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c-Axis Excitations in High-T_c Superconductors Detected by Grazing Incidence Reflectivity Measurements

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Measuring absolute reflectivity at an angle of incidence of 80° for $Tl_2Ba_2CuO_6$, $Bi_2Sr_2CuO_6$, $Nd_{1.85}Ce_{0.15}CuO_4$ with p-polarized light (plane of incidence normal to the CuO_2-planes), we were able to extract the c-axis longitudinal optical modes in the normal and superconducting state. Superconducting induced changes in the electronic c-axis properties will be discussed and compared to $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_y$ where a superfluid plasma edge is observed at low frequencies.

1. INTRODUCTION

The c-axis properties of high-T_c superconductors are important for the understanding of the charge dynamics, not only perpendicular to, but also in the CuO_2-planes. Whereas in several double layer compounds an absorption edge in the normal state far-infrared c-axis conductivity is observed and attributed to a spin gap [1] or an inter-band transition [2], the single layer compound $La_{2-x}Sr_xCuO_4$ exhibits a weakly frequency dependent electronic conductivity [3]. In the superconducting state a screened Josephson plasma frequency, $\omega_j/\sqrt{\epsilon_s}$, is found below 100 cm^{-1} for $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_y$. The frequency of this prominent plasma edge is strongly material dependent as is T_c. Of special interest is whether a universal relation exists between ω_j and T_c as suggested by Anderson for the single layer compounds [4]. We studied a series of single layer compounds with c-axis length of 100 μm. Due to this small dimension, conventional reflectivity measurements (normal incidence), are not possible. By p-polarized reflectivity measurements at a grazing angle of incidence we were however able to determine the c-axis longitudinal optical phonon modes and the superfluid plasma frequency.

2. EXPERIMENTAL

Plate-like crystals with typically 2×2 mm in the ab-plane of $Tl_2Ba_2CuO_6$ ($T_c \approx 85$ K), $Bi_2Sr_2CuO_6$ ($T_c \approx 12$ K), and $Nd_{1.85}Ce_{0.15}CuO_4$ ($T_c \approx 23$ K) were mounted on a cone in a cold finger cryostat with the ab-plane perpendicular to the plane of scattering. Using p-polarized light at an angle of incidence of 80° we are sensitive to the c-axis optical properties. The sample was Au-coated using in-situ evaporation to obtain absolute reflectivity.

3. RESULTS AND DISCUSSION

Using Fresnel equations for the reflectivity of a uniaxial crystal we derive the following expression for the c-axis pseudo-loss function in terms of the absorptivity $A_p = 1 - R_p$:

$$\frac{A_p|n_{ab}|\cos(\theta)}{2(2-A_p)} \approx \mathrm{Im}\, e^{i\eta}\sqrt{1-\frac{\sin^2\theta}{\epsilon_c}}$$

where θ is the angle of incidence with the surface normal (c-axis), n_{ab} the complex in-plane refractive index with $|n_{ab}|\cos(\theta) \gg 1$ and $\eta \equiv \pi/2 - \mathrm{Arg}(n_{ab})$. A more detailed description of this function is discussed elsewhere [5]. For a strongly anisotropic material like the cuprates with metallic ab-plane properties, *i.e.* $|\mathrm{Re}(\epsilon_{ab})| \gg 1$ and an almost insulating c-axis the absorptivity, A_p will peak at the longitudinal optical c-axis modes. By entering the superconducting state an additonal zero crossing due to the formation of the superfluid is expected if the normal state carrier contribution is overdamped like in $La_{2-x}Sr_xCuO_4$ [3],

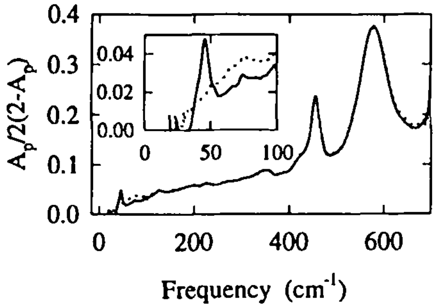

Figure 1: Generalized absorptivity $A_p/2(2-A_p)$ of $La_{1.85}Sr_{0.15}CuO_4$ measured at 80° of incidence with p-polarized light at 6 K (solid line) and at 35 K (dotted). The inset shows on an enlarged scale the appearance of the superfluid plasmon peak at 50 cm^{-1}.

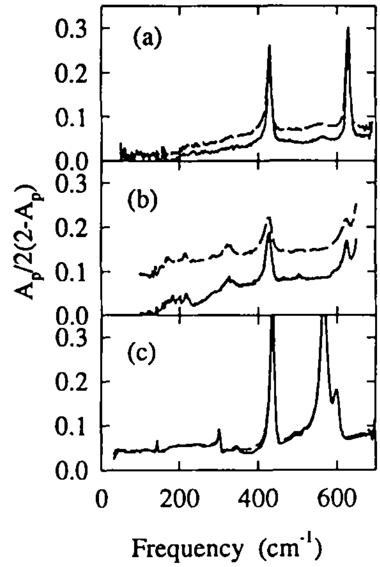

Figure 2: Generalized absorptivity $A_p/2(2-A_p)$ of (a) $Tl_2Ba_2CuO_6$ at 6 K (solid line) and 100 K (dashed), (b) $Bi_2Sr_2CuO_6$ 6 K (solid), 300 K (dashed) and (c) $Nd_{1.85}Ce_{0.15}CuO_4$ 6 K (solid), 30 K (dashed). There is no indication of a superfluid plasmon in the measured frequency ranges.

for which the screened superfluid plasma frequency is found at 50 cm^{-1} by normal incidence reflectivity measurements [3, 6]. As a confirmation of our technique we display in Fig. 1 the results for grazing incidence for $La_{1.85}Sr_{0.15}CuO_4$. Because the in-plane conductivity has a smooth frequency dependence [7], the two strong absorption peaks in the vicinity of 460 and 580 cm^{-1} can be attributed to c-axis longitudinal optical phonon frequencies. Below T_c ($\approx$ 30 K) an additional loss-peak appears which is caused by the superfluid plasmon. In Fig. 2 the results for the $Tl_2Ba_2CuO_6$, $Bi_2Sr_2CuO_6$ and $Nd_{1.85}Ce_{0.15}CuO_4$ single crystals are shown. For $Tl_2Ba_2CuO_6$ three LO modes are found at 157, 427 and 630 cm^{-1}. Below T_c there is no indication of an additional zero crossing in Re(ϵ_c). If the superfluid plasma frequency was located above 700 cm^{-1}, an apparent shift towards higher frequencies of the longitudinal modes, which have now mixed phonon-plasmon character, would be expected. Therefore the screened superfluid plasma frequency is located below 50 cm^{-1} and the corresponding penetration depth $\lambda_c \geq 10\mu m$. From the linewidth of the loss-peaks, which is determined by the intrinsic life-time and the c-axis electronic conductivity σ_c [5], an upper limit of $\sigma_c \approx 1$ S/cm is obtained. This indicates that the strong absorption edge as in the double layer compounds is not present, which might be related to the missing inter-band transition between CuO_2-bilayers. We note that clear evidence for a spin gap has been found in $Tl_2Ba_2CuO_6$ [8]. Similar considerations hold for $Bi_2Sr_2CuO_6$ and $Nd_{1.85}Ce_{0.15}CuO_4$ where the superfluid plasma frequency is not observed above 100 and 30 cm^{-1}, respectively, and also a shift of the longitudinal modes is absent.

4. CONCLUSIONS

Using p-polarized light at a grazing angle of incidence we have shown that the c-axis longitudinal modes can be extracted from extremely thin plate-like crystals. There is no indication of a c-axis superfluid plasma frequency in the measured frequency ranges except for $La_{1.85}Sr_{0.15}CuO_4$.

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