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Title

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Permalink

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Journal

Journal of Applied Physics, 69(8)

ISSN

0021-8979

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Publication Date

1991-04-15

DOI

10.1063/1.348208

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Peer reviewed

Observation of an unusual ESR signal in antiferromagnetic Eu_2CuO_4

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We report the observation of an unusual electron spin resonance (ESR) signal in single crystals of Eu_2CuO_4 . The signal appears to be associated with a resonance mode of the CuO_2 planes, similar to the midfield and low-field absorptions we have reported previously [Phys. Rev. B **41**, 1934 (1990)]. However, it is only observed when the projection of the applied dc magnetic field in the CuO_2 plane is within a few degrees of the $\langle 110 \rangle$ crystallographic direction. Additionally the sample must be field cooled in the CuO_2 plane, but with a component of the cooling field perpendicular to the $\langle 110 \rangle$ ESR observation direction. Both the field for resonance and the linewidth exhibit a $1/\cos \theta$ dependence, where θ is the angle of the applied dc field between the c axis and the $\langle 110 \rangle$ observation direction. Additional constraints for observation of the resonance are that the microwave rf magnetic field must have a component in the CuO_2 plane, but perpendicular to the dc field. The signal disappears above ~ 215 K, which we assume is associated with the antiferromagnetic ordering temperature.

We have previously reported on the unusual and diverse magnetic properties observed in single crystals of the rare-earth copper oxides, R_2CuO_4 with $\text{R} = \text{Eu}$ or Gd , below the Cu Neel ordering temperature T_N , which occurs at ~ 270 K.¹ In particular, we have observed two resonant absorptions, which we termed the low-field absorption (LFA) and midfield absorption (MFA). These absorptions exhibited a large out-of-plane anisotropy and were ascribed to resonant modes of the CuO_2 planes. They were not considered typical EPR signals in that the absorptions could be observed for any orientation of the microwave magnetic field h_{rf} to the applied dc magnetic field H_{dc} .

In this paper we report on a newly discovered microwave absorption signal (MAS) in single crystals of Eu_2CuO_4 which were prepared by standard techniques.¹ In contrast to the LFA and MFA, the MAS is not observable if h_{rf} is parallel to H_{dc} . The MAS exhibits the same out-of-plane anisotropy found for the low-field and midfield absorptions, but in addition, it exhibits a strong and unusual anisotropy within the CuO_2 plane. In particular, it is only observed when the dc magnetic field is oriented within a few degrees of the $\langle 110 \rangle$ crystallographic direction!

We find that the MAS is sample dependent. Although we have found it in both pure and Ce doped Eu_2CuO_4 , the signal is not observed in all of the Eu_2CuO_4 samples which we have investigated. We cannot rule out the possibility that the MAS arises from an impurity within the Eu_2CuO_4 compound, but if so, we are unaware of any prior report of an impurity response as unusual as that presented here.

The signal was detected with a standard EPR superheterodyne spectrometer operating at 9.2 GHz in the field derivative mode. Data was taken at temperatures ranging from 77–300 K. We have also observed the MAS at a frequency of 35 GHz and in Eu_2CuO_4 crystals doped with Gd and/or Ce. These preliminary results, as well as data taken at temperatures below 77 K, will be presented in a future publication.

In Fig. 1 we present spectra taken for a Eu_2CuO_4 single crystal at 77 K with H_{dc} applied along the CuO_2 plane at various angles near the $\langle 110 \rangle$ crystallographic direction. In order to observe the MAS, the sample must be cooled below T_N in a large dc magnetic field (discussed below). When H_{dc} is applied within the CuO_2 plane, the MAS

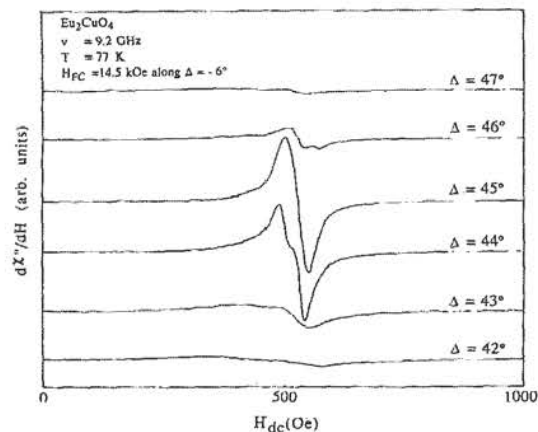


FIG. 1. Spectra for Eu_2CuO_4 taken at various angles, Δ , in the CuO_2 plane, near the $\langle 110 \rangle$ direction. The sample was cooled to 77 K in a field of 14.5 kOe applied along $\Delta = -6^\circ$.

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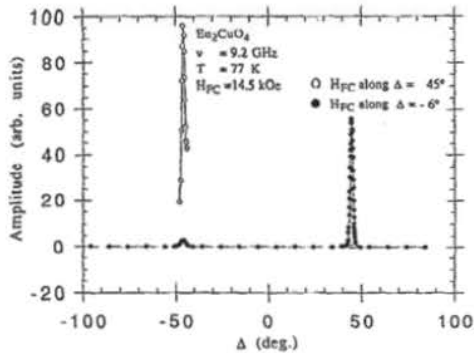


FIG. 2. Microwave absorption signal amplitude as a function of the angle Δ of H_{dc} within the CuO_2 plane. The sample was cooled to 77 K in a field of 14.5 kOe applied along either $\Delta = -6^\circ$ (solid circles) or $\Delta = 45^\circ$ (open circles). $\Delta = 0^\circ$ corresponds to the $\langle 100 \rangle$ direction and $\Delta = -45^\circ$ or 45° to the $\langle 110 \rangle$ direction.

exhibits a field for resonance, H_r , of ~ 500 Oe and a peak-to-peak linewidth ΔH of ~ 50 Oe. Note that the signal amplitude is a very sensitive function of the angle Δ of H_{dc} within the CuO_2 plane.

This dramatic in-plane anisotropy is further illustrated in Fig. 2, where we present the amplitude of the MAS as a function of Δ for two directions of cooling field, H_{FC} . When the sample is field cooled from above T_N to 77 K along any $\langle 110 \rangle$ direction, the MAS is only observed near the perpendicular $\langle 110 \rangle$ direction. In Fig. 2 the open circles correspond to cooling in a field H_{FC} along the $\langle 110 \rangle$ direction, $\Delta = 45^\circ$. In this case the signal is only observed near the perpendicular $\langle 110 \rangle$ direction, $\Delta = -45^\circ$. If the sample is cooled in a field pointing in an arbitrary direction (e.g., $\Delta = -6^\circ$) within the CuO_2 plane, the MAS is then observed in both $\langle 110 \rangle$ directions (solid circles in Fig. 2), with an amplitude which is largest for the $\langle 110 \rangle$ direction most nearly perpendicular to the direction of H_{FC} .

The MAS amplitude also depends strongly on the strength of the cooling field. This dependence is illustrated in Fig. 3. The sample was cooled in a field H_{FC} applied along a $\langle 110 \rangle$ direction and the maximum MAS amplitude was then measured along the perpendicular $\langle 110 \rangle$ direction. No signal was observed for $H_{FC} = 0$. From the figure it appears that a cooling field of about 14 kOe is necessary

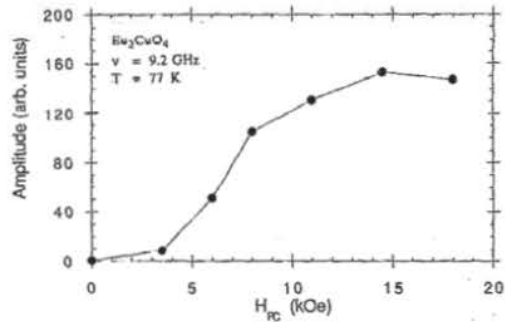


FIG. 3. Maximum microwave absorption signal amplitude as a function of the magnitude of the cooling field H_{FC} applied along a $\langle 110 \rangle$ direction.

to fully develop the MAS amplitude.

In addition to not observing the MAS if the Eu_2CuO_4 is cooled in zero field, the MAS is also not observed if the microwave magnetic field h_{rf} is either perpendicular to the CuO_2 plane or parallel to the measuring field H_{dc} .

The field for resonance H_r and the linewidth ΔH of the MAS are both found to be independent of the direction or strength of the cooling field. H_r , ΔH , and the signal intensity are also found to exhibit only a weak temperature dependence between 77 and ~ 150 K. However, as the temperature is increased above 150 K, H_r and ΔH both increase, and the signal intensity decreases, until the signal finally disappears at T_N .

H_r and ΔH are found to exhibit an out-of-plane anisotropy identical to the low-field and mid-field absorptions studied previously.¹ They both follow a $1/\cos \theta$ dependence where θ is the angle of the applied magnetic field H_{dc} to the CuO_2 plane. This out-of-plane anisotropy, and the nonobservability of the MAS for h_{rf} parallel to H_{dc} , is suggestive that the MAS is due to EPR of the Cu-O system.

Work partially supported at SDSU by NSF-DMR-88-01317 and NSF-INT-89-00851; at UCSD by NSF-DMR-86-13858 and ONR-N000-14-87-K-0338; and the work at Los Alamos National Laboratory under U.S. DOE.

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