $\Gamma_8^{(1)}$ quartet were observed in the single crystal alloy. The hyperfine lines belonging to a Dy^{163} isotope were observed in a separate Ag:Dy¹⁶³ sample (500 ppm). The hyperfine constant $A(\text{Dy}^{163})$ equals 82 ± 4 G (see Fig. 1).

B. Au:Er

The observation of the magnetic resonance spectrum of Au:Er has been reported previously (using the Er^{167} isotope, as well as natural-abundance crystals) in the liquid-He range (1.4–4.2 K). The thermal broadening was the smallest observed in any dilute alloy we have investigated to date: only 2.5 ± 0.3 G/K. This encouraged us to extend the measurements to higher temperatures (we followed the resonance to 25 K). We plot the measured linewidth in Fig. 3 for 700- and 1000-ppm samples. The insert in Fig. 3 represents previous low-concentration measurements³ in the temperature range $1.4 \leq T \leq 4.2$ K. It is clear from Fig. 3 that the slope of the thermal broadening at low temperatures is different from that at high temperatures, and therefore a relation of the form $a + bT$ cannot fit the data. As discussed in Secs. III and IV, the sharp change in slope at ~ 6 K will be interpreted as evidence for the presence of an excited level (in this case, the $\Gamma_8^{(1)}$). Detailed linewidth-versus-temperature calculations will confirm this assignment.

Only a single line was observed, with a Γ_7 g value in agreement with previous measurements.³ Further investigations are planned on single-cryst-

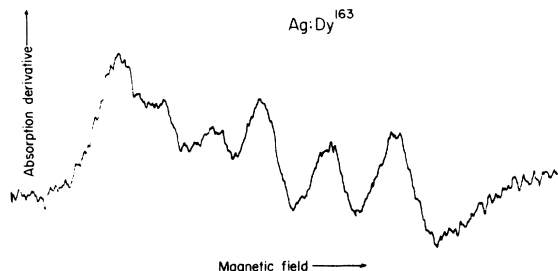


FIG. 1. Hyperfine spectrum associated with the Dy^{163} isotope ($I = \frac{5}{2}$) in 500-ppm Ag:Dy¹⁶³ at $T = 1.4$ K.

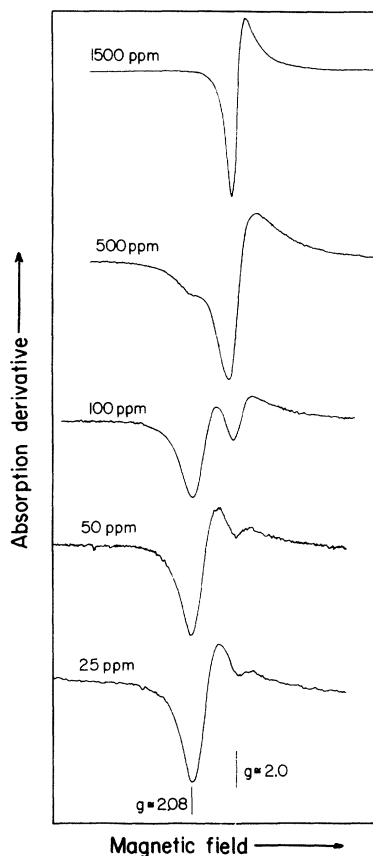


FIG. 2. EPR spectra (absorption derivative) for Ag:Gd alloys with various Gd concentrations at $T = 1.4$ K.

tal samples of Au:Er to see if the multiline pattern appropriate to the excited Γ_8 can be observed.

C. Ag:Gd

The Gd resonance in Ag:Gd dilute alloys was measured. At high concentrations ($c = 500$ ppm) two lines were observed, one at $g \approx 2$ and the other at $g \approx 2.08$.⁴ (See Fig. 2.) As the concentration was lowered the intensity of the $g = 2$ line appreciably diminished. For Gd concentrations of 50 and 25 ppm, the $g = 2$ line almost completely vanished.

We believe the $g = 2$ portion of the resonance spectrum, observed at high Gd concentrations, can be attributed to Gd clusters. The tendency towards clustering is a direct result of the very low solubility of Gd in Ag.⁵ The line at $g = 2.08 \pm 0.01$ is then identified as arising from isolated Gd impurities in a purely Ag environment. The similar g values observed previously for Gd in other nontransition-metal cubic hosts [$g(\text{Au}) = 2.045$,⁶ $g(\text{Th}) = 2.07$,⁷ and $g(\text{Al}) = 2.08$ ⁸] provide additional evidence for this identification.

Single crystals of Ag:Gd alloys were also examined. At high Gd concentrations (1000–800

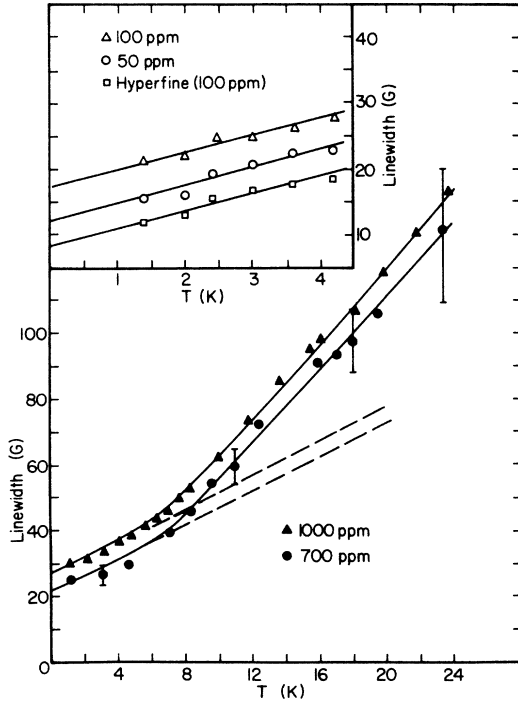


FIG. 3. EPR linewidth as a function of temperature for several Au:Er alloys. The solid line represents the theoretical linewidth (modified to include the experimental residual width) for $\Delta \approx 16$ K calculated according to (2). The dashed line is the linewidth expected for an isolated Γ_7 doublet. The insert exhibits the linewidth at low concentrations in the temperature range $1.4 \leq T \leq 4.2$ K. The slope of the thermal broadening agrees in this temperature range with the dashed line, i. e., is indistinguishable from an isolated Γ_7 . The squares in the insert represent the linewidth extracted from one of the hyperfine lines of the Au:Er¹⁶⁷ sample.

ppm), a single line with $g \approx 2$ was observed with no angular dependence. We were unable to detect a measurable signal for single crystals at low Gd concentrations (less than 100 ppm).

The thermal broadening (measured in the temperature range 1.4–4.2 K) of the $g \approx 2.08$ Ag:Gd line was observed to equal 19 ± 7 G/K for the 25 and 50 ppm samples. Addition of 1% of Au did not change either the g value or the thermal broadening. This indicates that the resonance is un bottlenecked at these low Gd concentrations in our samples. It should be stressed that the powdered samples were prepared by both arc melting and induction melting. No differences were observed between samples prepared in these two different manners.

III. THEORETICAL ANALYSIS

Conveniently, both Ag:Dy and Au:Er possess a ground Γ_7 level and a first excited $\Gamma_8^{(1)}$ level. The wave functions for the Γ_7 states are uniquely deter-

mined by the cubic symmetry alone; the wave functions and excitation energy of the $\Gamma_8^{(1)}$'s are a function of the fourth- and sixth-order crystalline-field coefficients for the particular alloy. In the presence of an external magnetic field (here equal to approximately 800 G for X-band measurements), the Γ_7 and $\Gamma_8^{(1)}$ are mixed by the Zeeman interaction, so that the effective g value will depart significantly from the isolated Γ_7 value if the $\Gamma_8^{(1)}$ lies close. Under these conditions, one must directly diagonalize the full crystal field plus the Zeeman Hamiltonian in order to obtain the correct field for resonance, and thus the effective g value. We use the form of Lea, Leask, and Wolf⁹ for the crystalline-field Hamiltonian:

$$\mathcal{H} = W[(O_4/F_4)x + (1 - |x|)(O_6/F_6)] + g_J \mu_B (\vec{H} \cdot \vec{J}). \quad (1)$$

Here F_4 and F_6 are numerical constants defined in Ref. 9; W is the over-all crystal-field splitting of the $J = \frac{15}{2}$ ground multiplet; O_4 and O_6 are the fourth- and sixth-order equivalent operators, respectively; x is proportional to the ratio of fourth- to sixth-degree cubic crystalline-field coefficients; g_J is the Landé factor; and μ_B is the Bohr magneton. The over-all splitting W was taken equal to 157 K for Ag:Dy and 105 K for Au:Er, as measured by Williams and Hirst.¹ The value of x then characterizes the energy separation between the ground Γ_7 and the $\Gamma_8^{(1)}$, as well as the $\Gamma_8^{(1)}$ wave functions themselves.

Because the susceptibility analysis¹ indicated a close proximity of the two levels for Ag:Dy, the field for resonance (in a powder, only the Γ_7 resonance would be observable) of the predominantly Γ_7 admixed states was calculated as a function of Δ (equivalent to the crystalline-field ratio x). The result is displayed in Fig. 4 for two field directions, [100] and [110]. The differences arise because of the anisotropy of the Γ_8 itself. The field for resonance must be expressible in terms of an effective g factor:

$$g_{\text{eff}} = g_0 + g_1 p, \quad (2)$$

where

$$p = 1 - 5(l^2 m^2 + m^2 n^2 + n^2 l^2) \\ = \frac{5}{2}(l^4 + m^4 + n^4 - \frac{3}{5}).$$

Here l , m , and n are the direction cosines the magnetic field makes with the cubic crystalline axes. The curves in Fig. 4 therefore determine g_0 and g_1 for a given value of Δ . In a single crystal, this anisotropy of g will result in an anisotropic field for resonance. More will be said about this effect in Sec. IV.

The thermal broadening of the Γ_7 level will be modified by the presence of the $\Gamma_8^{(1)}$ level. Labeling the ground levels by $|\pm\rangle$, and the excited $\Gamma_8^{(1)}$

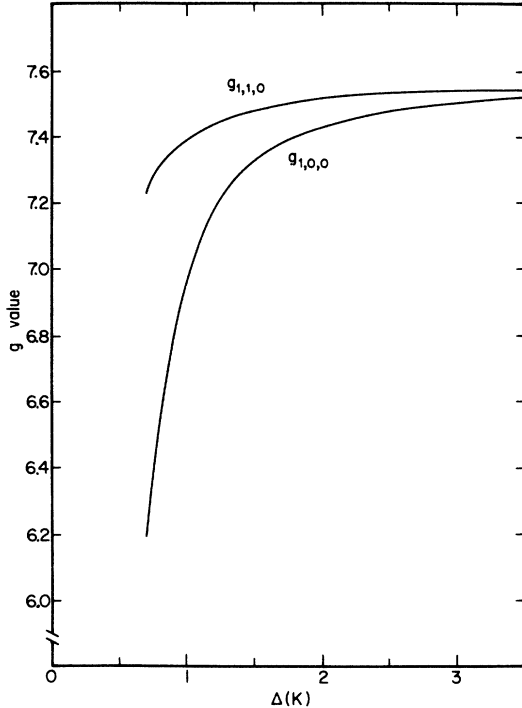


FIG. 4. Theoretical g value calculated by diagonalization of (1) for the external magnetic field along the [110] and [100] directions.

levels by $|i\rangle$, we find for the thermal broadening ΔH^{10}

$$\Delta H = (\pi/2 g_{\text{eff}} \mu_B) [(g_J - 1) \eta(E_F)]^2 \langle J^2 \rangle (A + B + C),$$

where

$$\begin{aligned} A &= (\langle + | J_x | + \rangle - \langle - | J_x | - \rangle)^2 k_B T, \\ B &= \frac{1}{2} |\langle + | J_x | - \rangle|^2 [f(\Delta_0) + f(-\Delta_0)], \\ C &= \frac{1}{2} \sum_i [(|\langle + | J_x | i \rangle|^2 + |\langle + | J_x | i \rangle|^2) f(\Delta_i^\dagger) \\ &\quad + (|\langle i | J_x | - \rangle|^2 + |\langle i | J_x | \tau \rangle|^2) f(\Delta_i^\ddagger) \\ &\quad + 2 |\langle + | J_x | i \rangle|^2 f(\Delta_i^\dagger) + 2 |\langle - | J_x | i \rangle|^2 f(\Delta_i^\ddagger)], \\ \Delta_0 &= E(\Gamma_7, +) - E(\Gamma_7, -), \\ \Delta_i^\dagger &= E(\Gamma_8, i) - E(\Gamma_7, \pm), \\ f(\Delta) &= \Delta / (e^{\Delta/k_B T} - 1). \end{aligned} \quad (3)$$

Here $\langle J^2 \rangle$ is the average on the Fermi sphere of the square of the wave-vector-dependent exchange integral $J(\vec{q})$; g_{eff} is the effective g value [for Ag:Dy, it would be the angular average of (2) or simply g_0]; $\eta(E_F)$ is the one-spin density of states at the Fermi surface; and Δ_0 is the Zeeman splitting of the ground Γ_7 doublet.

For isolated doublets, (3) reduces to only the first two terms. For a given host metal, this allows the ratio of b between various rare-earth so-

lutes to be used as a measure of the actual isolation. Thus, for Dy and Er, for example,

$$\frac{b(\text{Dy})}{b(\text{Er})} = \left(\frac{g_J(\text{Dy}) - 1}{g_J(\text{Er}) - 1} \right)^2 \frac{g_{\text{eff}}(\text{Er})}{g_{\text{eff}}(\text{Dy})} \left(\frac{\langle J^2 \rangle_{\text{Dy}}}{\langle J^2 \rangle_{\text{Er}}} \right). \quad (4)$$

If one assumes that the exchange is constant along the second half of the rare-earth series, then (4) and a completely analogous expression for Gd and Er yield for the Γ_7 ground states of Dy and Er

$$b(\text{Dy})/b(\text{Er}) = 2.5, \quad b(\text{Gd})/b(\text{Er}) = 2.6. \quad (5)$$

These relationships will be very useful in analyzing the experimental data in Sec. IV.

IV. ANALYSIS

A. Ag:Dy

According to Williams and Hirst,¹ the separation between the ground Γ_7 and first excited $\Gamma_8^{(1)}$ level is less than 1 K. For $x = 0.58$, we calculate $\Delta = 0.71$ K. Figure 3 then yields $g_{\text{eff}} = 6.19$, a value far outside of the error limits (7.63 ± 0.05). As Δ is increased (decreasing x) the effective g value comes closer to that observed experimentally. However, the curve in Fig. 4 varies so rapidly for small Δ that it is difficult to estimate a lower limit for Δ because of the experimental error in the observed g value [the polarization shift $\Delta g = [(g_J - 1)/g_J] g_{\text{eff}} J(0) \times \eta(E_F) = 0.05$, so that one should compare the theoretical values to 7.57 ± 0.05]. However, the anisotropy of the $\Gamma_8^{(1)}$ allows us to establish rather accurately a lower limit for Δ . We note that the single-crystal samples do not exhibit an anisotropic field for resonance to within our experimental errors, ± 5 G. If we were to assume that the excited-state admixture anisotropy has the maximum value (10 G) which would just escape our detection capability, we could determine a minimum separation Δ between the ground and excited levels. A 10-G shift of the field for resonance implies a maximum fractional shift in the g value of 0.01. From calculations (Fig. 4) this would require $\Delta \geq 5$ K.

An estimate of the upper limit on Δ can be ob-

TABLE II. Thermal broadening b , of the ESR linewidth of Er, Dy (Γ_7 ground state), and Gd in various cubic metal hosts. The thermal broadening was measured in the temperature range between 1.4 and 4.2 K.

Host	$b(\text{Dy})$ (G/K)	$b(\text{Gd})$ G/K	$b(\text{Er})$ G/K	$b(\text{Dy})/b(\text{Er})$	$b(\text{Gd})/b(\text{Er})$	
Th	34 ± 6	31 ± 4	14 ± 3	2.4	2.3	a
Rh	8 ± 3	9 ± 2	3.5 ± 1.5	2.3	2.6	b
Ir	7 ± 2	...	2.5 ± 1	2.8	...	c
Al	27 ± 5	60 ± 20^d	10 ± 2	2.7	d	e
Ag	23 ± 5	19 ± 7	7 ± 1	3.2	2.7	f

^aReference 7.

^bReference 11.

^cReference 12.

^dReference 13.

^eReference 8.

^fThis work.

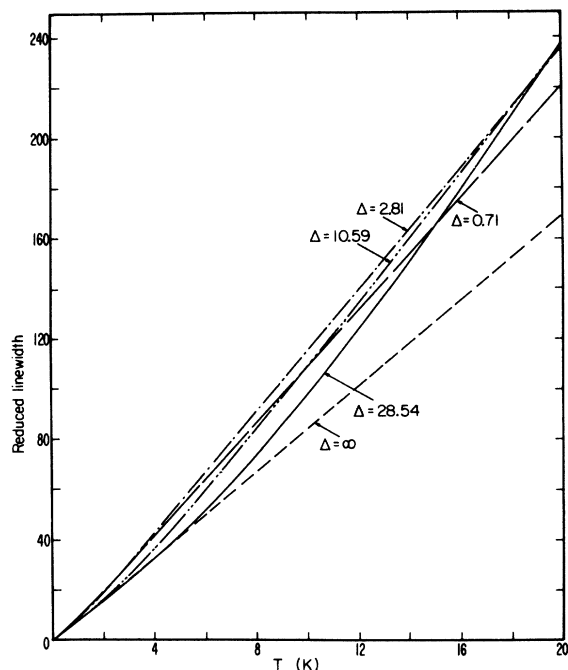


FIG. 5. Theoretical reduced thermal broadening for Ag:Dy for various values of Δ , the energy separation of the Γ_7 ground state from the $\Gamma_8^{(1)}$ first excited state. In order to focus on the effect of changes in crystalline field, $(A+B+C)/g_{\text{eff}}$, defined in (3), is plotted against temperature.

tained from the linewidth measurements. The argument goes as follows: Table II exhibits the experimental results observed for the ratio of the thermal broadening in various cubic host metals. It is clearly seen that the ratios $b(\text{Dy})/b(\text{Er})$ and $b(\text{Gd})/b(\text{Er})$ (apart from Al and Au) are close to 2.5 and 2.6, expected theoretically according to (5). However, for (5) to hold, two conditions must be satisfied: (a) The exchange interaction $\langle J^2 \rangle$ must be constant; (b) the isolation of the ground-state doublet from the first excited state must be at least 10 K. Thus, the systematics observed in various cubic host metals (with the exceptional case of Al:Gd and Au:Gd¹³ allows us to conclude that $\langle J^2 \rangle$ is roughly constant, going from Gd to Dy and Er in each of the cubic metals Th, Rh, Ir, and Ag. This constancy of exchange then gives significance to the departure of the experimental results for Ag:Dy [$b(\text{Dy})/b(\text{Er})=3$] from the predictions of (5) [$b(\text{Dy})/b(\text{Er})=2.5$]. This difference may be attributed to the influence of the excited $\Gamma_8^{(1)}$ level on the ground Γ_7 . In the limited temperature range at hand, and within experimental error, this effect appears as an increase in the slope of the Γ_7 linewidth with temperature.

To determine Δ quantitatively, we have computed the thermal broadening for Ag:Dy for a variety of

Δ (equivalently, a variety of x), and plotted the results against temperature in the temperature range 0–20 K in Fig. 5. It is seen from the plot that the slope of linewidth versus temperature can be at most 30% larger than the isolated ($\Delta=\infty$) value in the range $1.4 \leq T \leq 4.2$ K. Experimentally, the slope is some $(20 \pm 10)\%$ larger than the isolated value. This places Δ at a value less than 10 K. We must emphasize that this upper limit is based on the assumption of constance of exchange, and should therefore be viewed with some caution.

In summary, we estimate Δ to lie between 5 and 10 K, where we are reasonably sure of the lower limit, but the upper limit must be approached with caution.

B. Au:Er

Because the linewidth increases so slowly with increasing temperature, it was possible to measure the Au:Er resonance to 25 K. This allowed a rather large temperature region of measurement, and hence a more accurate determination of the excited-state splitting. We plot in Fig. 6 the theoretical reduced linewidth versus temperature for Au:Er using (3) for various values of Δ (equivalently, for various values of x). The theoretical linewidths for $\Delta \approx 16$ K, corrected to include the residual

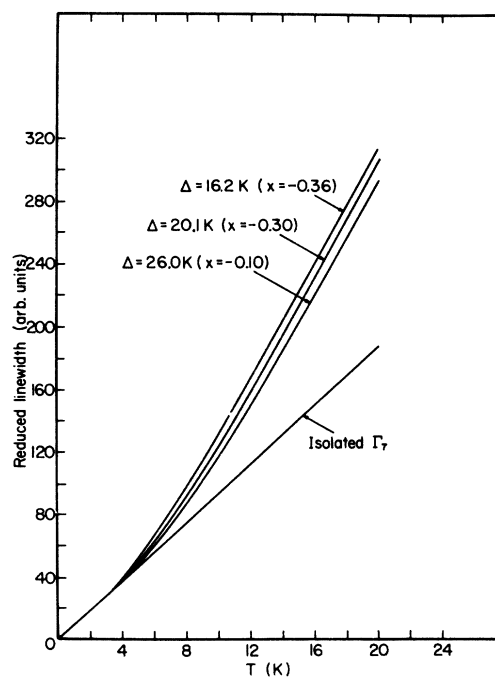


FIG. 6. Theoretical reduced thermal broadening from (3) for various values of Δ , the energy separation of the Γ_7 ground state from the $\Gamma_8^{(1)}$ first excited state, for Au:Er alloys. In order to focus on the effect of changes in crystalline field, $(A+B+C)/g_{\text{eff}}$, defined in (3), is plotted against temperature.

width Δs found from Fig. 6, are then included as the solid lines in Fig. 3. Although the theoretical linewidth for $\Delta \approx 16$ K is the best fit to the experimental values, the relatively large error bar in the latter as well as the insensitivity of the theoretical linewidth to small changes in Δ (see Fig. 6) yields a value $= 16 \pm 6$ K. This is in agreement with the magnetic susceptibility results of Williams and Hirst.¹ The fit is the more remarkable because of the rather complex character of the temperature dependence.

There is known to be a Γ_8 level somewhat above the $\Gamma_8^{(1)}$, at a splitting of about 23 K from the ground level. However, the exchange coupling has no matrix elements between the ground Γ_7 and the Γ_8 , so that off-diagonal transitions are not allowed, and the temperature dependence of the Γ_7 linewidth is a function only of the position and wave function of the $\Gamma_8^{(1)}$ level.

In summary, we have shown that, by the use of the systematics of the exchange coupling in the second half of the rare-earth series, it is possible to

utilize linewidth measurements to determine the separation between the ground and first excited levels in dilute magnetic alloys. For Ag:Dy it is higher than 5 K but probably less than 10 K, in substantial excess of the susceptibility interpretation. In Au:Er it lies at 16 ± 6 K, in agreement with the susceptibility interpretation. Knowledge of this splitting, and of the resonance parameters for the excited Γ_8 level, would be sufficient to completely determine the two cubic crystalline-field parameters.¹⁴ Unfortunately, where a Γ_8 resonance has been directly observed in other alloys, the crystalline splitting is unknown. Observation of the $\Gamma_8^{(1)}$ in Au:Er would provide this information, but as remarked in the Introduction, single-crystal measurements are necessary, and appear to be difficult because of sensitivity problems at the required elevated temperatures.

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⁵D. Davidov, R. Orbach, L. J. Tao, and E. P. Chock, *Phys. Lett. A* **34**, 379 (1971). In this reference, b was reported equal to 40 ± 15 G/K. In the present work (Table II) $b = 23 \pm 5$ G/K. The error in the former arises from the much larger residual width in the samples used in that work.

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⁷At still higher concentration ($c \geq 1000$ ppm) only the $g \approx 2$ line was observed. This is the spectra observed also by M. Peter, D. Shaltiel, J. H. Wernick, H. J. Williams, J. B. Mock, and R. C. Shaltiel [*Phys. Rev.* **126**, 1395 (1962)] for 5% Gd in Ag.

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¹⁵D. Davidov, R. Orbach, C. Rettori, L. J. Tao, and B. Ricks, *Phys. Lett. A* **40**, 269 (1972).

¹⁶The thermal broadening in Al:Gd was obtained by extrapolation to the unbottlenecked regime. In the extrapolation we fitted the g shift to the linewidth assuming the exchange interaction to be independent of wave vector. However, this assumption may not be correct. If we take the maximum observed thermal broadening to be the unbottlenecked value, we would find $b = 45 \pm 15$ G/K for Al:Gd. In the case of Au:Gd, new measurements (to be published) which extend over a much larger temperature range than those of Ref. 7, exhibit a much larger thermal broadening (approximately 14 ± 4 G/K). Because of this, and the fact that Au:Dy possesses a Γ_8 ground state, we omit reference to the Au data in Table II.

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