Electronic and phononic Raman scattering in detwinned YBa$_2$Cu$_3$O$_{6.95}$ and Y$_{0.85}$Ca$_{0.15}$Ba$_2$Cu$_3$O$_{6.95}$: 
$s$-wave admixture to the $d_{x^2−y^2}$-wave order parameter

M. Bakr, 1 A. P. Schnyder, 2 L. Klam, 1 D. Manske, 1 C.T. Lin, 1 B. Keimer, 1 M. Cardona, 1 and C. Ulrich 1

1Max-Planck-Institut f"ur Festk"orperforschung, Heisenbergstr. 1, D-70569 Stuttgart, Germany
2Kavli Institute for Theoretical Physics, University of California, Santa Barbara, CA 93106, USA
(Dated: July 27, 2009)

Inelastic light (Raman) scattering has been used to study electronic excitations and phonon anomalies in detwinned, slightly overdoped YBa$_2$Cu$_3$O$_{6.95}$ and moderately overdoped Y$_{0.85}$Ca$_{0.15}$Ba$_2$Cu$_3$O$_{6.95}$ single crystals. In both samples modifications of the electronic pair-breaking peaks when interchanging the $a$- and $b$-axis were observed. The lineshapes of several phonon modes involving plane and apical oxygen vibrations exhibit pronounced anisotropies with respect to the incident and scattered light field configurations. Based on a theoretical model that takes both electronic and phononic contributions to the Raman spectra into account, we attribute the anisotropy of the superconductivity-induced changes in the phonon lineshapes to a small $s$-wave admixture to the $d_{x^2−y^2}$ pair-wave function. Our theory allows us to disentangle the electronic Raman signal from the phononic part and to identify corresponding interference terms. We argue that the Raman spectra are consistent with an $s$-wave admixture with an upper limit of 20 percent.

PACS numbers: 74.20.Rp, 74.25.Gz, 74.25.Kc, 74.72.Bk

I. INTRODUCTION

The symmetry of the pair wavefunction provides important clues to the mechanism of high temperature superconductivity. Phase sensitive experiments have shown that the dominant component of the order parameter exhibits $d_{x^2−y^2}$ symmetry in cuprates of the YBa$_2$Cu$_3$O$_7$−$\delta$ type with orthorhombic crystal symmetry, an $s$-wave admixture resulting in an anisotropy of the superconducting (SC) energy gap, $2\Delta$, between the $a$- and $b$-axes is expected. A more general trend of increasing $s$-wave admixture with increasing doping level has also been reported.

In YBa$_2$Cu$_3$O$_{7−\delta}$ (YBCO$_{7−\delta}$), such an admixture has been confirmed by angle-resolved photoemission spectroscopy (ARPES) and flux measurements on Josephson junctions. However, the interpretation of the ARPES data is complicated because the surface properties of YBCO$_{7−\delta}$ may differ from those of the bulk. The phase-sensitive measurements, on the other hand, have yielded only lower bounds (9%) to the $s$-wave admixture. In addition, phenomenological calculations based on the two-dimensional Hubbard model have demonstrated that the in-plane anisotropy of the spin fluctuation spectrum determined by inelastic neutron scattering on detwinned YBCO$_{7−\delta}$ is consistent with a small isotropic $s$-wave admixture to the $d_{x^2−y^2}$-pairing symmetry. However, the same calculations show that the dispersion and spectral weight of the spin fluctuations are also influenced by anisotropic hopping parameters that are difficult to determine independently. Moreover, different interpretations of the neutron data have also been proposed.

Further experiments are therefore required to conclusively establish the magnitude of the $s$-wave contribution to the SC gap in YBCO$_{7−\delta}$.

Inelastic light (Raman) scattering is a powerful probe of electronic and lattice vibrational excitations in high-$T_c$ superconductors. Both electronic and phononic Raman scattering have been applied to study the superconducting gap anisotropy in YBCO$_{7−\delta}$. The anisotropy can be inferred from the energies of gap features of the electronic continuum in various polarization geometries but the accuracy of this method is limited by overlap with Raman-active phonons. Possible manifestations of the gap anisotropy and an isotropic electron-phonon coupling in the lineshape of a Raman-active out-of-plane vibration of the in-plane oxygen atoms of $B_{1g}$ symmetry have also been investigated. However, the analysis of these data has been contested and a quantitative estimate of the gap anisotropy has not been extracted from them.

The present work was in part motivated by a recent theoretical study that yielded quantitative predictions for the electronic Raman continua in a $(d_{x^2−y^2}+s)$-wave superconductor. In order to enable a detailed comparison with these predictions, we have performed Raman scattering measurements of the temperature dependence of the electronic continua and phonon modes in twin-free, slightly overdoped YBCO$_{6.95}$ and moderately overdoped Y$_{0.85}$Ca$_{0.15}$Ba$_2$Cu$_3$O$_{6.95}$ (henceforth YBCO$_{6.95}$:Ca) single crystals. We found that interference between the electronic and phononic scattering channels imposed severe limitations on our capability to extract information about the gap anisotropy from the electronic continuum alone, in accordance with previous work. We have therefore employed a phenomenological model that treats the electronic and phononic contributions to the Raman signal on equal footing. This formalism yields predictions for the energy, intrinsic linewidth, and Fano parameters of phonons coupled to the electronic continuum as a function of temperature. From a comparison to the experimentally determined lineshapes of two phonon modes, the $B_{1g}$ vibration of the in-plane oxygen at 340 cm$^{-1}$ as well as an apical-oxygen vibration at 501 cm$^{-1}$, we extract an upper bound of 20% on the $s$-wave admixture to the SC energy gap. This is consistent with a 10% to 15% admixture of $s$-wave to the $d_{x^2−y^2}$ pair-breaking peak observed in the electronic continuum in the superconducting state, and it coincides with the lower bound extracted from the phase-sensitive measurements. Similar effects have been recently reported for an organic superconductor with
same point group as $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ($D_{2h}$). In this case, however, the analysis led to the conclusion that only an isotropic $s$-wave state is present.\textsuperscript{22}

This paper is organized as follows. In Sec. II we describe experimental details and discuss the raw Raman spectra. An introduction to our theoretical model is given in Sec. III. In particular, we focus on a simultaneous description of both, the electronic Raman response and the phonon anomalies above and below $T_c$. Based on this description we are able to analyze the superconductivity-induced changes in the phonon lineshape in Sec. IV. A summary of our results and a comparison to prior work is contained in Sec. V.

II. EXPERIMENTAL DETAILS

High quality single crystals of $\text{YBCO}_{6.95}$ and $\text{YBCO}_{6.95}:\text{Ca}$ were grown by the top-seeded solution growth method.\textsuperscript{23} The orientations of the crystallographic axes were determined by Laue x-ray diffraction and polarized light microscopy. The samples were then cut into rectangular shapes of typical size $3 \times 3 \times 1 \text{ mm}^3$, annealed at 520°C in a flow of oxygen gas for 150 hours, and quenched in liquid nitrogen in order to avoid further oxygen diffusion. The magnetization curves of the $\text{YBCO}_{6.95}$ ($\text{YBCO}_{6.95}:\text{Ca}$) crystal show an onset of $T_c = 92 \text{ K}$ (75 K). A transition width $\Delta T_c$ of less than 3 K (6 K) indicates good homogeneity of the samples. Employing Tallon's phenomenological expression,\textsuperscript{24} the hole doping level of our samples is estimated as 0.17 (0.21) per planar Cu ion, i.e., they are slightly (moderately) overdoped. The crystals were detwinned individually using the procedure described in Refs. 23 and 24. Raman spectra measured at different spots of the $ab$-surface of each sample look basically the same, thus confirming the homogeneity of our single crystals.

The Raman scattering experiments were performed in backscattering geometry using a Dilor XY-triple grating Raman spectrometer with a CCD camera as detector. For excitation, the 514.5 nm line of an Ar$^+$ laser line ($\lambda = 514.5 \text{ nm}$) was used. The resolution of our spectrometers for this experiment was about 3 cm$^{-1}$. In order to avoid heating of the sample, the power of the incident laser was kept below 10 mW at the sample surface with a laser spot of 100 $\mu$m in diameter. The direction of the incident laser light was always parallel to the crystallographic $c$-axis. For the selection rule analysis, a polarizer and an analyzer were placed into the light path before and after the sample.

$\text{YBCO}_{6.95}$ and $\text{YBCO}_{6.95}:\text{Ca}$ are orthorhombic with the space group $Pmmm$ ($D_{2h}$ symmetry). In polarized Raman scattering experiments, excitations of $A_g$, $B_{1g}$, $A_g+B_{1g}$, and $A_g$ symmetries are accessible by using $xx$ (or $yy$), $xy$ (or $yx$), $x'x'$ (or $y'y'$), and $x'y'$ (or $y'x'$) polarizations of the incident and scattered light fields, respectively.\textsuperscript{17,27} In this notation $x$ and $y$ correspond to the direction of the electric field of light along the $a$- and $b$-axes, whereas $x'$ and $y'$ correspond to the diagonal directions, i.e., $x' \sim x + y$ and $y' \sim x - y$, respectively. As most previous work on YBCO has been performed on twinned specimens, we follow previous publications and use the tetragonal notation of the polarization symmetries throughout this paper. In this notation, the structure is described in terms of the closely related tetragonal point group $D_{4h}$, and the excitations of $A_g+B_{1g}$ symmetry are accessible for either $xx$ or $yy$ polarization, $B_{2g}$ for either $xy$ or $yx$ polarization, $A_g+B_{2g}$ for either $x'x'$ or $y'y'$ polarization, and $B_{1g}$ for either $x'y'$ or $y'x'$ polarization of the incident and scattered electric field vectors.

Figure 1 show Raman spectra of detwinned $\text{YBCO}_{6.95}$ crystals measured at various temperatures. As in other high-$T_c$ superconductors,\textsuperscript{16,28,29,30,31} the normal-state spectra exhibit a flat electronic continuum with superimposed phonons. Below $T_c$, a significant fraction of the electronic spectral weight is transferred to higher energies, resulting in a broad pair-breaking peak in the electronic continuum. In addition, the phonon lineshapes reveal characteristic changes. In $B_{1g}$ polarization this spectral-weight redistribution is most pronounced because electronic Raman scattering in this geometry is sensitive to the maximum of the SC gap along the anti-nodal direction of the two-dimensional SC gap. We therefore focus in this work on this geometry, along with the $xx$ and $yy$ polarization channels that provide direct information about the in-plane anisotropy of the electronic response.

Figure 1 gives an overview of the phonon modes of $\text{YBCO}_{6.95}$ in the $xx$ and $yy$ polarization geometries. As expected based on a group-theoretical analysis,\textsuperscript{17,22,33,34,35,36} five phonons appear in both polarization channels. The lowest-frequency phonon at 113 cm$^{-1}$ ($A_{1g}$) originates predominantly from vibrations of the barium atoms. The next low-
est frequency phonon at 148 cm\(^{-1}\) (\(A_{1g}\)) corresponds mainly to the vibrations of copper (Cu2 ions). The vibrations of the apical oxygen ions (O4) appears at 501 cm\(^{-1}\) (\(A_{1g}\)). The two modes at 340 cm\(^{-1}\) (\(B_{1g}\)) and 446 cm\(^{-1}\) (\(A_{1g}\)) originate from out-of-phase and in-phase vibrations of the planar oxygen ions (O2 and O3), respectively. Two additional defect-induced modes appear in the \(yy\) symmetry at 232 cm\(^{-1}\) and at 579 cm\(^{-1}\). They originate from vibrations of the copper (Cu1) and the oxygen (O1) ions, respectively, in the Cu–O chains, which are aligned along the crystallographic \(b\)-axis. In YBCO crystals with fully oxygenated chains, these two modes are Raman forbidden but infrared–allowed (\(B_{1g}\) symmetry). The vibrations of the copper (Cu2 ions). The vibrations of the (intraband) and phononic excitations. In order to disentangle these two contributions it is necessary to employ a proper fitting procedure. A phonon interacting with intraband excitations acquires a renormalized self-energy and exhibits an asymmetric, Fano-type Raman lineshape. To describe such an electron-phonon coupled Raman spectrum the following formula can be employed:

\[
|\chi(\omega)|^2 = \frac{g^2}{\Gamma(\omega)} \left( \frac{1 + \epsilon^2(\omega)}{1 + \epsilon^2(\omega)} \right)
\]

The measured Raman intensity \(I_\sigma(\omega)\) in a given polarization channel \(\sigma\) is related to the imaginary part of the response function \(\Im |\chi(\omega)|^2 \propto I_\sigma(\omega)\), where \(\Im |\chi(\omega)|^2\) denotes the Bose distribution and \(A\) is a coupling constant. The Raman response in cuprate superconductors in the optimally and overdoped regimes consists of electronic (intraband) and phononic excitations. In order to disentangle these two contributions it is necessary to employ a proper fitting procedure. A phonon interacting with intraband excitations acquires a renormalized self-energy and exhibits an asymmetric, Fano-type Raman lineshape. To describe such an electron-phonon coupled Raman spectrum the following formula can be employed:

\[
\Im |\chi(\omega)|^2 = \rho_\sigma(\omega) + \frac{g^2}{\Gamma(\omega)\left[1 + \epsilon^2(\omega)\right]} \times \left\{ S^0(\omega) - 2\epsilon(\omega)S(\omega)\rho(\omega) - \rho_\sigma^2(\omega) \right\},
\]

where \(\epsilon(\omega) = (\omega^2 - \Omega^2)/2\omega_0\Gamma(\omega)\) and \(S(\omega) = S_0 + R_\sigma(\omega)\). The renormalized phonon frequency and the renormalized phonon line width are given by \(\Omega^2 = \omega_0^2 - 2\omega_0g^2\rho(\omega)\) and \(\Gamma(\omega) = \Gamma_0 + g^2\rho_\sigma(\omega)\), respectively, with the intrinsic phonon frequency \(\omega_0\) and the bare phonon line width \(\Gamma_0\). The parameter \(S_0\) can be expressed in terms of the electron-phonon coupling \(g_\sigma\), the Raman phonon matrix element \(T_\sigma\), and the Raman electronic matrix element \(\gamma_\sigma\), that is, \(S_0 = T_\sigma/(\gamma_\sigma g_\sigma)\). In our model, \(g_\sigma, T_\sigma,\) and \(\gamma_\sigma\) are assumed to be real and therefore \(S_0\) is real. The Kramers-Kronig related functions \(R_\sigma(\omega)\) and \(\rho_\sigma(\omega)\) denote real and imaginary parts of the electronic response function, respectively.

III. ANALYSIS OF RAMAN SPECTRA

The measured Raman intensity \(I_\sigma(\omega)\) in a given polarization channel \(\sigma\) is related to the imaginary part of the response function \(\Im |\chi(\omega)|^2 \propto I_\sigma(\omega)\), where \(\Im |\chi(\omega)|^2\) denotes the Bose distribution and \(A\) is a coupling constant. The Raman response in cuprate superconductors in the optimally and overdoped regimes consists of electronic (intraband) and phononic excitations. In order to disentangle these two contributions it is necessary to employ a proper fitting procedure. A phonon interacting with intraband excitations acquires a renormalized self-energy and exhibits an asymmetric, Fano-type Raman lineshape. To describe such an electron-phonon coupled Raman spectrum the following formula can be employed:

\[
\Im |\chi(\omega)|^2 = \rho_\sigma(\omega) + \frac{g^2}{\Gamma(\omega)\left[1 + \epsilon^2(\omega)\right]} \times \left\{ S^0(\omega) - 2\epsilon(\omega)S(\omega)\rho(\omega) - \rho_\sigma^2(\omega) \right\},
\]

where \(\epsilon(\omega) = (\omega^2 - \Omega^2)/2\omega_0\Gamma(\omega)\) and \(S(\omega) = S_0 + R_\sigma(\omega)\). The renormalized phonon frequency and the renormalized phonon line width are given by \(\Omega^2 = \omega_0^2 - 2\omega_0g^2\rho(\omega)\) and \(\Gamma(\omega) = \Gamma_0 + g^2\rho_\sigma(\omega)\), respectively, with the intrinsic phonon frequency \(\omega_0\) and the bare phonon line width \(\Gamma_0\). The parameter \(S_0\) can be expressed in terms of the electron-phonon coupling \(g_\sigma\), the Raman phonon matrix element \(T_\sigma\), and the Raman electronic matrix element \(\gamma_\sigma\), that is, \(S_0 = T_\sigma/(\gamma_\sigma g_\sigma)\). In our model, \(g_\sigma, T_\sigma,\) and \(\gamma_\sigma\) are assumed to be real and therefore \(S_0\) is real. The Kramers-Kronig related functions \(R_\sigma(\omega)\) and \(\rho_\sigma(\omega)\) denote real and imaginary parts of the electronic response function, respectively.

\[
|\chi(\omega)|^2 = \frac{g^2}{\Gamma(\omega)\left[1 + \epsilon^2(\omega)\right]} \times \left\{ S^0(\omega) - 2\epsilon(\omega)S(\omega)\rho(\omega) - \rho_\sigma^2(\omega) \right\},
\]

where \(\epsilon(\omega) = (\omega^2 - \Omega^2)/2\omega_0\Gamma(\omega)\) and \(S(\omega) = S_0 + R_\sigma(\omega)\). The renormalized phonon frequency and the renormalized phonon line width are given by \(\Omega^2 = \omega_0^2 - 2\omega_0g^2\rho(\omega)\) and \(\Gamma(\omega) = \Gamma_0 + g^2\rho_\sigma(\omega)\), respectively, with the intrinsic phonon frequency \(\omega_0\) and the bare phonon line width \(\Gamma_0\). The parameter \(S_0\) can be expressed in terms of the electron-phonon coupling \(g_\sigma\), the Raman phonon matrix element \(T_\sigma\), and the Raman electronic matrix element \(\gamma_\sigma\), that is, \(S_0 = T_\sigma/(\gamma_\sigma g_\sigma)\). In our model, \(g_\sigma, T_\sigma,\) and \(\gamma_\sigma\) are assumed to be real and therefore \(S_0\) is real. The Kramers-Kronig related functions \(R_\sigma(\omega)\) and \(\rho_\sigma(\omega)\) denote real and imaginary parts of the electronic response function, respectively.

\[
\rho(\omega) = \frac{\omega}{\sqrt{\omega^2 + \epsilon_1^2(\omega)}} + \frac{C_1}{1 + \epsilon_1^2(\omega)} - (\omega \rightarrow -\omega),
\]

where \(\epsilon_1(\omega) = (\omega - \omega_1)/\Gamma_1\) and \(\epsilon_2(\omega) = (\omega - \omega_2)/\Gamma_2\). The first term in Eq. (4) models an incoherent electronic background, which dominates the response in the normal state of cuprate superconductors. It is linear in \(\omega\) at small frequencies and becomes constant for large Raman shifts. The second and third terms are Lorentzians describing the pair-breaking peak located below 2\(\Delta\), and the suppression of spectral weight at low frequencies, respectively. The latter is due to the opening of the SC gap. To reduce the number of free fitting parameters, we set \(\omega_2 = \Gamma_2 = (\omega_1 - \Gamma_1)/2\). The last two terms in Eq. (4) decrease in intensity and shift spectral weight to lower frequency as the superconducting transition temperature \(T_c\) is approached from below. When \(\omega \rightarrow -\omega\) terms, similar to the \(C_1\) and \(C_2\) terms but with \(\omega\) replaced by \(-\omega\), are essential to achieve the symmetry requirements for the Raman response. In the normal state, the electronic response function is entirely described by the incoherent contribution [first
solid curve the fitting result with formula (1) and four Lorentzians for the remaining data after phonon subtraction. Panels (c) and (d) show the phonon-subtracted spectra for the $XX$ and $YY$ (20K) Raman spectra of YBCO$_{6.95}$ (100K) and YBCO$_{6.95}$/Ca in the superconducting (normal) state. $C_n$ and $\omega_T$ describe the intensity and position of the maximum of the normal-state electronic background given by the square root of a rational function. In the superconducting state $C_1/\Gamma_2$, $\omega_{1,2}$, and $\Gamma_{1,2}$ describe amplitude, position and width of a Lorentzian function reflecting the region of the pair-breaking peak. Finally, $\Gamma_0$, $\omega_0$, and $S_0$ effectively characterize amplitude, width, position, and asymmetry of a generalized Fano function (describing the $B_{1g}$ phonon). Note that for not too strong frequency dependences of $\rho(\omega)$ and $R(\omega)$, the renormalization of the Fano formula due to the electronic background can be simply viewed as an offset of the parameters entering in the Fano formula [cf. Eqs. (1), (2), and (3)]. The results of this fit procedure are shown in Fig. 2 and Fig. 3 together with the experimental data. Table I lists the corresponding fit parameters. Note that on general grounds the extracted $\omega_0$ and $\Gamma_0$ parameter values need to be identical at a given temperature for all measurements of a given phonon. In Table I we show, however, the parameter values that correspond to the best (non-linear least squares) fit to the data. The differences reflect the error bars of our procedure. It is important to emphasize that the Fano profile of the $B_{1g}$-mode shown in Figs. 2(b) and 3(a) results from the interaction with the electronic Raman signal; both electronic and phononic contributions and their interdependence can be described by Eqs. 1 and 4. The final result agrees well with the measured data. Thus, our model allows us to some extent to disentangle the electronic and phononic parts of the Raman response, and to identify the shape of the electronic background. However, strictly speaking, only a combination of parameters such as $g^2\rho$ and $g^2R$ can be extracted. In the next subsection this procedure will be improved for the SC state by employing a microscopic description of the pair-breaking excitations.

| YBCO$_{6.95}$ | $S_0$ | $2\Gamma_0$ | $\omega_0$ | $\omega_0$(INS) | $\omega_0$(sqrt) | $\omega_0$(lin.) |
|----------------|------|-------------|-------------|-----------------|-----------------|-----------------|
| $B_{1g}$ (100K)| 1.5  | 15.3        | 343.1       | 343.9           | 342.8           | 341.9           |
| $XX$ (100K)    | 1.8  | 12.2        | 342.9       | 343.9           | 339.1           | 340.7           |
| $YY$ (100K)    | 1.8  | 12.2        | 342.9       | 343.9           | 339.5           | 338.7           |
| $B_{1g}$ (20K) | 2.6  | 14.8        | 338.6       | 338.4           | 335.8           | 335.7           |
| $XX$ (20K)     | 4.1  | 13.2        | 336.4       | 338.4           | 333.0           | 332.5           |
| $YY$ (20K)     | 2.1  | 13.8        | 336.3       | 338.4           | 334.2           | 334.2           |

TABLE I: Extracted parameter values of the $B_{1g}$ oxygen vibration in YBCO$_{6.95}$ measured at 20 K and 100 K, respectively. Left part: asymmetry parameter $S_0$, intrinsic phonon linewidth (FWHM) $2\Gamma_0$, and intrinsic phonon frequency $\omega_0$ using the generalized Fano approach of Eq. (1). Right part: $\omega_0$ extracted with Eq. (2) for different phenomenological electronic backgrounds [sqrt = first term in Eq. (2); lin. = linear background with offset at $\omega = 0$]. For comparison we display in the middle part $\omega_0$ obtained from inelastic neutron scattering (INS) experiments by Reznik et al.\textsuperscript{22}.
conducting order parameter. In particular, we are interested in information about the wavevector dependence of the super-Raman spectra based on Eq. (1) allows us to obtain precise microscopic approach in conjunction with the analysis of the model introduced in Ref. 21. The electronic Raman response with anisotropic hopping parameters and a superconducting gap with a mixture of

function in the SC state for a given symmetry channel \( \sigma \) is described by Eq. 5:

\[
\chi_{\sigma}(\omega) = \langle \gamma_\sigma^2 \theta_k(\omega) \rangle - \langle \gamma_\sigma \theta_k(\omega) \rangle^2 / (\theta_k(\omega)),
\]

(5)

\( \theta_k(\omega) \) being the Tsuneto function. The angular brackets denote the average over the Brillouin zone, that is,

\[
\langle (\cdots) \theta_k(\omega) \rangle = \frac{1}{V} \sum_k (\cdots) \Delta_k^2 \tanh \left( \frac{E_k}{2T} \right) \times \left( \frac{1/E_k^2}{\omega + i\eta + 2E_k} - \frac{1/E_k^2}{\omega + i\eta - 2E_k} \right).
\]

(6)

The quasiparticle dispersion \( E_k = \sqrt{\varepsilon_k^2 + \Delta_k^2} \) contains an effective one-band description of a single copper-oxygen plane

\[
\varepsilon_k = -2t \left[ (1 + \delta_0) \cos k_x + (1 - \delta_0) \cos k_y \right] - 4t' \cos k_x \cos k_y - \mu,
\]

(7)

and the superconducting gap

\[
\Delta_k = \frac{\Delta_d}{2} \left( \cos k_x - \cos k_y \right) + \Delta_s.
\]

(8)

For the computation of the electronic Raman response in the superconducting state, Eq. 5, we have assumed the same band structure parameters as in Ref. 21 together with \( \Delta_d = 30 \text{ meV} \) and \( \Gamma_c = 5 \text{ meV} \).

The Raman vertices \( \gamma_{\sigma} \) in Eq. (5) can be classified according to the irreducible representations of the symmetry group of the crystal. Since we shall consider a model with small distortions, \( \delta_0 \ll 1, \Delta_s \ll \Delta_d \), we use the notation corresponding to tetragonal symmetry, as in Sec. II above. We can thus express, for example, the \( B_{1g} \) Raman vertex as

\[
\gamma_{B_{1g}} \propto t \left[ (1 + \delta_0) \cos k_x - (1 - \delta_0) \cos k_y \right].
\]

(9)

To describe the combined electronic and phononic Raman response in the SC state, we use Eq. (1) together with the real and imaginary parts of the electronic Raman response in the SC state, Eq. 5. Furthermore, we assume that the bare fit parameters do not change as we go from the normal state to the SC state.

In Figure 3(a) we show numerical results obtained from our theoretical model and their comparison with the data on YBCO\(_{6.95}\) obtained in \( B_{1g} \)-polarization. The calculations have been performed at \( T = 20 \text{ K} \) and for various \( s \)-wave contributions. The best description for both the \( B_{1g} \)-mode and the electronic response is found if \( \Delta_s \) is assumed to be 10 percent of the maximum of the \( d_{x^2-y^2} \)-wave gap (short dotted line); this also accounts for the slight shift of the pair breaking peaks in \( xx \) and \( yy \) polarizations (Fig. 3(c and d). Furthermore, our numerical results solely for the electronic response, i.e. the \( 2\Delta \)-pair-breaking peak, are also displayed in Fig. 4 (dashed, dotted and dash-dotted lines). They are obtained by setting all phononic parts and the corresponding interference terms in Eq. (1) to zero. Interestingly, we find that the pair-breaking peak shifts to lower energies with increasing \( \Delta_s \), a
FIG. 4: (color online) (a) $B_{1g}$ Raman spectra of YBCO$_{6.95}$ in the superconducting state at $T = 20$ K ($\lambda=514.5$ nm). Open circles show the experimental data. The short dashed, short dotted, and short dash-dotted curves are the theoretical result obtained with Eq. 5. The $B_{1g}$ phonon was also taken into account. We show results for various $s$-wave contributions $\Delta_s/\Delta_d = 0$ (dashed); 0.1 (dotted); 0.2 (dash-dotted). The dashed, dotted, and dash-dotted curves show the calculated imaginary part of the electronic response, $\chi_{B_{1g}}''(\omega)$. (b) Subtracted spectra [20K (sc-state)-100K (n-state)] for the $B_{1g}$ polarization channel of YBCO$_{6.95}$. Before subtraction the spectra were divided by the Bose factor. The solid curve depicts the theoretical result for 10% $s$-wave contribution.

FIG. 5: (color online) (a) $B_{1g}$ Raman spectra of YBCO$_{6.95}$:Ca in the superconducting state at $T = 12$ K ($\lambda=514.5$ nm). Open circles show the experimental data. The short dashed, short dotted, and short dash-dotted curves are the theoretical result obtained with Eq. 5. The $B_{1g}$ phonon was also taken into account. We show results for various $s$-wave contributions $\Delta_s/\Delta_d = 0$ (dashed); 0.15 (dotted); 0.25 (dash-dotted). The dashed, dotted, and dash-dotted curves show the calculated imaginary part of the electronic response, $\chi_{B_{1g}}''(\omega)$. (b) Subtracted spectra [12K (sc-state)-80K (n-state)] for the $B_{1g}$ polarization channel of YBCO$_{6.95}$:Ca. Before subtraction the spectra were divided by the Bose factor. The solid curve depicts the theoretical result for 15% $s$-wave contribution.

fact which has been discussed in a previous paper by Schnyder et al. In addition, the cubic low-energy response, i.e. its $\omega/(2\Delta_0)^{3}$-behavior found for $\Delta_s = 0$ changes if $\Delta_s \neq 0$: we obtain linear correction terms which are, however, proportional to $\Delta_s/\Delta_d$, and thus barely observable.

Figure 4(b) shows the differences $(\chi_S'' - \chi''_N)$, which were obtained by subtracting the spectra at 20 and 100 K after dividing by the Bose factor. The solid line is obtained after subtracting the model function $\chi''_N$ for the normal state [see Fig. 3(a)] from the results for the SC state when $\Delta_s/\Delta_d = 0.1$ [dotted line in Fig. 4(a)]. We find that the position of the pair-breaking peak at $\sim 460$ cm$^{-1}$ (and partly its shape) is well described by our theory. This confirms that 10% $s$-wave contribution [short dotted line in Fig. 4(a)] yields an excellent description of the electronic Raman response. Furthermore, the temperature-dependence of the related $B_{1g}$ phonon is also reproduced (see below). Note that the difference between experimental and calculated data at small energies is likely to be due to elastic impurity scattering which is not taken into account in our theoretical model. 

Finally, we return to the moderately overdoped sample. Raman data on YBCO$_{6.95}$:Ca above and below $T_c$ in several polarization geometries are displayed in Fig. 3. In the absence of specific information about the electronic band dispersions of this material, we modeled these data by scaling the magnitude of the energy gap $\Delta_d$ by the ratio of transition temperatures, keeping all other model parameters identical to those used for YBCO$_{6.95}$. The best fit was obtained for $\Delta_s/\Delta_d = 0.15$ (however the estimated error bars are about $\pm 0.05$), slightly larger than the corresponding quantity.
IV. TEMPERATURE DEPENDENT PHONON LINESHAPES

A. Anisotropic Fano profile

Several phonons in YBCO display a pronounced asymmetric lineshape suggesting a strong interaction with the electronic continuum. As shown in Fig. 6 the asymmetry is most pronounced for the 340 cm\(^{-1}\) mode. Interestingly, the phonon peak reflecting the vibration of the apical oxygen at 501 cm\(^{-1}\) exhibits a strong asymmetry for a polarization of incident and scattered light along the \(a\)-axis, whereas along the \(b\)-axis the phonon appears to be almost symmetric (see Fig. 6). Two additional modes are present in this spectral range. The mode at 440 cm\(^{-1}\) originates from an in-phase vibration of the oxygen atoms O2 and O3. Additional modes are present at about 472 cm\(^{-1}\) and 480 cm\(^{-1}\) for polarizations parallel to the \(a\)- and \(b\)-axis, respectively. As mentioned above, these modes are Raman forbidden, but correspond to IR allowed vibrations involving the Cu1-O1 chains. Due to defects (oxygen vacancies) in the Cu1-O1 chains they become Raman active.

Similar to previous temperature dependent Raman experiments on YBCO\(_{7−}\) and HgBa\(_2\)Ca\(_2\)Cu\(_4\)O\(_{10+\delta}\) we have fitted the phonons of YBCO\(_{6.95}\) by using simple Fano profiles. The solid lines in Fig. 6 are the results of fits to the experimental data using Eq. (2), the dash dotted lines correspond to the resulting fitted lineshape of the entire spectrum. The calculated profiles agree well with the measured spectra. In order to obtain estimates of the intrinsic phonon positions and linewidths, we have corrected the peak position and linewidth by the procedure used in Ref. 20. We find for the \(xx\)-symmetry, i.e., polarizations along the \(a\)-axis:

\[
\omega_{xx} = \omega_0 + \Gamma_{xx}/q_{xx} \quad \text{and} \quad \Gamma_{FWHM}^{xx} = 2\Gamma \left| \frac{1 + q_{xx}^2}{1 - q_{xx}^2} \right|
\]

where \(\Gamma\) denotes the intrinsic full width at half maximum (FWHM). For the \(yy\)-polarizations one obtains an analogous equation.

Fits to the simplified Fano profile of Eq. (2) are much less complicated than the fitting procedure to the full spectrum discussed above. This is a key advantage especially in situations in which several closely spaced phonons partially overlap as is the case, for instance, for the apical-oxygen vibration in Fig. 6. The multi-parameter, global fit [Eq. (1)] yields unstable results for these phonons. It is important to note, however, that the quantities extracted from simple Fano fits are renormalized by the electronic response function, and therefore deviate slightly from the intrinsic phonon frequency \(\omega_0\) and the bare linewidth \(\Gamma_0\) of Table I. We will compare the results of both procedures in detail at the end of the next subsection.

B. Superconductivity-induced changes in the position and linewidth

Figure 7 shows the temperature dependence of the energies and linewidths of two particular phonons, the 340 cm\(^{-1}\) and 501 cm\(^{-1}\) modes, measured with light polarization along the crystallographic \(a\)- and \(b\)-axes. The spectra were taken at temperatures ranging from 20 to 300 K. The temperature dependence of the phonon energy and linewidth in the normal state arises from anharmonic phonon-phonon interactions, i.e., the decay of a high-energy optical phonon into two phonons of lower energy with opposite momenta. For simplicity, assuming the resulting phonons to have the same energy and using Bose-Einstein statistics, this decay process leads to

\[
\Gamma_{an}(T) = \Gamma_{an}(0) (1 + 2n(\omega_p/2))
\]

where \(\Gamma_{an}(0)\) is the anharmonic decay rate at zero temperature.

In Fig. 7 both linewidth and peak position were fitted simultaneously for the temperature range above \(T_c\) (solid lines). Both of these quantities show abrupt changes at the SC transition temperature due to the opening of the superconducting gap, as previously observed in YBCO (see for example...
The SC-induced changes in the phonon linewidth and peak position (for a phonon labeled as $\mu$) can be related to changes in the phonon self–energy $\Sigma_\mu(\omega) = |g_\mu^0|^2 \Pi(\omega + i\eta)$ (with $\eta \to 0$), resulting from the interaction between the phonons and the electronic system below $T_c$. The induced frequency shifts $\Delta \omega$ are related to the real part of the phonon polarization $\Pi$ by

$$\frac{\Delta \omega}{\omega} = \frac{1}{N(0)} \lambda \text{Re}\Pi,$$

while the induced changes in the linewidth can be calculated via

$$\frac{\Delta \Gamma}{\omega} = \frac{1}{N(0)} \lambda \text{Im}\Pi.$$  

Here, $N(0)$ denotes the electronic density of states at the Fermi level and $\lambda = 2 \sum_k \sum_\mu \int \frac{d^2 k}{(2\pi)^2} |g_{\mu,0}^k|^2 F^\mu_k(\omega) \delta(\epsilon_k)$ is the dimensionless electron-phonon coupling constant. $F^\mu_k(\omega)$ denotes the spectral function for phonon $\mu$ under consideration. Taking into account screening effects, i.e., the long-range Coulomb force, the self-energy is given by

$$\Sigma_\lambda(\omega) = -\left\langle \left( g_{\mu,0}^k \right)^2 |\theta_k(\omega)| + \left( g_{\mu,0}^k \theta_k(\omega) \right)^2 \right\rangle,$$

where the angular brackets are defined by Eq. (6). The symmetry of the optical phonons is reflected in the matrix element $g_{0,0}^0$. The electron-phonon coupling of phonons of $A_{1g}$ and $B_{1g}$ symmetry are in a first approximation given by

$$g_{B_1g} = g_{B_{1g}}(\cos k_x - \cos k_y)/2,$$

$$g_{A_{1g}} = g_{A_{1g}}(\cos k_x + \cos k_y)/2.$$  

The energy of the apical oxygen phonon (501 cm$^{-1}$) is right at the gap energy and is therefore sensitive to small changes in the energy of the SC gap. The 340 cm$^{-1}$ mode is well below the SC energy gap for $T < T_c$. With increasing temperature the energy of the 2$\Delta$ gap shifts to lower energies and moves through the energy of the 340 cm$^{-1}$ mode. This explains the maximum of the linewidth at 75 K. A similar behavior was observed by Limonov et al.

Finally, we discuss the role of the $s$-wave contribution to the superconducting gap which has been introduced in Eq. (5). Our numerical results obtained with Eqs. (5)–(8) and Eqs. (11)–(14) using the fits reported in Sec. III are also displayed in Fig. 7. We compare $\Delta \omega = 0$ (dashed line) with $\Delta \omega = 3$ meV (dash-dotted line) and $\Delta \omega = 6$ meV (dotted line). We find that the results obtained with 20 percent $s$-wave contribution cannot describe our data. This is clearly visible in Figs. 7a and c which show the SC-induced changes $\Delta \omega$ in the position of the corresponding phonon. In particular, for

References: 33, 53, and also other superconducting compounds like HgBa$_2$Ca$_2$Cu$_3$O$_{10}$ and Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$

Within the error bars of $\sim 0.8$ cm$^{-1}$ in (a)-(c) and 2.0 cm$^{-1}$ in (d), the phonon peak position and linewidth of the 340 cm$^{-1}$ mode is the same for the $xx$ and $yy$ polarizations (see Fig. 7b and b). Taking the phonon positions and linewidths in the SC state as $\omega_p$ and $\Gamma_p$, the maximum change in the phonon position and linewidth is obtained from $\Delta \omega = \omega_p^0(T) - \omega_p(T = 100 \, K)$ and $\Delta \Gamma = \Gamma_p^0(T) - \Gamma_p(T = 100 \, K)$, where $T$ denotes the temperature at which the SC-induced changes occur. The additional softening below $T_c$ due to the electron–phonon interaction is about -6 cm$^{-1}$. The change in linewidth (FWHM) is $+3$ cm$^{-1}$, reflecting a broadening. For the vibration of the apical oxygen, i.e., the 501 cm$^{-1}$ mode, the corresponding shift and broadening are +3 cm$^{-1}$ and +5 cm$^{-1}$, respectively. The SC-induced changes for the 340 cm$^{-1}$ and 501 cm$^{-1}$ modes are in good agreement with previous data obtained on twinned YBCO.

FIG. 7: (color online) Frequency and line width (FWHM) obtained using Eqs. (2) and (10) versus temperature of the 340 cm$^{-1}$ mode and the 501 cm$^{-1}$ mode of YBCO$_{0.95}$ for the $xx$- and $yy$-polarization, respectively ($\lambda$=514.5 nm). Below $T_c$, a comparison between calculations for $s = \Delta s / \Delta c = 0$ (dashed line), 0.1 (dash-dotted line), and 0.2 (dotted line) is displayed. The solid line corresponds to the temperature dependence of pure phonon–phonon interaction (interpreted as Klemens decay into two phonons of equal frequencies).
the 501 cm\(^{-1}\)-mode which is known to be very sensitive to the superconducting gap \(\Delta_s\). One predicts a softening for \(\Delta_s = 6\) meV, while instead a hardening is observed. Therefore, our data imply an upper limit of \(\Delta_s/\Delta_d = 0.2\). The best agreement is obtained for 10\% s-wave admixture.

At the end of this subsection, we are contrasting our generalized Fano theory [see Eq. (1)] with the standard Fano approach described by Eqs. (2) and (10). The main difference between both approaches is the theoretical description of the electronic Raman response. While in the standard Fano approach the background is assumed either to be linear (see Fig. [6]) or to follow a square-root behavior [first term in Eq. (4)] in both the normal and SC-state, our generalized Fano theory is able to take the rearrangement of spectral weight due to the opening of the superconducting gap into account. Within our microscopic description [Eqs. (5)–(8)] it is then possible to determine the ratio \(\Delta_s/\Delta_d\). Another difference between both Fano theories concerns the asymmetry parameter \(q\): in our generalized theory it becomes \(\omega\)-dependent which allows a self-consistent description of the \(B_{1g}\)-phonon and the electronic Raman response from 80 to about 1000 cm\(^{-1}\). Thus, in other words, no specific frequency interval close to the phonon peak position [see Fano lineshape analysis in Figs. 6(a) and (b)] needs to be selected.

We have summarized our comparison in Table I in which the intrinsic frequency of the \(B_{1g}\)-phonon extracted from both Fano theories is displayed: \(\omega_0\) obtained from Eq. (1) [left part, third column], \(\omega_0\) obtained from Eqs. (2) and (10) either for a square-root \([\omega_0(\text{sqrt})]\) or linear background \([\omega_0(\text{lin.})]\) (right part). These values have to be compared with \(\omega_0\) obtained from inelastic neutron scattering experiments \([\omega_0(\text{INS})]\) (middle column). One clearly sees that the values for \(\omega_0\) obtained from our generalized Fano theory are similar to those measured in INS experiments. On the other hand, the values extracted from the standard Fano approach differ by 1.1 [\(B_{1g}(100K), \omega_0(\text{sqrt})\)] to 5.9 [\(XX(20K), \omega_0(\text{lin.})\)] wavenumbers. Note, however, that these values can be substantially modified if a specific frequency interval around the peak position of the \(B_{1g}\)-phonon is defined and the corresponding lineshape analysis is then restricted to this interval (see Fig. 6). These improved values are shown in Fig. 7. In the case of the apical oxygen vibration analyzed in Figs. 6(c) and (d), a fit with Eq. (1) is hardly possible due to the multiple peak structure between 400 cm\(^{-1}\) and 540 cm\(^{-1}\). Therefore, we have used Eqs. (2), (4), and (10). On the whole, we find characteristic differences between our self-consistent, generalized Fano theory and the standard Fano approach, but our conclusions about the ratio \(\Delta_s/\Delta_d\) are independent of the theory used provided that Eqs. (2), (4), and (10) are (strongly) restricted to an interval around the peak position of the phonon being investigated.

C. Temperature dependence of the asymmetry parameter

Figure 8 shows the temperature dependence of the asymmetry parameters \(q_{xx}^{-1}\) and \(q_{yy}^{-1}\) in Eq. (2) as obtained from the fits of the Fano profile to the experimental spectra. We discuss these data in the framework of the approximate expression

\[
q \approx \frac{T_{\sigma}/\gamma_{\sigma}}{g_{\sigma}X_{\sigma}^{0}}, \quad (15)
\]

already applied in Raman work on other high \(T_c\) superconductors.\(^{32,54,65}\) We first focus on temperatures above \(T_c\) and note that \(q_{xx}^{-1} \approx q_{yy}^{-1} \approx -0.12\) for the 340 cm\(^{-1}\) mode, whereas a pronounced difference in the asymmetry parameters along \(xx\) and \(yy\) geometries is observed for the 501 cm\(^{-1}\) mode (\(q_{xx}^{-1} \approx -0.20, q_{yy}^{-1} \approx -0.08\)). As \(g_{\sigma}\) is a materials parameter independent of the light-field configuration and \(\chi_{\sigma}^{0}\) is weakly \(\omega\)-dependent, the strong variation of asymmetry parameters must be attributed to differences in \(T_{\sigma}/\gamma_{\sigma}\). Our results are in fair agreement with the phononic and electronic Raman efficiencies (proportional to \(|T_{\sigma}|^2\) and \(|\gamma_{\sigma}|^2\), respectively) calculated in refs. 66,67 using density functional methods. For instance, the calculated \(\gamma/T_{\sigma}X_{\sigma}^{0} (\sim q_{xx}^{-1}/q_{yy}^{-1})\) in the normal state is about 1.2 and 0.65 for the 340 cm\(^{-1}\) and 501 cm\(^{-1}\) modes, respectively. The changes of the asymmetry parameters with temperature in the normal state may reflect a temperature dependence of \(\gamma_{\sigma}\), i.e. parameters in the electronic band structure as well as that of the anharmonic linewidth, but their detailed origin cannot be disentangled.

Moving to the superconducting state, we note that the only parameter in Eq. (15) expected to change significantly across
$T_c$ is the response function $\chi''$. The temperature dependence of $q_{xx}^{-1}$ and $q_{yy}^{-1}$ below $T_c$ therefore also reflects the anisotropy in the 2$\Delta$-gap. Thus we have computed the temperature evolution of the asymmetry parameters in the superconducting state, using the same model parameters described in Sec. III. The reasonable agreement with the data (Fig. 8) demonstrates the self-consistency of our approach. However, since the superconductivity-induced modification of $q_{xx}^{-1}$ and $q_{yy}^{-1}$ is relatively subtle (compared to that observed in the normal state), this analysis does not provide constraints on the ratio $\Delta_g/\Delta_d$ beyond those already discussed above.

It is interesting to note that other $A_g$ modes also reveal a temperature-dependent asymmetry. In particular, we have observed that the $B_{1g}$ mode near 115 cm$^{-1}$ is symmetric (i.e. $q^{-1} = 0$) for $T \to 0$ and its asymmetry increases monotonically to $q^{-1} = 0.25$ (not shown). Its intensity, however, is rather weak compared with the $B_{1g}$ mode (and this applies even more so to the $C_u$ mode near 150 cm$^{-1}$), so we have not used these modes further in our discussion. Their asymmetry has been analyzed in more detail in Ref. 69, however without considering a possible $s$-component to the $d_{x^2-y^2}$-wave gap.

V. SUMMARY AND CONCLUSIONS

In this work we have revisited the superconductivity-induced effects on phononic and electronic Raman scattering in detwinned slightly overdoped YBa$_2$Cu$_3$O$_{6.95}$ and moderately overdoped Y$_{0.85}$Ca$_{0.15}$Ba$_2$Cu$_3$O$_{6.95}$ single crystals, both from an experimental and a theoretical point of view. In particular, we have performed a detailed study of the in-plane anisotropies in the electronic continuum and in the phonon lineshapes and assessed their implications for an $s$-wave admixture to the $d$-wave superconducting gap. To this end, we developed and applied a formalism that treats the frequency dependence of both electronic and phononic Raman scattering on equal footing. Thus we can disentangle both parts and clarify the role of interference terms. Due to this procedure we are able to extract the intrinsic frequency $\omega_0$ of the phonon position reflecting the intrinsic electron-phonon interaction. A comparison shows that $\omega_0$ agrees well with INS data at the $\Gamma$-point. The best agreement with the Raman data was obtained by model calculations based on admixtures of 10% and 15% ± 5% $s$-wave contribution for YBa$_2$Cu$_3$O$_{6.95}$ and Y$_{0.85}$Ca$_{0.15}$Ba$_2$Cu$_3$O$_{6.95}$, respectively. This agrees with values obtained by other experimental methods$^{34,5,6,7,8}$ and confirms the previously observed trend$^{14}$ of an increase in the $s$-wave contribution with increasing doping level. Our data do not show evidence of the previously reported$^{50}$ unusual quantum interference between electronic Raman scattering from planes and chains in overdoped Y$_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{7-\delta}$.

Acknowledgments

We would like to thank R. Zeyher and J. Unterhinninghofen for helpful discussions and K. Syassen for a critical reading of the manuscript. M.B. thanks V. Hinkov, R. Merkle, and B. Baum for their help in sample preparation and A. Schulz and H. Uhlig for technical support. This work was partially supported by the International Max Planck Research School for Advanced Materials (IMPRS). A. S. thanks the Swiss NSF for its financial support and the Max Planck Institute for hospitality.

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