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Relaxation of dc magnetization in Gd_2CuO_4

J. Mira[a], J. Castro[a,*], J. Mahia[a], C. Vazquez-Vazquez[a], M.A. Lopez-Quintela[a], J. Rivas[a], S.B. Oseroff[b]

[a] *Departamento de Física Aplicada, Facultad de Física, Campus Universitario, Universidad de Santiago de Compostela, 15706 Santiago de Compostela, Spain*

[b] *San Diego State University, San Diego, CA, USA*

Abstract

In this work, detailed relaxation measurements of dc magnetization in ceramic Gd_2CuO_4 are presented. This material presents a weak ferromagnetic order in the CuO_2 planar lattice below $T_N \approx 290$ K. From both the presence of irreversibilities and the slow decay of the remanent magnetization, it was concluded that the magnetic state below T_N shows a spin glass like behaviour.

1. Introduction

Rare earth cuprates of the R_2CuO_4 type R = Pr, Nd and Sm are basic compounds of interest in electron-doped high T_C superconductors R_{2-x}-(Ce, Th)$_x$CuO$_4$ [1,2]. An ordering of the Cu spins at temperatures between 250 and 280 K in simple antiferromagnetic configurations appears in these materials [3,4]. Instead, the heavier rare earths R = Gd, Tb, etc. present a more complex ordering with a weak ferromagnetic component and do not become superconductors when doped with Ce or Th [5]. Consequently, superconductivity (SC) and weak ferromagnetism (WF) seem to be mutually excluding in these materials.

Extensive research of several magnetic properties of solid solutions $(R, R')_2CuO_4$ suggest the existence of limits for WF associated with the size of CuO_2 planar lattice. Eu_2CuO_4 with $a \approx 3.910(1)$ Å is in this boundary, presenting indications of extremely weak ferromagnetism in pure compounds and difficulties for becoming a superconductor when doped with Ce.

A spin glass behaviour has been observed in measurements of ac susceptibility and time decay of dc magnetization in Tb_2CuO_4 [6]. In this work we present a detailed experimental study of the relaxation phenomena associated with dc magnetization measurements in Gd_2CuO_4 in the $77 < T(K) < 300$ temperature range.

2. Experimental techniques

For the ceramic synthesis of Gd_2CuO_4, Gd_2O_3 and CuO oxides from Aldrich p.a. were used as starting materials. A stoichiometric quantity of these materials was milled for several hours and

* Corresponding author. Tel: +34-81 563 100, ext. 4022. Telefax: +34-81 595 012.

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thermal treatments at 850°C with frequent intermediate millings were carried out afterwards. Around 400 h were required in order to obtain the pure Gd_2CuO_4 phase. The structural characterization of the samples was carried out by means of X-ray diffraction using a Philips PW-1710 diffractometer with Cu anode ($CuK_{\alpha 1}$ radiation, $\lambda = 1.54060$ Å). The analytic characterization was carried out using Inductively Coupled Plasma Atomic Emission Spectroscopy (ICPAES) with Ar plasma ICP (Perkin-Elmer 5000). The static magnetic measurements were performed using a SQUID magnetometer (Quantum Design) in the $2 < T(\mathrm{K}) < 300$ temperature range and the relaxation measurements were carried out by means of a vibrating sample magnetometer (digital measurement systems 1660) in the $77 < T(\mathrm{K}) < 300$ temperature range.

3. Experimental results

In Fig. 1 we show the dc magnetization measurements for a Gd_2CuO_4 sample when it was (a) zero field cooled (ZFC) from 300 to 4 K ($H_{\mathrm{cool}} < 1$ Oe) and subsequently measured at increasing temperatures with an applied field $H_{\mathrm{appl}} = 5$ Oe and (b) field cooled (FC), i.e. measured at an applied field while cooling down from 300 K at the same field

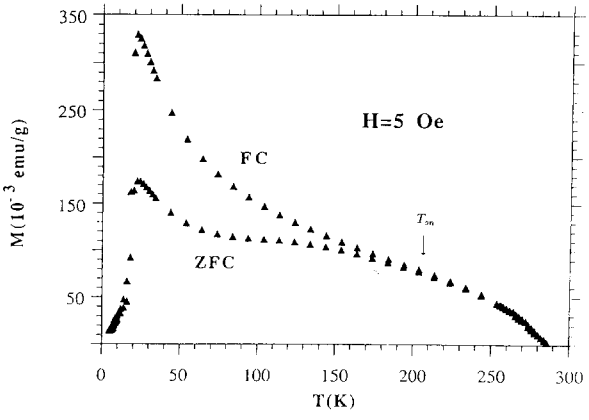

Fig. 1. FC and ZFC dc magnetization of Gd_2CuO_4 versus temperature, measured with an applied field of 5 Oe.

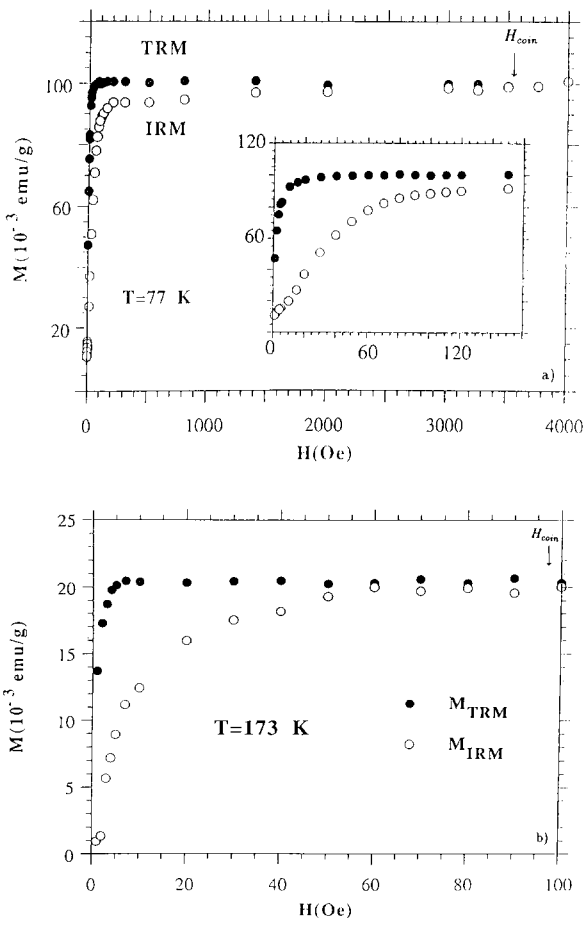

Fig. 2. TRM and IRM measurements for Gd_2CuO_4 sample at selected temperatures. (a) $T = 77$ K; the inset shows the detail of M versus H for low fields. (b) $T = 173$ K.

($H_{\mathrm{cool}} = H_{\mathrm{appl}} = 5$ Oe). The difference between the ZFC and FC curves indicate the onset of irreversibility at a temperature $T_{\mathrm{on}} \approx 200$ K for this field (Fig. 1).

The nature of the magnetic state below the temperature at which irreversibility sets in has also been examined by measurements of the thermoremanent (TRM) and isoremanent (IRM) magnetizations at selected temperatures (Fig. 2). These measurements show that the IRM and TRM curves increase with field at different rates until they reach the same saturated value $M_{\mathrm{Rs}}(T)$. We denote as H_{coin} the field where both magnetization components become coincident. For the magnetization

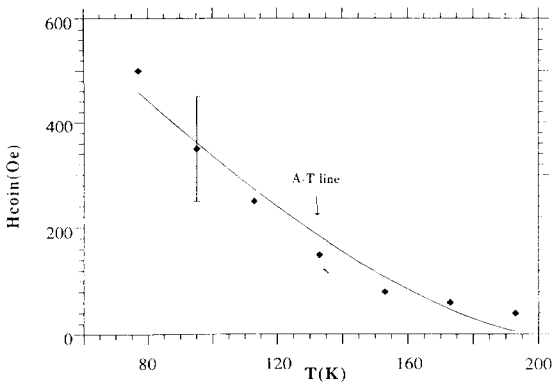

Fig. 3. T–H phase diagram for Gd_2CuO_4. The solid line shows the best fitting to the Almeida–Thouless model $H = t^{3/2}$.

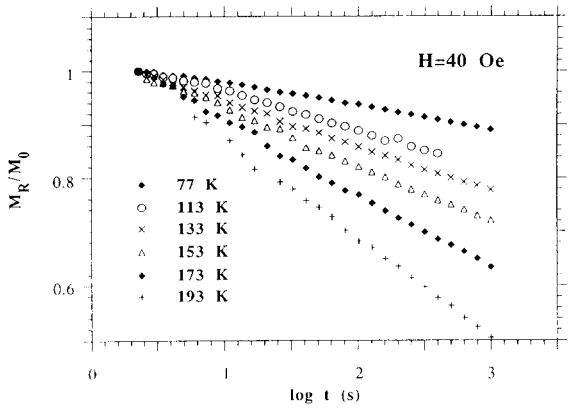

Fig. 4. Long time isotherms of the TRM at selected temperatures.

components we have checked the relationship: $M_{FC}(H, T) - M_{ZFC}(H, T) \approx M_{TRM}(H, T) - M_{IRM}(H, T)$ and we have found a good agreement in the range of temperatures and fields used in this work. Thus, at a given temperature, T, H_{coin} is the measuring field for which $T_{on} = T$.

In Fig. 3 we have plotted the measuring field versus the onset temperature. We interpret this figure as an Almeida–Thouless (AT) line in the sense that it marks the boundary between reversible and irreversible behaviours. The AT predicted dependence $H_{coin} = Ct^{3/2}$, where $t = \{1 - [T_{on}(H)/T_{on}(0)]\}$ is also shown.

4. Discussion

We also studied the hysteresis behaviour of a Gd_2CuO_4 sample below T_{on}. We have observed a wide variation in the shape of the hysteresis loop, which depends on the magnetic history of the sample. In the case of a ZFC sample we always obtain a symmetric and feautureless hysteresis loop with small but appreciable coercitivity and remanence. In the case of a field cooled sample, for $H_{cool} > 10$ kOe we observe a displaced hysteresis loop.

To measure the time dependence of the relaxation of the thermoremanent magnetization, the sample was cooled in an applied field from a reference temperature $T_R > 300$ K, above the Neel ordering temperature, to a measuring temperature $T_M > 77$ K, then the field was reduced to zero and the decay of the thermoremanent magnetization was observed up to 10^4 s. We have thus two different waiting times t_w in our experimental procedure, the cooling time from T_R to T_M (t_{w1}) and the time that the sample stays at the measuring temperature before the applied field is switched off (t_{w2}). In our case (t_{w1}) was always of the order of 100 s and in order to investigate the possible aging effects (t_{w2}) was varied from 30 to 90 min. We have also investigated the influence of the applied field on the relaxation behaviour. At a given temperature the applied fields were choisen beneath the AT line.

Two important experimental results are that the relaxation is almost independent of our applied field and that we have not encountered aging effect in the investigated region of fields, times and temperatures. Fig. 4 shows the dependence of the TRM with time at selected temperatures for an applied field of 40 Oe.

Several relaxation functions were proposed for the description of the time dependence of the TRM.

(i) Logarithmic law [7]:

$$M_{TRM}(t) = M_0 - S \ln t. \tag{1}$$

(ii) Power law [8]:

$$M_{TRM}(t) \propto \Delta M t^{-\alpha}. \tag{2}$$

(iii) Stretched exponential law [9]:

$$M_{TRM}(t) \propto \Delta M \exp[-(t/\tau)^{\beta}]. \tag{3}$$

Table 1.
Parameters of the fits

T(K)	S/M_0	α	β	$\tau(s)$
77	0.020	0.021	0.107	650
113	0.031	0.035	0.113	275
133	0.034	0.040	0.130	90
153	0.047	0.055	0.150	8
173	0.062	0.080	0.120	4
193	0.083	0.114	0.118	1

Preliminary fits of our experimental data to these three relaxation functions were performed. All these functions fit the data within the experimental error. The parameters obtained are shown in Table 1.

5. Conclusions

Gd_2CuO_4 below $T \approx 290$ K presents WF and also some spin glass characteristics:

(i) large differences between the ZFC and FC magnetization;

(ii) differences between the TRM and IRM magnetization;

(iii) the presence of very slow relaxation processes.

As an explanation for the microscopic origin of the WF in Gd_2CuO_4 the presence of Dzyaloshinky–Moriya (DM) interaction between neighbouring Cu moments have been suggested. Due the tetragonal symmetry of the lattice these are not allowed and crystal distortions are required in order to have a non-zero interaction. Several evidences for the presence of these distortions were presented [10]. As a possible origin of the spin glass like behaviour has been assumed that these distortions are random [11]. At the moment there is not clear indications of the actual existence of this randomness.

Further work is in progress in order to clarify the microscopic basis of the glassy features presented here.

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