Screening in \((d+s)\)-wave superconductors: Application to Raman scattering

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We study the polarization-dependent electronic Raman response of untwinned \(\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}\) superconductors employing a tight-binding band structure with anisotropic hopping matrix parameters and a superconducting gap with a mixing of \(d\)- and \(s\)-wave symmetries. Using general arguments we find screening terms in the \(B_{1g}\) scattering channel which are required by gauge invariance. As a result, we obtain a small but measurable softening of the pair-breaking peak, whose position has been attributed for a long time to twice the superconducting gap maximum. Furthermore, we predict superconductivity-induced changes in the phonon line shapes that could provide a way to detect the isotropic \(s\)-wave admixture to the superconducting gap.

I. INTRODUCTION

The symmetry of the superconducting (SC) order parameter in cuprate high-\(T_c\) superconductors is now agreed to be unconventional after early intense theoretical and experimental debates.\(^1\) Phase-sensitive experiments such as corner-junction superconducting quantum interference device experiments\(^2\) and tricrystal experiments on \(\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}\) (YBCO) (Ref.\(^3\)) show that the SC order parameter undergoes sign changes in the Brillouin zone (BZ) that are consistent with the \(d_{x^2-y^2}\) symmetry. A sign change of a SC order parameter with the \(d_{x^2-y^2}\) symmetry implies the existence of four nodal points in the Brillouin zone that have been observed using momentum-resolved probes such as inelastic neutron scattering (INS) or angle-resolved photoemission spectroscopy (ARPES). Correspondingly, the simple gap

\[
\Delta_k = \Delta_0 (\cos k_x - \cos k_y)/2 \tag{1.1}
\]

with a \(d_{x^2-y^2}\) symmetry on the Brillouin zone has often been used as a starting point of a quantitative interpretation of these experiments.

Of course, a sign change in the SC gap does not preclude a gap that is more complicated than the simple \(d_{x^2-y^2}\) gap\(^1,\)\(^1\). For those high-\(T_c\) cuprates with a tetragonal crystalline structure, higher \(d\)-wave harmonics have been invoked to explain ARPES measurements of the magnitude of the SC gap.\(^4\) For the cuprate family \(\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}\), which exhibits quite strong orthorhombic distortions, one expects corrections to the \(d_{x^2-y^2}\) gap\(^1,\)\(^1\) on symmetry grounds alone. This expectation has been confirmed by several experimental methods such as ARPES studies,\(^5\) INS studies,\(^6\) and measurements of Josephson currents,\(^7,\)\(^8\) and has also been investigated theoretically.\(^9,\)\(^10,\)\(^11\)

Polarization-dependent electronic Raman scattering also probes the momentum dependence of the magnitude of the superconducting order parameter and has provided yet another piece of evidence for the \(d_{x^2-y^2}\)-wave pairing scenario.\(^12\)\(^,\)\(^13\)\(^,\)\(^14\) In particular, for tetragonal high-\(T_c\) cuprates, one finds (a) various low-energy power laws in different polarization channels that are consistent with the existence of nodal points for the gap and (b) the pair-breaking peak in the \(B_{1g}\) channel at energy twice the superconducting gap maximum \(\Delta_0\) seen by other means.\(^15\)

In this paper, we are going to investigate the consequences for polarization-dependent Raman scattering of a subdominant admixture of an isotropic \(s\)-wave component to the gap\(^1,\)\(^1\) which should be of relevance to orthorhombic high-\(T_c\) superconductors of the YBCO family. Assuming the existence of well-defined SC quasiparticles, we compute the polarization-dependent electronic Raman-scattering cross section including the effects of (i) an orthorhombic tight-binding dispersion with \((d+s)\) gap, (ii) a long-range Coulomb interaction treated within the random-phase approximation (RPA), (iii) and the effective mass approximation for the Raman vertex. We show that the pair-breaking peak in the \(B_{1g}\) channel is softened by an amount proportional to the isotropic \(s\) component to the gap in that it occurs at a lower value than the absolute maximum of the gap on the “normal-state” Fermi surface. We also show that the \(A_{1g}\) channel develops a double peak structure with the peak separation proportional to the isotropic \(s\) component to the gap. Furthermore, we compute superconductivity-induced changes in the phonon line shapes, and argue how Raman scattering on phonons allows us to extract a signature of a subdominant and isotropic \(s\)-wave component to the gap.

II. THEORY

The differential cross section in a Raman-scattering experiment for a momentum transfer \(q\) that is small compared to the extension of the Brillouin zone is proportional to \([1 + n(\omega)] \chi_\gamma'(\omega)\), where \(n\) denotes the Bose distribution, \(\omega\) the frequency of the incoming plane wave, and

\[
\chi_\gamma(\omega) = (\chi_\gamma' + i\chi_\gamma'')(\omega) \equiv \chi_\gamma(q \approx 0, \omega + i\eta) \tag{2.1a}
\]

is the linear-response function for the density operator

\[
\rho_q = \sum_k \sum_{\sigma = \uparrow, \downarrow} \gamma_k c_{k+q, \sigma}^\dagger c_{k, \sigma}. \tag{2.1b}
\]
The coupling between the SC quasiparticles, linear superpositions of the fermionic creation \( c_{\mathbf{k},\sigma} \) and annihilation \( \bar{c}_{\mathbf{k},\sigma} \) operators, and the incoming (outgoing) plane wave with the polarization vector \( \hat{\epsilon}^I(\hat{\epsilon}^O) \) is here approximated by

\[
\gamma_{\mathbf{k}} \propto \sum_{\alpha,\beta} \epsilon^O_{\alpha} \frac{\partial^2 \epsilon_{\mathbf{k}}}{\partial k_{\alpha} \partial k_{\beta}} \hat{\epsilon}^I_{\beta} \tag{2.1c}
\]

in the nonresonant limit. The “normal-state” dispersion

\[
\epsilon_k = -2t \left[ (1 + \delta_0) \cos k_x + (1 - \delta_0) \cos k_y \right] - 4t' \sin k_x \sin k_y - \mu \tag{2.1d}
\]

(see Figs. 1 and 2 in Ref. [16] combines with the gap

\[
\Delta_k = \frac{\Delta_0}{2} \left( \cos k_x - \cos k_y \right) + \Delta_s \tag{2.1e}
\]

to give the SC quasiparticle dispersion \( E_k = \sqrt{\epsilon_k^2 + \Delta_k^2} \).

Both \( \Delta_0 \) and \( \Delta_s \) represent symmetry-breaking terms that lower the symmetry from tetragonal to orthorhombic in an effective one-hand description of a single copper-oxygen plane. The \( s' \)-wave component \( \Delta_s \) is isotropic [compared with the extended \( s \)-wave admixture from Eq. (3.8)]. The Raman vertices \( \gamma_{\mathbf{k}} \) can be classified according to the irreducible representations of the symmetry group of the crystal. For a crystal with tetragonal symmetry (point group \( D_{4h} \)), the relevant symmetries are the \( B_{1g}, B_{2g} \), and \( A_{1g} \) polarizations. As we shall consider a model with subdominant orthohombic distortions, \( \delta_0 \ll 1 \), \( \Delta_s \ll \Delta_0 \), we will use, in what follows, the notation of tetragonal symmetry. If so, we can identify the \( B_{1g}, B_{2g} \), and \( A_{1g} \) channels for Raman scattering with the Raman vertices

\[
\gamma_{B_{1g},\mathbf{k}} \propto t \left[ (1 + \delta_0) \cos k_x - (1 - \delta_0) \cos k_y \right], \tag{2.2a}
\]
\[
\gamma_{B_{2g},\mathbf{k}} \propto 4t' \sin k_x \sin k_y, \tag{2.2b}
\]
\[
\gamma_{A_{1g},\mathbf{k}} \propto t \left[ (1 + \delta_0) \cos k_x + (1 - \delta_0) \cos k_y \right] + 4t' \cos k_x \cos k_y, \tag{2.2c}
\]

respectively.

The electronic Raman response is calculated in the gauge invariant form assuming that the quasiparticles interact through the long-range Coulomb potential \( V_{\mathbf{q}} = \frac{4\pi e^2}{q^2} \). For the tetragonal symmetry, the RPA for the polarization-dependent Raman response function \( \chi_{\mathbf{q}}(\omega) \) was derived in Refs. [15, 18] and [14, 19, 20, 21] and shown to be well defined in the limit \( q \to 0 \).

Its generalization to orthorhombic symmetry leads to

\[
\chi_{B_{1g}}(\omega) = \left\langle \frac{\gamma_{B_{1g},\mathbf{k}}^2}{\theta_{\mathbf{k}}^2} \right\rangle_{\omega} - \left\langle \frac{\gamma_{B_{1g},\mathbf{k}}}{\theta_{\mathbf{k}}} \right\rangle_{\omega}^2, \tag{2.3a}
\]
\[
\chi_{B_{2g}}(\omega) = \left\langle \frac{\gamma_{B_{2g},\mathbf{k}}^2}{\theta_{\mathbf{k}}^2} \right\rangle_{\omega}, \tag{2.3b}
\]
\[
\chi_{A_{1g}}(\omega) = \left\langle \frac{\gamma_{A_{1g},\mathbf{k}}^2}{\theta_{\mathbf{k}}^2} \right\rangle_{\omega} - \left\langle \frac{\gamma_{A_{1g},\mathbf{k}}}{\theta_{\mathbf{k}}} \right\rangle_{\omega}^2. \tag{2.3c}
\]

in the \( B_{1g}, B_{2g} \), and \( A_{1g} \) channels for Raman scattering, respectively. As usual, the bracket (in a box of volume \( V \))

\[
\left\langle (\cdots)_{\mathbf{k}} \right\rangle_{\omega} = \frac{1}{V} \sum_{\mathbf{k}} (\cdots) \tanh (\frac{\omega}{2T}) \left(\frac{1}{\omega + i\eta + 2E_{\mathbf{n}}} - \frac{1}{\omega + i\eta + 2E_{\mathbf{n}}} \right) \tag{2.4}
\]
denotes an average over the BZ weighted by the Tsuneto function \( \theta_{\mathbf{k}} \). The second term in Eqs. (2.3a) and (2.3c) is commonly called screening term. It can be viewed as originating from the Goldstone mode of the superconductor and ensures gauge invariance of the Raman response. For tetragonal symmetry only screening terms in the \( A_{1g} \) scattering channel are possible. That is, the screening term in Eq. (2.3a) is vanishing identically if the SC quasiparticle dispersion is of the tetragonal symmetry. In the presence of orthorhombic distortions of the YBCO type screening terms can affect the \( B_{1g} \) channels on general symmetry grounds, but are absent in the \( B_{2g} \) channel. Similarly, in orthorhombic \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x \), whose crystallographic axes are rotated by 45° with respect to the CuO bonds, screening terms arise in the \( B_{2g} \) channel, but do not affect the \( B_{1g} \) channel. In the following we shall only consider orthorhombic superconductors of the YBCO type and disregard any possible screening terms in the \( B_{2g} \) channel. The presence of screening terms in the \( B_{1g} \) or \( B_{2g} \) channels for orthorhombic superconductors has been previously reported in the literature, see Refs. [10 and 21].

**III. DISCUSSION**

We have computed numerically the Raman response using the parameters \( t = 200 \text{ meV}, \delta_0 = -0.03, t' = -0.4, \)}
systematic transport measurements of the critical current density will be presented and discussed in more detail in a forthcoming publication. However, the argument we have given above in terms of the effective Josephson coupling and the doping dependence of the Tc is consistent with the reduced critical current density. We therefore conclude that the observed trend can be understood within the framework of the Josephson proximity effect.

In summary, we have presented evidence for a phase transition with a critical temperature Tc that decreases with increasing doping. The observed trend is consistent with the Josephson proximity effect. Additional systematic transport measurements will be presented and discussed in a forthcoming publication.
with these simplifications and in the zero-temperature limit the imaginary part of the BZ averages reduce to
\[
\langle \cdots | \theta_k^n | \cdots \rangle \simeq \int \frac{kd\phi}{2\pi} \int \frac{d\phi}{2\pi} \langle \cdots | \Delta_{\phi} \delta (\omega - 2E_{k,\phi}) \rangle \\
\simeq \int_{-\delta k}^{+\delta k} \frac{dk}{2\pi} \sum_{\phi_i} \frac{\phi}{2E_{k_F} + \phi_i} |\Delta_{\phi_i}|, 
\]
where we have restricted the BZ integration to a ring $-\delta k < k < -k_F < \delta k$ around the Fermi surface, the summation runs over all $\phi_i \in \{ \phi | \omega = 2E_{k,\phi} \}$, and $\Delta_{\phi}$ denotes the derivative of the gap function $\Delta \phi$. The imaginary part of the averaged Tsuneto function $\langle \theta_k^n \rangle$ is a positive function of $\omega$ and exhibits a positive divergence whenever $\Delta_{\phi_i} = 0$, i.e., at the frequencies
\[
\omega_c^{(1)} \simeq E_{k_F,\phi_i} = 2(\Delta_0 - \Delta_s), \\
\omega_c^{(2)} \simeq E_{k_F,\phi_i} = 2(\Delta_0 + \Delta_s),
\]
as can be seen from the last line of Eq. (3.6). On the other hand, $\langle \gamma_{B_{1g}} \theta \rangle$ possesses a negative divergence at $\omega_c^{(1)}$ and a positive divergence at $\omega_c^{(2)}$, and changes its sign at $\sim \Delta_0$, since the $B_{1g}$ vertex $\gamma_{B_{1g}} \simeq \cos 2\theta$ exhibits a sign change along the Fermi surface. Any divergence in the frequency dependence of $\langle \theta \rangle$ and $\langle \gamma_{B_{1g}} \theta \rangle$ results in a step-like discontinuity in $\langle \theta \rangle$ and $\langle \gamma_{B_{1g}} \theta \rangle$, respectively, due to the Kramers-Kronig relation. Vice versa, from the absence of any step-like discontinuity in the imaginary part of the BZ averages follows the absence of any divergence in the real part of the corresponding quantity. Furthermore, it turns out that both $\langle \theta \rangle$ and $\langle \gamma_{B_{1g}} \theta \rangle$ are positive in the frequency range $0 \leq \omega \leq 2(\Delta_0 + \Delta_s)$. Taking all these observations together, we find that the first term in the $B_{1g}$ screening function (3.4) features a negative divergence at $\omega = 2(\Delta_0 - \Delta_s)$, which is compensated by a positive divergence of the second term. But then, at $\omega = 2(\Delta_0 + \Delta_s)$ both terms in Eq. (3.4) show a positive divergence, which results in a strong screening of the second peak in $B_{1g}$. At $\omega \sim 2\Delta_0$ the first term in Eq. (3.4) is vanishing, whereas the second term is negative, which yields to antiscreening of the $B_{1g}$ scattering efficiency.

It is important to note that the effects induced by a nonvanishing isotropic s-wave gap $\Delta_s$ in Fig. 2 would not occur had we assumed a subdominant extended s-wave gap such as
\[
\Delta_k = \Delta_0 (\cos k_x - \cos k_y) / 2 \\
+ \Delta_{\text{ext}} (\cos k_x + \cos k_y) / 2.
\]
Contrary to the isotropic subdominant s-wave admixture $\Delta_s$ in Eq. (2.16), the extended s-wave admixture in Eq. (3.8) only leads to a shift of the nodal line, but does not give a different absolute gap value at $(\pi, 0)$ compared to $(0, \pi)$ (see Fig. 2(d) in Ref. 16). From the above discussion one infers that the property $|\Delta_{\phi=\pi/2}| \neq |\Delta_{\phi=0}|$ is crucial for the splitting of the $B_{1g}$ pair-breaking peak as well as for the appearance of antiscreeing in the $B_{1g}$ channel. Thus, we conclude that to a first approximation an extended s-wave admixture leaves the $B_{1g}$ response unchanged, an expectation that we have confirmed by numerical calculations.

IV. SUPERCONDUCTIVITY-INDUCED CHANGES IN PHONON LINE SHAPES

In principle, the existence of a subdominant and isotropic s-wave component $\Delta_s$ to the $d$-wave SC gap can be extracted from a line-shape analysis of the $A_{1g}$ and $B_{1g}$ electronic responses in a polarization-resolved Raman-scattering experiment. However, as part of the electronic Raman signal is in general masked by phonon excitations, it might be hard to detect experimentally these changes in the line shape. On the other hand, we argue in the following that polarization-resolved Raman scattering on phonons can be used to extract a signature of $\Delta_s$.

To substantiate this point, we calculate the superconductivity-induced changes in the self-energy of optical, zone-center ($q = 0$) phonons. Theretoe, we consider a linear coupling between electrons and phonons
\[
H_{eL-ph} = \frac{1}{\sqrt{3}} \sum_{q_\gamma \sigma} g_{q, \gamma} c_{q+q_\gamma, \sigma} c_{q, \sigma} (b_{q, \gamma} + b_{-q, \gamma}^\dagger),
\]
where $b_{q, \gamma}$ and $b_{q, \gamma}^\dagger$ are the creation and annihilation operators of phonons in a given branch $\gamma$ with phonon frequency $\omega_\gamma$, respectively, and $g_{q, \gamma}$ denotes the electron-phonon coupling. The form of the electron-phonon interaction (4.1) is the one for the nonresonant electronic Raman scattering provided the effective Raman vertex is replaced by the electron-phonon vertex $g_{q, \gamma}$. Hence, within an RPA treatment of the Coulomb interactions, we find that in $q \to 0$ limit the superconductivity-induced changes in the phonon self-energy are given by (29, 30, 31)
\[
\Sigma_\gamma (\omega) = -\left\langle \left\langle \frac{g_{q, \gamma}^\dagger}{\omega} \theta_k \right\rangle \omega \right\rangle + \left\langle \frac{g_{q, \gamma}^\dagger \theta_k}{\omega} \right\rangle, 
\]
where the angular brackets are defined by Eq. (2.4). The symmetry of the optical phonons is encoded in the matrix element $g_{q, \gamma}$. The electron-phonon coupling of the $A_{1g}$ and $B_{1g}$ phonons are in a first approximation given by
\[
g_{B_{1g}} = g_{B_{1g}} (\cos k_x - \cos k_y) / 2, \\
g_{A_{1g}} = g_{A_{1g}} (\cos k_x + \cos k_y) / 2.
\]
with the electron-phonon coupling constants $g_{B_{1g}}$ and $g_{A_{1g}}$. In Fig. 3 we have numerically evaluated the superconductivity-induced changes in the self-energy $\Sigma_\gamma (\omega)$ for $B_{1g}$ and $A_{1g}$ phonons in a $(d + s)$-wave superconductor. To estimate the size of these effects we have assumed some typical values for the phonon coupling strength $g_{\gamma}$ that are in
rough overall agreement with the observed Raman shifts in YBCO. A negative (positive) Re $\Sigma_\omega(\omega)$ corresponds to a softening (hardening) of the phonon below $T_c$, whereas a negative (positive) Im $\Sigma_\omega(\omega)$ leads to a broadening (sharpening) of the phonon linewidth below $T_c$.

For $B_{1g}$ phonons [see Fig. 3(a)] we find that the real part of the phonon self-energy crosses zero around $2\Delta_0$ irrespective of the value of $\Delta_s$. However, upon inclusion of a subdominant $s$-wave gap the maxima and minima of Re $\Sigma_{B_{1g}}$ are shifted to $2(\Delta_0 + \Delta_s)$ and $2(\Delta_0 - \Delta_s)$, respectively. Hence, the maximal softening of the phonon frequency occurs at a frequency $2\Delta_s$ smaller than the gap maximum $\Delta_0$. The inset of Fig. 3(a) displays the decomposition of the real part of the self-energy Re $\Sigma_{B_{1g}}(\omega)$ into its screened Re $\Sigma_{B_{1g}}^{sc}(\omega)$ and bare parts Re $\Sigma_{B_{1g}}^0(\omega)$. The frequency dependence of these two contributions mimics that of $B_{1g}^{sc}(\omega)$ and $B_{1g}^{sc}(\omega)$, which we discussed in the previous Section. A finite $s$-wave component splits the step in Re $\Sigma_{B_{1g}}(\omega)$ into two steps located at $2(\Delta_0 \mp \Delta_s)$. In between these steps the screening contribution $B_{1g}^{sc}(\omega)$ is negative, which leads to antiscreening. Neglecting the influence of a nonzero $\delta n$, one can infer the Kramers-Kronig transform of the curves in Fig. 3(a), directly from Fig. 2 and thereby obtain the imaginary part of the self-energy Im $\Sigma_{B_{1g}}(\omega)$. Due to the $s$-wave admixture, the frequency where the maximal broadening occurs is shifted from $2\Delta_0$ to $2(\Delta_0 - \Delta_s)$.

Similar effects occur for the self-energy of $A_{1g}$ phonons [see Fig. 3(b)]. An isotropic $s$-wave component splits the step in the real part Re $\Sigma_{A_{1g}}(\omega)$ into two steps, thereby shifting the crossing point of Re $\Sigma_{A_{1g}}(\omega)$ with the zero line to lower frequencies ($\Delta_s = 3 $ meV) or to higher frequencies ($\Delta_s = 6 $ meV). Contrary to $B_{1g}$ phonons, antiscreening effects are absent in the self-energy of $A_{1g}$ phonons.

Finally, we note that the effects on the superconductivity-induced changes of the phonon self-energy induced by an isotropic $s$-wave gap are again absent for a gap of pure $d$-wave character or for an extended $s$-wave admixture such as in Eq. (3.8). The effects on the phonon self-energy reported in this paper therefore constitute a fingerprint of an isotropic $s$-wave admixture to the pairing symmetry.

V. CONCLUSIONS

We have calculated the polarization-dependent Raman response for electrons and phonons in $(d + s)$-wave superconductors. We find that the screening terms in the $B_{1g}$ channel lead to a softening of the $B_{1g}$ pair-breaking peak of the order of $2\Delta_s$, i.e., twice the value of the isotropic $s$-wave component to the SC gap. This fact calls into question the long-standing interpretation that the $B_{1g}$ pair-breaking peak is located at $\omega = 2\Delta_0$ and thus can be directly used as a measure of the $d$-wave component of the SC gap when orthorhombicity is present. Secondly, we have estimated the effects of a subdominant $s$-wave admixture on the superconductivity-induced phonon renormalizations. These effects, although comparatively small, might serve as a fingerprint of an isotropic $s$-wave admixture to the pairing symmetry.

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