Czech. J. Phys. **46-S2**, 1127 (1996).

Probing the Mid-Infrared Spectrum of $YBa_2Cu_3O_{6.0}$ with High Magnetic Fields and Zinc Doping

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The mid-infrared phonon and spin-wave spectrum of antiferromagnetic $YBa_2Cu_3O_{6.0}$ was investigated by infrared transmission measurements ($\vec{k}$ ||c-axis) at T=4K. Peaks at 178 meV, 346 meV and 470 meV were previously interpreted as excitations of single magnons of the optical branch and of bimagnons, respectively. Infrared measurements in high magnetic fields up to 16.5 Tesla and on samples doped with 5% of Zinc have been performed to query this interpretation.

Quite some work in the field of high-T_c superconductivity has been dedicated to the magnetic properties of the insulating parent compounds. While neutron scattering has supplied many answers on e.g. magnetic structure or the exchange coupling constant J in the CuO_2 layers, there is no final agreement about the important issue of the actual size of the intrabilayer exchange coupling constant J_{12} in $YBa_2Cu_3O_{6+x}$ so far. Recently, the observation of a single optical magnon excitation has been claimed by some of us in mid-infrared transmission experiments on $YBa_2Cu_3O_{6.0}$ at 178 meV, together with two-magnon excitations at 346 meV and 470 meV [1]. These have been used to determine J to be 120 meV and $J_{12} \approx 0.5J$, in good agreement with a theoretical estimate of $J_{12} \approx 56$ meV [2]. On the other hand, recent inelastic neutron scattering experiments indicate the presence of an optical magnon branch at 67 meV in $YBa_2Cu_3O_{6.2}$ [3]. Moreover, similar looking multi-magnon spectra have been observed in single layer cuprates [4] and have been explained successfully as bimagnon-plus-phonon absorption processes [5]. In order to disentangle the magnetic excitations from e.g. higher order phonon excitations we performed MIR transmission experiments in an external magnetic field. A second approach is to introduce a non-magnetic impurity by substituting Zn on the Cu(2) sites. A third approach (oxygen isotope substitution) is still in preparation.

Using a Fourier spectrometer, MIR transmission experiments have been performed at T=4K on the same sample of $YBa_2Cu_3O_{6.0}$ as in [1], on a different sample of $YBa_2Cu_3O_{6+x}$ with ultra-low oxygen doping $x \ll 0.1$, and on two samples of $YBa_2(Cu_{0.95}Zn_{0.05})_3O_6$. The same technique has been used for the measurements in an external magnetic field up to 16.5 Tesla at T=1K, again on the same sample of $YBa_2Cu_3O_{6.0}$ as in [1]. All measurements were carried out with $\vec{k} \parallel c$ and the electric field vector parallel to the ab-plane, while different orientations of the sample with the external magnetic field parallel or perpendicular to the CuO_2 layers have been used.

Although the relevant excitations are too weak to be observable in reflectivity, they can be seen in transmission in the IR transparent window between the highest fundamental phonon line and the charge transfer gap. Changes in reflectivity are very small in this frequency range, and as $T \approx (1\text{-}R)^2 \exp(-4\pi kd/\lambda)$, the function $-\ln T/d$ plotted in Fig. 1 represents the optical conductivity $\sigma(\omega) = 2\omega\epsilon_0 nk$. The solid lines show the spectra of the two samples of $YBa_2Cu_3O_{6+x}$ (with $x \approx 0$ and $x \ll 0.1$), while the dotted lines belong to the two Zn-doped samples. The difference between the two solid lines in Fig. 1 is due to an incoherent increase of the optical conductivity in the MIR with oxygen doping. Both show two strong peaks at 2795 cm^{-1} (= 346 meV) and at 3800 cm^{-1} (= 470 meV), which we attributed to direct two-magnon absorption [1]. Upon Zn-substitution (dotted lines), both peaks broaden substantially. While the lower peak is shifted by about 20 cm^{-1} to 2775 cm^{-1} (see inset on the right hand side), the second peak has become so broad

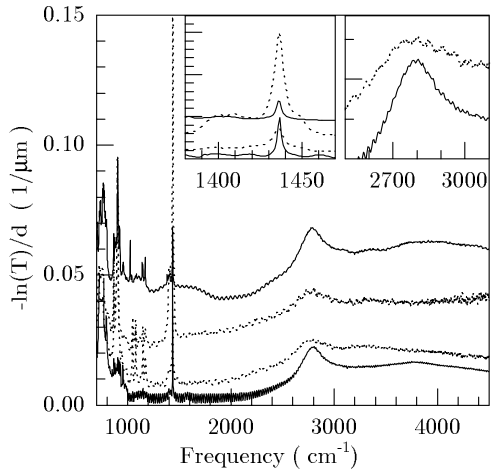

Figure 1: Solid lines: $YBa_2Cu_3O_{6+x}$ (with $x \approx 0$ and $x \ll 0.1$); dotted lines: $YBa_2(Cu_{0.95}Zn_{0.05})_3O_6$; T=4K.

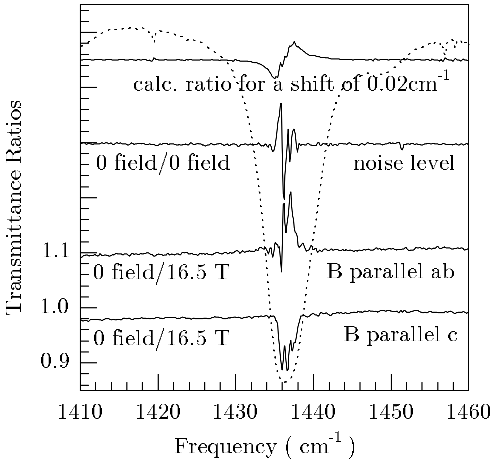

Figure 2: Dotted line: Transmittance of the sharp peak at 1436 cm^{-1} for T=1K. Solid lines: Calculated (topmost) and measured ratios of transmission data for different values of the external magnetic field.

that it is not possible to determine the peak energy. Both, broadening and shift to lower energies, are expected for the magnon absorption after substitution of the non-magnetic Zn-impurities, but this is also true for the bimagnon-plus-phonon process. The differences in the absolute value of $\sigma(\omega)$ might be due to an increase in oscillator strength of the multi-magnon absorption and/or to a finite oxygen doping also in the Zn-doped samples. The inset on the left hand side of Fig. 1 shows the sharp peak at 1436 cm^{-1} (= 178 meV), which has been attributed to the excitation of a single optical magnon at $\vec{k}=0$ by some of us [1]. The energy of this peak is too high for a usual two-phonon peak, but maybe it can be explained as a bound state of two interacting phonons. Similar to the two-magnon absorption peaks, this sharp line is broadening and shifting to lower energies (about 0.5 cm^{-1}) upon Zn-doping.

Measuring the Zeeman splitting in magnetic field should be the ultimate test for a magnon interpretation, but the insulating parent compounds are expected to undergo a transition to a spin-flop phase [6], in which all spins are mainly *perpendicular* to the external field and only canted by a small angle in the direction of the field. We calculated the shift for an optical magnon at 1436 cm^{-1} in this situation and obtained for B=16.5 Tesla ($B \parallel ab$) 0.06 cm^{-1} and 0.01 cm^{-1} for the in-plane and out-of-plane excitations, respectively (and vice versa for $B \parallel c$; in-plane and out-of-plane mode are separated from each other by 0.35 cm^{-1}). As the peak itself has a linewidth of 8 cm^{-1}, it is quite probable that such a small shift will be covered by other effects. The dotted line in Fig. 2 shows the original transmission data at T=1K. The topmost solid line shows a calculated ratio of the dotted line and the same line shifted by 0.02 cm^{-1}. The next line depicts the ratio of two different measurements both at zero field to show the error in the range where transmission is very low. A shift of the order of 0.02 cm^{-1} is thus at the detection limit. Finally, ratios of measurements at zero field and 16.5 Tesla are shown for different orientations of the sample in the two lower curves. As there is hardly any difference to the second curve (0 field/0 field) we can conclude that any shift has to be smaller than 0.02 cm^{-1}. This puts a severe experimental constraint on the magnon interpretation.

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