

Electron spin resonance of $Gd_xLa_{1-x}Pd_3$ intermetallic compounds

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Electron spin resonance of Gd in $Gd_xLa_{1-x}Pd_3$ intermetallic compounds annealed at 740 and 840°C exhibit a dependence of linewidths and g shifts on the annealing temperature. The linewidths and g shifts were measured as functions of temperature and concentration of Gd. Samples with high Gd concentration (1 at.% or more) were found to have bottleneck effects. All samples annealed at 740°C were found to exhibit Korringa-like thermal broadening between 20 and 40°K and those annealed at 840°C were found to be Korringa-like between 6 and 35°K. Below 20°K, samples annealed at 740°C were found to exhibit ordering effects whereas samples annealed at 840°C did not show such effects even at 6°K. These differences are indicative of the difference in the structural composition of the lattice due to annealing. We have deduced the average values for the exchange integral J to be 0.16 ± 0.03 eV and 0.15 ± 0.03 eV for the samples annealed at 740 and 840°C, respectively. We have obtained also the magnitudes of two contributions to the conduction-electron relaxation rates to the lattice (i) due to undisturbed host lattice and (ii) by spin-flip scattering due to Gd.

I. INTRODUCTION

Electron-spin-resonance (ESR) technique has been successfully used in the study of spin dynamics¹⁻⁴ of the conduction electrons and paramagnetic impurities in the intermetallic systems. The exchange interaction between the spins of the conduction electrons and those of the paramagnetic impurities can be expressed by the following Hamiltonian

$$\mathcal{H} = J\vec{S}_i \cdot \vec{S}_e, \quad (1)$$

where \vec{S}_i and \vec{S}_e are the spins of the paramagnetic ions and the conduction electrons, respectively, and J is the exchange interaction integral between the two spin systems. This exchange interaction gives rise to two important effects. Firstly, the electron-spin-resonance frequency of the impurity, which determines the g values of the ion, shifts from its free-ion resonance frequency to lower or higher values depending on the sign of the exchange integral.

The second effect provides a strong relaxation mechanism for the impurity ions in which the microwave energy absorbed by the resonating spins of the ions is transferred to the spin system of the conduction electrons, which in turn transfers it to the lattice. As a result of this relaxation process the ESR linewidth of the ions is broadened. This is known as the Korringa-like thermal broadening⁵ of the ESR line. However, it has been shown that the relaxation rate of the conduction electron to the lattice in some systems is not infinitely fast because of the finite capacity of the lattice thermal reservoir.

In addition, the conduction electrons take part in the resonance because of the proximity of the

resonance frequencies of two spin systems (having the same g value ≈ 2 for conduction electrons and impurities). As a result, the conduction electrons transfer back to the impurity ions part of the microwave energy they absorb. This relaxation rate, δ_{ei} has been shown to be comparable to, or even larger than the conduction-electron relaxation rate to the lattice, δ_{el} .

It has been shown that the so called "bottleneck effect" where $\delta_{ei} \gg \delta_{el}$ is present, will modify the g shift and the thermal broadening of the ions resulting from the exchange interaction. Furthermore, some systems may exhibit the temperature-dependent g shift, the so-called dynamic effect, as a result of the simultaneous excitation of the resonance of both the spin systems with the condition that the paramagnetic susceptibility of the ions be large at lower temperatures.

We have studied the electron-spin resonance of $Gd_xLa_{1-x}Pd_3$ intermetallic compounds annealed at 740 and 840°C which exhibit a dependence of linewidths and g shifts on the annealing temperature. The linewidths and g shifts were measured as functions of temperature and concentration of Gd. Samples with high Gd concentration (1 at.% or more) were found to have bottleneck effects. All samples annealed at 740°C were found to exhibit Korringa-like thermal broadening between 20 and 40°K and those annealed at 840°C were found to be Korringa-like between 6 and 35°K. Below 20°K, samples annealed at 740°C were found to exhibit minimum linewidth around 20°C, whereas samples annealed at 840°C did not show many values even at 6°K. We have deduced the average values for the exchange integral J for the samples annealed at 740 and 840°C, respectively. We have obtained also the magnitudes of two contributions to the

conduction-electron relaxation rates to the lattice due to undisturbed host lattice and by spin-flip scattering due to Gd.

II. THEORY

Such a system of two coupled resonators with equal g values was first treated by Hasegawa² by means of two coupled Bloch equations of motion. Assuming that the direct relaxation rate of the impurity ions to the lattice is negligible and the Pauli spin susceptibility χ_e of the conduction electrons is small compared to the paramagnetic susceptibility of the impurity ions χ_i , Hasegawa's, Davidov *et al.*'s,⁵ and Schmidt's⁴ results for the g shift Δg , and the linewidth ΔH can be expressed by the following formulas⁷:

$$\Delta g = \frac{x^2}{(1+x)^2 + y^2} \Delta g_{\max}, \quad (2)$$

$$\Delta g = f(x, y) \Delta g_{\max}, \quad (2a)$$

$$\gamma \Delta H = \frac{1+x+y^2}{(1+x)^2 + y^2} \delta_{ei} \frac{\chi_e}{\chi_i}, \quad (3)$$

$$\gamma \Delta H = \phi(x, y) \delta_{ei} \frac{\chi_e}{\chi_i}, \quad (3a)$$

where the bottleneck factor

$$\chi = \delta_{ei} / \delta_{ei}, \quad \Delta g_{\max} = J \chi_e / N g_e \mu_B^2,$$

where N is the number of lattice sites per unit volume, μ_B is the Bohr magneton, and $y = \gamma \lambda \chi_i H / \delta_{ei}$, where γ is the gyromagnetic ratio of the ion and H is the external magnetic field, while $\lambda = J / 2g \mu_B^2 N$ is the molecular-field constant.

In the above expressions we have made use of the detailed balance condition that

$$\chi_e \delta_{ei} = \chi_i \delta_{ie}, \quad (3b)$$

where δ_{ie} is the relaxation rate of the ions to the conduction-electron system. We may rewrite Eq. (3a) as

$$\frac{\Delta H}{T} = \phi(x, y) \frac{\delta_{ei}}{\delta_{ei}} \frac{\Delta H_k}{T}, \quad (3c)$$

where $\gamma \Delta H_k$ stands for δ_{ie} .

And δ_{ie} is given by the relation⁵

$$\delta_{ie} = (\pi / \hbar) \eta^2 J^2 k T, \quad (4)$$

where η is the conduction-electron density of states, k is Boltzmann's constant, and T is the absolute temperature. Whereas the relaxation rate δ_{ei} of the conduction-electron spins to the impurity spins is given by Overhauser⁸

$$\delta_{ei} = (2\pi / 3\hbar) \eta J^2 S_i (S_i + 1) C_i, \quad (5)$$

where C_i is the concentration of magnetic impurities. Combining the Eqs. (3c), (4), and (5)

we have

$$\frac{\Delta H}{T} = \frac{3\eta k}{2\gamma S_i (S_i + 1)} \phi(x, y) \frac{\delta_{ei}}{C_i}. \quad (6)$$

In Eq. (6) for $x \ll 1$ due to the large bottleneck effect in higher Gd^{3+} concentration samples we put $\phi(x, y) \approx 1$. It is obvious from Eq. (6) that the linewidths are inversely concentration (C_i) dependent in the presence of bottleneck.

It has been shown by Moriya⁹ and others^{10,11} that there can exist electron-electron Coulomb interactions amongst the conduction electrons which may result in the enhancement of the susceptibility χ_e . In the presence of such interactions, Eq. (2)–(3c) remain the same, but the various parameters are redefined in the following way¹⁰

$$\chi_e = \frac{\chi_e^{(0)}}{1 - U\chi(0)} = \frac{\chi_e^{(0)}}{1 - \alpha}, \quad (7a)$$

$$\Delta g_{\max} = \frac{J(0)\eta}{1 - U\chi(0)} = \frac{J(0)\eta}{1 - \alpha}, \quad (7b)$$

$$\delta_{ei} = \delta_{ei}^{(0)} \frac{K(\alpha)}{1 - \alpha}, \quad \delta_{ie} = \delta_{ie}^{(0)} \frac{K(\alpha)}{(1 - \alpha)^2}, \quad (7c)$$

and

$$\lambda = \frac{J(0)}{g^2 \mu_B^2 N}, \quad \alpha = U\chi(0), \quad (7d)$$

where U is the electron-electron Coulomb interaction. The parameters $\delta_{ei}^{(0)}$ and $\delta_{ie}^{(0)}$ are the unenhanced values, α and $K(\alpha)$ are the enhancement factors as defined in Ref. 9–11.

Furthermore, J and χ_e are q dependent [$J(q)$ and $\chi_e(q)$]. Assuming no q dependence of J , one may write the modified J in the following form¹²:

$$J_{\text{eff}}(0) = J(0) / (1 - \alpha). \quad (8)$$

III. EXPERIMENT

A. Sample preparation

The series of samples of $Gd_xLa_{1-x}Pd_3$ were prepared in an argon arc furnace. They were violently contracted by sudden cooling below their melting point.

The purities of the starting metals used are Gd(99.9%), La(99.99%), Th(99.9%), and Pd(99.99%). Samples with the lower concentration of Gd were then prepared from the master samples by successive dilution. This ensures homogeneous dispersion of the impurities in the host lattice.

In general, as the annealing temperatures range from $\frac{1}{2}$ to $\frac{2}{3}$ of the melting point of an intermetallic in Kelvin unit, we prepared two sets of samples, one set annealed at 740 °C and the other annealed at 840 °C. The melting point of $LaPd_3$ intermetallic is around 1600 °C. The structure of ordered $LaPd_3$

is AuCu_3 -type cubic.¹³ The x-ray analysis of these samples showed that they were single phase. Samples were ground to fine granular powders and dispersed in pure quartz powders to reduce the skin-effect problem one encounters when a microwave field is applied to a conductor. Quartz powders have been tested for extraneous paramagnetic impurities and centers, and were found to be free from such unwanted impurity resonances in the temperature range beginning at 300°K down to 4.2°K. Samples were sealed in evacuated 3-mm diameter (i.d.) quartz tubes which fit easily into the microwave cavity. The ESR measurements were carried out by the standard Varian X-band (V-4500-10A) model spectrometer. For low-temperature studies we have used a liquid-helium flow-cryostat similar to the one used by Haupt.¹⁴ Temperature was varied from 60 to 6°K and could be maintained steady to within 5%, during the entire length of each magnetic field scan of the resonance, which is about 2.5 min.

B. Experimental procedure

The ESR signals of Gd^{3+} in $\text{Gd}_x\text{La}_{1-x}\text{Pd}_3$ samples annealed at 740 and 840°C were measured in the concentration range from 0.12- to 3.0-at.% Gd, with respect to La. The measurements were taken in the temperature range from 40 to 6°K. Our ESR signals show the asymmetric line shapes typical of metallic samples. These signals are fitted to the Lorentzian line shape and were analyzed for ΔH and Δg using the procedure outlined by Peter *et al.*¹⁵ The linewidth ΔH in Figs. 1 and 2 is half-power halfwidth of the absorption part of the resonance line. The g shifts, Δg , in Figs. 3 and 4 were measured with respect to

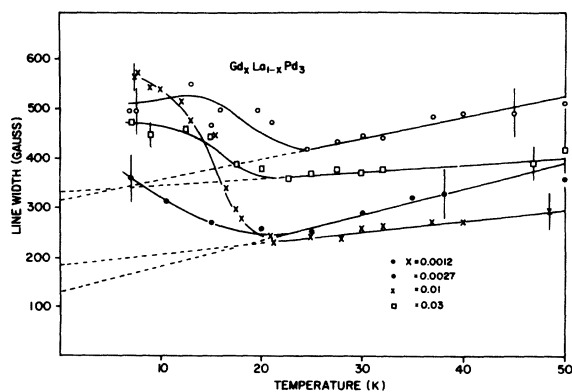


FIG. 1. ESR linewidths ΔH of Gd in $\text{Gd}_x\text{La}_{1-x}\text{Pd}_3$ (annealing temperature at 740°C) as a function of temperature for various Gd concentrations. (Below 20°K the linewidths increase with the decreasing temperature due to ordering effects.)

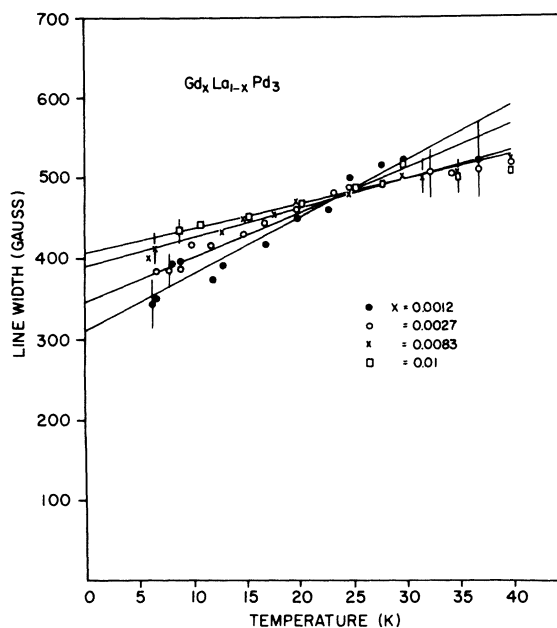


FIG. 2. ESR linewidths ΔH of $\text{Gd}_x\text{La}_{1-x}\text{Pd}_3$ (annealing temperature at 840°C) as a function of temperature for various Gd concentrations. (No ordering effects were observed down to 6°K.)

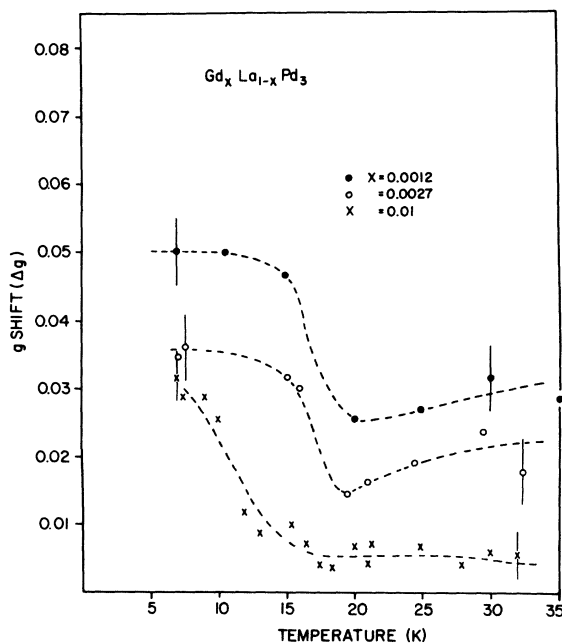


FIG. 3. g shift of Gd in $\text{Gd}_x\text{La}_{1-x}\text{Pd}_3$ annealed at 740°C as a function of temperature. (Below 20°K g shifts increase with the decreasing temperature due to ordering effects.)

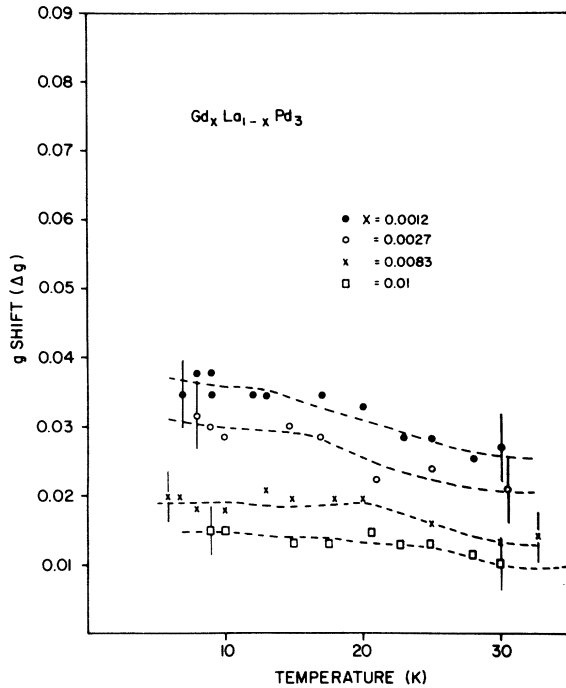


FIG. 4. g shift of Gd in $Gd_xLa_{1-x}Pd_3$ annealed at $840^\circ C$ as a function of temperature (there are no ordering effects).

the g value of pure Gd ion (Gd^{3+}) 1.993. The values on ΔH and Δg in Figs. 1 and 3 were obtained from the set of samples annealed at $740^\circ C$ and those in Figs. 2 and 4 were obtained from the set of samples annealed at $840^\circ C$. The minimum values of ΔH and Δg appear around $20^\circ K$ for the set of samples annealed at $740^\circ C$.

IV. INTERPRETATION OF THE RESULTS

A. Thermal broadening, ΔH

All the samples have shown Korringa-like thermal broadening of the line, exhibiting linear temperature dependence of the line width ΔH . Plots of linewidth versus temperature are presented in Figs. 1 and 2. To calculate the bottleneck factor x we have taken into account only the linewidths above $20^\circ K$. The function $\phi(x, y)$ in Eq. (3a) contains the parameter y which is temperature dependent and may become dominant at low temperatures. Therefore, we can ignore y at high temperatures such as $20^\circ K$. With that approximation we have estimated the bottleneck factor x for the two sets of samples. We have presented $\Delta H/\Delta T$ values for each concentration of Gd^{3+} in the two sets of samples in Table I. In the first set of samples we have observed ordering effects set-in in the low-temperature region. Therefore, the line-

TABLE I. ESR linewidth of Gd in $Gd_xLa_{1-x}Pd_3$ as a function of Gd concentrations. Results in the last two columns are calculated by solving for the coefficients of Eq. (3) using the values for the samples containing 0.12- and 0.27-at.% Gd.

Conc. (at.%)	$\Delta H/\Delta T$ (expt.)	$\Delta H/\Delta T$ (calc.)	x (δ_{ei}/δ_{ei})
(a) Samples (annealing temperature $740^\circ C$)			
0.12	5.2	5.2	3.66
0.27	4.1	4.1	1.63
1.0	2.2	2.0	0.44
3.0	1.3	0.85	0.15
(b) Samples (annealing temperature $840^\circ C$)			
0.12	7	7	4
0.27	5.6	5.6	1.78
0.83	3.6	3.2	0.57
1.0	3.0	2.84	0.48
3.0	1.3	1.2	0.16

widths in Fig. 1 have started to increase in the low-temperature region (T below $20^\circ K$), deviating from the Korringa-like behavior. We have not taken into account the low-temperature results on ΔH in our calculations. The linewidth in Fig. 2, however, did not show such a deviation even at $6^\circ K$. The results in the last two columns of Table I are calculated by solving for the coefficients of Eq. (3) using the values for the samples containing 0.12- and 0.27-at.% Gd. The parameter x was found to be 3.66, and $\Delta H_k/\Delta T = 6.6$ for the sample with 0.12-at.% Gd^{3+} annealed at $740^\circ C$. Likewise the x was found to be 4 and $\Delta H_k/\Delta T = 8.7$ for the sample of the same concentration as above but annealed at $840^\circ C$. The minimum value of $\Delta H/\Delta T$ is $1.3 G^\circ K$ for the samples with 3.0-at.% Gd in both temperatures. The results presented in Figs. 1 and 2 indicated the presence of bottleneck in high impurity samples, as seen from Eq. (6). The straight-line plots of Figs. 1 and 2, when extrapolated to $0^\circ K$ exhibit residual linewidths in each and every sample which may be interpreted as being due to the dipolar broadening and the unresolved hyperfine structure.

Equation (3a) for small values of x , with $\phi(x, y) = 1$ gives

$$\gamma \Delta H \approx \chi \delta_{ie} = (\pi/\hbar) \chi \eta^2 J^2 k \Delta T. \quad (9)$$

Using Eq. (9) and the $\Delta H/\Delta T$ values from Figs. 1 and 2 we have calculated as follows:

$$J = +0.16 \pm 0.03 \quad \text{for samples annealed at } 740^\circ C,$$

$$J = +0.15 \pm 0.03 \quad \text{for samples annealed at } 840^\circ C.$$

(10)

J was positive because of the positive g shifts.

From the measurements of the magnetic susceptibility¹⁶ and the specific heat¹⁷ in LaPd₃ systems as reported by Hutchens *et al.* there is some evidence that the enhancement effects may be present in these systems. Since the density of states, $\eta = 0.71$ states/eV atom spin, computed from the magnetic susceptibility data¹⁶ is larger than $\eta = 0.12$, obtained from the specific-heat data,¹⁷ the enhancement factor α is 0.8 and $K(\alpha)$ is 0.24.¹¹ From Eq. (8), $J_{\text{eff}}(0)$ values are

$$J_{\text{eff}}(0) = 0.8 \text{ eV for samples annealed at } 740^\circ\text{C},$$

$$J_{\text{eff}}(0) = 0.75 \text{ eV for samples annealed at } 840^\circ\text{C}.$$

One must not put too much emphasis on the accuracy of the enhanced values because those samples (LaPd₃ system) usually contain other paramagnetic impurities.

B. g shifts, Δg

In Table II we have presented the g shifts as a function of concentration of Gd³⁺ for the two sets of samples. The results in the last columns are calculated using Eq. (2) with $y = 0$, $x = 3.66$ in Table II(a), $x = 4$ in Table II(b), and fitting the values for the samples containing 0.12-at.% Gd. Our measurements of the g shifts of Gd as a function of temperature in the LaPd₃ system annealed at 740 and 840 °C are presented in Figs. 3 and 4. The g shifts for the two samples (0.12- and 0.27-at.% Gd³⁺) annealed at 740 °C, are found to be dependent on the concentration of Gd³⁺, whereas the sample with 1.0-at.% Gd³⁺ exhibits very little g shifts from that of the free ion around 30 °K. From these results in Fig. 3 we conclude that the latter sample suffers from a bottleneck effect. Similar analysis

of the g shifts for the second set of samples (annealed at 840 °C) indicates that here again the sample with high Gd³⁺ concentration (1.0-at.% Gd³⁺) suffers from the bottleneck effect (in Fig. 4). The maximum g shift measured at 30 °K was found to be 0.046 for the sample with 0.12-at.% Gd annealed at 740 °C and 0.04 for the sample of the same concentration annealed at 840 °C.

So far only two systems,⁴ Gd³⁺ in LuAl₂ and Eu²⁺ in Yb, were reported to have exhibited the dynamic effect. Our samples with the high impurity concentrations do not have g shifts and no dynamic effects are expected in them. Those with lower impurity concentrations exhibiting the g shifts such as samples with 0.12- and 0.27-at.% Gd³⁺ are expected to show such dynamic effects, but they did not. In these latter samples, g shifts should have decreased with decreasing temperature according to Eq. (2) if there exists a dynamic effect. Actually the g shifts observed have increased with decreasing temperature below 20 °K. Therefore, these g shifts cannot be attributed to the dynamic effect. They are due to ordering effects. Above 20 °K, within the experimental error, the samples were found to exhibit no change in g shift as a function of temperature in Figs. 3 and 4.

C. Conduction-electron relaxation rate

As explained earlier in the paper the δ_{e1} consist of the relaxation rate δ_{e1}^0 , characteristic of the undisturbed host lattice and due to the spin-flip scattering by the magnetic and nonmagnetic impurity. The latter relaxation rate δ_{e1s} is dependent on the concentration of the magnetic impurity Gd³⁺. We then have

TABLE II. (a) g shift^a of Gd in Gd_xLa_{1-x}Pd₃ (annealing temperature at 740 °C) as a function of Gd concentrations. Results in the last column are calculated using Eq. (2) with $y = 0$ and $x = 3.66$, and fitting the value for the sample containing 0.12-at.% Gd. (b) g shift^a of Gd in Gd_xLa_{1-x}Pd₃ (annealing temperature at 840 °C) as a function of Gd concentrations. Results in the last column are calculated using Eq. (2) with $y = 0$ and $x = 4$, and fitting the value for the sample containing 0.12-at.% Gd.

Conc. (at.%)	Δg (expt. at 10 °K)	Δg (expt. at 30 °K)	Δg (calc. at 30 °K)
(a)			
0.12	0.049	0.028	0.028
0.27	0.034	0.020	0.017
1.0	0.025	0.005	0.004
(b)			
0.05	0.04	0.034	0.032
0.12	0.034	0.025	0.025
0.27	0.03	0.020	0.016
0.83	0.019	0.013	0.005
1.0	0.014	0.010	0.004

^a Experimental error in Δg is ± 0.005 .

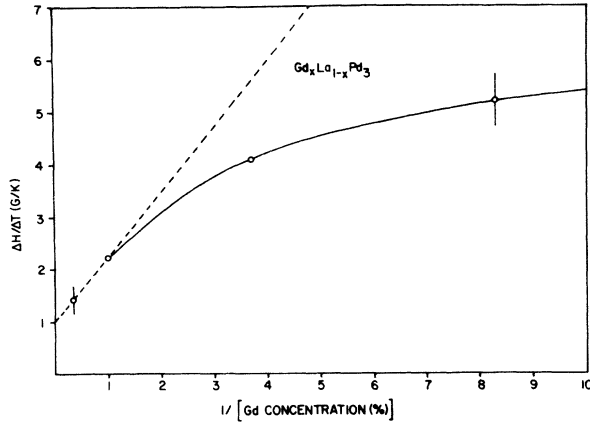


FIG. 5. Thermal broadening $\Delta H/\Delta T$ of Gd in $Gd_xLa_{1-x}Pd_3$ (annealing temperature at $740^\circ C$) as a function of $1/C_i$ (C_i : Gd concentration). Slope of dashed line gives δ_{ei}^0 and its intercept with the vertical axis yields $\partial(\delta_{ei})/\partial(C_i)$ according to Eq. (10).

$$\delta_{ei} = \delta_{ei}^0 + \delta_{eis} = \delta_{ei}^0 + \frac{\partial(\delta_{ei})}{\partial(C_i)} C_i. \quad (11)$$

From the values of intercept of the straight-line plots in Figs. 5 and 6, we have estimated the relaxation rates of the conduction electrons to the lattice due to the spin-flip scattering by Gd^{3+} . In the above calculation we have used $\eta = 0.12 \text{ eV}^{-1}$ per atom and per spin direction for the density of states. This value was computed using the electronic specific-heat data of Hutchens *et al.*¹⁷

$$\frac{\partial(\delta_{ei})}{\partial(C_i)} = 5.2 \times 10^{11} \text{ sec}^{-1}/\text{at.}\%Gd,$$

for the samples from the first set ($740^\circ C$).

$$\frac{\partial(\delta_{ei})}{\partial(C_i)} = 3.9 \times 10^{11} \text{ sec}^{-1}/\text{at.}\% Gd,$$

for the samples from the second set ($840^\circ C$). Also we estimated from the slopes of the straight lines in Figs. 5 and 6 that the relaxation rates of the conduction electrons due to the undisturbed host lattice are

$$\delta_{ei}^0 = 6.5 \times 10^{11} \text{ sec}^{-1}$$

for samples annealed at $740^\circ C$,

$$\delta_{ei}^0 = 9.3 \times 10^{11} \text{ sec}^{-1}$$

for samples annealed at $840^\circ C$.

The conduction-electron relaxation rate δ_{ei} increases in the presence of additional nonmagnetic impurities. They produce additional spin-flip scattering. Therefore, this will result in the increase of x and may open up the bottleneck present in high Gd^{3+} concentration samples. Thorium

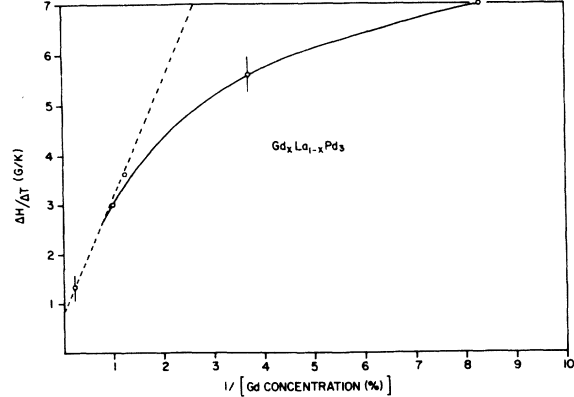


FIG. 6. Thermal broadening $\Delta H/\Delta T$ of Gd in $Gd_xLa_{1-x}Pd_3$ (annealing temperature at $840^\circ C$) as a function of $1/C_i$. (C_i : Gd concentration.)

is highly soluble in La and nonmagnetic. We added 4- and 8-at. % Th to 1.0- and 2.0-at. % Gd^{3+} doped samples of $LaPd_3$ to form the intermetallic $Gd_xTh_yLa_{1-x-y}Pd_3$. Samples with 1.0- and 2.0-at. % Gd^{3+} concentration, which were bottlenecked in the absence of Th, are found to exhibit g shifts and an increase in their linewidths in the presence of Th, thereby indicating the opening up of the bottleneck.

V. CONCLUSIONS

Although the experimental parameters such as ΔH and Δg were found to be sensitive to the annealing temperature of the samples, the average J values obtained for both of the sets are essentially the same within the experimental error. The different annealing temperatures (740 and $840^\circ C$) seem to have a significant effect in the structural composition of the lattice. This indeed can be seen from the changes in ΔH and Δg . But with such structural change, there seems to be a change in $x (= \delta_{ei}/\delta_{ei}^0)$ also, which contains the electron-electron lattice relaxation rate δ_{ei} . This may be resulting in the same average J values for both sets of samples. The J values calculated are comparable to these obtained in $Gd_xLa_{1-x}Al_z$ and $Gd_xLu_{1-x}Al_z$ systems. We have not taken into account the q dependence of J because of the uncertainty involved in the purity of La and the resulting discrepancy in the density of states η obtained from the susceptibility and the specific-heat measurements. The contribution by the host lattice to the conduction-electron relaxation rates δ_{ei}^0 are $6.5 \times 10^{11} \text{ sec}^{-1}$ for the first set of samples and $9.3 \times 10^{11} \text{ sec}^{-1}$ for the second set. This clearly demonstrates the change in the lattice structure due to annealing.

The intermetallic systems of LaPd₃ doped with Gd are found to exhibit no dynamic effects above 20 °K. The temperature dependence of the g shift observed in samples annealed at 740 °C below 20 °K is very pronounced and in reverse direction to the one expected from dynamic effects. This may be due to the ordering effects that set in at low temperature. The changes in linewidth with temperature in the samples annealed at 740 °C show similar ordering effects. The linewidths of samples annealed at 840 °C were found to exhibit Korringa-like behavior down to 6 °K. The g shift of the second set as a function of temperature below 20 °K did not exhibit such a pronounced change as these samples annealed at 740 °C, and within experimental error no dynamic effect can

be attributed to them in the range between 6 and 20 °K. This indicates that the samples annealed at 740 °C may be structurally ordered to a lesser extent than the ones annealed at 840 °C. Therefore, annealing temperature does play a significant role in such studies. Studies are in progress on the samples annealed at 940 °C. In spite of such changes, it is just a coincidence that we have obtained the same average J values in both sets of samples.

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