

Magnetic order and weak ferromagnetic transition in Gd_2CuO_4

J. Mira and J. Rivas

Departamento de Física Aplicada, Universidade de Santiago, E-15706 Santiago de Compostela, Spain

A. Butera^{a)} and L. B. Steren^{a)}

Centro Atómico Bariloche-Instituto Balseiro, Bariloche, 8400 Río Negro, Argentina

J. M. García-Beneytez and M. Vázquez

Instituto de Magnetismo Aplicado, RENFE-UCM-CSIC, Apdo. 155, E-28230 Las Rozas, Spain

The weak ferromagnetic transition of T' -phase Gd_2CuO_4 single crystals has been studied by dc-magnetization measurements. Weak ferromagnetism is generated in this material by a very slight canting of the Cu moments from a perfect antiferromagnetic structure. This canting enables the inspection of the magnetic state of the Cu sublattice by pure magnetic methods. The critical exponents of the transition are calculated, $\gamma=1.32(2)$, $\beta=0.34(1)$. Nevertheless, a rapid evolution of β to a 2D XY behavior is observed below the critical temperature. It is interpreted as evidence of a transition in the 2D system, namely CuO_2 planes, that behave as 3D at the critical point due to the effect of the interlayer interaction along the c -axis. © 2000 American Institute of Physics.

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I. INTRODUCTION

R_2CuO_4 materials (R =rare-earth) have increased their condition as a target of scientific research since the discovery of high- T_c superconductivity.¹ When suitably doped, many of them exhibit a superconductive phase.^{2,3} They mainly crystallize in the tetragonal T' -phase, with $\text{CuO}(1)_2$ planes separated by $\text{R}_2\text{O}(2)_2$ blocks.⁴ The magnetic interactions are therefore strongly restricted to such planes, yielding a possible example of a two-dimensional (2D) system. These antiferromagnetic (AF) interactions, by superexchange, have been estimated to be very intense,⁵ $J \sim 1500$ K. Nevertheless, at temperatures in the range of 250–290 K, the Cu sublattice of all these systems undergoes a three-dimensional (3D) ordered antiferromagnetic phase,^{6,7} whose origin has given rise to many theoretical discussions.^{8,9} The analysis of their 3D transition through pure magnetic measurements is usually difficult, due to the small signal obtained from the Cu AF sublattice. Gd_2CuO_4 occupies an unique place among the series. In this compound, the small size of the Gd^{3+} ion causes a distortion of the tetragonal phase.^{10–12} Such distortion, which consists of the deviation of the O(1) atoms from the line defining the Cu–O(1)–Cu bond,¹² allows the onset of the antisymmetric Dzyaloshinskii–Moriya exchange interactions between the Cu atoms.¹³ As result, the Cu moments cant away from their perfect AF alignment and a weak ferromagnetic (WF) component appears, like that first observed by Borovik-Romanov and Ozhogin in CoCO_3 .¹⁴ The magnetic contribution of the Cu sublattice then becomes measurable. In the present work this circumstance is used to analyze the 3D magnetic transition by dc magnetic measurements, in order to gain insight into the nature of the Cu ordering of the R_2CuO_4 materials.

II. EXPERIMENTAL DETAILS

A Gd_2CuO_4 single crystal, 4.7 mg weight, was used for the magnetic measurements. The crystal was grown in a Pt crucible using a flux technique.¹⁵ It had a platelike shape ($3 \times 2 \times 0.1$ mm³) with the crystallographic c -axis oriented perpendicular to the plate. Dc-magnetization measurements (usually magnetization vs field loops at a fixed temperature) were made in a Quantum Design SQUID magnetometer. In all cases the external magnetic field was applied perpendicular to the c -axis and parallel to the in-plane easy axis.

III. RESULTS

By low-field dc-magnetization measurements vs temperature a first estimation of the transition temperature at 268 K was obtained. This value differs appreciably from that obtained in ceramic samples,¹⁶ probably due to Pt contamination of the crystal during the synthesis process.¹⁷ Near the transition point a cusp appears¹⁵ as in other compounds of the family.¹⁰ Initial susceptibility $\chi_i(T) = (dM_{dc}(H))/(dH)|_{H \rightarrow 0}$ was measured above T_C . From this signal we subtracted the contributions of the paramagnetic susceptibility of Gd^{3+} ions, $[2C/(T + \theta)]$, $C = 7.94 \mu_B/\text{atom}$, $\theta \sim 12$ K] and the diamagnetic susceptibility contributed by the different ions. Because of the log–log nature of the plots used to obtain the critical exponents it was not necessary to subtract the term originated by the polarization induced by the Cu sublattice on the Gd^{3+} sites.¹⁸

As in the critical zone $\chi_i \propto |T - T_C|^{-\gamma}$ for $T > T_C$, $\log \chi_i^{-1}$ vs $\log(T/T_C - 1)^\gamma$ was plotted (Fig. 1), and from the resulting curves the critical exponent γ was obtained, $\gamma = 1.32(2)$. In order to obtain the critical exponent β we have used the modified Arrott plot technique,¹⁹ based on the relations given by Arrott and Noakes,²⁰ $\log(M^{1/\beta + 1/\gamma})$ vs $\log H^{1/\gamma}$ isotherms were plotted in the vicinity of the critical point, introducing the previously calculated value of γ (Fig.

^{a)}Also at Consejo Nacional de Investigaciones Científicas y Técnicas, Argentina.

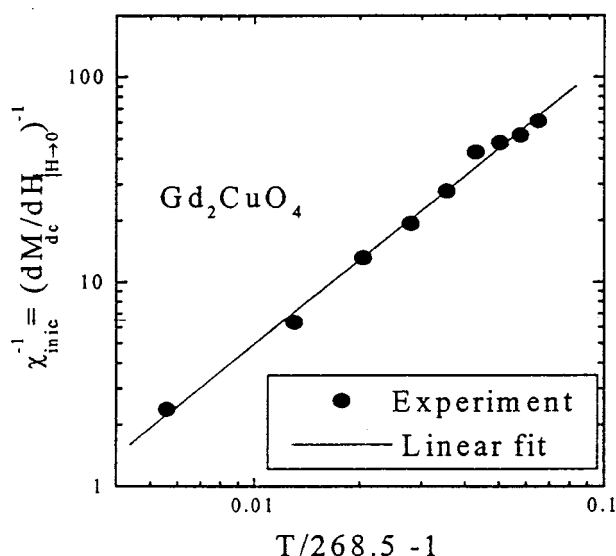


FIG. 1. Log-log plot of the inverse initial susceptibility [$\chi_i(T) = (dM_{dc}(H))/(dH)_{H \rightarrow 0}$] of Gd_2CuO_4 versus the reduced temperature. From the linear fit of the data a value for the critical exponent $\gamma = 1.32(2)$ is obtained.

2). In this way we obtained values of β for several temperatures below the critical point, shown in the inset of Fig. 2.

IV. DISCUSSION

The WF is manifested macroscopically once the 3D order sets in; for this reason the weak ferromagnetic transition temperature will be associated with the 3D order of the Cu

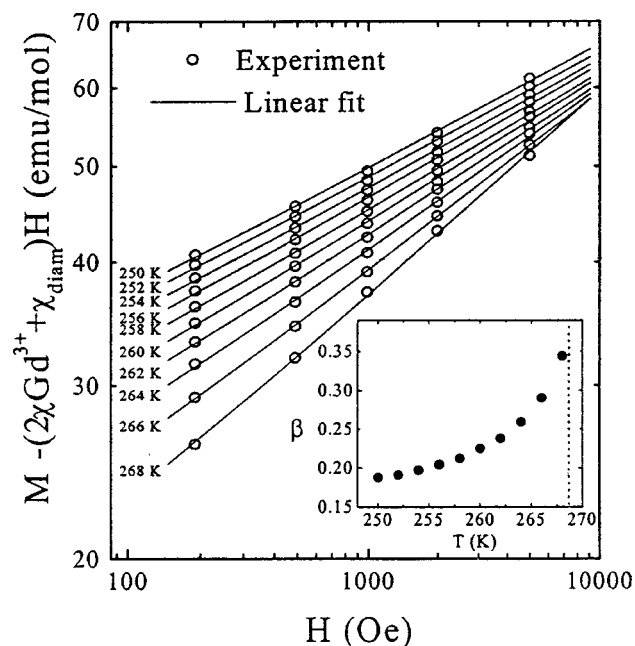


FIG. 2. Log-log plots of the dc magnetization of a Gd_2CuO_4 single crystal versus magnetic field. From linear fits of the data β is obtained from each isotherm, using the previously calculated value of γ . The polarization of the Cu sublattice on the R site does not affect the slope of the log-log lines (from which β is calculated). If this contribution is represented as $\lambda 2\chi_{Gd^{3+}}$ (Ref. 18), such term will only modify the cut of the log-log line with the M axis. Inset: Temperature dependence of β calculated from the log-log plots.

system. The nature of the coupling of Cu atoms in R_2CuO_4 materials has been a matter of controversy. The separation between CuO_2 planes makes the difference between intra-layer interactions, $J_{\parallel}$, and interlayer ones, $J_{\perp}$, very important. De Jongh and Miedema have signaled a ratio $J_{\parallel}/J_{\perp} > 100$ to have an effective 2D behavior.²¹ For Gd_2CuO_4 , estimations of $J_{\parallel}$ and $J_{\perp}$ are 1500 K and 2.3 mK, respectively,²² i.e., $J_{\parallel}/J_{\perp} > 6 \times 10^5$. This means that the system is a good candidate to exhibit 2D behavior. Non-WF members of the family are even more clear 2D systems, with higher ratios due to the cancellation of the exchange fields between the nearest-neighbor planes.⁸ In this way, 2D correlations begin to develop well above T_N^{3D} , until at temperatures between 240 and 290 K a global AF arrangement is present.^{6,7} In the prototypical K_2NiF_4 , a similar process was observed three decades ago.²³ It was interpreted as a crossover from 2D to 3D behavior. By analogy a similar crossover has been proposed for R_2CuO_4 materials. According to the classical theory of weak ferromagnetism,²⁴ T_{WF} is not exactly the Néel temperature of the ideal undistorted Cu sublattice (pure AF alignment). Actually, if $T_N = C'J$ (C' a constant and J the superexchange coupling value) then $T_{WF} = \sqrt{J^2 + D^2}$, with $D = 2J\phi$, ϕ the canting angle of the Cu moments. In the case of weak ferromagnetic cuprates this angle is about $0.5(2)^\circ$.²⁵ Introducing an approximate value of $J \sim 1500$ K, a correction of less than 1% for the ideal T_N^{3D} is obtained. Therefore the WF component scarcely affects the ordering point of the Cu sublattice, and T_N^{3D} can be discussed inspecting T_{WF} . Like in non-WF R_2CuO_4 compounds, 2D correlations are appreciable in Gd_2CuO_4 above T_{WF} . It is detected because they are responsible for the induction of WF above T_{WF} with an external applied magnetic field. Such prolongation of WF under the external magnetic field above the critical point can be of 50 K.²⁶ This extension of the temperature range is only possible due to the appreciable 2D correlations among the Cu moments after the breaking of the 3D arrangement.

The critical exponents, $\gamma = 1.32(2)$ and $\beta = 0.34(1)$ (at $T = 268$ K), differ from those given by Rouco *et al.*²⁵ for Y_2CuO_4 (1.10 and 0.53, respectively) and indicate that the system can be described with a 3D-Heisenberg model in the critical region. If the system were 2D around T_{WF} , an anisotropy term, described by a field H_A , should be present in order to allow long-range order above T_{WF} .²⁷ If $g\mu_B H_A \gg J_{\perp}$, it would behave as a 2D-Ising type system, but near the critical temperature $J_{\perp}$ would play an active role and the system would behave as a 3D one. Nevertheless, the most appropriate scenario could be a 2D XY one. The variation of β seems to confirm this hypothesis if we take into account that in the range $1 - T/T_{WF} < 0.06$ log-log plots give a value of β that can be compared to theory: at 250 K $\beta = 0.19$, but when approaching T_{WF} it changes over to a 3D-Heisenberg value, $\beta \sim 1/3$. Bramwell and Holdsworth²⁸ have signaled that $\beta = 0.23$ (close to the value we obtain) is a universal signature of finite-size 2D XY behavior and Als-Nielsen *et al.*²⁹ have found a behavior similar to the one reported here for Rb_2CrCl_4 (i.e., a 2D XY regime below a zone, around a 3D ordering point, exhibiting three-dimensional critical behavior). Chattopadhyay *et al.*³⁰ found for Gd_2CuO_4

$\beta=0.23$, which coincides with the theoretically proposed value, supporting the 2D XY hypothesis. The 3D character is active by means of $J_{\perp}$ at the point at which the phase transition occurs. This phase transition might be from a pure 2D behavior to a 2D-XY one. At the moment this 2D-XY regime is active, the 3D AF order would immediately follow, due to the weak interlayer coupling, $J_{\perp}$. Matsuda *et al.*⁸ have already invoked a similar mechanism, signaling values of intraplanar anisotropy, J_{XY} , on the order of $J_{XY}/J_{\parallel}=1.4 \times 10^{-4}$ for Nd_2CuO_4 , for example, in order to explain the similar 3D ordering points of the Cu sublattices in the R_2CuO_4 series, despite the differences in their $J_{\perp}$'s.

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