FREQUENCY SHIFTS AND LINEWIDTH CHANGES OF INFRARED-ACTIVE PHONONS IN DOUBLE-LAYERED HIGH-TEMPERATURE SUPERCONDUCTORS

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(Received )

We calculate frequency shifts and changes in linewidths of infrared-active phonons within a shell model for the bare phononic system coupled to an electronic double-layer structure with inter-layer charge transfer. The theoretical concept is applied to YBa$_2$Cu$_3$O$_7$ yielding a good description of experimental results in the normal state as well as at the transition to the superconducting state.
1 Introduction

Raman- [1, 2] and infrared-experiments [3] – [8] show a strong influence of the superconducting transition on phonon frequencies and linewidths in YBa$_2$Cu$_3$O$_7$ and similar compounds. The experimental results on Raman-active phonons are explained by the theory of Zeyher and Zwicknagl [9, 10]. Complementary to their work the even greater variety of effects found on infrared-active phonons, e. g. narrowing of phonon-lines [7] and softening of high-frequency phonons [3] – [8] in connection with the transition into the superconducting state, has been explained qualitatively [11, 13] within a double-layer model for high-temperature superconductors.

In this paper we link the approaches of refs. [12], where a microscopically based model for a Coulomb-type electron-phonon interaction in layer systems was developed, with the calculation of the electronic susceptibility for the double-layer model from [13]. This results in a microscopically founded description of phonon line width changes and frequency shifts which fits to the experimental data in the normal state as well as in the superconducting state.

2 Infrared absorption in coupled phonon-layer systems

We are going to investigate the infrared absorption of an essentially ionic system coupled to conduction electrons which are confined to two-dimensional planes within a model presented in [12, 13]: Bare phonons are described by a shell-model of lattice dynamics, bare electrons are allowed to move almost freely within the conducting CuO$_2$-planes; we assume hopping in c-direction between different planes within one unit cell but neglect transport between different double-layers for simplicity. We consider electron-phonon coupling due to the Coulomb-interaction between vibrating ions and conduction electrons.

In order to determine the response function for IR absorption, the dielectric function, of a coupled electron-phonon system it is not sufficient to calculate a renormalized phononic Green’s function and, independently, an — also renormalized — electronic susceptibility. Electron-phonon coupling yields “off-diagonal” terms in
the dielectric function which may change lineshapes of absorption lines. This can be described by a generalized susceptibility of the coupled electron-phonon system depending on frequency $\omega$ and wavevector $q$. Its inverse is given by

$$
\chi_{\text{gen}}^{-1}(q, \omega) = \left( \begin{array}{cc} G_{\text{ph}}^{-1} & \gamma^+ \\ \gamma & \chi^{-1} \end{array} \right) .
$$

(1)

Here, $G_{\text{ph}}$ is the bare phonon Green’s function which is determined by

$$
[1(\omega + i\delta)^2 - D_0] M^{1/2} G_{\text{ph}} M^{1/2} = 1
$$

(2)

with the dynamical Matrix $D_0(q)$ and diagonal matrices $M$ containing the vibrating masses. For reasons of simplicity we do not make any formal distinction between cores and shells in the lattice dynamical shell-model used for the description of the bare phonon system. Instead, we use shell masses about $10^6$ times smaller than core masses. Thus, for a system of $N$ ions $D_0$ is a $(6N \times 6N)$ matrix whose $3N$ lowest eigenvalues describe the dispersion of the bare vibrational system.

$\chi(q, \omega)$ is the susceptibility of the bare electronic system. In general this is a $(n \times n)$ matrix where $n$ is the number of conducting layers per unit cell. In the case of a double-layer system and $q$ parallel to the planes this matrix can be transformed into diagonal form with elements describing intra- and interband excitations, respectively [13].

Finally, $\gamma(q)$ in (1) is a $(n \times 6N)$ matrix containing the electron-phonon matrix elements. We calculate these matrix elements according to our ionic approach described in detail in [12] (with $X_{\mu \kappa}$ from [12] and ionic charge numbers $Z_{\kappa}$ we have $\gamma_{\mu \kappa} = Z_{\kappa} X_{\mu \kappa}$ for $\mu = 1, 2, \kappa = 1, \ldots , 6N$).

In order to calculate the infrared absorption we still have to multiply the matrix elements of $\chi_{\text{gen}}$ by the coupling constants to the external electric field. For the case of infrared active phonons with eigenvectors perpendicular to the $a$-$b$-plane and charge fluctuations between the double layers the dielectric function reads

$$
\Delta \varepsilon(\omega) = 4\pi A^+ \chi_{\text{gen}}(q \to 0, \omega) A
$$

(3)

with

$$
A_{\kappa} = e Z_{\kappa} \quad \text{f"ur} \quad \kappa = 3\nu; \nu = 1, \ldots 2N
$$

(4)
\[ A_{6N+1} = \pm 0.5ed \]  
\[ A_{6N+2} = \mp 0.5ed. \]

All other components of the external field coupling vectors \( A \) are zero. The electrical field couples to the polarization in the crystal. For lattice vibrations this polarization is given by the product of ionic charge (or shell charge) and the corresponding displacement. The latter is contained in \( \chi_{\text{gen}} \). For the electronic excitations the polarization is determined by the distance \( d \) between the two planes connected by the electronic hopping process and the charge transfered between these layers which depends on the electronic susceptibility. The signs and distances to be used in (5) and (6) depend on the way the inter-layer hopping process is taking place physically.

3 Infrared absorption of \( \text{YBa}_2\text{Cu}_3\text{O}_7 \)

In the following we apply the theoretical concepts described above to \( \text{YBa}_2\text{Cu}_3\text{O}_7 \) using a lattice-dynamical shell model with parameters from [14]. As we are mainly interested in general trends we refrain from a "fine-tuning" of the original shell-model parameters.

The electronic double-layer system is modelled by two bands crossing the Fermi surface [13]. For simplicity the bands are assumed to have cylindrical symmetry but an energy distance varying from zero to about 1000 cm\(^{-1}\) in the vicinity of the Fermi surface. The mean density of states is 0.85 eV\(^{-1}\) for each band. In the superconducting state we use a BCS-type approximation with equal gaps \( 2\Delta = 320 \text{ cm}^{-1} \) in both bands.

The bare electronic susceptibility is calculated numerically as described in [13] taking into account vertex corrections and screening effects in a random-phase approximation. We assume that the electronic hopping process takes place via the copper-oxygen chains, while the yttrium layer is considered to be completely insulating. Thus, the distance \( d \) to be used in (5) and (6) is about 8.3 rA.

The electron-phonon coupling at large wavelengths is completely determined by the Coulomb interaction between ionic and electronic charges without additional parameters as described in [12].

Fig. 1a shows the result of the calculation for the normal state. The dashed line represents the shell model
calculation (with an artificial broadening of the phonon lines), the solid line represents the results for the coupled system including the electron-phonon interaction. Three prominent effects are to be observed: softening of phonon frequencies, broadening of absorption lines and strong changes of oscillator strengths for different modes.

Most pronounced are these effects for the mode at 365 cm\(^{-1}\) in the uncoupled system, where the yttrium-layer moves against the conducting CuO\(_2\)-planes [14]. The frequency of this mode is lowered by about 85 cm\(^{-1}\) due to the electron-phonon coupling, the linewidth increases as well as the oscillator strength. Actually, such a frequency shift for this mode is observed experimentally going from YBa\(_2\)Cu\(_3\)O\(_6\) to YBa\(_2\)Cu\(_3\)O\(_7\) [15]. Similar effects are found also for the modes at 154 cm\(^{-1}\) and 508 cm\(^{-1}\) (in the bare model) which both become softer by 13 cm\(^{-1}\).

These softenings result from the fact that ionic displacements perpendicular to the conducting double layer induce a charge transfer between the planes of this double layer. This charge transfer is accompanied by a polarization field [12] which reduces the restoring forces onto the ion, the frequency of the vibration becomes lower. The line broadening is caused by the damping of lattice vibrations due to interband particle-hole excitations in the electronic system.

Somewhat less evident is the situation concerning the changes in the oscillator strength. Here, one has to distinguish two cases: firstly, vibrations of ions which are located between the two planes connected by the electronic hopping process, in our case this would be e. g. the barium ions; here the direction of the electronic polarization due to the hopping process is antiparallel to the polarization resulting from ionic displacements, therefore the oscillator strength of the mode (around 150 cm\(^{-1}\)) decreases. If on the other hand e. g. yttrium ions move perpendicular to the planes, the induced electronic polarization is in the same direction as the phononic polarization, the oscillator strength therefore increases. Experiments also show large oscillator strengths for barium modes [4] in contradiction to our results. This may be due to additional charge transfer processes, e. g. in the CuO-chains, which are not contained in our model.

Our numerical results for the oscillator strength depend to a large extent on the details of the electronic band
structure, while the results for the frequency shifts are more independent of such details. Our calculations for the dielectric function also yield a direct contribution from collective electronic charge fluctuations between the layers. It appears at much higher frequencies and therefore is not shown in Fig.1.

What happens at the transition to the superconducting state? The infrared absorption spectrum in the superconducting state is shown in Fig. 1b. In Figs. 2a and b we show the relative changes in frequency and linewidth due to this transition calculated within the framework of our model. The small symbols refer to different experimental results, the circles are the results of our numerical calculations and fit well the experimental frequency shifts. All phonons — even those at high frequencies — become softer in the superconducting state, phonon lines below 500 cm$^{-1}$ become narrower due to the loss of decay channels resulting from the gap formation in the superconducting state. Above 500 cm$^{-1}$ damping increases and phonon lines may become broader in the superconducting state. The strongest effects are observed for the modes around 300 cm$^{-1}$.

We have also calculated phonon spectra at finite wave vectors parallel to the planes. Here we find that the relative frequency shifts and line width changes at the transition to the superconducting state become much smaller. This is confirmed by recent neutron scattering experiments [17].

4 Summary

In this work we have presented calculations for the infrared absorption of YBa$_2$Cu$_3$O$_7$ using a microscopically based double-layer model for high-temperature superconductors.

The approximations used to describe the conduction electron system are rather crude in view of strong-correlation effects. Nevertheless, they allow for a treatment of realistic systems and we regard the results to be qualitatively correct in the long wavelength limit.

The electron-phonon coupling was described without any additional parameters by the Coulomb interaction between the ions inside and outside the CuO$_2$-planes and the mobile electrons. Again, we believe that this type of interaction will dominate the interband electron-phonon
coupling in the limit of small wavevectors.
Our model calculations explain experimental results on
frequency shifts and line width changes of infrared-active
c-axis phonons. In particular the softening of modes at
the transition to the superconducting state is well de-
scribed by our model. Concerning the absolute position
of the absorption peaks the agreement with experi-
tmental results could be improved if we would adjust the
parameters of the original shell-model. We did not do
this, because we think it more important to first improve
the approximations for the electronic system. In some
experiments it seems, that the softening of phonon
frequencies sets in already at temperatures above the su-
perconducting transition temperature. At the moment
it is not clear wether this effect can be explained by su-
perconducting fluctuations or by some other mechanism
like opening of a spin gap.

Acknowledgement — This work was supported by the
Deutsche Forschungsgemeinschaft.
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**Figure Captions**

**Fig. 1.** Calculated infrared absorption for YBa$_2$Cu$_3$O$_7$ without electron-phonon interaction (dashed line) and with electron-phonon interaction (solid line); a) results in the normal state ($T=100$K) and b) in the superconducting state $T=1$K.

**Fig. 2.** Frequency shifts (a) and line width changes (b) of infrared-active c-axis phonons in $RBa_2Cu_3O_7$ ($R =$ rare earth). The small symbols are experimental results from refs. [3] – [8]. The circles mark the frequency shifts and line width changes calculated in this work.