Gd concentration dependence of the spin reorientation critical field in Eu$_{2-x}$Gd$_x$CuO$_4$

A. Butera, A. Fainstein, and M. Tovar
Centro Atómico Bariloche and Instituto Balseiro, Comisión Nacional de Energía Atómica, 8400 San Carlos de Bariloche, Rio Negro, Argentina

Z. Fisk
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

S. B. Oseroff
Physics Department, San Diego State University, San Diego, California 92182

ESR measurements of the microwave absorption signal associated with weak ferromagnetism in single crystals of Eu$_{2-x}$Gd$_x$CuO$_4$ are presented for X band (9.5 GHz) and L band (1.2 GHz) as a function of the Gd concentration. The strong absorption observed at low magnetic fields was interpreted, for samples with low Gd concentration, as due to a field-induced spin reorientation transition occurring at a critical field $H_c$, coincident with the in-plane magnetic anisotropy effective field $H_{c\text{eff}}$. For larger x the Cu-Gd magnetic interaction needs to be considered leading to smaller $H_c$ values. Our measurements show that for Gd concentrations in the range 0 ≤ x ≤ 1 the experimental data can be very well fitted with parameters derived from previous measurements. This fact indicates that these compounds have nearly the same in-plane anisotropy effective field, in spite of the small changes in lattice parameters. For x = 2 lattice distortions increase causing an $H_c$ larger than the expected one.

I. INTRODUCTION

The rate earth cuprates RE$_2$CuO$_4$ (RE=Pr,..., Tm), parent compounds of the so-called n-type high $T_c$ superconductors, crystalize in the tetragonal Nd$_2$CuO$_4$ $T^1$ structure. Pr$_2$CuO$_4$ has the largest a lattice parameter which decreases monotonically for the heavier RE compounds due to the smaller rare earth ion size. This lattice reduction causes a distortion of the CuO$_2$ planes for Eu$_2$GdCuO$_4$ single crystals, slightly doped with Gd. The Cu-Gd magnetic interaction needs to be considered leading to smaller $H_c$ values. Our measurements show that for Gd concentrations in the range 0 ≤ x ≤ 1 the experimental data can be very well fitted with parameters derived from previous measurements. This fact indicates that these compounds have nearly the same in-plane anisotropy effective field, in spite of the small changes in lattice parameters. For x = 2 lattice distortions increase causing an $H_c$ larger than the expected one.

II. MODEL

The following expression for the magnetic free energy of the coupled PM–WF system was proposed:

\[ F_{\text{eff}} = -K_{\text{eff}}^x m_{\text{WF}}^2 + K_{\text{eff}}^y m_{\text{WF}}^2 - m_{\text{WF}}^2 H_{\text{0}} + \left( \frac{1}{2\chi_{\text{Gd}}} \right) M_{\text{Gd}}^2 - M_{\text{Gd}}^2 H_{\text{0}} - \lambda \mathbf{m}_{\text{WF}} \cdot \mathbf{M}_{\text{Gd}}, \]  

where $m_{\text{WF}}$ and $M_{\text{Gd}}$ are the Cu-WF and Gd-PM magnetizations, $2K_{\text{eff}}^x m_{\text{WF}} = H_{\text{eff}}^x$ and $2K_{\text{eff}}^y m_{\text{WF}} = H_{\text{eff}}^y$ are in-plane and out-of-plane magnetic anisotropy fields, $\lambda$ is the Cu-Gd coupling constant and $\chi_{\text{Gd}} = C/(T + \Theta)$ is the Gd molar magnetic susceptibility. This effective free energy describes the equilibrium and the low-energy excitations of the system. The resonance modes can be obtained solving a 6×6 dynamical matrix for $m_{\text{WF}}$ and $M_{\text{Gd}}$. Two modes are obtained: a high-energy WF-like mode, and a low-energy PM-like one.

In Fig. 1 we show both modes, as calculated for $\lambda \chi_{\text{Gd}} = 0$ (dashed curves) and $\lambda \chi_{\text{Gd}} \neq 0$ (solid curves). For $\lambda \chi_{\text{Gd}} = 0$ a softening of the WF mode would occur at a critical field $H_c = H_{\text{eff}}^y$, coincident with the field-induced spin reorientation transition when H is applied perpendicular to the easy axis (i.e., $\varphi = 90^\circ$). Note that for X and L bands a resonance absorption is expected only for $\varphi = 90^\circ$. For $\lambda \chi_{\text{Gd}} \neq 0$ the critical field is reduced by a factor $1 + \lambda \chi_{\text{Gd}}$ giving

\[ H_c = \frac{H_{\text{eff}}^y}{1 + \lambda \chi_{\text{Gd}}}, \]
and the WF mode does not soften to zero. This would imply that no resonance arising from the WF ordered Cu lattice should be observed at X and L bands. However, a maximum in the microwave absorption may be expected due to nonresonant losses as described in Ref. 5. In addition an energy gap opens in the PM-like branch which softens to zero at $H_c^x$. For $T>T_N$ the energy gap becomes zero again. Because of the anticrossing of the coupled modes an “anomaly” is also predicted in the PM-like branch which softens to zero at $H_c^x$.

III. RESULTS AND DISCUSSION

Eu$_{2-x}$Gd$_x$CuO$_4$ single crystals were grown following standard flux techniques in Pt crucibles. In all cases crystals grew in the shape of small platelets with the $c$ crystallographic axis perpendicular to the axis. EPR measurements were made in a Bruker ESP 300 spectrometer at X band (9.5 GHz) and L band (1.2 GHz) between 120 and 300 K.

Although the softening of the WF mode at $H_c^x$ is not complete for $\varphi=90^\circ$ (see Fig. 1), originating a strong reduction in the intensity of the WF line at the $X$ band, it could still be clearly detected in all samples due to nonresonant losses. In L band, however, no line was found for samples with low Gd content (0 $\leq x \leq 0.2$) indicating that the gap was large enough to prevent even the observation of nonresonant losses. We have determined the Gd concentration dependence of $H_c^x$, coincident with the X-band resonance field of the WF line, at $T = 120$ K and with $H_L$ [110]$_{FC}$. The experimental data, presented in Fig. 2, were fitted using Eq. (2) in the range 0 $\leq x \leq 1$. We obtain the following values for the in-plane anisotropy effective field and the Cu-Gd coupling constant: $H_{c0}^x = 425(5)$ G and $\lambda' = 1.2(1) \times 10^5$ G/(μμB/Cu-atom). These are consistent with the values found in Refs. 5 and 8 (for samples slightly doped with Gd) and in Ref. 9, respectively. Note that the experimental data can be explained with a single value of the in-plane anisotropy field for all compounds (0 $\leq x \leq 1$), although the lattice size varies and consequently the displacement of the oxygen ions might change. We did not include the value measured for Gd$_2$CuO$_4$ in the fit because it was proposed$^{10}$ that for this compound Eq. (2) should be corrected due to the presence of a metamagnetic-like transition at low fields. In Ref. 10 the value of $H_{c0}^x$ for $x = 2$ was estimated, from dc magnetization measurements, to be $\sim 1200$ G at $T = 120$ K. This value is nearly three times larger than the one measured for samples with lower Gd concentrations$^8$ probably due to the larger lattice distortions. Correspondingly, the measured $H_c^x$ value is almost three times larger than that predicted assuming a constant $H_{c0}^x$ (see Fig. 2).

In Fig. 3 we show the EPR spectra of Gd$_2$CuO$_4$ mea-
measured at $L$ band for $T = 250$ K. The PM mode in absence of coupling to the Cu lattice would occur at $\omega_{PM}/\gamma_{Gd}=410$ G. Due to the large linewidth of the Gd$^{3+}$ EPR line in Gd$_2$CuO$_4$ measured at X band$^4 (\Delta H_{pp} \approx 1500$ G) a superposition of the absorptions occurring at negative and positive fields would cause a strongly asymmetric signal with a broad minimum at $\approx 750$ G. Surprisingly a narrow and very intense line is observed at a lower field, $H_r \approx 100$ G. The origin of this absorption may be explained looking at the behavior of the PM-like mode in Fig. 1. From the X-band results we estimate the energy gap of the PM-like mode at $T = 120$ K, $(\omega_{PM}/\gamma_{Gd})H_0 \approx 300$ G, lower than the $L$-band frequency $\omega_{L}/\gamma_{Gd}=410$ G. Thus the microwave absorption should occur at fields lower than that corresponding to $g = 2$. When the temperature is increased $H_r$ moves to lower fields (see Fig. 4) and the line disappears at $T = 265$ K, i.e., 20 K below $T_N$. As we have mentioned above, the value of the energy gap of the PM-like mode is expected to increase as $T$ rises and hence $T = 265$ K would indicate the temperature where $\omega_{PM}$ equals $\omega_{L}$.

In this description the reduced linewidth of the PM-like absorption may be associated with coupled excitations. In fact, a strong mixture of modes is expected especially when an anticrossing of modes occurs at $H_r$.

In summary, we have analyzed the variation of the spin reorientation critical field, $H_c = H_{Gd}(1 + \lambda Gd)$, as a function of Gd concentration. We have found that the decrease of $H_c$ for increasing $x$ may be explained (in the range $0 < x < 1$) in terms of the magnetic coupling between the Cu-WF and the Gd-PM lattices. We have also discussed the origin of the absorption line measured at $L$ band and suggested that it is due to the PM-like branch of the PM-WF coupled modes.

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