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Magnetic properties of very dilute Pd(Gd) alloys

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Measurements of the magnetic moment $\sigma(H, T)$ of $\mathrm{Pd}_{1-x}\mathrm{Gd}_x$ alloys were made for $0.0003 \leq x \leq 0.02$ in applied fields H up to 215 kG and for $1.3 \leq T \leq 4.2$ K. The data were analyzed using four different formulas which permitted extrapolation of σ to very large fields so that the saturation moment μ_s and matrix susceptibility χ_{mat} could be determined. Detailed data analysis shows that: (a) $\mu_s \simeq 7\mu_B/(\mathrm{Gd\ atom})$ for all the alloys studied, (b) $\chi_{\mathrm{mat}} \leq \chi_{\mathrm{Pd}}$ for all the alloys and (c) the Gd moment is more difficult to saturate at large x. The most satisfactory fits to the data were obtained with the expression $\sigma = \{\alpha H/[H(H+H_0)]^{1/2}\}B_{7/2}(J\beta[H(H+H_0)]^{1/2}) + \chi_{\mathrm{mat}}H$, where α, β, H_0, and χ_{mat} are fitted parameters and $B_{7/2}(y)$ is the Brillouin function for $J = 7/2$. A model is presented in which $(HH_0)^{1/2}$ is associated with an average anisotropy-like field transverse to the applied field. A paramagnetic Curie temperature is extracted from the parameter β, which also includes s-f exchange effects. Some discrepancies with previously published results are discussed.

I. INTRODUCTION

In a recent paper[1] we presented results of high-field measurements of the magnetic moment of a series of Pd–rare-earth alloys, with rare-earth concentrations of about 1 at.%. The general features of the magnetization versus magnetic field could be described by a semiempirical formula which permitted determination of the saturation moments of the rare-earth ions. The derived saturation moments generally were equal to the free-ion rare-earth values. Earlier studies[2] (for which the applied magnetic field $H \leqslant 20$ kG did not completely saturate the Gd moment) had suggested that the magnetic moment per Gd atom in Pd decreased with decreasing Gd concentration, implying a negative conduction-electron polarization. We found the Gd moment in Pd was not fully saturated at 4.2 K even at fields up to 215 kG; thus, as indicated in Ref. 1, no evidence for negative polarization was detected. Furthermore, studies of the electrical resitivity[3] did not show the anomaly that should result from a negative electron polarization. However, the magnetic properties of Gd in Pd for Gd concentrations well below 1 at.% remained a problem; analysis of our magnetic measurements using the semiempirical formula indicated that the saturation moment per Gd atom seemed to increase with decreasing Gd concentration.[4] Because such a behavior would suggest a giant moment for very dilute concentrations of Gd in Pd, we have reexamined alloys of $\mathrm{Pd}_{1-x}\mathrm{Gd}_x$ for $0.0003 \leqslant x \leqslant 0.02$.

In this paper we summarize the results of these measurements. Within the present limitations of our experiments, we find that there is no convincing evidence for an increased moment above the Gd^{3+} free-ion value. The limitations of the analysis of the present data for $x \leqslant 0.001$ involve the competition of the relatively large Pd host susceptibility with the very small contribution of the dilute Gd moment, as well as the limits of the determination of the Gd concentration. Although the data unambiguously indicate that the free-ion value of Gd^{3+} is a lower bound for the lower Gd concentrations, the upper bound is set by experimental uncertainties. The results obtained employing other empirical formulas in addition to the one used in Ref. 1 are also compared, and the limitations of the present studies are summarized.

In Sec. II we very briefly summarize the experimental details. In Sec. III the experimental results are analyzed by employing three other empirical formulas in addition to the one used in Ref. 1. Also the magnetic properties for $T < 4.2$ K are discussed. Based on these studies, we find that the results obtained with one of the formulas (involving a modified Brillouin function and an *ad hoc* anisotropic contribution) yield the most consistent analysis. In Sec. IV, the various parameters that enter into this formula are determined and values for the paramagnetic Curie temperature Θ are obtained. In Sec. V we discuss magnetic measurements performed in $\mathrm{Pd}_{1-x}\mathrm{Gd}_x$ alloys by others.

II. EXPERIMENTAL DETAILS

The polycrystalline $\mathrm{Pd}_{1-x}\mathrm{Gd}_x$ alloys were prepared by arc-melting in an argon atmosphere.[1] Based on the weight loss incurred during fabrication (and assuming all the weight loss was due to Gd loss), we estimate that the Gd concentration could be in error by as much as 10% in some cases, but in general was less than 5%. In this paper we quote the nominal concentration, x, based on the measured weights of the constituents used for arc-melting.

The measurements of the magnetic moment were carried out as described in Ref. 1. High-magnetic-field data to 215 kG were obtained in high-power, watercooled Bitter solenoids with a very low-frequency vibrating-sample magnetometer.[5] High-resolution data up to 55 kG were obtained with a conventional vibrating-sample magnetometer adapted to a superconducting magnet. The low-field data also were used to normalize the lower-resolution high-field data. An average of 30 points were used to fit the data for each sample.

III. DISCUSSION OF RESULTS

Because the fields necessary to saturate the Gd moment were in excess of 200 kG at 4.2 K, it was imperative to develop an approximate analytical expression in order to fit the data and allow an analytical extrapolation to very large fields.[1] A semiempirical expression developed earlier,[6] and used extensively by us, is

$$\sigma = \frac{AH}{1+B|H|} + \chi_{\text{mat}} H, \tag{1}$$

where σ is the magnetic moment, χ_{mat} is the matrix susceptibility for the Pd(Gd) alloy, A is the initial susceptibility of the Gd, and $\mu_s = A/B$ is the saturation magnetization of the Gd. Application of this formula to magnetic data[1] for $\mathrm{Pd}_{1-x}\mathrm{Gd}_x$, as well as for other rare-earth–Pd alloys in the concentration range $0.005 \leqslant x \leqslant 0.02$ showed that Eq. (1) fit the measured results well, and gave $\mu_s \simeq 7\mu_B/$(Gd atom) and $\chi_{\text{mat}} < \chi_{\text{Pd}}$.

Equation (1) also provided a good fit to the magnetic data for more dilute $\mathrm{Pd}_{1-x}\mathrm{Gd}_x$ ($0.0003 \leqslant x \leqslant 0.005$), but a general trend was noted[4]: as x decreased, μ_s and χ_{mat} increased. The increase of χ_{mat} is understandable since χ_{mat} will approach χ_{Pd} when $x \to 0$. However, the increase in the value of μ_s was substantial and exceeded the uncertainty of x. We obtained $\mu_s \approx 10.5\ \mu_B/$(Gd atom) for $x = 0.0003$. This value is 50% higher than the Gd^{3+} free-ion value of $7\ \mu_B/$(Gd atom).

A more detailed analysis of the data presented here suggests that the increase in the value of μ_s with decreasing concentration [when Eq. (1) is employed] is spurious, and that within our experimental error, for $0.0003 \leqslant x \leqslant 0.02$, the saturation moment of Gd in Pd is $\mu_s \simeq 7\mu_B/$(Gd atom). This conclusion is based on the examination of (i) more detailed measurements and reexamination of the high-field magnetization at 4.2 K, (ii) further measurements at temperatures down to 1.3 K, and (iii) fits of the data using other analytic expressions.

We measured the magnetization up to 150 kG for $x = 0.0003$ and $x = 0.0005$ for $T = 4.2$ K and 1.3 K. The analysis of the 1.3 K data for $x = 0.0003$ and 0.0005, using Eq. (1), produced a lower value for μ_s and a higher χ_{mat} than the 4.2 K data. Clearly, this result, which is based on Eq. (1), is not reasonable. In order to observe the temperature dependence of the approach to saturation, the high-field magnetization (from 20 to 215 kG) was measured for $x = 0.0005$ at 4.2 K and 1.5 K. For these measurements the sample was not removed from the apparatus; the 4.2 K data was taken as a function of the applied field, and subsequently we pumped on the He and repeated the measurements as a function of field at 1.5 K. In this way we ensured that ratios of the raw data at $T = 4.2$ K and 1.5 K showed only the T dependence of the magnetization. At 1.5 K more rapid saturation occurred; the moment at 28.5 kG was 12% larger at 1.5 K than at 4.2 K, but at high fields (from 75 to 215 kG) the 1.5 and 4.2 K results were identical within the noise of the measurements.

In order to study the limitations of Eq. (1), three other analytical expressions were employed:

$$\sigma = \frac{AH}{1+B(H^2+H_0^2)^{1/2}} + \chi_{\text{mat}} H, \tag{2}$$

$$\sigma = \frac{\alpha H}{(H^2+H_0^2)^{1/2}} B_J\left(J\beta(H^2+H_0^2)^{1/2}\right) + \chi_{\text{mat}} H, \tag{3}$$

and

$$\sigma = \frac{\alpha H}{[H(H+H_0)]^{1/2}} B_J\left(J\beta[H(H+H_0)]^{1/2}\right) + \chi_{\text{mat}} H, \tag{4}$$

where $\boldsymbol{B}_J(y)$ is the Brillouin function for angular momentum J, and A, B, α, β, and H_0 are parameters which are fit separately for each equation. The rationale for the alternative expressions can be understood by comparing them with Eq. (1). Equation (2) simply removes the $H = 0$ singularity in the simple Padé approximation given by Eq. (1). This is done by replacing the term $|H|$ by $(H^2+H_0^2)$.$^{1/2}$ Both equations are identical when $H_0 = 0$. Equations (3) and (4) may be explained if we assume that the conduction-electron–$4f$-electron (s-f) interaction produces an average anisotropy field, $\langle H_{\text{an}}^2\rangle^{1/2}$, which is transverse to the external field $\boldsymbol{H}$. (The source of $\langle H_{\text{an}}^2\rangle$ may also be Gd-Gd interactions.) Figure 1 shows the relative orientations of $\langle H_{\text{an}}^2\rangle^{1/2}$, H_{eff}, and the applied field H. In the molecular-field approximation the Gd magnetization is along $H_{\text{eff}} = (H^2 + \langle H_{\text{an}}^2\rangle)^{1/2}$. For a random, polycrystalline, dilute alloy, there is no preferred transverse direction, and the transverse component of the magnetization should average to zero. Thus only the component along H is detected, and this component is given by

$$\sigma_{\mathrm{Gd}}=\frac{\alpha H}{(H^2+\langle H_{\mathrm{an}}^2\rangle)^{1/2}}\,B_J\,(J\beta(H^2+\langle H_{\mathrm{an}}^2\rangle)^{1/2})\,. \tag{5}$$

Here we also assume that the magnetization follows a Brillouin function. For Eq. (3) we choose $\langle H_{\mathrm{an}}^2\rangle = H_0^2$, independent of the Pd matrix magnetization, whereas for Eq. (4) we choose $\langle H_{\mathrm{an}}^2\rangle = HH_0$, which is proportional to the Pd matrix magnetization. Although this latter assumption may be more reasonable, both Eqs. (3) and (4) are based on the introduction of an anisotropy field which is transverse to the applied field.

It is interesting to note that if $H \gg H_0$, Eqs. (2) and (4) reduce to Eq. (1). Furthermore, for $H \to \infty$, Eqs. (2)-(4) all reduce to Eq. (1).

When considering the application of Eqs. (1)-(4) to the analysis of the magnetization data for the most dilute samples ($x = 0.0003$, and $x = 0.0005$), it should be noted that the Pd host magnetization is much larger than the Gd contribution. Therefore, the parameters A, B, α, β, and H_0 are deduced from the very small curvature (the nonlinear Gd contribution) superimposed on the linear dependence of σ versus H for the Pd matrix.[7] For $x > 0.005$ the Gd contribution is large and, therefore, our confidence in these parameters is much higher.

Our data cover a wide range of applied fields and concentrations and we would expect that they would permit a clear choice between the four analytic expressions used to fit the data. An extensive compilation of the results (not given in detail here) shows that: (a) For all the data from 16 to 215 kG, good fits are obtained for Eqs. (1)-(4); the rms errors of the fits at 4.2 K are about 0.3%. Thus, for this field range, the quality of a fit as reflected in the rms errors gives no guide for selecting a best expression. (b) Equation (4) gives the most satisfactory description of all the data when *all values* of x, H, and T are considered. When Eqs. (1)-(4) were fit to the data over the wider field interval, $4 \leqslant H \leqslant 215$ kG, the rms errors increased, but the rms errors for fits of Eq. (4) were substantially smaller than for Eqs. (1)-(3). This feature of Eq. (4) was especially noticeable when the results for high-resolution data up to 50 kG were compared. In this field range, the data were taken with the high-resolution vibrating-sample magnetometer for which we expect the data to have a higher degree of internal consistency. This was confirmed by the smaller rms errors compared to those obtained for data up to 215 kG. Similarly, Eq. (4) gave the best fits when data taken at 1.3 K were analyzed over the field range of $4 \leqslant H \leqslant 150$ kG. (c) Although in some circumstances it was difficult to determine a set of parameters that optimized the fit of Eq. (3) to the data, Eq. (4) always allowed an optimum fit. (d) For $x = 0.01$ and 0.02, Eqs. (1) and (2) gave identical results, i.e., the best fit for Eq. (2) occurs for $H_0 = 0$.

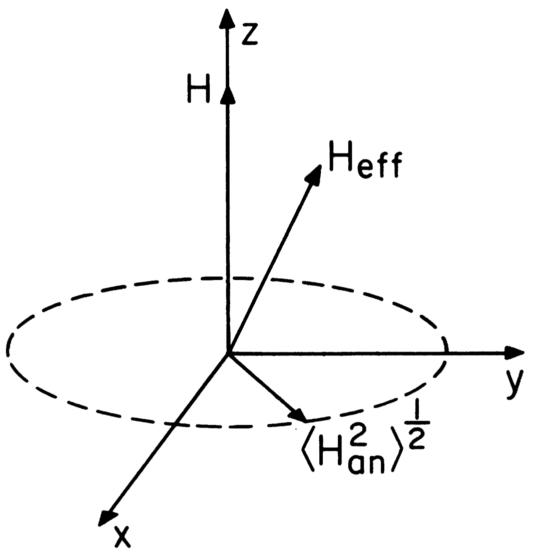

FIG. 1. Illustration of the applied magnetic field H, transverse anisotropy field $\langle H_{\mathrm{an}}^2\rangle^{1/2}$, and resultant effective field H_{eff}. The average Gd moment is assumed to be aligned along the instantaneous H_{eff} and to have a temperature and magnetic-field dependence given by a modified Brillouin function [see Eqs. (3) and (4)]. Because $\langle H_{\mathrm{an}}^2\rangle^{1/2}$ can have an arbitrary direction in the xy plane, the measured time-averaged Gd moment is the z component of the effective moment along H_{eff}.

In order to compare the results of the analyses using Eqs. (1)-(4), derived values of μ_s and χ_{mat} are shown in Table I for four representative concentrations. These particular analyses have been performed using magnetization data for $16 \leqslant H \leqslant 215$ kG. This low-field cutoff was made in order to emphasize the high-field results, and because Eqs. (1)-(4) do not give equally good fits to the data at low fields. The general features of the results in Table I show that the presence of the H_0 term always reduces μ_s and the reduction of μ_s is always accompanied by an increase in χ_{mat}. For $x = 0.0003$ and 0.0005 this increase in χ_{mat} is very small, but nonetheless this small increase is reflected in a very significant reduction of μ_s.

In Fig. 2 we show the values of μ_s obtained from the fits of Eqs. (1)-(4) as well as the limits of errors of μ_s. The indicated error ranges of μ_s (dashed lines) reflect estimates of uncertainties in the parameters derived from the fits as well as uncertainties in concentration. Note the larger errors at lower x. It is clear that the weight of the results points to $\mu_s \simeq 7\mu_B$/(Gd atom), independent of Gd concentration for $x \leqslant 0.02$. This conclusion

TABLE I. Saturation moment μ_s, and matrix susceptibility χ_{mat}, calculated with Eqs. (1)–(4). The results for four representative $\mathrm{Pd}_{1-x}\mathrm{Gd}_x$ alloys are tabulated.

	μ_s [μ_B/(Gd atom)]				χ_{mat} (10^{-6} emu/gG)			
x	Eq. (1)	Eq. (2)	Eq. (3)	Eq. (4)	Eq. (1)	Eq. (2)	Eq. (3)	Eq. (4)
0.0003	10.7	8.7	7.4	8.2	6.40	6.48	6.54	6.50
0.0005	9.8	8.0	7.2	7.6	6.55	6.63	6.68	6.67
0.0035	8.2	7.2	6.4	6.9	5.43	5.89	6.28	6.06
0.01	7.0	⋯	5.8	6.5	4.42	⋯	5.79	5.01

is reinforced by the evidence mentioned above about the overall applicability of Eq. (4) to the present data.

IV. FURTHER DISCUSSION OF EQ. (4)

Because we have found that Eq. (4) gives the best analytical description of the data, we will examine its coefficients in more detail. According to our simple phenomenological derivation of Eq. (4), the coefficient α is given by

$$\alpha = N g_J J \mu_B , \tag{6}$$

where N is the number of Gd atoms, $g_J = 2$ is the Landé g factor, $J=\frac{7}{2}$ is the angular-momentum quantum number for Gd^{3+}, and μ_B is the Bohr magneton. Our justification for writing Eq. (6) is based on the conclusions of Sec. III where we showed that $\mu_s \simeq g_J J \mu_B = 7\mu_B/(\mathrm{Gd\ atom})$.

For no Gd-Gd interaction and no s-f interaction, the coefficient β equals $g_J \mu_B / k_B T$, where k_B is the Boltzmann constant and T is the absolute temperature. For $x=0.0003$ and 0.0005 and at a temperature $T=4.2$ K, the value of β determined from the fit is $\beta \simeq 0.028/$ kG, whereas the value that results from the expression above is $\beta = 0.032/$ kG. The results of the EPR of very dilute Gd in Pd gave a value for the exchange[8] of -10% of the applied field. When this exchange correction is included, the two values of β agree within experimental error. This result suggests the following expression for β:

$$\beta = g_J \mu_B (1-\delta)/k_B (T-\Theta), \tag{7}$$

where the factor $1-\delta$ accounts (to first order) for the s-f exchange interaction, and Θ is the paramagnetic Curie temperature that results from Gd-Gd interactions. The parameters $1-\delta$ and Θ can be determined by other measurements: the concentration dependence of the EPR g factor[9,10] of Gd in Pd determines $1-\delta$, and temperature-dependent magnetic-susceptibility measurements[6] may be used to determine Θ.

For $x \leqslant 0.02$, the measured g factor, g_{eff}, is approximately linear with concentration,[9,10]

$$g_{\mathrm{eff}} \approx 1.800 + 4.5x , \tag{8a}$$

so that

$$1-\delta = g_{\mathrm{eff}}/g_J = g_{\mathrm{eff}}/2 . \tag{8b}$$

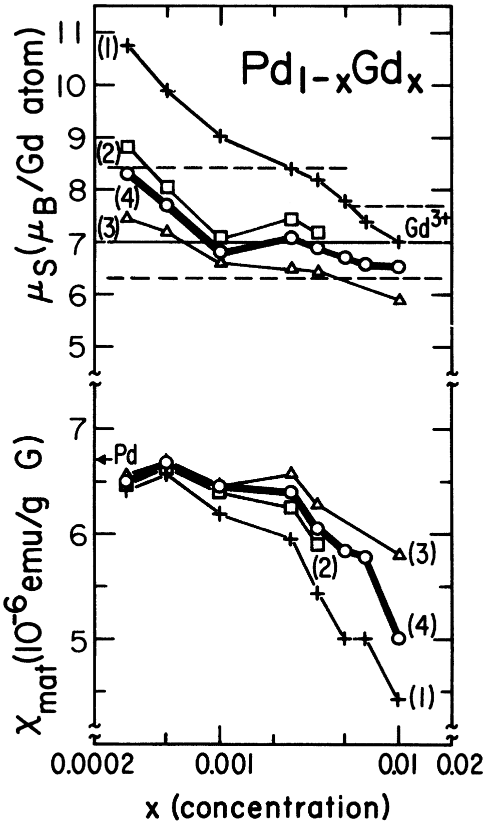

FIG. 2. Results of fits of the magnetization data at 4.2 K for $\mathrm{Pd}_{1-x}\mathrm{Gd}_x$ to Eqs. (1)-(4). Upper curves: saturation moment μ_s vs concentration x. The fits derived from Eq. (4) are the most satisfactory and these results are shown with a heavier line. The dashed lines show our estimates of the uncertainties of μ_s arising from errors in the fit and the concentration. Note the larger uncertainties at lower concentrations. The solid horizontal line indicates the free-ion value for Gd^{3+}. Lower curves: matrix susceptibility χ_{mat} vs concentration x. The value of χ_{Pd} is also indicated.

TABLE II. Parameters β and H_0 derived from fits of Eq. (4) to the magnetization data at 4.2 K for $Pd_{1-x}Gd_x$ alloys. The parameter Θ was calculated from Eqs. (7) and (8).

x	0.0003	0.0005	0.0010	0.0025	0.0035	0.0050	0.0065	0.01	0.02
β (1/kG)	0.028	0.028	0.031	0.037	0.039	0.043	0.049	0.061	0.127
Θ (K)	···	···	0.3	0.9	1.1	1.4	1.7	2.2	3.2
H_0 (kG)	7	8	3	11	13	14	14	17	16

In Table II we list the calculated values of β and Θ for all our samples. The values of Θ have been determined by the use of Eqs. (7) and (8) using the values of β determined from the fit of Eq. (4) to magnetization data taken at $T=4.2$ K and $4 \leqslant H \leqslant 215$ kG. (For the purposes of this paper we have analyzed measurements on paramagnetic samples only, and have not concerned ourselves with the magnetic ordering temperature T_c. However, for completeness we discuss some results of T_c measurements in Ref. 11.)

The parameter H_0 also given in Table II is concentration as well as temperature dependent. It is also possible that the spread in the values of H_0 as a function of x may be dependent on sample preparation. We have not investigated this possibility. The effect of the general increase of H_0 with increasing x is evident in the high-field properties of the alloys; the Gd is more difficult to saturate as x increases. This trend is contrary to that expected for the increase of Θ with x.

H_0 is most elusive to theoretical interpretation. Our assumption that the anisotropy field is proportional to the band susceptibility led to the expression $\langle H_{an}^2 \rangle = HH_0$. We expect that for Pd this linear analytical form applies for fields of at least 300 kG, because it was shown[7] that χ_{Pd} was independent of H up to 150 kG, and in Ref. 12 it was shown that the field dependence of χ_{Pd} is very small up to 325 kG.

As indicated previously, χ_{mat} is the band susceptibility of the alloys. As seen in Table I, χ_{mat} varies appreciably with x. Because the band susceptibility of Pd is strongly exchange enhanced, the changes of χ_{mat} must reflect both changes in the exchange enhancement as well as band-filling effects.

V. SUMMARY OF OTHER EXPERIMENTAL RESULTS

The results of magnetic-moment measurements on dilute rare-earth–palladium alloys have been used in attempts to interpret the results of EPR measurements[13-15] on similar alloys. One recurring problem has been the explanation of the large negative g shift for dilute Gd in Pd, and a negative polarization of the conduction electrons at the Gd site has been invoked to explain this effect. Because there have been some problems in interpreting published magnetic data for Pd(Gd), we will briefly discuss these data here.

Peter *et al.*[13] and Shaltiel *et al.*[14] first reported magnetic data for $x=0.03$ for which values of paramagnetic Curie temperature Θ between +12 K (Ref. 13) and +3 K (Ref. 14) and $\mu_{eff}=6.28\mu_B$/(Gd atom) (Ref. 14) were presented $\{\mu_s=\mu_{eff}[J/(J+1)]^{1/2} = 5.5\mu_B/(\text{Gd atom})\}$.

A more extensive range of Gd concentrations $(0.02 \leqslant x \leqslant 0.10)$ was studied by Crangle[2] for fields $H \leqslant 20$ kG, and a concentration dependent μ_s was reported. In Ref. 1 we criticized this apparent concentration dependence of μ_s and showed that the reduced values of μ_s were the result of incomplete magnetic saturation of the Gd for $H \leqslant 20$ kG. Despite our discussion, Crangle's results continue to be quoted in the literature as evidence for a reduced Gd moment.

Schaller *et al.*[16] measured the magnetic moments of high-concentration $Pd_{1-x}Gd_x$ alloys for $0.025 \leqslant x \leqslant 0.12$ (0.12 is the limit of Gd solubility in Pd). They concluded that μ_s was *independent* of x and about $6.5\mu_B$/(Gd atom). From their discussion, a magnetic-moment deficiency, $\Delta = \mu_s - g_J J \simeq -0.5$ μ_B/(Gd atom), is assumed to meaningful. However, we believe that the scatter of the data (Fig. 2, Ref. 16) and their questionable correction for the matrix contribution to the total moment place the Gd moment deficiency within experimental error.

Taylor and Coles[15] recently compared the normalized g shift for Gd in Pd with the apparent magnetic-moment deficiencies in the limit $x \to 0$. They quote moment deficiencies of −22%, −18%, and −20% for Refs. 14, 16, and 2, respectively. (However, for Ref. 16 the correct deficiency is −7%.) It is puzzling why Taylor *et al.* (Ref. 15, p. 311) compare Crangle's magnetic data for $x=0.075$ at 1.75 K (for which the alloy is ferromagnetic) with our low-x alloy results, for which the alloys are paramagnetic. Furthermore, it is difficult to understand why these and other authors use magnetic data for large-x alloys in order to interpret EPR results for extremely dilute alloys.

Most recently, Moret *et al.*[17] compare a calculated conduction-electron polarization of $-0.4\mu_B$/

(Gd atom) with $\Delta = -1.4\mu_B$/(Gd atom) obtained by extrapolation from Crangle's[2] low-field data. They say that $-0.4\mu_B$/(Gd atom) "... is a little smaller than that found by Crangle [Ref. 2], but exceeds the more recent value (zero) extracted by Guertin *et al.*[Ref. 1] ..." These authors also quote the results of Taylor and Coles[15] which were discussed above.

The results presented in this paper clearly indicate that $\Delta = 0$ within our experimental error. Thus we conclude that the conduction-electron polarization effects invoked so far to explain the g shifts are not supported by the magnetic data for $Pd_{1-x}Gd_x$.

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[11] J. Crangle (Ref. 2) and L. L. Isaacs and J. Crangle (private communication) determined T_c for $0.007 \leq x \leq 0.10$ using plots of H/σ vs σ^2, and their values of T_c are about equal to the calculated values of Θ given in Table II. Recently, Cannella *et al*. [V. Cannella, T. J. Burch, and J. I. Budnick, AIP Conf. Proc. 24, 464 (1975)] reported values of T_c for $0.02 \leq x \leq 0.07$ using low-field ac susceptibility measurements; these values of T_c are significantly lower than those obtained by Crangle *et al*. Specific-heat measurements in a highly homogeneous $x = 0.06$ sample [H. A. Zweers, H. W. M. Van der Linden, and G. J. Nieuwenhuys, Physica B 79, 83 (1975)] give a T_c in good agreement with the results of Cannella *et al*. [We have determined T_c of $x = 0.01$ and $x = 0.05$ samples using low-field ($H \lesssim 300$G) magnetic-moment measurements; our results are also in agreement with the results of Cannella *et al*.] These discrepancies in the values of T_c will be discussed in a later publication.

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