Superconductive Phonon Anomalies in High-$T_c$ Cuprates

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Abstract

We consider the effects on phonon dynamics of spin-lattice coupling within the slave-boson mean-field treatment of the extended $t$-$J$ model. With no additional assumptions the theory is found to give a semi-quantitative account of the frequency and linewidth anomalies observed by Raman and neutron scattering for the $340\text{cm}^{-1}$ $B_{1g}$ phonon mode in $YBa_2Cu_3O_7$ at the superconducting transition. We discuss the applicability of the model to phonon modes of different symmetries, and report a connection to spin-gap features observed in underdoped YBCO. The results suggest the possibility of a unified understanding of the anomalies in transport, magnetic and lattice properties.

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Anomalies in the transport and magnetic properties of high-temperature superconductors, observed in both normal and superconducting states, have attracted much interest \[1\], since these phenomena are considered to be manifestations of a metallic state arising near the Mott transition due to strong correlations. Although there is as yet no general consensus on the appropriate theoretical description of this anomalous metallic state, the approach based on the \(t-J\) model \[2\] treated by the slave-boson mean-field theory \[3\] has given important clues to the understanding of both transport \[1\] and spin excitation properties \[3 - 8\]. These include the temperature-dependences of the resistivity and Hall coefficient, and the different temperature-dependences \[3 - 10\] of the shift and rate of nuclear magnetic resonance between high- and low-doping regions, especially the “spin-gap” phenomenon \[3 - 8\] first noted by Yasuoka \[9\]. There have been various experimental reports \[11 - 13\] which indicate anomalous temperature-dependences also in the frequency shift and linewidth of phonons around the superconducting transition. Further, studies by neutron scattering \[14, 15\], EXAFS \[16\], ion-channeling \[17\] and ultrasound measurements \[18\] all provide strong evidence of a link between lattice anomalies and superconductivity. Although some aspects of the phonon problem have been studied theoretically \[19\], to our knowledge there have not so far been any efforts to understand all of these interesting low-lying excitations on a unified basis.

We consider the spin-phonon coupling arising naturally within the extended \(t-J\) model of a single \(CuO_2\) layer, which has been shown \[3, 6\] at the mean-field level to give a good account of many features of the spin excitations in both \(La_{2-x}Sr_xCuO_4\) (LSCO) and \(YBa_2Cu_3O_{7-\delta}\) (YBCO). The Hamiltonian is

\[
H = -\sum_{ij} t_{ij} a_{is}^\dagger a_{js} + \sum_{(ij)} J_{ij} S_i \cdot S_j, \tag{1}
\]

where the Hilbert space is that without double occupancy, \(t_{ij}\) corresponds to the transfer integrals used to reproduce the Fermi surface, and the superexchange interaction \(J_{ij}\), which is assumed to be finite only between nearest neighbours, has been taken as a constant in previous treatments.

In YBCO, the \(CuO_2\) layer is “buckled”, by which is meant that the oxygen atoms \(O(2)\)
and \(O(3)\) lie out of the plane of the \(Cu\) atoms, as shown schematically in Fig. 1. Then \(t_{ij}\) and \(J_{ij}\) between the nearest-neighbor \(Cu\) sites have contributions linear in the magnitude of the oxygen displacement along the \(c\)-axis, \(u_i^\alpha\) (\(i\) refers to the \(Cu\) sites on the square lattice, and \(\alpha\) to either \(O(2)\) (\(\alpha = x\)) or \(O(3)\) (\(\alpha = y\)), given by \(t_{ij} = t[1 - \lambda_t(u_i^\alpha/a)]\), where \(a\) and \(\lambda_t\) are the distance between \(Cu\) sites and the coupling constant, respectively, and similarly for \(J_{ij}\) with coupling \(\lambda_J\). A microscopic estimate of \(\lambda_t\) and \(\lambda_J\) requires several steps. From the structural data of Ref. [20], the equilibrium \(O\) displacement is \(u_0 = 0.256\ \text{Å}\) (neglecting henceforth the 5% anisotropy between \(\hat{x}\) and \(\hat{y}\)); although the degree of buckling is small, \(u_0/a \ll 1\), its inclusion is crucial in providing a coupling which is strong and linear.

Following Ref. [21], the dependence on interatomic separation of the transfer integral \(t_\sigma\) \((t_\pi)\) between \(Cu: d_{x^2-y^2}\) and \(O: p_\sigma\) \((O: p_\pi)\) is given by

\[
t_\sigma = \frac{\sqrt{3}}{2} V_{pd\sigma} \left[ 1 - \frac{3}{2} \left( \frac{2u}{a} \right)^2 \right] + \left( \frac{2u}{a} \right)^2 V_{pd\pi},
\]

\[
t_\pi = \frac{\sqrt{3}}{2} \frac{2u}{a} V_{pd\sigma} \left( 1 - \frac{2}{\sqrt{3}} \frac{V_{pd\pi}}{V_{pd\sigma}} \right),
\]

where \(\frac{\sqrt{3}}{2} V_{pd\sigma} \ (V_{pd\pi})\) is the transfer integral between \(d_{x^2-y^2}\) \((d_{xz})\) and \(p_\sigma\) \((p_z)\) orbitals with separation \(d = \left( \left( \frac{1}{2}a \right)^2 + u^2 \right)^{1/2}\) along the \(\hat{x}\)-direction. Taking \(V_{pd}(d) \propto d^{-7/2}\) and writing \(u = u_0 + \delta u\), where \(\delta u\) is the oscillation amplitude, gives \(t_\sigma(u)/t_\sigma(u_0) = 1 - 2.03\delta u/a\) and \(t_\pi(u)/t_\sigma(u_0) = 1.53\delta u/a\). The final requirement is the dependence on \(t_\sigma\) and \(t_\pi\) of \(t_{ij}\) and \(J_{ij}\). For \(J_{ij}\), the perturbative expression \(J = 4 \left( t_{\sigma}^4 - 2t_{\sigma}^2 t_{\pi}^2 \right) / \Delta^2 U \) (cf. [22]) would give a large coupling constant \(\lambda = 10.4\). However, use of the lowest-order perturbation form is not well justified, and investigations of the influence of higher-order terms find for the parameters of the \(CuO_2\) layer the effective relationships \(J \propto t_{pd}^x\) with \(x \simeq 2.3\) [22], and \(t \propto t_{pd}^y\), with \(y \simeq 1.0\) [23]; the result for \(y\) requires consideration of the many inter-site transfer integrals which contribute to the hopping of a Zhang-Rice singlet. The exact values of \(x\) and \(y\), and particularly of the powers \(y'\) and \(y''\) corresponding to the extended hopping terms \(t'\) and \(t''\), are quite sensitive to both the parameter choice and to the symmetry of the phonon mode [23]. This treatment disregards changes in \(\Delta\), which we believe to be appropriate for the local, screened deformation processes under investigation. By contrast,
a $d$-dependence of $\Delta$ is required to account for the weak relation $J \propto d^{-\alpha}$, $4 < \alpha < 6$, found by pressure- and substitution-induced variation of the bond length [24], where in addition the Madelung energy is altered [25]. Mindful of these uncertainties, we proceed by taking the values $x = 2.0$, $y = 1.0$ so that $\lambda_J = 5.2$ and $\lambda_t = 2.6 = \frac{1}{2} \lambda_J$, and neglect modulation of the extended $t_{ij}$ terms for the purposes of the current study. These estimates have also been found to give good agreement with the measured isotope shift in YBCO [26], a result (deferred to a future publication) which provides an independent indication of their validity.

The terms in $H$ (Eq. (1)) describing the coupling of $u_\alpha^\alpha_i$ to the spin degrees of freedom may be rewritten in the slave-boson, mean-field approximation as

$$-\sum_i \sum_{\alpha=x,y} \left\{ t\lambda_t \left( \frac{u_\alpha^\alpha_i}{a} \right) (b_i b_{i+\alpha}^\dagger) \chi_{i,i+\alpha} + \frac{1}{8} J\lambda_J \left( \frac{u_\alpha^\alpha_i}{a} \right) \left[ \langle \chi_{i,i+\alpha}^\dagger \rangle \chi_{i,i+\alpha} + 2 \langle \Delta_{ij} \rangle \Delta_{ij} + h.c. \right] \right\},$$

where $\chi_{i,j} = \sum_s f_{i,s}^i f_{j,s}$ and $\Delta_{ij} = (f_{i,\uparrow} f_{j,\downarrow} - f_{i,\downarrow} f_{j,\uparrow}) / \sqrt{2}$. We do not consider the phonon-holon coupling vertex which is also contained in the $t$ term, because in the normal state this will vanish at $q = 0$, while in the Bose-condensed state the holon has no dynamics.

Here we will be concerned mainly with the $T$-dependence of the frequency shift and linewidth of the $340\text{cm}^{-1} B_{1g}$ mode in $YBa_2Cu_3O_7$, which is an out-of-phase oscillatory motion of only the planar oxygen atoms ($u_x^x_i = -u_y^r_i$), and is also out of phase between the planes of the bilayer [27]. This mode has attracted experimental interest because it shows the largest effects at the superconducting transition, and there are available detailed Raman [11–13] ($q = 0$) and inelastic neutron scattering [28] (also $q \neq 0$) data. We will also show results for $A_{1g}$- or $A_{2u}$-symmetric (in-phase, $u_x^x_i = u_y^r_i$) oscillations, and discuss the relevance of the model for these.

The effect of the coupling on the dynamical properties of the phonon is calculated from the lowest-order spinon correction to the phonon self-energy $\Pi_{ph}$ (Fig. 2). Second-order perturbation theory in terms of the coupling results in a frequency shift $\delta\omega = \text{Re}\Pi_{ph}$ for $|\delta\omega| \ll \omega_0$, which is given at $q = 0$ by
\[ \delta \omega = c \left( \lambda J J \right)^2 \frac{4}{N} \sum_k B_k \frac{1}{\omega^2 - (2E_k)^2} \frac{\tanh \left( \frac{E_k}{2T} \right)}{E_k} \]  

(5)

where \( c = \left( \frac{3}{4a} \right)^2 \langle u^2 \rangle = 1.18 \times 10^{-4} \), \( \langle u^2 \rangle = \frac{\hbar}{2M\omega_0} = (0.055 \text{Å})^2 \), in which \( \omega_0 = 340 \text{cm}^{-1} \) is taken for the phonon frequency of interest and \( M \) is the mass of the \( O \) atom, \( E_k \) is defined by \( E_k = \left[ \xi_k^2 + \Delta_k^2 \right]^{1/2} \), with \( \xi_k \) the spinon band energy relative to the chemical potential and \( \Delta_k = -\frac{3\sqrt{2}}{4} J \Delta (\cos k_x - \cos k_y) \) the singlet order parameter, and \( B_k \) is the form factor

\[
B_k = 2\Delta^2 \left( \gamma_k \xi_k + \frac{3J}{4} \bar{\chi} \eta_k \right)^2 \quad \text{\( B_{1g}, B_{2u} \) modes}
\]

\[
B_k = 2\Delta^2 \eta_k^2 \left( \xi_k + \frac{3J}{4} \bar{\chi} \gamma_k \right)^2 \quad \text{\( A_{1g}, A_{2u} \) modes,}
\]

(6)

with \( \gamma_k = \cos k_x + \cos k_y \), \( \eta_k = \cos k_x - \cos k_y \), \( \Delta = \langle \Delta_{ij} \rangle \) and \( \bar{\chi} \equiv \langle \chi_{ij} \rangle + \frac{2J}{3} \). One observes in Eq. (5) that \( B_k \propto \Delta^2 \), and so there is no phonon energy correction due to spin coupling in the normal state; this result is a special feature of the \( \omega \neq 0 \) phonon mode we consider at \( q = 0 \), and of the current level of approximation. The superconductivity-induced correction to the linewidth \( \Gamma \) is computed similarly from \( \text{Im} \Pi_{ph} \).

A detailed numerical analysis of the self-consistent mean-field equations has been carried out \[5\] to compute the temperature-dependences of \( \Delta \), \( \bar{\chi} \) and chemical potential for several choices of doping \( \delta \), for the transfer integrals \( t = 4J \), \( t' = -\frac{J}{6} \) and \( t'' = \frac{J}{5} \), appropriate to YBCO. With these parameters, the frequency-dependence of \( \delta \omega \), given by Eq. (5), has been evaluated numerically for several temperatures, and the results are shown in Fig. 3 for \( q = 0 \) and \( \delta = 0.2 \) at \( T/T_{RVB} = 0.2 \), where \( T_{RVB} = 0.069J \) is the onset temperature for the singlet RVB order parameter \( \Delta \). All calculations were performed with an assumed Lorentzian broadening of the spinon spectrum \( \Gamma = 0.12k_BT_{RVB} \), and are found to be quite insensitive to the value of this parameter. From the \( \omega \)-dependent denominator in Eq. (5), the frequency where \( \delta \omega \) of the \( B_{1g} \) mode changes sign is an approximate measure of the value of \( 2\Delta_k(T) \) near the \( (\pi,0) \) points, where the gap is maximal: at higher temperatures (but below \( T_{RVB} \)) the \( \omega \)-dependence of \( \delta \omega \) is qualitatively the same, but the sign-change occurs at lower frequencies. The experimental mode frequency \( \omega_0 \) for the \( B_{1g} \) mode is near, but just below, the low-\( T \) crossover, so this mode can be expected to show a maximal effect.
The linewidth broadening $\delta \Gamma$ has the form to be expected for the corresponding imaginary part, namely a peak at the crossover frequency.

The temperature-dependence of $\delta \omega$ is shown in Fig. 4(a) for $q = 0$ and $\omega/J = 0.25$, a value in reasonable quantitative agreement with the $340\text{cm}^{-1}$ mode: while modes with frequencies considerably less than $2\Delta$ have a sharp transition, those close to it show clearly that as $\omega$ approaches the maximal value of $2\Delta_k(T = 0)$, the shift in frequency occurs at a temperature somewhat below $T_{RVB}$. This corresponds well to the observations of Ref. [12], where the full frequency shift develops over a range of temperatures below the onset. Comparison with the experimental result [12] for the broadening of the $B_{1g}$ mode again shows a good agreement in sign and magnitude ($\delta \omega \simeq \delta \Gamma \simeq 0.01\omega_0$) for a frequency close to $\omega_0$ (Fig. 4(b)), while at lower frequencies (“off resonance”) $\delta \Gamma$ is suppressed. We have extended our analysis to the case where the phonon has a finite wavevector $\mathbf{q}$, finding smaller but sharper anomalies in further qualitative agreement with experiment; these calculations will be presented elsewhere. Quantitatively, the magnitude of the effects given by the model with the chosen values of $\lambda_t$ and $\lambda_J$ is within a factor of 1.5 of the Raman measurements [12] on $YBa_2Cu_3O_7$; this degree of correspondence, as well as that in $\omega_0$ above, can be regarded as satisfactory within a mean-field treatment using no adjustable parameters.

In Figs. 3 and 4 are shown not just the $B_{1g}$ mode, but also the results for a mode of in-phase $O(2)$ and $O(3)$ oscillations, which would correspond to $A_{1g}$ (Raman active, $440\text{cm}^{-1}$) or $A_{2u}$ (infrared-active, $307\text{cm}^{-1}$) symmetry. At the level of the current approximation these have negligible anomalies, a qualitative difference from $B_{1g}$ which arises from the form factors $B_k$ with $d$-wave singlet pairing; we note that extended $s$-pairing gives an immeasurably small frequency shift for both types of mode symmetry. However, experimental studies of the $A_{2u}$ mode [11], whose frequency is close to that investigated, reveal a strong shift $\delta \omega/\omega_0 \simeq 1\%$. We believe that the model cannot reproduce this result because it contains nothing to account for the charge transfer between the $CuO_2$ planes of a bilayer which accompany this mode in YBCO, and so is inapplicable. Developing the theory to include interplane charge motion [29] is beyond the basic $t$-$J$ framework, and will be pursued in a
subsequent publication.

The 193$cm^{-1} B_{2u}$ mode is an out-of-phase oscillation of planar $O$ (in-phase between the planes of the bilayer) involving little charge motion, and thus a single-layer model is expected to be valid. Because this mode is i.r.-silent, it may be studied only by neutron scattering, and investigations on $YBa_2Cu_3O_7$ have recently been performed [30]. The observed anomalies are sharp and occur close to $T_c$, with a relative magnitude $\delta \omega/\omega_0 \approx 1\%$ similar to those of the $B_{1g}$ mode, features which are indeed well described by the current model at this lower frequency. Finally, the effects studied should be strongly suppressed in the $E$-symmetric phonon modes, where $O$ displacements are parallel to the plane and so have little influence on the interaction $J_{ij}$, a feature also largely in agreement with experiment.

These results raise the interesting possibility of probing the spin-gap behavior observed in the temperature-dependence of the NMR relaxation rate by considering the frequency shifts of particular phonons in members of the YBCO class in the low-doping regime, where the rate has been found to be maximal at some value $T_0$ above the superconducting critical temperature $T_c$. There exist already several experimental reports [11–13] of anomalies in the frequency shift well above $T_c$, and near the temperature where the NMR rate exhibits a maximum, while recent, highly accurate Raman studies of phonon anomalies in underdoped YBCO compounds [31] show clearly the onset of a frequency shift at some $T_0 \approx 150K$, followed by growth of this shift as temperature is lowered, until a saturation below $T_c$. Such features may be understood on the basis of the mean-field phase diagram of the extended $t$-J model [3], in which $T_{RVB}$ is indeed higher than $T_c$ only in the low-doping region, and they would be interpreted as the onset of singlet-RVB order around $T_0$, i.e. its identification with the crossover temperature $T_{RVB}$ [4]. This result is consistent with the observation that the specific heat anomalies at $T_c$ in these cases are qualitatively different from those near optimal doping [32,33]. We suggest that the existence of two temperature scales is the most likely explanation for the contrasting low-$q$ behavior of the energy shifts in nominal $O_{6,92}$ and $O_7$ compounds observed in Ref. [28].

In summary, we have proposed a theory of spin-phonon coupling which accounts very
well for the anomalies observed in phonon modes in typical high-$T_c$ cuprates of the YBCO class, based on the mean-field approximation to the extended $t$-$J$ model. We believe that this model is a useful step in constructing a coherent theory of the spin, transport and lattice properties in the anomalous metallic state, which has a strong bearing on high-temperature superconductivity.

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**FIGURE CAPTIONS**

Fig. 1: Schematic representation of CuO$_2$ layer, showing “buckling” deformation of the equilibrium positions of O(2) and O(3) atoms out of the plane of the Cu atoms, appropriate for YBCO. $a$, $b$ and $c$ represent crystal axes.

Fig. 2: Diagrammatic representation of the lowest-order contribution to the phonon self-energy $\Pi_{ph}$ due to coupling to spinons.

Fig. 3: Phonon frequency shift $\delta \omega$ for $B_{1g}$ ($\circ$) and $A_{2u}$ ($\times$) modes at $q = 0$, as a function of frequency at $T = 0.2 T_{RVB}$. The arrow indicates the frequency whose temperature-dependence is illustrated in Fig. 4.

Fig. 4: Phonon frequency shift $\delta \omega$ (a) and linewidth broadening $\delta \Gamma$ (b) for $B_{1g}$ ($\circ$) and $A_{2u}$ ($\times$) modes at $q = 0$, as functions of temperature for mode frequency $\omega_0 = 0.25 J$. 
