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J. G. S. Duque, C. Adriano, R. Lora-Serrano, C. Rettori, R. R. Urbano, J. L. Sarrao, S. B. Oseroff, and P. G. Pagliuso

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Electron spin resonance of Gd^{3+} in $Gd_mM_nIn_{3m+2n}$ (M=Rh,Ir; n=0,1; m=1,2) antiferromagnets

J. G. S. Duque,[1] C. Adriano,[1] R. Lora-Serrano,[1] C. Rettori,[1] R. R. Urbano,[2] J. L. Sarrao,[2]
S. B. Oseroff,[3] and P. G. Pagliuso[1,a)]
[1]*Instituto de Fisica "Gleb Wataghin," UNICAMP, Campinas-SP, 13083-970, Brazil*
[2]*Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[3]*San Diego State University, San Diego, California 92182, USA*

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We report electron spin resonance experiments of Gd^{3+} in the $Gd_mM_nIn_{3m+2n}$ (M=Rh,Ir; n=0,1; m=1,2) intermetallic compounds. For $T>T_N\sim 45$ K, all compounds present a single Dysonian resonance and show a Korringa-like temperature dependence of the linewidth, $\Delta H=a+bT$. The residual linewidth a is strongly affected by the transition metal M=Rh or Ir and/or by the layering (m=1 or 2) or change in structure (n=0,1). The residual linewidth is associated with an unresolved crystalline electrical field (CEF) fine structure. Consequently, a systematic evolution of the CEF in the $Gd_mM_nIn_{3m+2n}$ compounds is inferred. A discussion to what extent our results can explain to the CEF effects observed in isostructural R-based compounds will be given. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2839592]

I. INTRODUCTION

$Gd_mM_nIn_{3m+2n}$ (M=Rh,Ir; n=0,1; m=1,2) are tetragonal compound variants of the Cu_3Au structure.[1–6] Their structure can be viewed as m layers of RIn_3 units sequentially stacked along the c axis with n layers of MIn_2.[6] The Ce-based compounds in the $R_mM_nIn_{3m+2n}$ (R=rare-earth, M=Co,Rh,Ir; n=0,1; m=1,2) family include a class of heavy-fermion superconductors (HFS) with remarkable physical properties,[7–12] such as the interplay between antiferromagnetism and unconventional superconductivity (USC), non-Fermi-liquid behavior, and quantum criticality (QC).

The variety of interesting physical properties in structurally related series represent a great opportunity to explore systematically the role of the Ruderman–Kittel–Kasuya–Yoshida (RKKY) magnetic interaction, Kondo effect, crystalline electrical field (CEF), Fermi surface (FS) effects, and QC in determining their properties, specially, in favoring USC in many Ce-based members of these series. As the properties of the HFS in their family are presumably magnetically mediated, studies of non-Kondo isostructural $R_mM_nIn_{3m+2n}$ (R=Nd,Gd,Tb) magnetic materials have been used to elucidate the role of the RKKY interactions and CEF effects in the evolution of the magnetic properties.[13–16]

Along the R-series, the Gd-based compounds are particularly interesting because the Gd^{3+} ground state is a S-state ($S=7/2$, $L=0$) where CEF effects are small. Gd-based materials are commonly taken as a reference where the magnetic properties reflect the details of the RKKY interactions and FS effects.[4,14,15] Within the $R_mM_nIn_{3m+2n}$ (M=Rh,Ir; n=0,1; m=1,2) family the studies for R=Gd have shown that the magnetic properties (T_N and magnetic structure) are nearly unaffected by changing M (Rh or Ir) in the MIn_2 or by the number of layers m=1,2 of $GdIn_3$.[4,14,15] This suggests that the CEF is the main cause for the evolution of the magnetic properties observed in the other non-S members of the series (R=Nd or Tb).[5,16]

In the Gd-compounds, the Gd^{3+} ions are excellent electron spin resonance (ESR) probes to reveal details about the microscopic interaction between the Gd^{3+} $4f$ electrons and the conduction electrons (c-e). Furthermore, although the CEF effects are small for the Gd^{3+} ions, the evolution of the CEF may be inferred from ESR studies.

In this work we present ESR studies in the $Gd_mM_nIn_{3m+2n}$ (M=Rh,Ir; n=0,1; m=1,2) intermetallic compounds for $T>T_N\sim 45$ K. For all compounds we observe a single Dysonian resonance with a Korringa-like temperature dependence of the linewidth. The residual linewidth is strongly dependent on the transition metal M=Rh or Ir, on the layering (m=1 or 2) and on the change in structure (n=0,1). We attribute the evolution of the residual linewidth to changes in the tetragonal CEF, following the trend found for the CEF in the isostructural compounds with R=Nd and Tb.[13,16,17]

II. EXPERIMENTAL RESULTS AND DISCUSSION

Single crystals of $Gd_mM_nIn_{3m+2n}$ (M=Rh,Ir; n=0,1; and m=1,2) were grown using a flux method. The structure and phase purity were confirmed by x-ray powder diffraction. The powder ESR spectra were taken in crushed single crystals in a Bruker X (9.48 GHz) and Q-bands (34.4 GHz) spectrometers, using appropriate resonators coupled to a T-controller of a helium gas flux system for $4.2\lesssim T\lesssim 300$ K. Figure 1 presents the X-band ESR spectra of Gd^{3+} in $Gd_mM_nIn_{3m+2n}$ (M=Rh,Ir; n=0,1; m=1,2) measured at room-T. The solid lines are fits to the spectra using Dysonian analysis. The asymmetric shape of the resonance spectra is due to a skin depth smaller than the size of the particles. The best fits of the ESR linewidths (ΔH) and g-values are shown

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[a)] Electronic mail: pagliuso@ifi.unicamp.br.

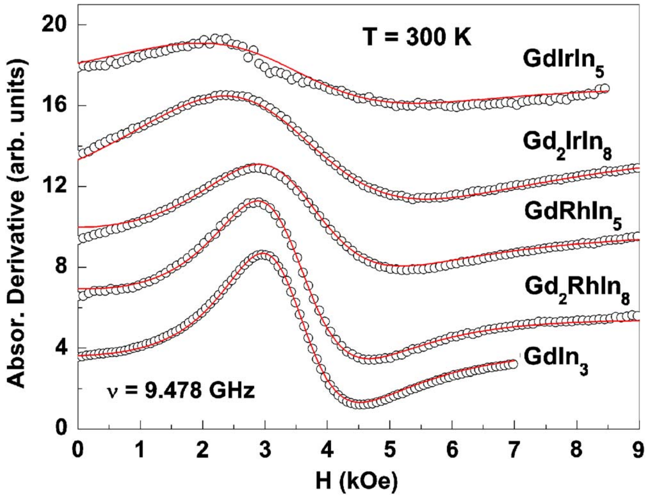

FIG. 1. (Color online) ESR powder spectra of Gd^{3+} in $Gd_mM_nIn_{3m+2n}$ (M =Rh, Ir; $n=0,1$; $m=1,2$) measured at room-T. The solid lines are the best fits to the Dysonian lineshape.

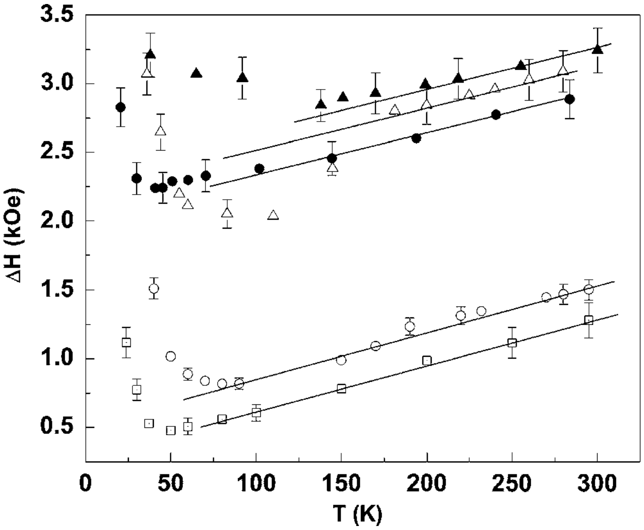

FIG. 2. Temperature dependence of the ESR ΔH for (□) $GdIn_3$, (○) Gd_2RhIn_8, (●) $GdRhIn_5$, (△) Gd_2IrIn_8, and (▲) $GdIrIn_5$. The solids lines are the best fit to $\Delta H = a + bT$.

in the Table I. These g-values are nearly T-independent for all studied compounds in the range of $50 \leqslant T \leqslant 300$ K.

The thermal broadenings of ΔH for $GdIn_3$, $GdMIn_5$, and $GdMIn_8$ (M=Ir or Rh) are shown in Fig. 2. This behavior is typical for all compounds. Their linear dependence was fitted to the expression $\Delta H = a + bT$ for $50 \leqslant T \leqslant 300$ K. The best fitted parameters are also given in Table I. Figure 2 shows that ΔH increases at lower-T ($T \leqslant 50$ K) due to the increase of magnetic correlations as T approaches $T_N \cong 50$ K. This behavior was observed for all compounds.

To compare the dependence of the residual linewidth a with the structure of the studied compounds, we present in Fig. 3(a) the evolution of a for $GdMIn_5$ and Gd_2MIn_8 (M =Rh and Ir) normalized to that of $GdIn_3$. This behavior looks similar to the one observed for the evolution of T_N for $NdMIn_5$ and Nd_2MIn_8 (M=Rh and Ir) normalized to that of $NdIn_3$ shown in Fig. 3(b).

Our results show that a is strongly affected by changing the transition metal M=Rh or Ir and by the number of layering (m=1 or 2) or the change in structure from cubic to tetragonal ($n=0,1$). However, their magnetic properties (T_N and magnetic structure) are nearly unaffected by these changes,[4,14,15] in contrast to the evolution of the magnetic properties found in the non-S members with R=Ce, Nd, or Tb.[5,13,16]

Table I shows that ΔH measured at room-T and a increase in the sequence $GdIn_3-Gd_2RhIn_8-GdRhIn_5-Gd_2IrIn_8-GdIrIn_5$ similar to the evolution of T_N in $NdMIn_5$ and Nd_2MIn_8 (M=Rh and Ir) compounds. The increase in T_N was ascribed to the splitting of the Γ_8 quartet ground state into two Kramer doublets due to the tetragonal structure of $NdIn_3$.[5,13,18,19] This similarity suggests that the CEF trends observed for T_N in the non-S members also affects a of Gd^{3+} in a S-ground state. However, to confirm this claim one needs to properly consider the different effects that may contribute to the ESR linewidth. There are two types of

TABLE I. Experimental and fitting parameters: ESR (ΔH) linewidths and g-values measured at room-T and linear broadening terms, a and b for Gd^{3+} between $50 \leqslant T \leqslant 300$ K in $Gd_mM_nIn_{3m+2n}$ (M=Rh, Ir; n=0, 1; m=1, 2).

	ΔH (kOe)[a]	g[a]	a (kOe)[b]	b (Oe/K)[b]
$GdIrIn_5$	3.0(3)	2.0(4)	2.5(3)	2.4(5)
Gd_2IrIn_8	2.9(3)	2.0(4)	2.2(3)	2.6(5)
$GdRhIn_5$	2.3(2)	1.9(2)	2.1(2)	2.6(5)
Gd_2RhIn_8	1.4(1)	1.9(2)	0.7(2)	2.5(5)
$GdIn_3$	1.3(1)	1.9(2)	0.30(6)	3.0(5)

[a] T=300 K.

[b] $50 \leqslant T \leqslant 300$ K.

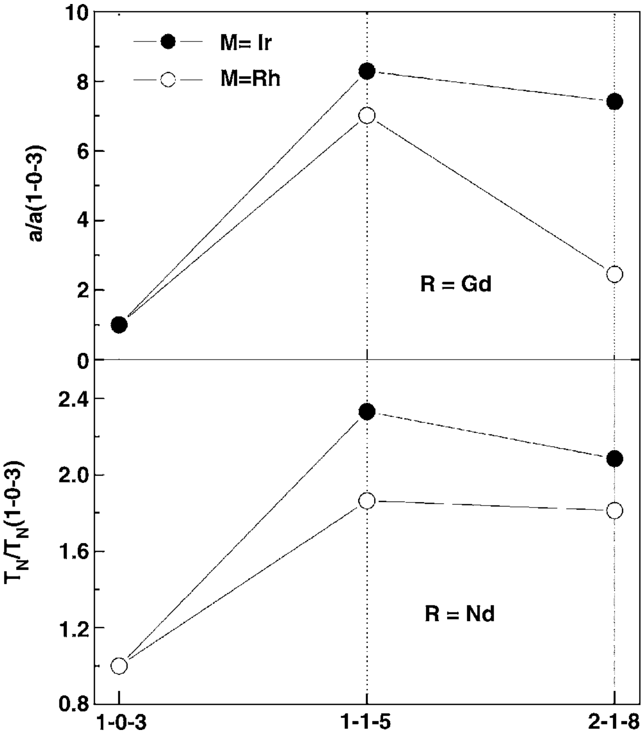

FIG. 3. Evolution of the relative (a) residual ESR linewidth a for the R =Gd and (b) T_N, for the R=Nd compounds in the R_mMIn_{3m+2} (m=1,2; M =Rh and Ir) as compared to the cubic relatives RIn_3.

ESR line broadening in solids: homogeneous and inhomogeneous broadening. Homogeneous ESR linewidth is inversely proportional to the so-called *spin-spin* relaxation time T_2.[20] It occurs when the resonance results from a transition between two spin levels which are not sharply defined, but instead are intrinsically broadened. The homogeneous broadening that may contribute to ΔH in our samples are dipolar interaction between like spins and spin-lattice interaction (Korringa relaxation in metals).[20] On the other hand, an inhomogeneously broadened line consists in a distribution of individual lines merged into an overall line or envelope. For instance, a distribution of local fields caused by unresolved fine (CEF) and/or hyperfine structure, g-value anisotropy, strain distribution, and/or crystal irregularities that exceed the natural linewidth ($2/\gamma T_2$, γ is the gyromagnetic factor),[20] will make the spins in various parts of the sample feel different field strengths. In the cases of inhomogeneous broadening caused by g-value anisotropy and related strain distribution and/or crystal irregularities, the ESR linewidths are expected to increase as a function of magnetic field. However, the ESR linewidth is usually field (frequency) independent, when it is homogeneously or inhomogeneously broadened by unresolved fine (CEF) and/or hyperfine structures. The spectra of Fig. 1 were indeed found to be frequency (field) independent when measured at Q-band (34 GHz) (not shown). On the other hand, the Korringa thermal broadening (see Fig. 2) and T_N are roughly the same for all studied compounds. This suggests that the two main contributions for the homogeneous broadening of our ESR spectra may be dipolar interaction between like spins and spin-lattice relaxation, which are expected to be nearly the same in these compounds. Therefore, we propose that the evolution of a, found for Gd compounds in these series, reflects the changes in the unresolved fine structure following the trend of the tetragonal CEF found in this family.[4,5,13,16,17]

Furthermore, our results allow us to estimate the exchange interaction, between a localized Gd^{3+} $4f$ electron spin ($\mathbf{S}$) and the free c-e's spin ($\mathbf{s}$) of the host metal for the studied compounds. In the simplest treatment for $\mathbf{q}$-dependent exchange interaction, $J_{fs}(\mathbf{q})\mathbf{S}\cdot\mathbf{s}$ the ESR Korringa rate,[21,22] can be written as

$$b = \frac{d(\Delta H)}{dT} = \frac{\pi k}{g\mu_B}\langle J_{fs}^2(\mathbf{q})\rangle \eta^2(E_F), \quad (1)$$

where $\langle J_{fs}^2(\mathbf{q})\rangle$ is the effective exchange interaction between the Gd^{3+} local moment and the c-e in the presence of c-e momentum transfer ($0 \leqslant \mathbf{q} \leqslant 2k_F$) averaged over the Fermi surface, $\eta(E_F)$ the *bare* density of states for one spin direction at the Fermi surface, k the Boltzman constant, μ_B the Bohr magneton, and g the Gd^{3+} g-value.[23] Due to the large ESR ΔH observed for all Gd-based compounds, we were not able to determine the g-shift of the Gd^{3+} resonance relative to the g-value of Gd^{3+} in insulators [$g=1.993(2)$ (Ref. 22)]. We found $b \approx 2.5(5)$ Oe/K for all samples of $\mathrm{Gd}M\mathrm{In}_5$ and $\mathrm{Gd}_2M\mathrm{In}_8$ (M=Rh and Ir). Thus, the product $\langle J_{fs}^2(\mathbf{q})\rangle \eta^2(E_F)$ is the same for these materials. To calculate $\langle J_{fs}^2(\mathbf{q})\rangle^{1/2}$ for the studied compounds, we have used the value of $\eta(E_F) = 1.0(5)$ states/eV mol spin extracted from the electronic contribution to the specific heat of the isomorphous compounds $\mathrm{La}_m M_n \mathrm{In}_{3m+2n}$. Using this value for $\eta(E_F)$ and Eq. (1), we obtain $\langle J_{fs}^2(\mathbf{q})\rangle^{1/2} \approx 10$ meV for the $\mathrm{Gd}M\mathrm{In}_5$ and $\mathrm{Gd}_2M\mathrm{In}_8$ (M=Rh and Ir) materials.

III. CONCLUSIONS

In this work we measured the evolution of a for $\mathrm{Gd}M\mathrm{In}_5$ and $\mathrm{Gd}_2M\mathrm{In}_8$ (M=Rh and Ir). The results were attributed to changes in the unresolved (CEF) fine structure for the different compounds. From the Korringa rate, $b \approx 2.5(5)$ Oe/K we have estimated $\langle J_{fs}^2(\mathbf{q})\rangle^{1/2} = 10$ meV, between a localized $4f$ electron spin ($\mathbf{S}$) and the free c-e's spin ($\mathbf{s}$) for all compounds.

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