Observation of an unusual ESR signal in antiferromagnetic Eu$_2$CuO$_4$

D. C. Vier and S. Schultz
Department of Physics 0319, University of California, San Diego, 9500 Gilman Drive, La Jolla, California 92033

C. Rettori, D. Rao, S. B. Oseroff, and M. Tovar
Department of Physics, San Diego State University, San Diego, California 92182

Z. Fisk and S-W. Cheong
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

We report the observation of an unusual electron spin resonance (ESR) signal in single crystals of Eu$_2$CuO$_4$. 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 (110) 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 (110) ESR observation direction. Both the field for resonance and the linewidth exhibit a $1/\cos \theta$ dependence, where $\theta$ is the angle of the applied dc field between the c axis and the (110) 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$_2$CuO$_4$ with R = Eu or Gd, below the Cu Neel ordering temperature $T_N$, which occurs at ~270 K. 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 $h_F$ 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$_2$CuO$_4$ which were prepared by standard techniques. In contrast to the LFA and MFA, the MAS is not observable if $h_F$ 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 (110) crystallographic direction.

We find that the MAS is sample dependent. Although we have found it in both pure and Ce doped Eu$_2$CuO$_4$, the signal is not observed in all of the Eu$_2$CuO$_4$ samples which we have investigated. We cannot rule out the possibility that the MAS arises from an impurity within the Eu$_2$CuO$_4$ 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$_2$CuO$_4$ 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$_2$CuO$_4$ single crystal at 77 K with $H_{dc}$ applied along the CuO$_2$ plane at various angles near the (110) 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

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![FIG. 1. Spectra for Eu$_2$CuO$_4$ taken at various angles, $\Delta$, in the CuO$_2$ plane, near the (110) direction. The sample was cooled to 77 K in a field of 14.5 kOe applied along $\Delta = -6^\circ$.](image-url)
FIG. 2. Microwave absorption signal amplitude as a function of the angle \( \Delta \) 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 \( 45^\circ \) to the \langle 110 \rangle direction.

A sample is cooled in a field pointing near the perpendicular \langle 110 \rangle direction. No signal was observed for the microwave magnetic field \( h_{zz} \) is either perpendicular to the CuO\(_2\) plane or parallel to the measuring field \( H_{dc} \).

This dramatic in-plane anisotropy is further illustrated in Fig. 2, where we present the amplitude of the MAS as a function of \( \Delta \) 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 to fully develop the MAS amplitude.

In addition to not observing the MAS if the Eu\(_2\)CuO\(_4\) is cooled in zero field, the MAS is also not observed if the microwave magnetic field \( h_{zz} \) 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 \( \Delta H \) of the MAS are both found to be independent of the direction or strength of the cooling field. \( H_r \), \( \Delta H \), and the signal intensity are also found to exhibit only a weak temperature dependence between 77 and \( \sim 150 \) K. However, as the temperature is increased above 150 K, \( H_r \) and \( \Delta H \) both increase, and the signal intensity decreases, until the signal finally disappears at \( T_N \).

\( H_r \) and \( \Delta H \) are found to exhibit an out-of-plane anisotropy identical to the low-field and midfield absorptions studied previously.\(^1\) They both follow a \( 1/\cos \theta \) dependence where \( \theta \) is the angle of the applied magnetic field \( H_{dc} \) parallel to the CuO\(_2\) plane. This out-of-plane anisotropy, and the nonobservability of the MAS for \( H_{dc} \) parallel to \( H_{dc} \) is suggestive that the MAS is due to EPR of the Cu–O system.

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\(^1\) S. B. Oseroff, D. Rao, F. Wright, D. C. Vier, S. Schultz, J. Thompson, Z. Fisk, S.-W. Cheong, M. F. Hundley, and M. Tovar, Phys. Rev. B \textbf{41}, 1934 (1990).