

## Far- and Mid-Infrared Spectrum of $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$ in High Magnetic Fields

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*The far- and mid-infrared spectrum of antiferromagnetic  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  was investigated by infrared transmission measurements ( $\vec{k} \parallel c$ -axis) in high magnetic fields up to 16.5 Tesla at  $T=1\text{K}$ . A peak at  $1436\text{ cm}^{-1}$  which previously was assigned to the excitation of single optical magnons did not show a measurable shift with magnetic field. In the far-infrared, no signature of acoustic magnon absorption has been observed in the magneto transmittance. However, at  $83\text{ cm}^{-1}$  the sixth phonon of symmetry  $E_u$  in  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  has been found.*

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The insulating parent compounds of high- $T_c$  superconductors are antiferromagnets characterized by an unusually strong spin-exchange coupling in the  $\text{CuO}_2$  layers. Considerable effort has been taken to investigate the magnetic properties of these materials. While neutron scattering has supplied insight into the magnetic structure of the layers and allowed to determine the in-plane exchange coupling  $J$ , no final consensus has been reached on the important issue of the actual size of the intrabilayer exchange coupling constant  $J_{12}$  in  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  so far. Recently, the observation of a single optical magnon excitation has been claimed by some of us in mid-infrared transmission experiments on  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  at  $1436\text{ cm}^{-1}$  (178 meV), together with two-magnon excitations at  $2795\text{ cm}^{-1}$  (346 meV) and  $3800\text{ cm}^{-1}$  (470 meV).<sup>1</sup> This yields a value for  $J$  of 120 meV and  $J_{12} \approx 0.5J$ , in good agreement with a theoretical estimate of  $J_{12} \approx 56\text{ meV}$ .<sup>2</sup> On the other hand, recent inelastic neutron scattering experiments indicate the presence of an optical

magnon branch in  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  at  $67 \pm 5$  meV for  $x=0.2$  and at  $74 \pm 5$  meV for  $x=0.15$ .<sup>3,4</sup> Moreover, similar looking multi-magnon spectra have been observed in single layer cuprates<sup>5</sup> and have been explained successfully as bimagnon-plus-phonon absorption processes.<sup>6</sup> In order to disentangle the magnetic excitations in the MIR from e.g. higher order phonon excitations, we performed infrared transmission experiments on  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  in high magnetic fields. This also allowed us to search for excitations of acoustic magnons in the FIR. The mid-infrared magneto transmittance of  $\text{Y}_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_6$  has been studied in Ref. 7.

Using a Fourier spectrometer, infrared transmission experiments were performed in an external magnetic field up to 16.5 Tesla at  $T=1\text{K}$  on the same sample of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  as in Ref. 1. All measurements were carried out with  $\vec{k} \parallel c$  and the electric field vector parallel to the  $ab$ -plane, while the external magnetic field was applied either parallel or perpendicular to the  $\text{CuO}_2$  layers.

The solid line in Fig. 1 shows the optical conductivity  $\sigma(\omega) = 2\omega\epsilon_0nk$  between the highest fundamental phonon line and the charge transfer gap.

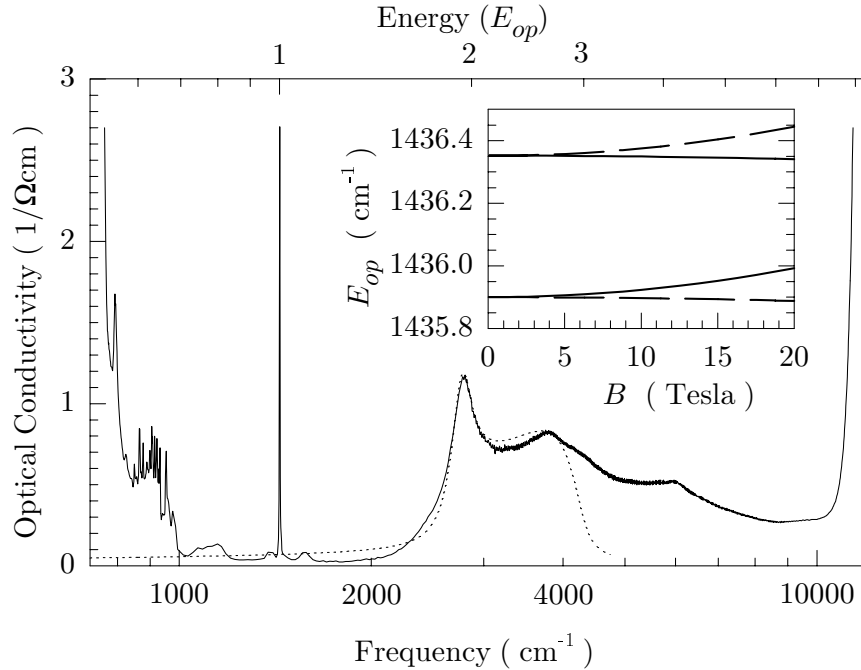


Fig. 1. Solid line: Optical conductivity of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  at  $T=4\text{K}$ . Dotted line: Calculated two-magnon spectrum for  $J = 120$  meV and  $J_{12} \approx 0.5J$ . Inset: Magnetic field dependence of a single optical magnon at  $E_{op} = 1436 \text{ cm}^{-1}$  in the spin-flop phase (Solid lines:  $B \parallel ab$ , dashed lines:  $B \parallel c$ ).

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$\sigma(\omega)$  has been obtained directly from the measured transmittance and reflectance spectra (in the absence of an external field). However, the information about the features in this spectral region is almost entirely contained in the transmittance spectrum because these features are too weak to be observable in reflectance. Peaks below  $1400 \text{ cm}^{-1}$  can be attributed to two-phonon excitations. The sharp line at  $E_{op} = 1436 \text{ cm}^{-1}$  (178 meV) and the two strong peaks at  $2795 \text{ cm}^{-1}$  (346 meV) and at  $3800 \text{ cm}^{-1}$  (470 meV) have been explained as spin-wave excitations by some of us.<sup>1</sup> In this picture, absorption at  $E_{op}$  takes place due to the excitation of a single optical magnon at  $\vec{k}=\mathbf{0}$ , while the two broad peaks both are resonances in the two-magnon spectrum. The single optical magnon energy  $E_{op}$  and the position of e.g. the lower resonance of the two-magnon spectrum are sufficient to determine the in-plane exchange coupling constant  $J \approx 120 \text{ meV}$  and the bilayer coupling  $J_{12} \approx 0.5J$  in linear spin-wave theory. The two-magnon absorption spectrum that we obtain with these values is in good agreement with experiment (dotted line in Fig. 1). Note that the position of the higher resonance of the two-magnon spectrum is described correctly. This picture depends on the verification of the magnon interpretation of the peak at  $E_{op}$ , which relies on the fact that the energy of this peak is too high for a usual two-phonon peak. However, it is possible that this peak can also be explained as a bound state of two interacting phonons.

Measuring the Zeeman splitting in magnetic field should be the ultimate test for a magnon interpretation. On the other hand, the insulating parent compounds are expected to undergo a transition to a spin-flop phase,<sup>8</sup> in which all spins are mainly *perpendicular* to the external field and only canted by a small angle in the direction of the field. In this situation much smaller effects have to be expected. The inset of Fig. 1 shows the shift with magnetic field of an optical magnon at  $1436 \text{ cm}^{-1}$  in the spin-flop phase. Due to the in-plane/out-of-plane anisotropy of the exchange constant  $J$ , the magnon branch is split into two different modes already for zero field. The upper mode contains 70% (30%) of out-of-plane (in-plane) character. The solid lines in the inset of Fig. 1 show the behavior of the modes in an external magnetic field for  $B \parallel ab$ , while the dashed lines are valid for  $B \parallel c$ . The size of the splitting  $\Delta E$  can be obtained from the value of the gapped acoustic mode at  $\vec{k}=\mathbf{0}$ , which is known to be  $E_{ac} = 36 \text{ cm}^{-1}$  (4.5 meV) from neutron scattering.<sup>9</sup> With  $\Delta E = E_{ac}^2/2 * E_{op}$  the splitting turns out to be  $0.45 \text{ cm}^{-1}$ , much too small to be observable due to the linewidth of about  $8 \text{ cm}^{-1}$ .

The dotted line in the lower panel of Fig. 2 shows the original transmission data around  $E_{op}$  at  $T=1\text{K}$ . The curves in the upper panel of the figure show calculated transmittance ratios (zero field/16.5 Tesla) which have been obtained by distributing the measured spectral weight into the two modes

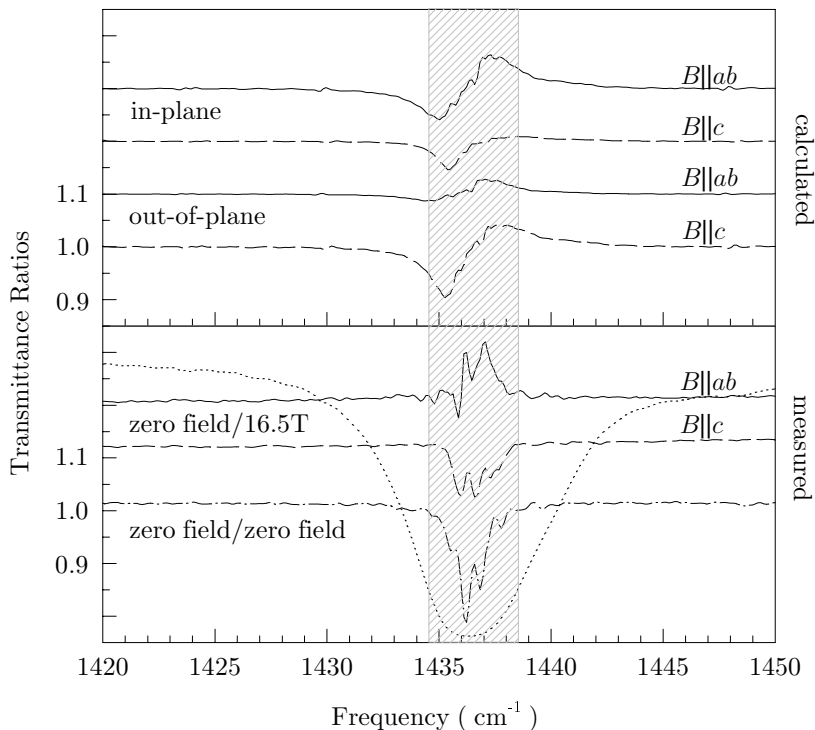


Fig. 2. Upper panel: Calculated transmittance ratios (zero field/16.5 Tesla) in the spin-flop phase for coupling to the in-plane (two upper curves) or out-of-plane (lower curves) mode. Lower panel: Dotted line: Measured transmittance. Other lines: Ratios of measured transmittance. Solid lines:  $B \parallel ab$ , dashed lines:  $B \parallel c$ . The measurements are only reliable *outside* the hatched area, inside they are dominated by noise due to the very low signal (note the ratio of two measurements at zero field).

and shifting them according to the inset of Fig. 1. The predicted changes are very small for any orientation of the external magnetic field. The upper two curves show the case where the infrared light couples only to the in-plane spin-wave excitations, while the lower two curves are valid for a coupling to out-of-plane excitations. The actual measurements are depicted in the lower panel. The lowest (dashed-dotted) curve shows the ratio of two different measurements obtained at zero field. It shows that the ratios are only reliable *outside* the frequency range indicated by the hatched area because the measured transmittance in this range — around the absorption maximum — almost goes to zero. Outside the hatched area, the calculated curves (upper panel) show only small effects. Nevertheless, in the case of coupling to in-plane (out-of-plane) excitations these should be observed at least for  $B \parallel ab$  ( $B \parallel c$ ). However, the ratios of the spectra measured at zero field

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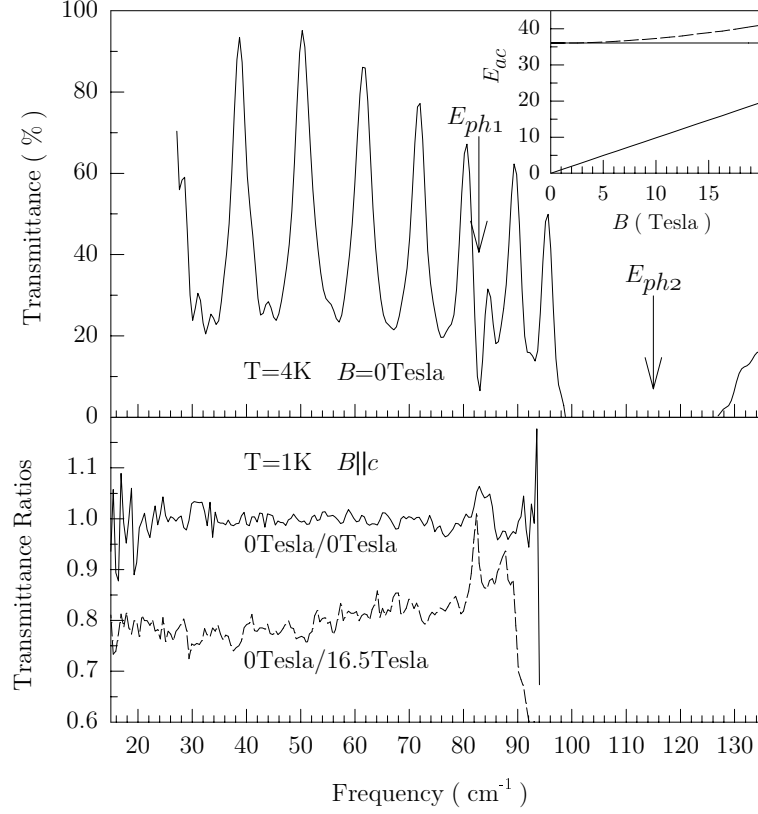


Fig. 3. Upper panel: FIR transmittance at  $T=4\text{K}$ , dominated by interference fringes (sample thickness  $d = 125\mu\text{m}$ ). The arrows indicate phonon absorption lines. Inset: Magnetic field dependence of the acoustic magnon modes in the spin-flop phase (Solid lines:  $B \parallel ab$ , dashed line:  $B \parallel c$ ). Lower panel: Ratios of measured transmittance at  $T=1\text{K}$  for  $B \parallel c$  (Solid line: zero field/zero field, dashed line: zero field/16.5 Tesla).

and at 16.5 Tesla (the two upper curves in the lower panel) do not show any measurable effect of the magnetic field for both orientations of the sample. On the one hand, this makes the magnon interpretation questionable, but on the other hand it is quite probable that the predicted shifts, which are much smaller than the linewidth, will be covered by other effects.

The FIR transmittance measurements are plotted in Fig. 3. The upper panel shows the zero field transmittance which is dominated by interference fringes of the  $125\mu\text{m}$  thick sample. A group theoretical analysis predicts six phonons of symmetry  $E_u$  for  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$ , of which only five have been observed in infrared measurements so far.<sup>10,11</sup> The frequency of the lowest of these observed modes is  $115\text{ cm}^{-1}$  ( $E_{ph2}$  in Fig. 3), and it is due to

absorption of this phonon that the transmittance becomes smaller with increasing frequency in Fig. 3. In our data the sixth phonon of symmetry  $E_u$  has been observed at  $83\text{ cm}^{-1}$  for the first time in infrared measurements. The frequency and the small oscillator strength agree both with theoretical predictions<sup>11</sup> and with neutron scattering results.<sup>12</sup>

From neutron scattering data we know that the gapped acoustic magnon mode at  $\vec{k}=\vec{0}$  can be found at  $E_{ac} = 36\text{ cm}^{-1}$  (4.5 meV).<sup>9</sup> But, the intensity of the infrared signal at such low frequencies is quite small and the spectrum becomes noisy. On the other hand, if there is finite absorption due to acoustic magnons, this should be easily detectable as a change in the magneto transmittance for  $B \parallel c$ , because the gapped acoustic magnon mode is expected to shift about  $3.5\text{ cm}^{-1}$  (for  $B_{max} = 16.5\text{ Tesla}$ , see inset of Fig. 3) even in the spin-flop phase. But the lower panel of Fig. 3 indicates that there is no magnetic field dependence, thus we are probably not coupling to acoustic magnons. (The change in absolute value is due to the field dependence of the detector; and the structure in the transmittance ratios at  $83\text{ cm}^{-1}$  which is already present in the ratio of two different zero field measurements (solid line) can be explained as noise due to the small intensity of the signal at the peak frequency.)

To conclude, we were not able to detect any signature of single magnon absorption neither in the FIR nor in the MIR magneto transmittance of single crystal  $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$  for values of the external magnetic field up to 16.5 Tesla both with  $B \parallel ab$  and  $B \parallel c$ . This puts a severe experimental constraint on the magnon interpretation of the peak at  $1436\text{ cm}^{-1}$ . However, the effect of an external magnetic field is expected to be very small and might easily be covered by other effects, at least in the MIR.

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