J. Phys.: Condens. Matter 4 (1992) 9643–9650. Printed in the UK

Plasmon–phonon coupling in $YBa_2Cu_3O_7$

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Received 28 January 1992, in final form 1 May 1992

Abstract. The $E \| c$ far-infrared reflection spectra of a $YBa_2Cu_3O_7$ single crystal were fitted for three different temperatures (10 K, 60 K and 100 K) using phononic oscillators, a mid-infrared excitation and a low-energy plasmon. The plasmon frequency changes from 600 cm^{-1} in the normal state to 365 cm^{-1} in the superconducting state. Some consequences of strong plasmon–phonon interaction for the pairing mechanism are discussed.

1. Introduction

Several models of the mechanism of superconductivity in $YBa_2Cu_3O_7$ have been developed which invoke the phononic properties of this material. Such models are supported by the growing experimental evidence that lattice effects are important in the mechanism of high-T_c superconductivity (see e.g. [1]).

The direct experimental evidence from the observation of the temperature evolution of the frequencies of optically active phonons [2–9] is that softening occurs for some modes with a relative decrease of 2% in the real part of the phonon self-energy in the superconducting state compared with the normal state. Most modes seem to follow the predictions of strong coupling theory [10] although high-frequency phonons soften in contrast to the predicted hardening [7–9]. These phonon anomalies observed in $YBa_2Cu_3O_7$ with a high T_c are substantially larger than those in $La_{1.85}Sr_{0.15}CuO_4$ with a lower T_c [11–12]. On purely thermodynamic grounds the phonon softening in $YBa_2Cu_3O_7$ leads to a decrease in the Gibbs energy (i.e. stabilization) of the superconducting phase and hence increases T_c . Although the observed phonon renormalization contributes to an increase in the transition temperature, the effect is presumably not the primary cause of the superconductivity.

While it is generally agreed that the two-dimensional CuO_2 layer is crucial for the high T_c, it seems that some kind of interlayer coupling is necessary both to provide an additional mechanism for the T_c enhancement [13, 14] and to stabilize the long-range order by raising the dimensionality of the system to $(2+\epsilon)$. The dimensionality of the system is critical because fluctuations destroy the long-range order in purely 1D and 2D systems [15, 16]. The marginal dimension is 2 so long-range superconducting order can be established in quasi-two-dimensional (i.e. $(2+\epsilon)$-dimensional) structures [17]. Recent experiments on $YBa_2Cu_3O_7$–$PrBa_2Cu_3O_7$ superlattices [18, 19] seem to confirm this assumption: superconductivity is observed in a single CuO_2 layer with a T_c of 10 K, but interlayer coupling along the c-axis is required to achieve a T_c

0953-8984/92/489643+08$07.50

of 90 K. Evidence from infrared and Raman measurements such as the anomalous oscillator strength of the 155 cm^{-1} mode in the infrared spectrum [2, 20, 21] and the Fano profile of the 115 cm^{-1} Raman line [22], indicate the existence of strong interactions between c-axis phonons and electronic degrees of freedom.

In the model for superconductivity in layered structures, proposed in 1964 [23], the coupling between layers was mediated by excitons. In this paper we suggest that the interlayer coupling in the high T_c compounds may be mediated by a low-lying c-axis plasmon. We draw attention to the possibility of resonance coupling between this plasmon excitation and the 310 cm^{-1} (TO) phonon which might be strong enough to contribute directly to the pairing mechanism.

2. Experimental evidence

The experimental evidence for plasmon–phonon excitations is based on the $E \| c$ infrared reflection spectra of $YBa_2Cu_3O_7$ single crystals [24, 25]. The LO phonon damping of the 310 cm^{-1} mode changes with temperature and the minimum near 110 cm^{-1} sharpens and decreases when the sample is cooled into the superconducting phase. We find that these effects, which are illustrated in figures 1–3, are fully consistent with LO phonon–plasmon coupling [26, 27]. The plasma minimum is not restricted to $YBa_2Cu_3O_7$ as a similar feature exists in the reflection spectrum of $La_{2-x}Sr_xCuO_4$ (x=0.175) [11, 28].

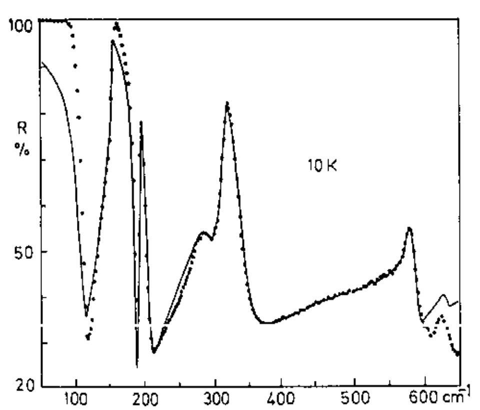

Figure 1. $E \| c$ far-infrared reflection spectrum of a $YBa_2Cu_3O_7$ single crystal at 10 K. The experimental data are taken from [24], while the full line represents the model fit (1).

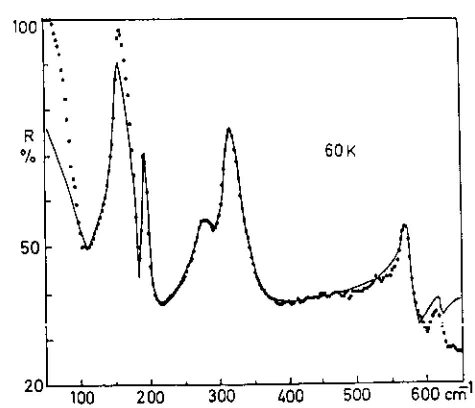

Figure 2. As figure 1, but at 60 K.

Attempts to analyse the spectra of $YBa_2Cu_3O_7$ in terms of the three-parameter phonon model without a plasmon–phonon coupling failed to reproduce the experimental findings. Instead, we now analyse the data from [24] using the four-parameter phonon model with a low-lying plasmon and the usual mid-infrared term [29, 30]:

$$\varepsilon(\omega) = \varepsilon(\infty) \left[\prod_{j=1}^{6} \frac{\omega_{j\mathrm{LO}}^2 - \omega^2 - \mathrm{i}\omega\Gamma_{jLO}}{\omega_{jTO}^2 - \omega^2 - \mathrm{i}\omega\Gamma_{j\mathrm{TO}}} - \frac{\omega_\mathrm{p}^2}{\omega(\omega + \mathrm{i}\Gamma_\mathrm{p})} \right] + \frac{S_\mathrm{e}\omega_\mathrm{e}^2}{\omega_\mathrm{e}^2 - \omega^2 - \mathrm{i}\omega\Gamma_\mathrm{e}} \qquad (1)$$

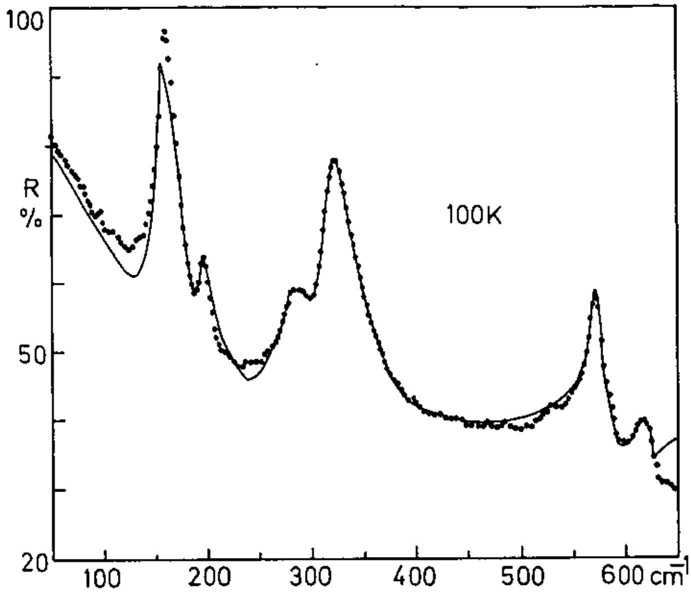

Figure 3. As figure 1, but at 100 K.

The four-parameter model [31] for the phonons is necessary because there is large LO–TO splitting similar to that observed in other partly ionic oxides [32, 33] and because there is plasmon–phonon coupling, as is the case in certain degenerate semiconductors [34, 35]. In our case, the splitting of the 360 cm^{-1} ($E \| c$) band in $YBa_2Cu_3O_6$, which corresponds to the 310 cm^{-1} phonon in the orthorhombic phase, is greater than 100 cm^{-1} [36]. A large splitting was also predicted theoretically from lattice dynamics calculations [37–39]. The fitted spectra are shown together with the observed data in figures 1-3 and the parameters are listed in table 1. The agreement between the theoretical and experimental spectra is good in the spectral range from 100 cm^{-1} to 600 cm^{-1} . Discrepancies occur at low frequencies for $T < T_c$ because no attempt was made to model the reflectivity related to the superconducting gap. Also it is worth mentioning the anomalous oscillator strength of the 155 cm^{-1} mode [2], and the rise in oscillator strength of the 195 cm^{-1} phonon with lowering temperature. At the high-energy end of the spectrum, the data could be influenced by experimental uncertainties (end of the spectral range of the beamsplitter, sample misalignment etc) and we did not try to achieve better agreement between the model and the observation. The relevant part of the spectrum is the plasma minimum around 120 cm^{-1} , the region around the 310 cm^{-1} profile and the flat part of the spectrum between 400 cm^{-1} and 550 cm^{-1} which are directly related to a low-lying plasmon.

The change in the reflection spectrum during the phase transition is well reproduced by the change in the plasmon frequency which drops from 600 cm^{-1} in the normal state to 365 cm^{-1} in the superconducting state with an approximately linear drop of the plasma damping Γ_p with temperature (table 1).

We now simplify the dispersion equation in order to estimate the frequencies of the coupled plasmon–phonon excitations. As the damping shows no anomalous increase and the coupling between the different phonon branches is not essential for our argument, we ignore the damping parameters and concentrate on a one-phonon term. Taking into account that $\omega \ll \omega_e$ the dielectric function is then:

Table 1. The parameters used in our fit.

Parameters	10 K	60 K	100 K
$\omega_{\mathrm{TO}}(1)$	153	153	155
$\omega_{\mathrm{LO}}(1)$	184	186	186
$\Gamma_{\mathrm{TO}}(1)$	1.5	3	3
$\Gamma_{\mathrm{LO}}(1)$	1.5	6.5	5
$\omega_{\mathrm{TO}}(2)$	193	193	195
$\omega_{\mathrm{LO}}(2)$	212	212	212
$\Gamma_{\mathrm{TO}}(2)$	2	3	12
$\Gamma_{\mathrm{LO}}(2)$	12	18	18
$\omega_{\mathrm{TO}}(3)$	285	285	287
$\omega_{\mathrm{LO}}(3)$	296	297	303
$\Gamma_{\mathrm{TO}}(3)$	30	28	32
$\Gamma_{\mathrm{LO}}(3)$	23	23	29
$\omega_{\mathrm{TO}}(4)$	313	313	319
$\omega_{\mathrm{LO}}(4)$	437	437	453
$\Gamma_{\mathrm{TO}}(4)$	6	9	9.5
$\Gamma_{\mathrm{LO}}(4)$	120	200	200
$\omega_{\mathrm{TO}}(5)$	573	575	576
$\omega_{\mathrm{LO}}(5)$	613	613	613
$\Gamma_{\mathrm{TO}}(5)$	14	14.5	12
$\Gamma_{\mathrm{LO}}(5)$	18	25	45
$\omega_{\mathrm{TO}}(6)$	622	622	627
$\omega_{\mathrm{LO}}(6)$	635	635	639
$\Gamma_{\mathrm{TO}}(6)$	8	8	11
$\Gamma_{\mathrm{LO}}(6)$	30	30	33
ω_p	365	365	600
Γ_p	30	100	230
ω_e	3000	3000	3000
Γ_e	2500	2500	2500
S_e	19	18	18.5
ε_∞		4.8	

$$\varepsilon(\omega) = \varepsilon(\infty) + \left[\left(\varepsilon(0) - \varepsilon(\infty)\right) / \left(\omega_{\mathrm{TO}}^2 - \omega^2\right)\right]\omega_{\mathrm{TO}}^2 - \varepsilon(\infty)\omega_{\mathrm{p}}^2/\omega^2 + S_{\mathrm{e}} \tag{2}$$

where $\varepsilon(0)$ and $\varepsilon(\infty)$ are the dielectric constants without the electronic terms. A simple rearrangement along with the parameters for 10 K gives:

$$\varepsilon(\omega) = \tilde{\varepsilon}(\infty) + \left[\left(\tilde{\varepsilon}(0) - \tilde{\varepsilon}(\infty)\right) / \left(\omega_{\mathrm{TO}}^2 - \omega^2\right)\right]\omega_{TO}^2 - \tilde{\varepsilon}(\infty)\tilde{\omega}_{\mathrm{p}}^2/\omega^2 \tag{3}$$

where

$$\tilde{\varepsilon}(\infty) = \varepsilon(\infty) + S_{\mathrm{e}} \qquad \tilde{\varepsilon}(0) = \varepsilon(0) + S_{\mathrm{e}} \qquad \tilde{\omega}_{\mathrm{p}} = \omega_{\mathrm{p}}\sqrt{\varepsilon(\infty)/\tilde{\varepsilon}(\infty)} \simeq 164\ \mathrm{cm}^{-1}.$$

Using the Lyddane–Sachs–Teller relation, we find $\varepsilon(0)$ and $\tilde{\varepsilon}(0)$ and hence the renormalized frequency of the LO phonon:

$$\tilde{\omega}_{LO} = \omega_{TO}\sqrt{\tilde{\varepsilon}(0)/\tilde{\varepsilon}(\infty)} \simeq 342\ \mathrm{cm}^{-1}. \tag{4}$$

The mid-infrared term has reduced ω_{LO} from 437 cm^{-1} to 342 cm^{-1} . The coupled plasmon–phonon modes are defined as the zeros of the dielectric function (3) given by

$$2\omega_{\pm}^2 = \tilde{\omega}_{LO}^2 + \tilde{\omega}_{\mathrm{p}}^2 \pm \sqrt{(\tilde{\omega}_{LO}^2 + \tilde{\omega}_{\mathrm{p}}^2)^2 - 4\tilde{\omega}_{\mathrm{p}}^2\omega_{TO}^2}. \tag{5}$$

The estimated numerical values are $\omega_- \cong 147$ cm^{-1} and $\omega_+ \cong 349$ cm^{-1} . When the full dielectric function (1) is taken into account, these values change slightly to 110 cm^{-1} and 345 cm^{-1} . Our simplified analysis showed that the coupled plasmon–phonon frequency ω_+ at 345 cm^{-1} is close to the renormalized value of the LO mode (a plasma excitation is absent) in the superconducting state. In the normal state, the coupled mode occurs at $\omega_+ = 379$ cm^{-1} while the $\tilde{\omega}_{LO}$ does not change significantly.

3. Discussion

Our analysis points towards the existence of a low-energy plasmonic excitation. Additional support for the idea that a low-energy electronic excitation exists in $YBa_2Cu_3O_7$ is supplied by the following experimental results. Firstly, Raman spectra of the electronic background of $YBa_2Cu_3O_7$ in the superconducting state with A_{1g} symmetry [40, 41] showed an increase in background scattering near 340 cm^{-1} which grows with decreasing temperature. Secondly, the plasmon–phonon coupled mode is in agreement with results from HREELS [42] where a broad peak appeared around 500 cm^{-1} in the normal state.

We find that this low-energy plasmonic excitation is coupled to a LO phonon. Such an effect was invoked as a possible mechanism to increase the critical temperature in superconducting semiconductors [43–45]. In this case, the effective potential of the carrier interaction via the plasmon–phonon modes can be written as:

$$V_{\text{eff}}(q,\omega) = 4\pi e^2/q^2\varepsilon(q,\omega). \tag{6}$$

We now approximate ε by our dielectric function (3) and obtain [43]:

$$V_{\text{eff}}(q,\omega) = -(4\pi e^2/\varepsilon(\infty)q^2)\left[\omega^2(\omega_{\text{TO}}^2 - \omega^2)/\left[(\omega^2 - \omega_+^2)(\omega^2 - \omega_-^2)\right]\right]. \tag{7}$$

This potential is negative for $\omega < \omega_-$ or $\omega_{\text{TO}} < \omega < \omega_+$ and poles exist for $\omega = \omega_\pm$. If the pole ω_+ is close to the LO phonon, the kernel in the Linhard approximation based on the above potential becomes strongly attractive [43], which can substantially raise T_c .

Electron–LO phonon coupling has already been discussed as a possible pairing mechanism for high T_c superconductivity [46, 47]. It was found that the LO phonons strongly couple to charge fluctuations generating Cu–O charge-transfer oscillations with frequencies close to those of the LO phonons [46]. This causes the effective dielectric function to be negative for some wavenumbers and accordingly T_c is enhanced. The calculation also showed that the isotope effect must be weakened as T_c rises. As the c-axis phonons are not strongly screened, the interaction between electrons at low wavenumbers is via the coupled plasmon–phonon modes, whereas for large wavenumbers the coupling is possibly also via the LA phonons as in classical superconductors. We find an interesting analogue between the behaviour of optical modes in high T_c superconductors and that of the acoustic modes in classical superconductors concerning both their softening and mediation [48].

It is interesting to note that the resonance depends sensitively on the chemical composition and the carrier concentration via the dependence of both the LO frequency and the plasma frequency. Quantitative observations of the variation of the critical temperature with carrier concentration reported for $SrTiO_3$ [49] bear close resemblance to the behaviour of high-temperature oxide superconductors.

The role of the mid-infrared excitations is to reduce the LO–TO splitting of the 310 cm^{-1} phonon band which indicates a reduction of the electron–polar-phonon coupling. In a picture based on the condensation of bipolarons in the superconducting phase, the effect of the smaller LO–TO splitting is to reduce the effective mass of the bipolarons facilitating the condensation (see e.g. [50–52]). Recently it was found that in the region where $\omega_p \simeq \omega_{LO}$ there is a large enhancement of the bipolaron binding energy [62]. In these theories the long-range electron–phonon coupling may have an important role [53, 54]. Self-localized polarons have already been observed in the semiconducting parent compounds with photoinduced infrared absorption [55, 56].

Using the relation $\omega_p^2 = 4\pi n e^2/(\varepsilon(\infty)m^*)$ we estimate that approximately 60% of the charge carriers involved in the plasma oscillations in the normal state (600 cm^{-1}) are condensed below T_c (365 cm^{-1}). If we correlate these excitations with the normal state carriers, we have to conclude that such carriers exist in the superconducting state simultaneously with the condensate. This idea is clearly compatible with Bose condensation of bipolarons and similar theories [50, 57]. It is also compatible with all two-fluid-type models (see e.g. [58, 59]), including some aspects of strong coupling theory [60, 61] as long as it allows for normal state carriers below T_c .

4. Conclusion

We fitted the $E||c$ far-infrared spectra of $YBa_2Cu_3O_7$ using the four-parameter model for the phonons along with a low-lying plasmon and the mid-infrared term.

We found that the plasma frequency changed at T_c from 600 cm^{-1} to 365 cm^{-1} (the renormalized values are 269 cm^{-1} and 164 cm^{-1} respectively). This causes the renormalized LO phonon frequency to be close to the upper coupled plasmon–phonon mode ω_+ in the superconducting state. This coupling might provide an additional mechanism to raise T_c .

The presence of the mid-infrared term caused a substantial reduction in the LO–TO splitting from 100 cm^{-1} to 30 cm^{-1} approximately. This may cause a transition from a small to a large polaronic state, which is a necessary condition for bipolaronic superconductivity.

We believe that a low-lying plasmon in the superconducting state (10 K) represents oscillations of normal carriers and therefore evidence for the coexistence of normal state carriers with superconducting pairs in the superconducting state.

Acknowledgments

RG would like to thank Dr M Bauer for sending his PhD thesis and the permission to use the data.

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