# Antiferromagnetic resonance in $La_{2-x}CuO_{4-y}$

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We report the first observation of antiferromagnetic resonance in $La_{2-x}CuO_{4-y}$. Transmitting far-infrared radiation through our polycrystalline samples we observe a resonance at 9 $cm^{-1}$ from which we extract an anisotropy energy of $\simeq 0.02$ $cm^{-1}$. This is more than an order of magnitude smaller than in the quadratic-layer spin-1 antiferromagnet $K_2NiF_4$, in which anisotropy has been shown to be responsible for the observed Néel transition. Nevertheless, using the same analysis we find that, because of the very large intraplanar exchange coupling, the small anisotropy that we measure is sufficient to account for the observed Néel temperatures of up to 300 K in $La_{2-x}CuO_{4-y}$.

Following the recent discovery of high-temperature superconductivity in $La_{2-x}Ba_xCuO_4$,[1] an intense scrutiny of both the superconducting and normal-state properties of related compounds has naturally begun. In studies of the normal state, one hopes to find some unique property of the layered oxides that is responsible for, or in some way related to, the occurrence of high-temperature superconductivity. Particular attention has been paid to the antiferromagnetic behavior of $La_{2-x}CuO_{4-y}$,[2-5] and interest in magnetic mechanisms of superconductivity has been encouraged by recent reports of a large antiferromagnetic exchange energy in this material.[6-8] These approaches derive credibility from the belief that superconductivity mediated by spin-fluctuation exchange occurs (at much lower temperatures) in some heavy-fermion systems.

Antiferromagnetism in $La_{2-x}CuO_{4-y}$, initially suggested by susceptibility anomalies,[2] was confirmed by elastic neutron scattering.[3,4] More recently, inelastic neutron measurements[6] and two-magnon Raman scattering[7,8] have indicated that the intraplanar antiferromagnetic exchange interaction in this system is quite strong. Complementary to these techniques is the measurement of the spin-wave gap, or antiferromagnetic resonance (AFMR),[9] which probes the anisotropy in the underlying Hamiltonian. The existence of a spin-wave gap is particularly important to the magnetic properties of quadratic-layer antiferromagnets[10-12] because they are close to the two-dimensional Heisenberg limit in which long-range order is unstable with respect to thermal fluctuations. In the prototypical quadratic layer antiferromagnet, $K_2NiF_4$, a reasonably accurate estimate of the Néel temperature ($T_n = 97$ K) is provided by the calculation of Lines[10] in which the existence of $T_n > 0$ is wholly dependent on a nonvanishing spin-wave gap (anisotropy energy). Except for a small orthorhombic distortion, $La_{2-x}CuO_{4-y}$ is structurally isomorphic to $K_2NiF_4$.

In this paper we report a far-infrared measurement of the spin-wave gap (antiferromagnetic resonance) in polycrystalline $La_{2-x}CuO_{4-y}$ at temperatures from 0.4 to 20 K and in magnetic fields from 0–12 T. From the zero-field spin-wave gap of $\sim 9$ $cm^{-1}$ we deduce an anisotropy energy of about 0.02 $cm^{-1}$, which is more than an order of magnitude smaller than in $K_2NiF_4$,[12] and which demonstrates that $La_{2-x}CuO_{4-y}$ is close to the ideal Heisenberg limit. Nevertheless, we find that given the large value of $J$, this small anisotropy is sufficient to account for the observed transition to long-range order in $La_{2-x}CuO_{4-y}$.

The samples used in this study were cold-pressed pellets, approximately 1 mm thick and 5 mm in diameter, of $La_{2-x}CuO_{4-y}$ powder. The susceptibility of these powders as a function of $T$, given in Fig. 1, shows a peak near 200 K which has previously been associated with the onset of antiferromagnetic order.[2,5] To observe the antiferromagnetic resonance, we transmit far-infrared radiation from a polarizing interferometer through the sample at normal incidence and measure the transmitted intensity using a bolometric detector. We study the far-infrared transmission in magnetic fields of 0–12 T, applied parallel to the direction of propagation of the incident radiation, with the sample at temperatures between 0.4 and 20.0 K.

An absorption line at 9 $cm^{-1}$ is observed in transmission with no applied magnetic field. As the field is in-

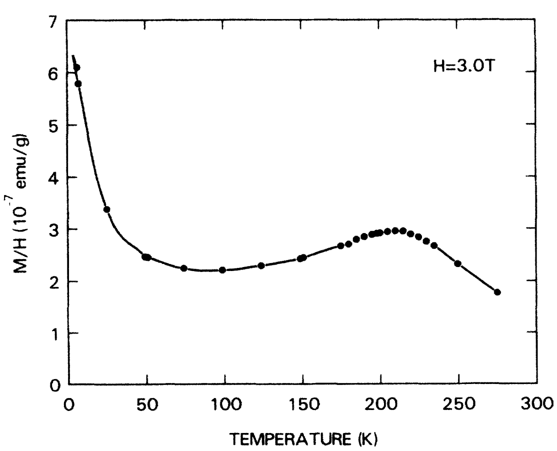


FIG. 1. The susceptibility of our $La_{2-x}CuO_{4-y}$ powder sample as a function of temperature. The broad maximum near 200 K is associated with antiferromagnetic order.

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creased the absorption line gradually disappears into the noise. This is to be expected in a polycrystalline (powder) sample, because an applied magnetic field causes the resonance to shift (or split) by different amounts in crystallites with different orientations with respect to the field.[9] Thus, the absorption broadens and drops in amplitude. In Fig. 2 we show spectra from 0–4 T, each divided by an 8 T reference spectrum for normalization. In the $H=0$ T ratio the antiferromagnetic resonance occurs at $\simeq 9$ cm$^{-1}$ with a width of about 3 cm$^{-1}$ and an integrated intensity of $\simeq 2$ cm$^{-2}$. For comparison, the $K_2NiF_4$ resonance frequency, width, and intensity are reported to be 18 cm$^{-1}$, $\simeq 0.5$ cm$^{-1}$, and 2.4 cm$^{-2}$, respectively, at 4.2 K.[12] Given the large effect oxygen (and possibly lanthanum) stoichiometry has on $T_n$,[5] the greater linewidth of the $La_{2-x}CuO_{4-y}$ resonance may indicate lifetime broadening due to scattering induced by oxygen defects. Sample inhomogeneity or the presence of overlapping resonances (an in-plane and out-of-plane mode as discussed below) may also contribute to the width.

The antiferromagnetic resonance frequency (spin-wave gap) for $H=0$ can be written as

$$\omega_c = (2\omega_e \omega_a)^{1/2} , \tag{1}$$

where $\omega_a$ is the anisotropy energy, $\omega_e = SzJ$ is the exchange energy, $S$ is the spin ($S=\frac{1}{2}$), $z$ is the number of nearest neighbors ($z=4$), and $J$ is the nearest-neighbor intraplanar antiferromagnetic exchange interaction. Respective values for $J$ of about 500 and 1000 cm$^{-1}$ have recently been reported from two independent studies of two magnon Raman scattering,[7,8] and a lower bound of $J \gtrsim 500$ cm$^{-1}$ is obtained from an analysis of neutron data.[6] Apparently some questions regarding the interpretation of the Raman peaks remain to be resolved, while the neutron data is consistent with either result. Using, for example, the larger value (1000 cm$^{-1}$) for $J$ we obtain an anisotropy energy $\omega_a \simeq 0.02$ cm$^{-1}$ (see Table I).

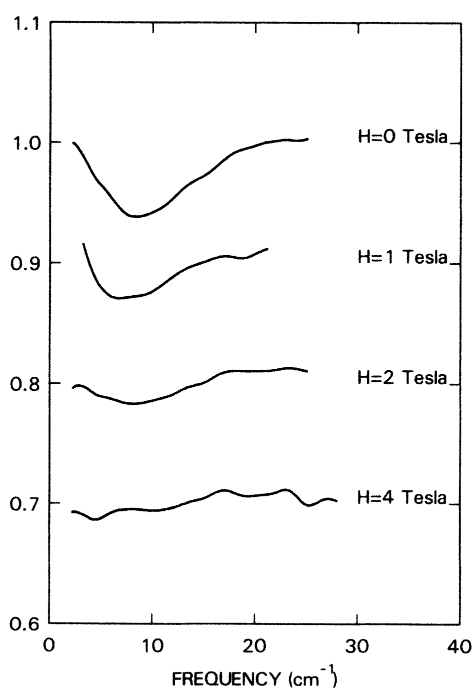


FIG. 2. The transmission though a polycrystalline $La_{2-x}CuO_{4-y}$ sample at 0, 1, 2, and 4 T, divided by the transmission at 8 T. The base lines have been shifted by 0.1 in each successive spectrum. Deviations of the base lines from unity are caused by drift between scans and are not significant. The antiferromagnetic resonance occurs at $\simeq 9$ cm$^{-1}$ for $H=0$.

Interpreting the anisotropy energy, $\omega_a$, is somewhat more difficult in $La_{2-x}CuO_{4-y}$ than in $K_2NiF_4$ because of the absence of uniaxial symmetry. In $K_2NiF_4$ the spins are perpendicular to the $a$-$b$ plane,[11] while in orthorhombic $La_{2-x}CuO_{4-y}$ the spins lie in the plane (along the $b$ axis),[3] in which case there are two potentially nondegenerate spin-wave modes at $q=0$.[9–13] Presumably the $c$ axis is the hard axis in this system and, thus, the out-of-plane mode has the higher frequency. Since our measurements were made on a powder, it is possible that we observe either one of these modes and that the other is outside the frequency range of our measurements. If the in-plane and out-of-plane anisotropy energies differ by roughly a factor of 2 or less, however, both modes would lie within the broad resonance which we observe at $\sim 9$ cm$^{-1}$. We believe this to be the case. This point of view is consistent with experimental results from other nonuniaxial, $s=\frac{1}{2}$ antiferromagnets[9,13] [e.g., $CuCl_2(2H_2O)$]. This interpretation could be directly tested by far-infrared measurements of single-crystal $La_{2-x}CuO_{4-y}$ samples.

In Table I a summary of some of the parameters of $K_2NiF_4$ and $La_{2-x}CuO_{4-y}$ is presented for comparison. If we use the value of $J$ reported by Lyons *et al.*,[8] then the exchange energy $\omega_e = SzJ$, which is characteristic of the zone-boundary magnon energy, is about seven times larger in $La_{2-x}CuO_{4-y}$ than in $K_2NiF_4$, while the anisotropy energy, $\omega_a$, is about 30 times smaller. (The anisotropy ratio $\omega_a/\omega_e$ is thus $\simeq 200$ times smaller in $La_{2-x}CuO_{4-y}$.) Presumably, in $La_{2-x}CuO_{4-y}$ this small anisotropy originates from dipolar anisotropy and anisotropic exchange interactions, since the crystal-field (single-ion) contribution, which is dominant in the $S=1$ antiferromagnet $K_2NiF_4$, should be absent for $S=\frac{1}{2}$.[10–13]

Of crucial importance in understanding a layered antiferromagnet is the question of whether a 2D or 3D ordering mechanism is dominant.[6,10–12] In the former case the

TABLE I. Characteristic frequencies for the quadratic-layer antiferromagnets $K_2NiF_4$ and $La_{2-x}CuO_{4-y}$. $J$ is the intraplanar exchange interaction, while $\omega_e$, $\omega_c$, and $\omega_a$ are the exchange energy, antiferromagnetic resonance frequency, and the anisotropy energy, respectively.

| | $K_2NiF_4$[a] | $La_{2-x}CuO_{4-y}$ | |
|---|---|---|---|
| $J$ (cm$^{-1}$) | 75 | 500[b] | 1000[c] |
| $\omega_e$ (cm$^{-1}$) | 300 | 1000 | 2000 |
| $\omega_c$ (cm$^{-1}$) | 18 | 9 | 9 |
| $\omega_a$ (cm$^{-1}$) | 0.5 | 0.04 | 0.02 |

[a]References 10–12.
[b]Reference 7.
[c]Reference 8.

anisotropy energy plays a crucial role, whereas in the latter case it is the interplanar coupling which drives the transition. For a 2D system (no interplanar coupling) Lines has shown that[10]

$$k_B T_n = \frac{S(S+1)}{3\langle \mu_k/(\mu_k^2-\lambda_k^2)\rangle_k} , \qquad (2)$$

where $(\mu_k^2-\lambda_k^2)$ is related to the magnon spectrum[10] and $\langle \ldots \rangle_k$ represents an average over the Brillouin zone. [In the absence of anisotropy there is no spin-wave gap (at $k=0$) and this average diverges, hence $T_n=0$.] For $K_2NiF_4$ measured values of $\omega_c$ and $J$ have been used in this approach to calculate $T_n$, and the agreement between theory and experiment has been interpreted as evidence for a two-dimensional mechanism.[10-12] With the same approach using the measured values of $\omega_c=9$ cm$^{-1}$ (13 K) and $J=1000$ cm$^{-1}$ ($\sim$1400 K) we obtain (from Fig. 7 of Ref. 10)

$$k_B T_n \simeq 0.3JS(S+1) \simeq 325 \text{ K} , \qquad (3)$$

which is adequate to explain the experimentally observed ordering temperatures of up to 300 K. Thus we find that while the anisotropy energy is extremely small, it may, nevertheless, play an important role in the ordering transition of $La_{2-x}CuO_{4-y}$. We also note that the small size of the anisotropy energy compared to the exchange energy $(\omega_a/\omega_e < 10^4)$ indicates $La_{2-x}CuO_{4-y}$ is close to the ideal 2D Heisenberg limit. Near this limit 2D critical fluctuations with large correlation lengths can be expected to persist at temperatures far in excess of $T_n$ as has been observed.[6] The exchange of such fluctuations provides the mediating interaction in a number of magnetic models of superconductivity (see, for example, Ref. 14).

The dependence of $T_n$ on oxygen deficiency[5] is not well understood. Perhaps an interplay between interplanar coupling (3D) and anisotropy (2D) ordering mechanisms[10] is involved. The former may be quite sensitive to oxygen defects that can break the symmetry which otherwise tends to cancel the exchange interaction between adjacent planes.[10,11]

In conclusion, we have measured the antiferromagnetic resonance frequency in $La_{2-x}CuO_{4-y}$ and observe a resonance at 9 cm$^{-1}$ with a width of $\sim$3 cm$^{-1}$ at $H=0$. The anisotropy energy deduced from this measurement is more than an order of magnitude smaller than in $K_2NiF_4$, demonstrating that $La_{2-x}CuO_{4-y}$ is very close to the ideal Heisenberg limit. Nevertheless, within the theoretical framework of Lines,[10] the anisotropy is sufficient to account for the observed Néel temperatures of up to 300 K in $La_{2-x}CuO_{4-y}$.

*Note added.* As R. J. Birgeneau and M. A. Kastner have recently pointed out, the lowering of the symmetry of $La_{2-x}CuO_{4-y}$ due to the rotation of the copper-oxygen octahedra makes allowed an antisymmetric exchange term in the spin Hamiltonian.[15] In this case, Eq. (1) for the in-plane residence frequency becomes $\omega_c = (2\omega_e\omega_a + \omega_{dm}^2)^{1/2}$, where $\omega_{dm}$ is the antisymmetric exchange energy. Thus, our experiment probes both $\omega_a$ and $\omega_{dm}$. Depending upon the size of $\omega_{dm}$, $\omega_a$ may be even smaller than the estimate given above which now provides an upper bound on $\omega_a$.

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