

Spin Disorder Resistivity and Electronic Specific Heat of $Gd_4(Co_{1-x}Cu_x)_3$ Compounds

T.M. Seixas^{1,2,a}, M.A. Salgueiro da Silva², O.F. de Lima^{3,b}, J. Lopez³, H.F. Braun^{4,c}, G. Eska⁴

¹Instituto de Física dos Materiais da Universidade do Porto (IFIMUP)

²Departamento de Física da Universidade do Porto, Rua do Campo Alegre, 687, 4169-007 Porto, Portugal

³Instituto de Física Gleb Wataghin, Universidade Estadual de Campinas, 13083-970, Campinas, SP, Brazil

⁴Physikalisches Institut, Universitaet Bayreuth, D – 95440 Bayreuth, Germany

^atmseixas@fc.up.pt, ^bdelima@ifi.unicamp.br, ^chans.braun@uni-bayreuth.de

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Abstract. In this work, we present a study of the spin disorder resistivity ($\rho_{m\infty}$) and the electronic specific heat coefficient (γ) in $Gd_4(Co_{1-x}Cu_x)_3$ compounds, with $x = 0, 0.05, 0.10, 0.20, 0.30$. The experimental results show a strongly non-linear dependence of $\rho_{m\infty}$ on the de Gennes factor which, in similar intermetallic compounds, is usually attributed to the existence of spin fluctuations on the Co 3d bands and its amplification by the thermal disorder of the Gd magnetic moments through the Gd-Co exchange coupling. Using a novel combined analysis of $\rho_{m\infty}$ and γ , we show, however, that only electron band structure changes are involved in the anomalous behaviour of $\rho_{m\infty}$ and that a linear dependence of $\rho_{m\infty}$ on the de Gennes factor is obtained when the variation of the effective mass is properly taken into account.

Introduction

Among the R-Co family, Gd-Co compounds are those that have the highest Curie temperatures due to the direct dependence of the 4f-3d exchange coupling on the spins of the 4f and 3d elements and due also to the fact that Gd has the highest spin among the rare earths. Moreover, Gd-Co compounds are also, at least in first order approximation, free from crystal electric field effects, which simplifies the analysis of their magnetic behavior. As the Co:Gd concentration ratio increases, the itinerant character of the magnetism is reinforced, whereas compounds with a low Co:Gd concentration ratio are expected to exhibit a magnetic behavior much closer to the 4f localized magnetism typical of rare earths.

Having a low (3/4) Co:Gd concentration ratio and crystallizing in the hexagonal Ho_4Co_3 type structure [1], Gd_4Co_3 is a special system. It orders ferrimagnetically below $T_C \approx 220$ K [2-4] with the Co magnetic moments antiparallelely coupled to those of Gd. Below $T_{SR} \approx 163$ K [2], and somehow similarly to pure Gd [5] it exhibits a spin-reorientation process in which the Gd and Co magnetic moments tilt rigidly away from the initial easy magnetic direction, presumably the c-axis.

In this study, we focus on the behaviour of the spin disorder resistivity across the series of compounds $Gd_4(Co_{1-x}Cu_x)_3$ ($x = 0.05, 0.10, 0.20, 0.30$) in order to investigate the role of spin fluctuations and band structure effects on magnetic scattering. A successful evaluation of the importance of these two usual explanatory mechanisms will be made possible through a combined analysis involving also the de Gennes factor and the electronic specific heat coefficient.

Experimental details

The $\text{Gd}_4(\text{Co}_{1-x}\text{Cu}_x)_3$ ($x = 0.05, 0.10, 0.20, 0.30$) compounds were prepared by arc-melting stoichiometric quantities of Gd (3N), Co (4N) and Cu (5N) under a purified argon atmosphere. In order to increase homogeneity, the ingots were remelted several times. Given the very small mass losses of the order of 0.04%, final stoichiometric compositions can be assumed. The resulting ingots were then encapsulated in quartz tubes, under an argon atmosphere, and annealed at 600 °C for 2 h, then at 635 °C for 24 h and at 650 °C for 48 h. X-ray diffraction on the annealed materials revealed a single phase with the Ho_4Co_3 crystal structure [1].

The determination of the average magnetic moment of Co atoms for all studied compounds through magnetisation data has been made as described in [2] for the case of Gd_4Co_3 .

A conventional dc four-probe technique has been used for electrical resistivity measurements in a closed-cycle refrigerator operating from 13 K to 300 K. The magnetic contribution to the electrical resistivity for all compounds was calculated using the relation $\rho_m(T) = \rho(T) - \rho_{\text{ph}}(T) - \rho_0$, where ρ_0 is the residual resistivity and $\rho_{\text{ph}}(T)$ is the electron-phonon scattering contribution given by the Bloch-Grüneisen model [6]. The spin disorder resistivity ($\rho_{\text{m}\infty}$) is taken as the saturation value of $\rho_m(T)$ at high temperatures.

Specific heat measurements were made on samples of $2.5 \times 2.5 \times 1.5 \text{ mm}^3$ with a Quantum Design PPMS calorimeter that uses a two-relaxation times technique, and data was always collected during sample cooling. The electronic specific heat coefficient (γ) of all compounds was determined through linear fittings to the C/T versus T^2 curves between 3.8 K and 10 K.

Results and discussion

Fig. 1 shows the compositional dependence of the spin disorder resistivity. It is observed that the replacement of Co by Cu produces a large and almost exponential decrease of $\rho_{\text{m}\infty}$ with x . In order to shed light into the possible origin of this interesting result, we show, in Fig. 2, the corresponding variation of the saturation magnetic moment of Co atoms. An exponential decrease is also apparent in $M_{\text{Co}}(x)$, which may be attributed to a strong dependence of Co moment on the number of local Co neighbouring atoms. The ratio of the decaying constants of $\rho_{\text{m}\infty}(x)$ and $M_{\text{Co}}(x)$ is 1.8. It should be noticed that, within the mean field approximation for localized magnetism [7], the spin disorder resistivity is proportional to the concentration and effective magnetic moment of the atoms responsible for the electron scattering. Neglecting, for the moment, the distinction between saturation and effective moments of Co atoms, a ratio of 2 would be expected if the spin disorder resistivity behaviour could be linked exclusively to the compositional dependence of Co magnetic moment. The good agreement found between these two values clearly stresses the important role of the Co 3d electrons on magnetic scattering.

In order to further analyse the variation of the spin disorder resistivity, we will proceed by investigating its correlation with the variation of the de Gennes factor across the series of compounds. The spin-disorder resistivity of a system consisting of a single species of localised magnetic moments can be written as [8]:

$$\rho_{\text{m}\infty} = \frac{3\pi N}{2\hbar e^2} \frac{m^*}{\varepsilon_{\text{F}}} |\Gamma|^2 G, \quad (1)$$

where N is the atomic concentration, m^* is the effective electron mass, ε_{F} is the Fermi energy, Γ is a parameter describing the interaction between conduction and localised electrons, and $G = (g-1)^2 J(J+1)$ is the de Gennes factor.

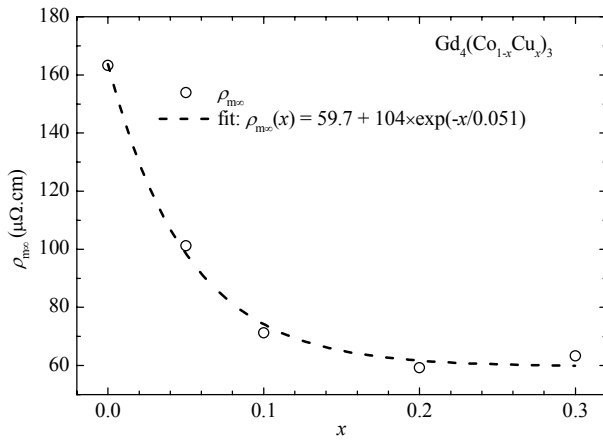


Fig. 1 Variation of the spin disorder resistivity in $Gd_4(Co_{1-x}Cu_x)_3$ compounds. An exponential curve fit is also shown for the $\rho_{mso}(x)$ curve.

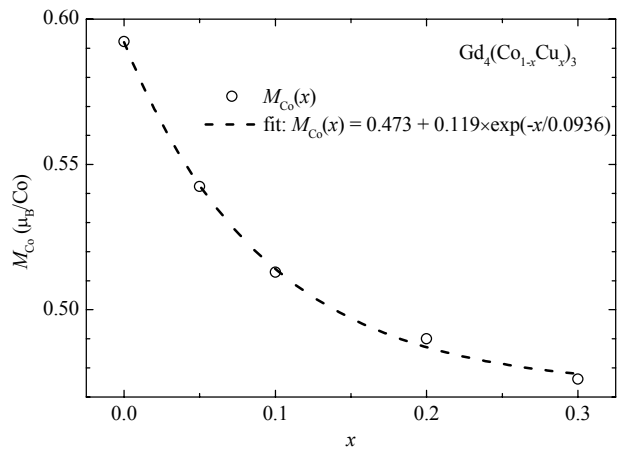


Fig. 2 Variation of the Co saturation magnetic moment in $Gd_4(Co_{1-x}Cu_x)_3$ compounds. An exponential curve fit is also shown for the $M_{Co}(x)$ curve.

In the case of the $Gd_4(Co_{1-x}Cu_x)_3$ compounds, in which two magnetic species (Gd, Co) are present, G must be properly averaged taking into account the respective concentrations ($c_{Gd} = \frac{4}{7}, c_{Co} = \frac{3}{7}(1-x)$), Landé factors ($g_{Gd} = g_{Co} = 2$) and angular momenta ($J_{Gd} = \frac{7}{2}, J_{Co} = M_{Co}/(g_{Co}\mu_B)$). Following Williams [9] we have:

$$G = c_{Gd}^2 (g_{Gd} - 1)^2 J_{Gd} (J_{Gd} + 1) + c_{Co}^2 (g_{Co} - 1)^2 J_{Co} (J_{Co} + 1) + 2c_{Gd}c_{Co}(g_{Gd} - 1)(g_{Co} - 1)J_{Gd}J_{Co} \quad (2)$$

As seen in Fig. 3, ρ_{mso} has a strongly non-linear dependence on the average de Gennes factor. Usually, in 4f-nd intermetallics, such non-linear dependence is attributed to spin fluctuations in the nd band enhanced by thermal disorder of the 4f magnetic moments. In the case of Gd_3Co , e. g., it has been proposed [10] that the enhancement of spin fluctuations by the 4f-3d exchange coupling was responsible for the observed anomalously large values ($109 - 170 \text{ mJ mol}^{-1} \text{ K}^{-2}$) of the electronic specific heat coefficient (γ) in this compound, comparatively to the case of the isostructural nonmagnetic Y_3Co compound ($14 \text{ mJ mol}^{-1} \text{ K}^{-2}$), and that γ should depend on the de Gennes factor. In principle, such interpretation could be consistent with our present results, which show also large values of γ with respect to the non-magnetic Y_4Co_3 compound ($\gamma \approx 38.4 \text{ mJ mol}^{-1} \text{ K}^{-2}$), if γ was found to be an increasing function of G . However, as seen in Fig. 3, this is clearly not the case, since γ does not even change monotonically with G . Accordingly, the large γ -enhancement in the present compounds cannot be ascribed to spin fluctuations, but rather to electron-phonon or electron-magnon couplings, or, more likely, to just band structure effects. As will be shown below, the latter hypothesis proves to be more realistic.

It must be noticed that besides the dependence on the de Gennes factor, ρ_{mso} also depends on electron band parameters that may change with the composition. More specifically, and independently of the particular electron band model to be considered, we have $\varepsilon_F \propto (m^*)^{-1}$ and thus $\rho_{mso} \propto (m^*)^2$. The variation of the effective electron mass across the series $Gd_4(Co_{1-x}Cu_x)_3$ can be inferred from the corresponding variation of the electronic specific heat

coefficient (γ), since $\gamma \propto m^*$. Hence, the quantity $\rho_{m\infty}/\gamma^2$ should be virtually independent of the electron band structure and scale linearly with G . Effectively, as also shown in Fig. 4, $\rho_{m\infty}/\gamma^2$ exhibits a statistically well-defined linear dependence on G .

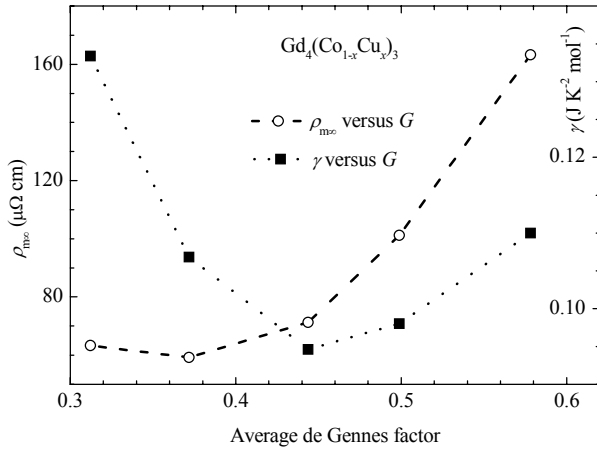


Fig. 3 Variation of $\rho_{m\infty}$ (left axis, open circles) and γ (right axis, solid squares) with the de Gennes factor in $Gd_4(Co_{1-x}Cu_x)_3$ compounds. The dashed and dotted lines are guides to the eye.

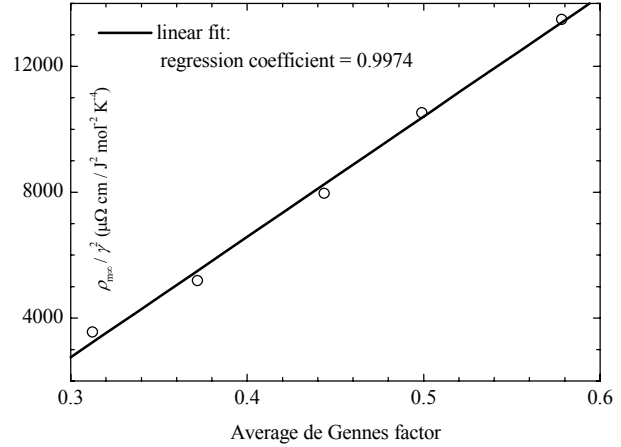


Fig. 4 Variation of $\rho_{m\infty}/\gamma^2$ with the de Gennes factor in $Gd_4(Co_{1-x}Cu_x)_3$ compounds. The solid line represents a linear fit to $\rho_{m\infty}/\gamma^2(G)$.

This important result, originated from the combination of two independent types of measurements, suggests that band effects rather than spin fluctuation effects are the main cause for the strong electron scattering and γ -enhancement observed in the present compounds, and possibly in other 4f-nd compounds. The dilution of Co by Cu is expected to induce changes in the 3d band structure, which is generally characterized by a high and peaked density of states. This may occur directly through the displacement of the Fermi level as a result of the filling of the 3d band by Cu-originated electrons. We may also invoke the change in the lattice parameters and eventual local disorder effects associated with the substitution of Co by Cu as concurrent mechanisms for band structure effects.

Acknowledgements

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