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Gd concentration dependence of the spin reorientation critical field in $Eu_{2-x}Gd_xCuO_4$

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ESR measurements of the microwave absorption signal associated with weak ferromagnetism in single crystals of $Eu_{2-x}Gd_xCuO_4$ 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^y_{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 $0 \leqslant x \leqslant 1$ 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. © *1996 American Institute of Physics.* [S0021-8979(96)03008-X]

I. INTRODUCTION

The rate earth cuprates RE_2CuO_4 (RE=Pr,..., Tm), parent compounds of the so-called n-type high T_C superconductors, crystallize in the tetragonal Nd_2CuO_4 T' structure.[1] Pr_2CuO_4 has the largest a lattice parameter which decreases monotonically for the heavier RE compounds due to the smaller rare earth ion size.[2] This lattice reduction causes a distortion of the CuO_2 planes for Eu_2CuO_4 ($a=3.910$ Å) and heavier compounds, consisting of a displacement of the in-plane oxygens [called O(1)] from its centrosymmetrical position. These displacements seem to be ordered, and several superstructures have been found in single crystals with diffraction techniques.[2,3] Copper moments order antiferromagnetically (AF) below room temperature for the whole series. However, coincident with the boundary for lattice distortions, a weak ferromagnetic (WF) component in the magnetization develops, that was attributed to a canting of the copper moment away from a perfect AF alignment.[4] Several experimental techniques have been used to characterize the WF behavior. Among them, microwave absorption proved to be very useful to elucidate the magnetic behavior of the compounds near the WF boundary.[5] X- and Q-band experiments in Eu_2CuO_4 single crystals, slightly doped with Gd, have shown that the WF moment lays in the CuO_2 plane pointing parallel to an easy axis defined by the field cooling (FC) magnetic field and coincident with a [110] crystallographic direction ($[110]_{\text{FC}}$). When the external field is applied perpendicular to this axis a field-induced spin reorientation transition occurs at a critical field H_c.[5] More recently[6] it was shown that, for samples with larger amounts of Gd, the interaction between the WF ordered Cu lattice and the Gd paramagnetic (PM) lattice must be taken into account in order to explain several anomalies found in the EPR spectra. In this article we present the Gd concentration dependence of H_c obtained from X-band measurements. We also discuss the L-band results within the proposed model.

II. MODEL

The following expression for the magnetic free energy of the coupled PM–WF system was proposed:[6]

$$F_{\text{eff}} = -K^y_{\text{eff}} m^2_{\text{WF},y} + K^z_{\text{eff}} m^2_{\text{WF},z} - \mathbf{m}_{\text{WF}} \cdot \mathbf{H}_0 + \left(\frac{1}{2\chi_{\text{Gd}}}\right) M^2_{\text{Gd}} - \mathbf{M}_{\text{Gd}} \cdot \mathbf{H}_0 - \lambda' \mathbf{m}_{\text{WF}} \cdot \mathbf{M}_{\text{Gd}}, \qquad (1)$$

$\mathbf{m}_{\text{WF}}$ and $\mathbf{M}_{\text{Gd}}$ are the Cu-WF and Gd-PM magnetizations, $2K^y_{\text{eff}} m_{\text{WF}} = H^y_{\text{eff}}$ and $2K^z_{\text{eff}} m_{\text{WF}} = H^z_{\text{eff}}$ are in-plane and out-of-plane magnetic anisotropy fields, λ' is the Cu-Gd coupling constant and $\chi_{\text{Gd}} = xC/(T+\Theta)$ 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 $\mathbf{m}_{\text{WF}}$ and $\mathbf{M}_{\text{Gd}}$. 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 $H_c = H^y_{\text{eff}}$, coincident with the field-induced spin reorientation transition when $\mathbf{H}$ is applied perpendicular to the easy axis (i.e., $\varphi=90°$). Note that for X and L bands a resonance absorption is expected only for $\varphi=90°$. For $\lambda'\chi_{\text{Gd}} \neq 0$ the critical field is reduced by a factor $1+\lambda'\chi_{\text{Gd}}$ giving

$$H_c = \frac{H^y_{\text{eff}}}{1+\lambda'\chi_{\text{Gd}}}, \qquad (2)$$

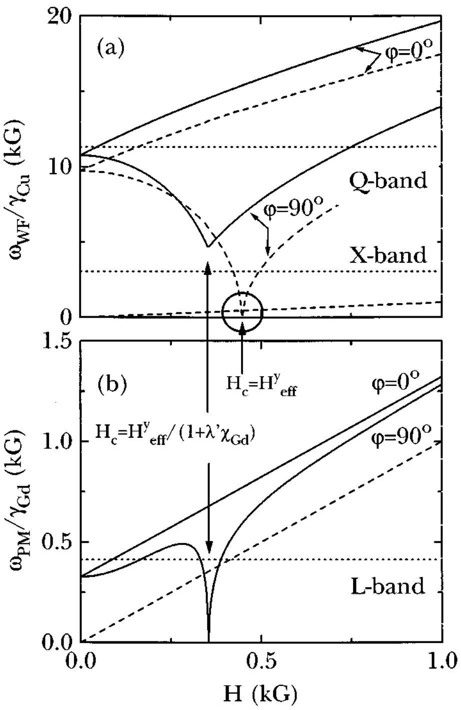

FIG. 1. Magnetic field dependence of (a) the WF-like and (b) the PM-like resonance modes when H is applied within the easy plane (solid curves). The dashed lines, calculated for $\lambda' \chi_{Gd} = 0$, illustrate the crossing of modes occurring at H_c. Note the shift of H_c to lower fields when $\lambda' \chi_{Gd} \neq 0$.

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 non-resonant loses as described in Ref. 5. In addition an energy gap opens in the PM-like mode for $H=0$ given by

$$\left(\frac{\omega_{PM}}{\gamma_{Gd}}\right)_{H=0} = \lambda' m_{WF} \sqrt{H^y_{eff}/[\lambda'^2 \chi_{Gd}(T) m_{WF} + H^y_{eff}]}. \quad (3)$$

Due to the temperature dependence of $\chi_{Gd}(T)$ the gap tends to zero for $T=0$ K and increases for $T>0$ because other parameters are only weakly temperature dependent.[6] 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.

III. RESULTS AND DISCUSSION

$Eu_{2-x}Gd_xCuO_4$ single crystals were grown following standard flux techniques in Pt crucibles.[7] 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 is not complete for $\varphi=90°$ (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 loses.[5] 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

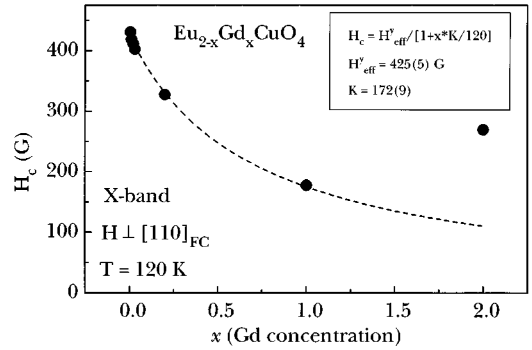

FIG. 2. Spin reorientation critical field, determined from the X-band low field absorption, as a function of x, the Gd concentration. The dashed line is a best fit of the experimental data in the range $0 \leq x \leq 1$ using Eq. (2).

losses. We have determined the Gd concentration dependence of H_c, coincident with the X-band resonance field of the WF line, at $T=120$ K and with $H \perp [110]_{FC}$. 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: $H^y_{eff} = 425(5)$ G and $\lambda' = 1.2(1) \times 10^5$ G/(μ_β/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_2CuO_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^y_{eff} for $x=2$ was estimated, from dc magnetization measurements, to be ≈ 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 value is almost three times larger than that predicted assuming a constant H^y_{eff} (see Fig. 2).

In Fig. 3 we show the EPR spectra of Gd_2CuO_4 mea-

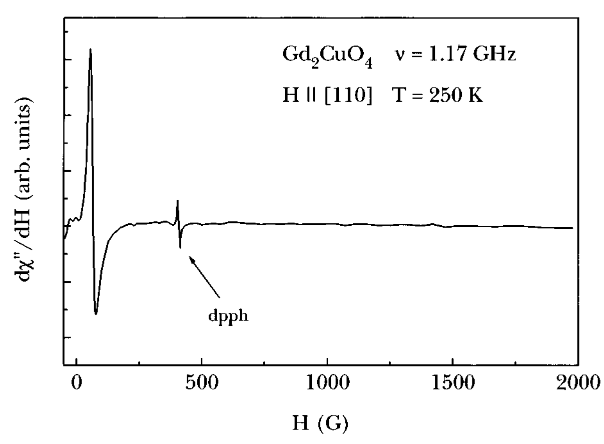

FIG. 3. L-band microwave absorption spectra of a Gd_2CuO_4 single crystal. Note the shift of the ω_{PM} line from $g=2$ (dpph).

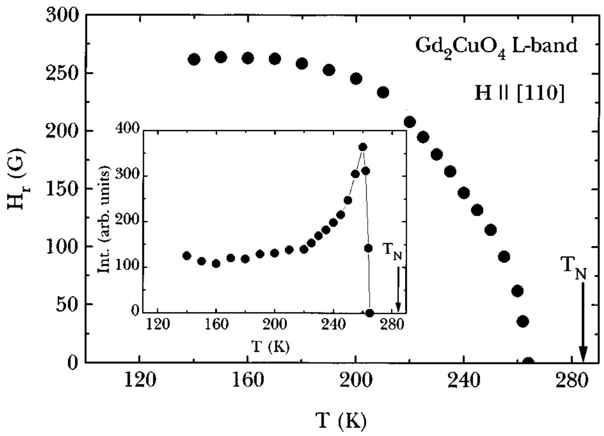

FIG. 4. Temperature dependence of the L-band Gd_2CuO_4 low field resonance line. Note the sharp decrease of the intensity at $T=260$ K, below the Néel temperature (inset).

sured 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_2CuO_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 ω_{PM} equals ω_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_c.

In summary, we have analyzed the variation of the spin reorientation critical field, $H_c = H^y_{eff}/(1+\lambda'\chi_{Gd})$, as a function of Gd concentration. We have found that the decrease of H_c for increasing x may be explained (in the range $0 \leq x \leq 1$) 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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