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Magnetic properties of frustrated Y-doped $GdInCu_4$

J.G.S. Duque, E. Miranda, A.M.O. Belon, L. Bufaiçal, C. Rettori, P.G. Pagliuso*

Instituto de Física, IFGW, Universidade Estadual de Campinas (UNICAMP), C.P. 6165, 13083-970, Campinas S.P., Brazil

Abstract

We report temperature dependent magnetic susceptibility and electron spin resonance (ESR) measurements on single crystals of the frustrated antiferromagnet $Gd_{1-x}Y_xInCu_4$ ($x = 0, 0.01$, 0.05 and 0.1). The magnetic susceptibility data revealed, as expected, a decreasing in the antiferromagnetic ordering temperature as function of the Y-concentration, while the high-T Curie–Weiss temperatures remain nearly unaffect by the doping. For $T \gtrsim 100$ K, a single Dysonian Gd^{3+} ESR line with a nearly temperature independent $g \sim 1.989(8)$ is observed, and its linewidth follows a Korringa-like ($\Delta H/\Delta T \sim 0.75(5)$ Oe/K) behavior as a function of temperature. Below $T \approx 100$ K both the ESR g-value and linewidth are affected by the presence of strong short-range magnetic correlation in the paramagnetic phase well above T_N, consistently with the frustrated character of this compound. The role of chemical disorder and dilution effects induced by Y-doping in the properties of this highly frustrated material is discussed.
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1. Introduction

The study of the magnetic frustration phenomena has been focus of intense and interesting theoretical and experimental research in solid state physics because frustrated spin fluctuations may play a role in many fascinating physics phenomena such as, quantum fluctuations, Quantum Griffith Phases spin glass, cluster glass, non-fermi-liquid (NFL) and even non-conventional superconductivity [1–3]. In geometrically frustrated solids the frustration is explained by the incompatibility between the local symmetries and the global arrangement of the interacting spins in the lattice. On the other hand, the competition among various kinds of magnetic interactions, such as Ruderman–Kittel–Kasuya–Yoshida (RKKY), crystalline electrical field (CEF) and Kondo effects may lead to magnetic frustration mechanism in complex materials such as heavy Fermion systems [4].

In this work, we study the effect of the Y-substitution in the Gd site for the highly frustrated antiferromagnet $GdInCu_4$. This material crystallizes in the cubic C15b $AuBe_5$-like where Gd ions form an FCC corner-sharing tetrahedral lattice which may lead to geometrical frustration [3].

Combining ESR of Gd^{3+} and magnetic susceptibility experiments we have explored the local and global effects of Y-doping in the frustrated magnetic interactions of $GdInCu_4$.

2. Experiment

Single crystals of $Gd_{1-x}Y_xInCu_4$ ($x = 0.01$, 0.05 and 0.1) were grown from a CuIn-flux method [3] and checked by X-ray powder diffraction. The ESR experiments were done in single crystals using a Bruker X-band ($\nu \approx 9.48$ GHz) spectrometer with TE_{102} cavity coupled to a T-controller using a helium gas flux. The magnetic susceptibility measurements were performed in a Quantum Design MPMS SQUID DC-magnetometer.

3. Results analysis

Fig. 1 presents the magnetic susceptibility as a function of temperature measured at $H = 1$ kOe for single crystals

*Corresponding author. Tel./fax: +55 193 788 5501.
E-mail address: pagliuso@ifi.unicamp.br (P.G. Pagliuso).

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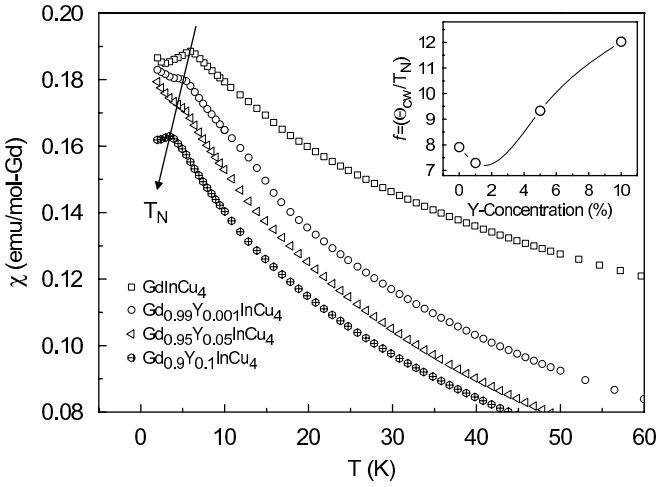

Fig. 1. Temperature dependence of the magnetic susceptibility at an applied field $H = 1$ kOe for the $Gd_{1-x}Y_xInCu_4$. Inset: The frustration parameter, f, as function of Ytrio concentration.

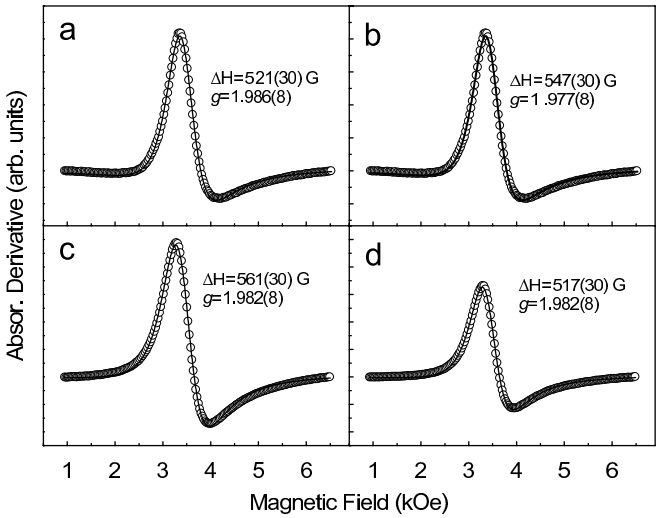

Fig. 2. ESR spectra at $T = 300$ K for (a) $GdInCu_4$, (b) $Gd_{0.99}Y_{0.01}InCu_4$, (c) $Gd_{0.95}Y_{0.05}InCu_4$ and (d) $Gd_{0.9}Y_{0.1}InCu_4$. The solid lines are the fits to the Dysonian lineshape.

of $Gd_{1-x}Y_xInCu_4$. For the pure compound, the peak associated with the antiferromagnetic (AFM) phase transition at $T_N = 5.9$ K is clearly observed and it shifts to lower temperature as a function of doping.

From Curie–Weiss fits to the data of Fig. 1 for $T > 150$ K, we have extracted for all samples an effective moment $p = 7.8(2)\,\mu_B$ for Gd^{3+} in $GdInCu_4$ in agreement with its theoretical value. The Curie–Weiss temperatures, Θ_{CW}, obtained from these fits confirm the presence of magnetic frustration for these materials, given a frustration parameter ($f = \Theta_{CW}/T_N$) larger than 8 for all compounds.

Fig. 2(a)–(d) shows the X-Band ($f = 9.45$ GHz) ESR spectra of Gd^{3+} at room-temperature for $Gd_{1-x}Y_xInCu_4$. The ESR spectra consist in a single line with Dysonian lineshape which is characteristic of localized moments in a metallic host with a skin depth smaller than the size of the sample. The ESR g-value and linewidth for the Gd^{3+} resonance were obtained from best fits to spectra using Dysonian lineshape.

Figs. 3(a) and 3(b) display, respectively, the T-dependence of the Gd^{3+} ESR linewidth, ΔH, and g-value as function of temperature. The thermal broadening of the linewidth were fitted using $\Delta H = a + bT$, with $a = 290(30)$ Oe and $b = 0.75(5)$ Oe/K.

As one can see from the values in Table 1 the frustration parameter f, for the $Gd_{1-x}Y_xInCu_4$ samples are roughly 10 which classify these materials as highly frustrated. The f parameter rarely exceeds 10 [5] and examples of large f in three-dimensional magnets are $MnSc_2S_4$ ($f = 11$) and $FeSc_2S_4$ ($f > 900$) [6], $ZnCr_2O_4$ ($f = 24$) [7], K_2IrC_{l6} ($f = 10$) [8] and FeF_3 ($f = 16$) [9]. Interestingly, the f parameter increases as a function of Y-doping as a consequence of the decreasing of T_N while $f = \Theta_{CW}/T_N$ remain nearly unchanged. Since dilution effects should affect directly the strength of the Gd–Gd RKKY interaction and reduce both Θ_{CW} and T_N, we speculated that the subtle disorder introduced by Y-doping is responsible for

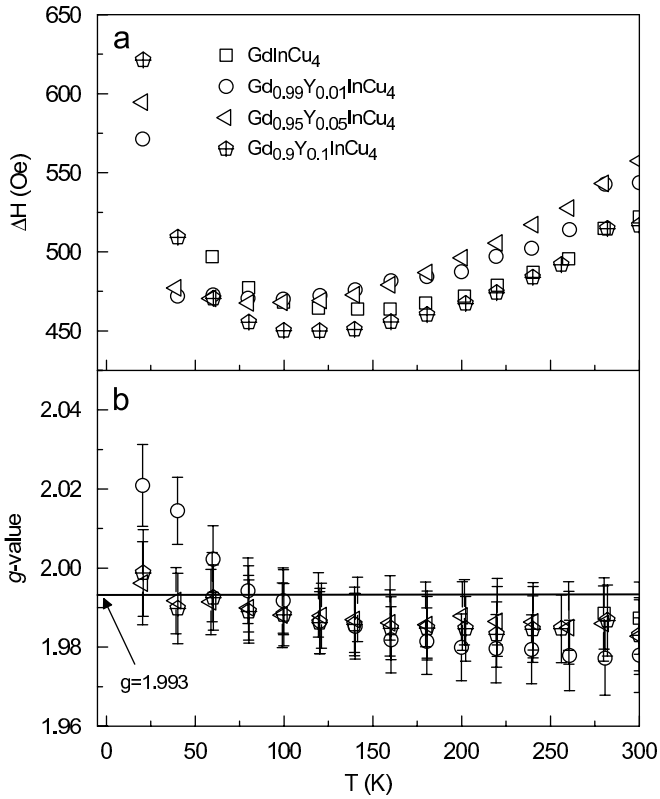

Fig. 3. T-dependence of the X-band ESR (a) linewidth, ΔH, and (b) g-value of Gd^{3+} in $Gd_{1-x}Y_xInCu_4$ ($x = 0.01, 0.05$ and 0.1) single crystals.

the T_N suppression. These results suggest that in presence of high spin frustration subtle disorder can more effectively avoid the on set of long range ordering.

Concerning the ESR measurements, both ΔH, and g-value revealed the presence of strong short range magnetic correlations for $T < 100$ K consistently with the frustrated

Table 1
Experimental parameters for Gd^{3+} in $Gd_{1-x}Y_xInCu_4$ extracted from Curie–Weiss fits

	T_N (K)	Θ_{CW} (K)	f
$GdInCu_4$	5.9	- 46(5)	8(1)
$Gd_{0.99}Y_{0.01}InCu_4$	5.4	- 40(4)	7(1)
$Gd_{0.95}Y_{0.05}InCu_4$	5.0	- 44(4)	9(1)
$Gd_{0.9}Y_{0.1}InCu_4$	3.5	- 42(4)	12(1)

character in these compounds. For all samples, the increasing of both ΔH, and g-value starts around the same $T\sim100$ K, suggesting again that the Y-doping is not strongly affect the Gd–Gd magnetic interaction in the studied range of concentration. It is important to point out that we have not observed any anisotropic behavior of ESR spectra of Gd^{3+} for the whole range of temperature studied. This result indicates the presence of exchange narrowing effects of the fine structure combined with spin–spin interaction in the temperature dependence of the ESR linewidth.

For a more detailed analysis of the ESR data we focus in the high-T data of Fig. 3 where the Gd^{3+} spins behave nearly independently since the g-value is temperature independent and ΔH follows a Korringa-like thermal broadening. In this case, a simple treatment of a **q**-dependent exchange interaction, $J_{fs}(\mathbf{q})\mathbf{S}.\mathbf{s}$, between a localized 4f electron spin (**S**) on a solute atom (Gd^{3+}) and the free c-e's spin (**s**) of the host metal, the ESR g-shift (Knight shift) [10] and the thermal broadening of the linewidth (Korringa rate) [11], when "*bottleneck*", "*dynamic*" effects and electron-electron exchange enhancement are not present [12], can be written as

$$\Delta g = J_{fs}(\mathbf{0})\eta(E_F), \tag{1}$$

and

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

where $J_{fs}(\mathbf{0})$ is the effective exchange interaction between the Gd^{3+} local moment and the c-e in the absence of c-e momentum transfer [13], $\langle J_{fs}^2(\mathbf{q})\rangle$ is that effective exchange interaction in the presence of c-e momentum transfer $0\leqslant\mathbf{q}\leqslant2k_F$) 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.

Unfortunately, because of the relatively large ESR linewidth (and associated error in the experimental g-value) observed for all $Gd_{1-x}Y_xInCu_4$ compounds we were not able to determine the g-shift of the Gd^{3+} resonance related the g-value of Gd^{3+} in insulators ($g = 1.993(2)$ [14]). However, we note that the Korringa rate ($\Delta H/\Delta T\sim0.75(5)$ Oe/K) obtained for all samples of $Gd_{1-x}Y_xInCu_4$ is in great agreement with the value of the Korringa rate obtained for highly dilute (0.2–0.5%) Gd^{3+} in $YInCu_4$. According to Eq. (2), this result indicates that the product $\langle J_{fs}^2(\mathbf{q})\rangle\eta^2(E_F)$ is roughly independent of Gd concentration. As Gd^{3+} and Y^{3+} are isovalent, one does not expect $\eta(E_F)$ to very dramatically as a function of doping.

Hence, in order to calculate $\langle J_{fs}^2(\mathbf{q})\rangle^{1/2}$ for $Gd_{1-x}Y_xInCu_4$ using Eq. (2), we have used the value of $\eta(E_F) = 0.34(3)$ states/eV.mol.spin extracted from the electronic contribution to the specific heat of the Y-concentrated isomorphous compound $YInCu_4$ [15]. Using this value of $\eta(E_F)$ and Eq. (2), we calculated the **q**-independent effective exchange interaction between Gd^{3+} local moments and the c-e, $\langle J_{fs}^2(\mathbf{q})\rangle^{1/2} \approx 17(2)$ meV for the $Gd_{1-x}Y_xInCu_4$ samples.

As the value of $\langle J_{fs}^2(\mathbf{q})\rangle^{1/2}$ was found to be independent of Gd-concentration is not unreasonable to assume that the total J_{fs} effective interaction between the Gd^{3+} local moment and the c-e is in fact independent of concentration for these materials. As such, taking into account the RRKY interaction is a long range interaction by nature and that J_{fs} is independent of Gd-concentration it is reasonable that our results indicate that the strength of the Gd–Gd magnetic correlations are not strongly modified by Y-doping, as the average first neighbors Gd–Gd distance are not strongly increased for the $Gd_{1-x}Y_xInCu_4$ samples in the studied concentration range. As such, it is a suggestion of our data, that the suppression of the long range ordering in this range of concentration is mainly given by disorder. Further experiments for samples with different Y-concentrations (up the critical concentration where $T_N \to 0$) are in progress to confirm this supposition.

4. Conclusions

In this work we report temperature dependent magnetic susceptibility and electron spin resonance (ESR) measurements on single crystals of $Gd_{1-x}Y_xInCu_4$ ($x = 0$, 0.01, 0.05 and 0.1) grown by flux method. The T-dependence of the magnetic susceptibility showed a continuous decreasing of the on set of long range ordering in the Neel temperature as function Y-doping. In contrast, the frustration parameter, f, increases as a function of Y-doping as a consequence of the decreasing of T_N while $f = \Theta_{CW}/T_N$ remain nearly unchanged. In the ESR studies we have found, at high-T, a single Dysonian Gd^{3+} ESR line with a nearly temperature independent $g\sim1.989(8)$ and a Korringa-like behavior as a function of temperature for the ESR linewidth. From the obtained Korringa rate ($\Delta H/\Delta T\sim0.75$ Oe/K) we have extracted the q-dependent effective exchange parameter $\langle J_{fs}^2(\mathbf{q})\rangle^{1/2} = 17(2)$ meV between the Gd^{3+} local moments and the c-e in this compound. This value was found to be independent of Gd-concentration and our results indicate that the suppression of the long range ordering is mainly given by disorder induced by Y-doping.

Acknowledgments

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