Electron-Phonon Coupling Deduced from Phonon Line Shapes

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Abstract

We investigate the Fano-type line shape of the Ba mode of Y$_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{6+y}$ films observed in Raman spectra with A$_{1g}$ symmetry. The line shape is described with an extended Fano formula that allows us to obtain the bare phonon parameters and the self-energy effects based on a phenomenological description of the real and imaginary part of the low-energy electronic response. It turns out that the phonon intensity originates almost entirely from a coupling to this electronic response with negligible contributions from interband (high-energy) electronic excitations. In the normal state we obtain a measure of the electron-phonon coupling via the mass-enhancement factor $\lambda$. We find $\bar{\lambda} = 6.8 \pm 0.5$ % around optimum doping and only weak changes of the self-energy in the superconducting state. With increased disorder at the Ba site we find a decreased intensity of the Ba mode which we can relate to a decreased electron-phonon coupling.

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Fano-type phonon line shapes and continua of electronic excitations are rather common observations in Raman experiments of doped high-temperature superconductors. Using extended Fano models like those presented by Chen et al. and Devereaux et al. the self-energy contributions to the bare phonon parameters as a consequence of the electron-phonon interaction can in principle be obtained. Moreover, knowledge of the self-energy effects allows one to obtain the mass-enhancement factor \( \lambda \) as a measure of the electron-phonon coupling strength. This, however, requires a simultaneous description of real and imaginary part of the electronic response function \( \chi^e(\omega) = R^e(\omega) + ig^e(\omega) \). Such a description has recently been presented by us and applied to the \( B_{1g} \) Raman-active phonon of \( Y_{1-x}(\text{Pr}, \text{Ca})_x \text{Ba}_2 \text{Cu}_3 \text{O}_7 \) [\( Y_{1-x}(\text{Pr}, \text{Ca})_x \text{-123} \)] films. Here, we will use our description in order to investigate the Fano-type line shape of the Ba mode observed in \( A_{1g} \) Raman spectra of \( Y_{1-x} \text{Ca}_x \text{-123-}O_{6+y} \) films near optimum doping. This mode exhibits a pronounced asymmetry indicative of a strong electron-phonon coupling which is in fact obtained with our description.

We study epitaxial \( Y_{1-x} \text{Ca}_x \text{-123-}O_{6+y} \) films grown by pulsed laser deposition on \( \text{SrTiO}_3(100) \) substrates in a process described elsewhere. Standard films (\( x=0 \); \( y=1 \)) are slightly overdoped, increased (decreased) doping levels are obtained by Ca substitution (oxygen reduction). Some properties of the films are given in Table 1. Raman spectra have been taken using the Ar\(^+\) laser line at 458 nm in a setup described elsewhere with the spectral resolution (HWHM) set to 3 cm\(^{-1}\). They have been corrected for the spectral response of spectrometer and detector and are normalized such that the \( B_{1g} \) spectrum of the film \#O4 at 18 K approaches unity above 650 cm\(^{-1}\). \( A_{1g} \) spectra are obtained by subtracting the \( B_{2g} \) data from those measured in \( z(x'x')\bar{z} \) geometry. All temperatures are spot temperatures with typical heatings of 2 K.

In order to describe the Fano-type line shape of the Ba mode in \( A_{1g} \) symmetry we subdivide the Raman efficiency \( I_0(\omega) \) into the electronic response and an electron-phonon interference term:

\[
I_0(\omega) = \frac{\rho_s(\omega) + C}{\gamma(\omega) [1 + \epsilon^2(\omega)]} \left\{ \left[ \frac{R_{\text{tot}}(\omega)}{C} \right]^2 - 2\epsilon(\omega) \frac{R_{\text{tot}}(\omega)}{C} \frac{\rho_s(\omega)}{C} - \left[ \frac{\rho_s(\omega)}{C} \right]^2 \right\}. \tag{1}
\]

The constant \( C = A\gamma^2/g^2 \) is a fit parameter for the intensity where \( \gamma \) represents the symmetry element of the electron-phonon vertex projected out by the incoming and outgoing polarization vectors and \( g \) is the lowest order expansion coefficient of the electron-phonon vertex describing the coupling to non-resonant intraband electronic excitations. \( \rho_s(\omega) = C g^2 g^e(\omega) \) and \( R_s(\omega) = C g^2 R^e(\omega) \) are the measured electronic response and the real part of the electronic response function which are connected by Kramers-Kronig relations. \( R_{\text{tot}}(\omega) = R_s(\omega) + R_0 \) with \( R_0 = C g(g_{pp}/\gamma) \) where \( g_{pp} \) is a constant which represents a mass-enhanced “phonon-phonon” vertex that describes the coupling to resonant interband electronic excitations. The renormalized phonon parameters in the above equation are \( \gamma(\omega) = \Gamma + \rho_s(\omega)/C \) and \( \omega_p^2(\omega) = \omega_p^2 - 2\omega_p R_s(\omega)/C \) with \( \epsilon(\omega) = [\omega^2 - \omega_p^2(\omega)]/[2\omega_p \gamma(\omega)] \). Note, that the interference term in Eq. (1) can be negative in contrast to the Raman efficiency.

The measured electronic response (background) is modeled by two contributions: \( I_{\infty} \tanh(\omega/\omega_T) \) and \( I_{\text{red}}(\omega, \omega_{2\Delta}, \Gamma_{2\Delta}, I_{2\Delta}, I_{\text{supp}}) \). The first term models the incoherent background using a hyperbolic tangent and the second the redistribution below \( T_c \) using two Lorentzians. For the two contributions analytic expressions of the real part of the electronic
response function exist. In the present work, the hyperbolic tangent is cut off at \( \omega_{\text{cut}} = 8000 \text{ cm}^{-1} \). \( R_\ast(\omega_p)/C \) increases by typically 10 \% when the cutoff is increased to 12000 \text{ cm}^{-1}.

With the description according to Eq. (1) a measure of the electron-phonon coupling can be obtained via the mass-enhancement factor \( \lambda \), defined by \( \lambda \cdot \omega_p = 2R_\ast(\omega_p)/C \). In reference to the conventional Fano mechanism, the total and the bare phonon intensity are

\[
I_{\text{tot}} = \pi C R^2_{\text{tot}}(\omega_p) \quad \text{and} \quad I_{\text{phon}} = \pi C R^2_0.
\]

To give an example, Fig. 1 displays the results of the analysis of the A\(_{1g}\) efficiency of the film #Ox4 taken at 18 K. We describe this figure from top to bottom: At the top, the measured efficiency as well as its description (solid line) is displayed. For the description we used interference terms given in Eq. (1) for the Ba and the O(4) mode, Lorentzians for the Cu(2) and the O(2)+O(3) mode, a simple Fano formula for the B\(_{1g}\) phonon, and the two background contributions stated above. In the second trace from the top, the phononic signal, i.e. the Lorentzians, the Fano profile, and the interference terms are given. Obviously, the interference term of the Ba mode becomes negative in a region above \( \sim 120 \text{ cm}^{-1} \). The electronic response \( \rho_\ast(\omega) \) that remains after subtraction of the phonons is shown below the phononic signal. It exhibits a 2\( \Delta \) peak at \( \sim 280 \text{ cm}^{-1} \) as well as a monotonically decreasing intensity for \( \omega \to 0 \) and is well described with our background model (solid line). At the bottom of Fig. 1 the real part of the electronic response function \( R_\ast(\omega) \) is shown. In order to obtain \( R_\ast(\omega) \) we have performed a numerical Hilbert transformation of \( \rho_\ast(\omega) \). For the transformation the measured spectrum is taken as constant for high frequencies up to \( \omega_{\text{cut}} \) and is interpolated to zero intensity at \( \omega = 0 \); for negative frequencies the antisymmetry of \( \rho_\ast(\omega) \) has been used. Evidently, the description of \( R_\ast(\omega) \) used in the fit agrees well with the numerically obtained data. This is important for the determination of the self-energy effects.

At 152 K, i.e. considerably above \( T_c \) and below room temperature, our description of the Ba mode in the film #Ox4 yields \( \omega_p = 121.6 \text{ cm}^{-1} \) and \( \lambda = 6.3 \% \). Similar values are obtained in the other films as given in Table I. More specifically, we find a somewhat increasing bare phonon frequency with increasing doping with a mean value of \( \bar{\omega}_p = 121.9 \pm 1.4 \text{ cm}^{-1} \) and a mean value of the mass-enhancement factor of \( \bar{\lambda} = 6.8 \pm 0.5 \% \). The disordered film #Ca1 clearly deviates from the others exhibiting a low bare phonon frequency compared with its doping value and an almost 50 \% smaller mass-enhancement factor. The low frequency is in fact one indication for the presence of disorder at the Ba site as enlarged \( c \)-axis parameters are expected in this case.

In order to look more closely at the peculiarities appearing in the disordered film we compare the temperature dependencies of the fit parameters of the Ba modes in the disordered film #Ca1 with that of the ordered one #Ca2 in Fig. 2. Beside the bare and renormalized phonon parameters also the self-energy contributions at \( \omega = \omega_p \) are depicted. Dashed lines are fits to anharmonic decays. Except for sharpenings of the bare and the renormalized phonon linewidth in the ordered film #Ca2, clear superconductivity-induced changes of the self-energies are not observed. This is in good agreement with earlier results obtained on a Y-123 single crystal. The self-energy contributions are weaker in the disordered film compared to the ordered one. In particular, they differ by a factor of two above \( T_c \) which is the same value by which the mass-enhancement factors are apart. Noteworthy, the sharpening of the renormalized linewidth in the ordered film cannot be related to a suppression of the measured electronic response. This indicates the presence of an additional decay channel for
the phonon. A similar effect has been observed in case of the $B_{1g}$ phonon in $Y_{1-x}(Pr, Ca)_x$-123 films where it was suspected that not Raman-active electronic excitations may be present in these compounds.

In the lowest panel in Fig. 2 the total intensities $I_{tot}$ as well as the bare phonon intensities $I_{phon}$ are given. It turns out that the bare phonon intensities are negligibly small in both films at least for temperatures below 250 K. The same finding is also observed in the other films studied. The total intensities, on the other hand, are 30% stronger in the ordered film #Ca2 compared to the disordered one. This appears to be related to the stronger mass-enhancement factor in the ordered film which is further supported when comparing with the factors and intensities of the other films given in Table 1. Regarding Table 1, one finds an increasing intensity of the Ba mode with decreasing doping in the studied doping regime. This increase is carried by an increasing linewidth which rises from $\Gamma = 4$ cm$^{-1}$ in the film #Ca2 up to $\Gamma = 8$ cm$^{-1}$ in the film #Ox2. At even lower dopings, however, the Ba mode eventually diminishes and is no longer observed in plane-polarized Raman spectra in the parent compound Y-123-O6.

Isotope experiments of Y-123 have shown that the mode, which we have called the Ba mode so far, is indeed dominated by vibrations of the Ba atom with less than 20% admixture from the Cu(2) site. This experimental eigenvector has recently been obtained in a linearized-augmented-plane-wave (LAPW) frozen-phonon calculation within a generalized gradient approximation (GGA). Using the LAPW method within the local-density approximation (LDA), Cohen et al. find large non-local contributions to the electron-phonon coupling from the Ba site with $\lambda \leq 5.4$ % in the Brillouin zone. The coupling appears to be in good agreement with the results of our extended Fano description of the ordered films. This is somewhat surprising as the observed background is believed to be a consequence of the strong electronic correlations which are not included in LDA-type calculations. Results of the electron-phonon coupling within the GGA are therefore of interest for a comparison as that method includes correlation effects.

To conclude, we investigate the Ba mode of $Y_{1-x}Ca_x$-123-O$_{6+y}$ films observed in Raman spectra with $A_{1g}$ symmetry. Our Fano-type analysis reveals that this mode is entirely described by a coupling to low-energy electronic excitations for $T < 250$ K. The absence of this mode in antiferromagnetic Y-123-O6 might therefore simply be a consequence of the vanishing low-energy electronic response. Our analysis yields mass-enhancement factors which appear to be in agreement with the result of the LAPW method within the LDA. With increasing disorder at the Ba site, the intensity of the Ba mode diminishes as a consequence of a reduced coupling strength.

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FIGURES

FIG. 1. In the uppermost spectrum the Raman efficiency $I_0(\omega)$ of the film #Ox4 taken at $T = 18$ K in $A_{1g}$ symmetry (dots) and the fit result (solid line) are shown. Below the fitted phonon profiles (solid line), the electronic response $\rho_\star(\omega)$ obtained after subtraction of the phonons (dots), and the numerically determined real part of the electronic response function $R_\star(\omega)$ (dots) are given. The analytical descriptions of the response used in the fit are included as solid lines. The spectra are offset as indicated in brackets, all intensities are given in the same units.

FIG. 2. Temperature dependence of fit parameters of the Ba mode mode for two Ca-doped films. Left (right) panel: disordered (ordered) film. Closed circles represent the bare phonon parameters $\omega_p$, $\Gamma$, and $I_{phon}$, open circles the renormalized values $\omega_{\nu}(\omega_p)$, $\gamma(\omega_p)$ and $I_{tot}$. Diamonds and crosses are the self-energy contributions $R_\star(\omega_p)/C$ and $\rho_\star(\omega_p)/C$, respectively. Dashed lines are fits to anharmonic decays and dash-dotted lines indicate the respective $T_c$’s of the films. Marker sizes represent the vertical accuracies.
TABLE I. Names and properties of the investigated Y_{1-x}Ca_x-123-O_{6+y} films. In contrast to the others, the film #Ca1 exhibits site-substitution disorder at the Ba site. The critical temperature $T_c$ is defined by zero resistance with $\Delta T_c \leq 2$ K. The hole doping per copper-oxygen plane $p$ has an accuracy of $\pm 0.008$ holes. $\omega_p$, $\lambda$, and $I_{tot}$ are the bare phonon frequency, the mass-enhancement factor, and the total intensity of the Ba mode at 152 K, respectively.

| Film | x (%) | y | $T_c$ (K) | $p$ | $\omega_p$ (cm$^{-1}$) | $\lambda$ (%) | $I_{tot}$ (arb. units) |
|------|-------|---|--------|----|-------------------|--------|-------------------|
| #Ox2 | 0 | 0.85 ± 0.05 | 87.0 | 0.145 | 120.0 | 6.2 | 58 |
| #Ox3 | 0 | 0.93 ± 0.05 | 89.8 | 0.163 | 122.2 | 7.5 | 47 |
| #Ox4 | 0 | 1.00 ± 0.05 | 88.0 | 0.180 | 121.6 | 6.3 | 40 |
| #Ca1 | 5 | 1.00 ± 0.05 | 85.0 | 0.186 | 120.0 | 3.8 | 27 |
| #Ca2 | 5 | 1.00 ± 0.05 | 82.7 | 0.198 | 123.8 | 7.1 | 40 |
