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ELECTRODYNAMICAL PROPERTIES OF HIGH TC SUPERCONDUCTORS STUDIED WITH POLARIZED ANGLE RESOLVED INFRARED SPECTROSCOPY

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Using infrared spectroscopy at grazing angle of incidence we study the electrodynamical properties of high temperature superconductors. We review some of our experiments where transverse polarized light is absorbed by longitudinal optical modes with their mode of oscillation perpendicular to the plane. This is particularly useful for the study of the plasmons and phonons perpendicular to the plane, and allows us to study in detail the c-axis dynamical properties of flux grown single crystals for which usually no samples with large dimensions in the c-direction exist.

1 Introduction and motivation

Recently P. W. Anderson pointed out[1], that for single layer superconductors the following correlation should exist between the bare Josephson plasmon energy (in in units of meV) and T$_c$ (in K) if superconductivity is caused by the Anderson-Chakraverty interlayer-tunneling mechanism:

$$\hbar\omega_J = 2.9\ T_c N(0)^{1/2} d^{1/2} a^{-1}$$

where a (in $\mathring{A}$) is the in-plane lattice parameter, d (in $\mathring{A}$) is the spacing between CuO$_2$ planes, and $N(0)$ (in eV^{-1}) is the density of states at the Fermi energy per unit of CuO$_2$. For all cuprate superconductors $N(0)$ is approximately 1 eV, as follows *e.g.* from specific heat data. Experimentally one observes the plasma resonance at a reduced value $\omega_J/\sqrt{\epsilon_S}$ due to screening. In the cuprates this reduction is a factor 3 to 5 depending on the compound considered. This relation between measurable quantities is a unique feature of this mechanism, and thus provides an experimental test of this theory. Using the above expression we constructed the following table:

Compound	T_c (K)	a $(\AA)$	d $(\AA)$	$\hbar\omega_J$ (meV)	λ_c (μm)
$Bi_2Sr_2CuO_6$	9	3.79	12.2	22	10
$Nd_{2-x}Ce_xCuO_4$	24	3.95	6.035	43	4.5
$La_{2-x}Sr_xCuO_4$	32	3.79	6.64	63	3.1
$Tl_2Ba_2CuO_6$	85	3.86	11.57	216	0.91
$Hg_1Ba_2CuO_5$	98	3.86	9.51	225	0.88

For optimally doped $La_{2-x}Sr_xCuO_4$ the value of the *un*screened Josephson plasmon energy is 25-30 meV [4], which is not too far below the prediction based on the Anderson-Chakravarty model. For the other systems no direct observations of the Josephson-plasmon energy have been reported yet, possibly due to the fact that samples of sufficient thickness for conventional normal incidence reflectivity experiments are not available.

2 The PARIS method

In this paper we discuss the reflection properties at grazing angles of incidence of anisotropic materials. In particular we consider the situation where the light is p-polarized, *i.e.* with the electric field vector parallel to the reflection-plane, and where the dielectric tensor component of the material along the crystal surface is metallic-like (Reϵ is large and negative). In this case the absorptivity A_p displays a series of resonance peaks at frequencies corresponding to the longitudinal optical modes with polarization perpendicular to the sample surface[2]. Using the Fresnel equations

$$\frac{A_p|n_x|\cos\theta}{2(2-A_p)} = \frac{\mathrm{Im}(\alpha_p)}{1+|\frac{\alpha_p}{n_x\cos\theta}|^2} \quad \text{with} \quad \alpha_p(\omega) = e^{i\eta}\sqrt{1-\frac{\sin^2\theta}{\epsilon_z}}$$

we obtain the pseudo-loss function $\mathrm{Im}(\alpha_p)/(1+|\alpha_p/n_x\cos\theta|^2)$ directly from the experimental data, without the need of a Kramers-Kronig analysis. In this expression θ is the angle of incidende with the surface normal (the z-direction), $\epsilon_i = n_i^2$ is the dielectric tensor component along x_i, and $\eta \equiv \pi/2 - \mathrm{Arg}(n_x)$. If $|\epsilon_z| \gg \sin^2\theta$, this is the loss function $\mathrm{Im}(-e^{i\eta}/\epsilon_z)$ with a Fano-type phase factor. The limiting behaviour $\eta = 0$ is reached for a superconductor or for a metal with $\omega\tau \gg 1$. In the low frequency limit of a metal $\eta = \pi/4$. For a metal with θ sufficiently far below the critical angle we have $|\alpha_p| \ll |n_x|\cos\theta$, so that the pseudo-loss function becomes $\mathrm{Im}(\alpha_p)$. Note that for grazing angles of incidence A_p is enhanced with a factor $1/\cos\theta$. We recently took advantage of this fact to study the in-plane conductivity of $La_{2-x}Sr_xCuO_4$ below T_c in detail.[3]

3 The pseudo-loss function of $\mathbf{Tl_2Ba_2CuO_6}$

Let us now consider $Tl_2Ba_2CuO_6$ (T_c = 85 K). To simulate the pseudo-loss function at a grazing angle of incidence we use the following parameters for the electronic c-axis dielectric function: $\epsilon_\infty = 4, \hbar\omega_{pc} = 200$ (meV), $\rho_{DC} =$ 1 (Ωcm), and transverse (longitudinal) optical phonons at 16.0 (17.7), 51.2 (55.9) and 74.6 (80.3) meV with $\hbar/\tau = 0.6 meV$. The result of this simulation is displayed in Fig. 1. In the normal state the electronic contribution is overdamped and does not give rise to an additional zero-crossing of ϵ'. If we model the superconducting state with BCS theory, as is shown in the middle curve of Fig. 1, a Josephson plasmon appears at 6 meV due to transfer of spectral weight of $\sigma(\omega)$ in the gap-region to the δ-function at $\omega = 0$. The effect of the appearance of an unscreened Josephson plasma energy of 200 meV in the superconducting state is shown in the lower curve: All longitudinal modes are now of mixed phonon/plasmon character, and the longitudinal phonons are pushed to a higher frequency compared to the normal state.

In Fig. 2 the experimentally measured pseudo-loss function is displayed above and below the phase transition. The half-width of the loss-peaks is $1/\tau_{ph} + 4\pi\sigma_e\epsilon_\infty^{-1} S_{ph}/(\epsilon_\infty + S_{ph})$, where τ_{ph} is the intrinsic phonon life-time, S_{ph} the oscillator strength, and σ_e the electronic optical conductivity. Hence the line-width of the longitudinal phonons can be used to obtain an upper-limit of the electronic contribution to the optical conductivity in the c-direction. From the experimental data we obtain an upper-limit for σ_c of 1 S/cm near the two dominant longitudinal phonon-peaks. From comparison with Fig. 1 we conclude that the shift of longitudinal frequencies below T_c, as well as the occurance of an extra peak at 48 meV, are both absent in the experiment. This implies that the unscreened Josephson plasmon energy is below 20 meV, or $\lambda_c > 10\mu m$ in this material.

4 Conclusions

The method of polarized angle dependent infrared spectroscopy was used to measure the c-axis infrared properties of thin single-crystalline platelets. The Josephson plasmon energies were calculated for a number of single layer compounds adopting an expression suggested by Anderson, and compared to experimental values for $La_{2-x}Sr_xCuO_4$ and $Tl_2Ba_2CuO_6$. The agreement with $La_{2-x}Sr_xCuO_4$ is within 50 percent. For $Tl_2Ba_2CuO_6$ no shift in longitudinal phonon frequencies was observed below T_c. This may be taken as an indication, that, as is also the case in conventional BCS theory, the electronic part of the dielectric function remains almost unaffected on an energy scale larger than $3.5 k_B T_c$.

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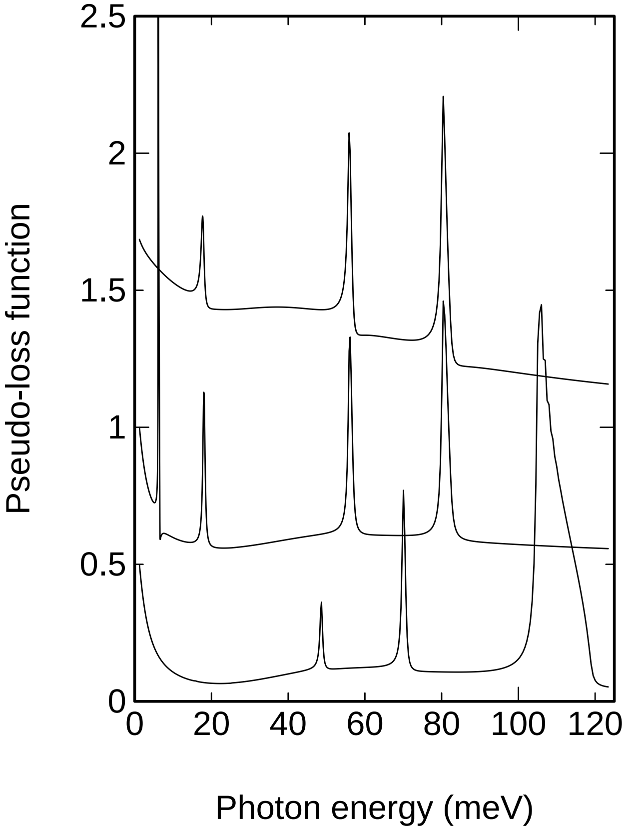

Figure 1: Simulation of the pseudo loss-function for $Tl_2Ba_2CuO_6$. Top curve: 300 K. Middle curve: 4 K using BCS theory. Lower curve: 4K, with $\omega_J = 200$ meV.

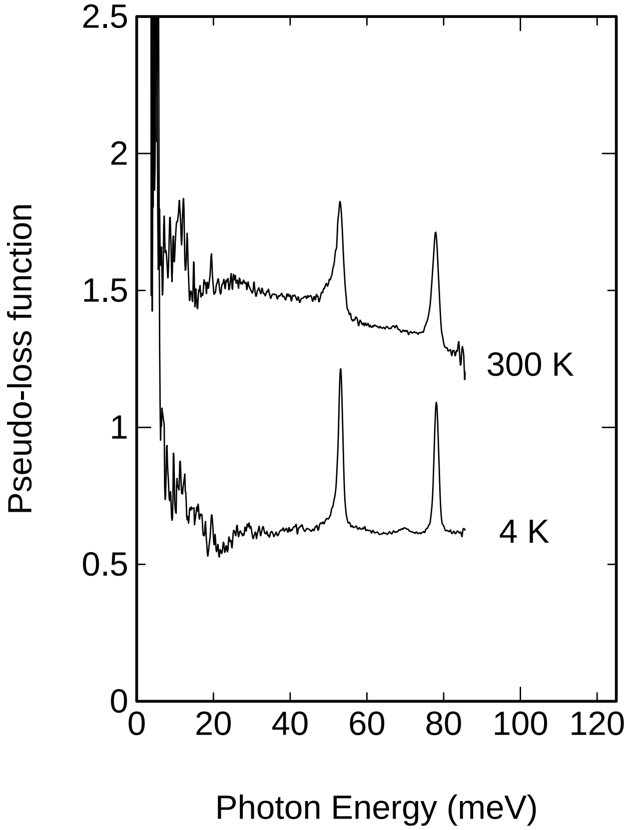

Figure 2: Experimental plot of the pseudo-loss function from ab-plane surface of $Tl_2Ba_2CuO_6$ (T_c=85 K) using p-polarized light with $\theta = 80^o$.