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Authors
Vier, DC
Schultz, S
Rettori, C
et al.

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Observation of an unusual ESR signal in antiferromagnetic Eu_2CuO_4

D. C. Vier and S. Schultz
Department of Physics 0319, University of California, San Diego, 9500 Gilman Drive, La Jolla, California 92093

C. Rettori,[a] D. Rao, S. B. Oseroff, and M. Tovar[b]
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_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 ⟨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 θ 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 $\sim$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 R = Eu or Gd, below the Cu Neel ordering temperature T_N, which occurs at $\sim$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 ⟨110⟩ 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 ⟨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

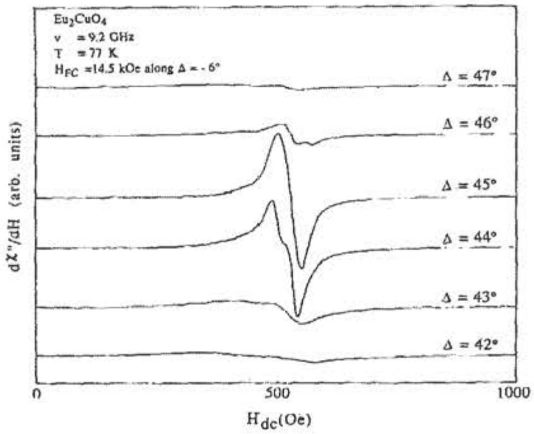

FIG. 1. Spectra for Eu_2CuO_4 taken at various angles, Δ, in the CuO_2 plane, near the ⟨110⟩ direction. The sample was cooled to 77 K in a field of 14.5 kOe applied along $\Delta = -6°$.

[a] Universidade Estadual de Campinas, Instituto de Fisica, CAIXA Postal 1170, 13.100 Campinas, Sao Paulo, Brazil.
[b] Central Atomico Bariloche, 8400 Bariloche, Rio Negro, Argentina.

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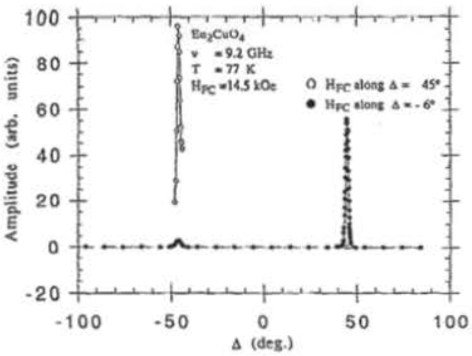

FIG. 2. Microwave absorption signal amplitude as a function of the angle Δ of H_{dc} within the CuO_2 plane. The sample was cooled to 77 K in a field of 14.5 kOe applied along either $\Delta = -6°$ (solid circles) or $\Delta = 45°$ (open circles). $\Delta = 0°$ corresponds to the $\langle 100 \rangle$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

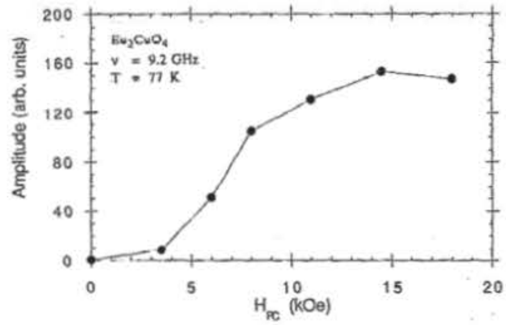

FIG. 3. Maximum microwave absorption signal amplitude as a function of the magnitude of the cooling field H_{FC} applied along a $\langle 110 \rangle$ direction.

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, $\Delta = 45°$. In this case the signal is only observed near the perpendicular $\langle 110 \rangle$ direction, $\Delta = -45°$. If the sample is cooled in a field pointing in an arbitrary direction (e.g., $\Delta = -6°$) 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_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 midfield 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.

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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 **41**, 1934 (1990).