Scaling of transition temperature and $CuO_2$ plane buckling in the cuprate superconductors

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A universal feature of all high temperature superconductors is the existence of a chemical composition that gives maximum $T_c$, separating so called under and over-doped region. Recently it is seen that both $T_c$ and the buckling angle of the $CuO_2$ planes goes through a maximum at the same doping level. We show that only for optimal doping concentration the Fermi surface touches the $M(0, \pi)$ point in the BZ, where the matrix element for interlayer pair tunneling amplitude is largest, so that the gain in delocalization energy by tunneling(in pairs) along the $c$ axis is largest. Buckling of the planes on the other hand modulates the separation between the planes and thereby modulates the interlayer pair tunneling amplitude. That is why both $T_c$ and buckling angle( Oxygen atom displacement out of the plane) scales the same way with doping concentration. We have calculated $T_c$ and buckling angle for various doping concentration. The agreement with experiment is remarkably good. We also point out the possible reason for large(about 1 percent) change of the buckling mode phonon frequency, across the transition temperature for optimal doping concentration, observed in neutron scattering experiments.

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I. INTRODUCTION

Since the discovery of High $T_c$ cuprates a wide variety of experiments have shown strong evidence of a link between anomalies in lattice structure and the onset of superconductivity in these materials. For a review of experiments and analysis please see [1]. Ion channeling experiments [2] show anomalous changes in the oscillation amplitudes of Cu and O atoms at and above $T_c$. Ultrasonic measurements [3] shows anomalies in elastic constants. Infrared reflectivity measurements [4] of high quality samples have shown clear connection between $c$-axis phonon modes and the charge carriers.

Recently it is seen [5] that both $T_c$ and the buckling angle of the $CuO_2$ planes goes through a maximum as the doping is varied. The buckling corresponds to a static displacement of the oxygen atoms out of the plane, with $u_{i \pm x} = -u_{i \pm y}$, where $u_{i \pm x}$ is the displacement of the oxygen atoms at $i \pm x$ along the $c$ axis, $i$ being the $Cu$ atom position.

In inelastic neutron scaterring experiments [6] it is also seen that the buckling mode phonon at 340 cm$^{-1}$ ( called $A_{1g}$ ) softens by more than 1 % across the transition temperature. On the other hand, other modes having frequency close to the 340 cm$^{-1}$ mode like $A_2$ ( 307 cm$^{-1} $ ) and $E_{2u}$ ( 347 cm$^{-1} $ ) and $B_{1g}$ ( 285 cm$^{-1} $ ) are studied in detail by inelastic neutron scattering experiments [6]. The $A_{1g}$ mode corresponds to the vibrations of the planar oxygen atoms out of the plane, and out of phase with the adjacent CuO plane(buckling mode). The $B_{1g}$ ( here the oxygen atoms in two adjacent planes vibrate in phase and out of the plane) and the $E_{2u}$ mode( here the atoms vibrate in the plane) shows much less softening ( estimated to be less than .4% and .3% respectively). This compares well with the optical data though for the $E_{2u}$ there is no optical data available to compare with.

In this paper we shall be exploring the phenomena of static buckling of the planes as well as the phonon mode softening within the interlayer tunneling mechanism of Wheatley, Hsu and Anderson(WHA) [7] and its more refined version proposed by Chakraborty et al. [8]. We will show that $T_c$ and the buckling angle varies with doping concentration the same way. This and the large softening of the buckling mode phonon can all be understood in a unifying way. The main ingredient of this mechanism is that the normal state is a non Fermi liquid and hence the single particle hopping out of planes becomes incoherent. However coherent propagation of pairs of particles becomes favourable at low enough temperatures leading to coherent pair fluctuation and the consequent superconductivity. The induced Josephson coupling between the planes, or the amplitude for pair tunneling is quadratic in $t_{\perp}$ (bare single particle hopping amplitude between the planes).

Any static displacement or dynamic fluctuations of the atoms in the unit cell that modulates the distance between the planes effectively modulates the pair tunneling amplitude between the planes. The main thesis of our paper, being, since the $A_{1g}$ mode effectively brings the planes closer together leading to a decrease of the hopping paths for the electrons between the planes. This in turn makes the Josephson coupling stronger, the electronic system gains energy by condensation and the corresponding optical phonon mode($q = 0$) frequency gets softer, leading also to an
average static buckling. We find that the greater part of the large frequency shift observed for the $A_{1g}$ mode (buckling mode) is due to the screening by interlayer pair propagation, or due pair current-phonon coupling. The $B_{1g}$ mode on the other hand, being in phase oscillations of the atoms keeps the distance between the planes unchanged and so it does not couple with the pair-current flow across the planes.

II. INTERLAYER TUNNELING MODEL

We begin by writing the gap equation from interlayer tunneling

$$
\Delta_k = T_J(k) \frac{\Delta_k}{2E_k} \tanh \frac{\beta E_k}{2} + \sum_{k'} V_{kk'} \frac{\Delta_{k'}}{2E_{k'}} \tanh \frac{\beta E_{k'}}{2},
$$

This equation can be obtained by considering two close Cu-O layers as in BI 2212 coupled by a Josephson tunneling term of the form

$$
H_J = -\frac{1}{t} \sum_k t^2 \left( c^\dagger_k c^\dagger_{-k} d_{k\uparrow} d_{-k\uparrow} + h.c. \right),
$$

where $t_\perp(k)$ is the bare single electron hopping term between the two coupled layers $c$ and $d$ and $t$ is a band structure parameter in the dispersion of electrons along the Cu-O plane. Finally, $T_J(k)$ in the right hand side of equation (1) is given by $T_J(k) = \frac{\Delta_0}{2}$. where $t_\perp(k) = \frac{\mu}{2} (\cos(kx) - \cos(ky))^2$. The dispersion of electrons along the Cu-O plane is chosen to be of the form

$$
\epsilon(k) = -2t \left( \cos k_x + \cos k_y \right) + 4t' \cos k_x \cos k_y - 2t'' \cos 2k_x \cos 2k_y,
$$

with $t = 0.25$ eV, $t'/t = 0.45$, $t''/t = 0.2$. We also choose $\epsilon_F = -0.45$ eV corresponding to a Fermi surface which is closed around the $\Gamma$ point. These choices are inspired by band structure calculations. Note that the Josephson coupling term in $H_J$ conserves the individual momenta of the electrons that get paired by hopping across the coupled layers. This is as opposed to a BCS scattering term which would only conserve the center of mass momenta of the pairs. This is the origin of all features that are unique to the interlayer tunneling mechanism. This term has a local $U(1)$ invariance in $k$-space and cannot by itself give a finite $T_c$. It needs an additional BCS type non local interaction in the planes which could be induced by phonons or residual correlations. Here we assume the inplane pairing interaction to be d-wave kind i.e, $V_{kk'} = V_0 f_k f_{k'}$ with $f_k = \cos k_x - \cos k_y$. $T_J(k)$ can be inferred from electronic structure calculations. As shown in reference 4, it is adequate to choose $t_\perp(k) = \frac{\mu}{2} (\cos k_x - \cos k_y)^2$. According to Anderson, it is the $k$-space locality that leads to a scale of $T_c$ that is linear in interlayer pair tunneling matrix element. He finds that in the limit $T_J > V_{kk'}, k_B T_c \approx \frac{\mu}{t}$ and in the other limit, $k_B T_c \approx h \omega_D e^{-\frac{\mu}{\hbar \omega_D \rho_0}}$, where $\omega_D \rho_0$ and $V_0$ are Debye frequency, density of states at the fermi energy, and fermi surface average matrix element $V_{kk'}$. The important point being that even with a little help from the in plane pairing interaction the interlayer tunneling term can provide a large scale of $T_c$. In our naive analysis with a fixed cut off $\omega_D$ we find that the $T_c$ drops when the fermi surface moves away from the $M(0, \pi)$ point in a parabolic fashion. This because of (1) the density of states is large near the $M$ points, and only for the optical concentrations, the fermi surface touches these regions, and (2) The momentum dependence of both the inplane BCS pairing interaction(d-wave kind) and the interlayer pair tunneling amplitude are such that they have large values near the $M$ also. So in a way the $T_c$ going through a maximum near the optimal doping concentration, is expected for both the interlayer tunneling mechanism and as well as for ordinary d-wave superconductor. Now we shall discuss the variation of the buckling angle with the doping concentration.

Some other feature of the WHA mechanism are, (1). The gap values near the $\Gamma - X(0, \pm \pi)$ points are very much temperature insensitive at low temperatures, and drops rapidly to zero near the $T_c$. It is also very sensitive to small doping variations, near the optimal doping concentrations, i.e as the fermi surface moves away from the $\Gamma - X$ points because it cannot then take advantage of the large pair tunneling amplitudes near those points in the BZ. (2). The gaps along the diagonal direction in the BZ($\Gamma - M$ directions) are on the other hand very sensitive to temperature variations, falling faster than BCS with temperature. It is also insensitive to small doping variations near the optimal doping concentrations. We shall discuss the possible consequences of these features of the WHA gap later.
III. BUCKLING OF COPPER OXYGEN PLANES

The interlayer pair hopping term for a bilayer system is,

$$ \sum_k \frac{t^2(k)}{t} (c_{k\uparrow}^{\dagger} c_{-k\downarrow} c_{-k\downarrow} c_{k\uparrow} + \text{h.c.}) $$

Now the interlayer hopping matrix element $t_{\perp}$ will be modulated due to the vibration out of plane of the oxygen atoms. We emphasize here again, that for the $B_{1g}$ mode the atoms all vibrate out of the CuO planes but the vibrations in two adjacent layers are in phase and hence the effective tunneling matrix element of the Zhang-Rice orbitals (a linear combination of the oxygen $p$ orbitals) from plane to plane dont change, but for the $A_{1g}$ mode $t_{\perp}$ will change. We can do a Taylor expansion of $t_{\perp}$ in small oxygen atom displacements. $t_{\perp}(u) = t_{\perp}(0) + \alpha \frac{(u)}{u}$ where $c$ is the $z$ direction layer separation and $\alpha$ is a constant of the order of one. Putting this back in the interlayer term we get the extra piece,

$$ \frac{u^2}{c^2} \sum_k \frac{t^2(k)}{t} (c_{k\uparrow}^{\dagger} c_{-k\downarrow} c_{-k\downarrow} c_{k\uparrow} + \text{h.c.}) + \frac{2\alpha u}{c} \sum_k \frac{t^2(k)}{t} (c_{k\uparrow}^{\dagger} c_{-k\downarrow} c_{-k\downarrow} c_{k\uparrow} + \text{h.c.}) $$

With these modifications the total effective Hamiltonian for the phonon modes corresponding to oxygen vibration out of plane, will be given by:

$$ H_{eff} = \sum_k \frac{m}{2} \omega^2_{0k} u^2 - \frac{u^2}{c^2} \sum_k \frac{t^2(k)}{t} (\langle c_{k\uparrow}\rangle c_{-k\downarrow} c_{-k\downarrow} c_{k\uparrow} + \text{h.c.}) - \frac{2\alpha u}{c} \sum_k \frac{t^2(k)}{t} (\langle c_{k\uparrow}\rangle c_{-k\downarrow} c_{-k\downarrow} c_{k\uparrow} + \text{h.c.}) \quad (3) $$

where we have taken the anomolous averages. As we can see that the second term renormalises the frequency of the buckling mode phonon, and the term linear $u$ shifts the harmonic oscillators without affecting its energy(this will correspond to static displacement of the oxygen atoms causing buckling of the CuO$_2$ planes).

The renormalised frequency of the buckling mode phonon $\omega_{0k}$ and the static displacement of the oxygen atoms out of the plane $u_0$ is given by,

$$ \omega^2_{0k} = \omega^2_{0} - \frac{2A}{m \omega^2_{0k} c^2} \quad ; \quad u_0 = \frac{2\alpha c A}{m \omega^2_{0k} - A} . \quad (4) $$

where, $A = \sum_k \frac{t^2(k)}{t} (\langle c_{k\uparrow}\rangle c_{-k\downarrow} c_{-k\downarrow} c_{k\uparrow} + \text{h.c.})$

We take $\omega^2_0$ to be equal to 340 cm$^{-1}$ or 10.2 THz for $k = 0$. In Fig: 1 and 2, we show the doping variation of both the superconducting $T_c$ and the buckling angle $\theta = \tan^{-1}(2u_0/a)$ where $a$ is the lattice spacing(Cu-O-Cu distance). We see that both $T_c$ and angle $\theta$ scales the same way with the doping concentration. As we have seen before, due to the momentum dependence of the pair tunneling amplitude at the optimal doping concentration, large number of pairs can delocalize by tunneling from plane to planes(thereby reducing the kinetic energy). The buckling mode phonon helps in this process. That is why the buckling mode phonon frequency softens and also develops a static buckling distortion.

IV. PHONON MODE SOFTENING

Now let us take a look at the phonon frequency softening below $T_c$ for different phonon modes. We shall specifically look at the $A_{1g}$ and $B_{1g}$ modes( corresponding to out of phase and inphase vibrations of the oxygen atoms at adjacent bilayers).

Superconductivity induced change in frequency and linewidths have been used to estimate the superconducting gap function. This is accomplished by determining the phonon self energy changes that occur below the $T_c$. The imaginary part of the phonon self energy is proportional to the phonon linewidth (inverse lifetime) which is measured as a function of temperature. If the phonon energy is less than twice the gap energy, it cannot decay by pair breaking and thus a decrease in linewidth below $T_c$ can result because of a decrease in the number of available decay channels. If the phonon energy is greater than the gap, it has sufficient energy to dissociate an electron pair and the number of possible decay routes can increase below $T_c$, resulting in an increase in linewidth.

The real part of the self energy is proportional to the phonon energy and the superconductivity induced changes are determined from a measurement of the phonon frequency as a function of temperature below $T_c$. Qualitatively one
expects that, for phonon energies below or near the gap, the electron phonon interaction will push the frequencies to smaller values. Conversely for larger energy phonons (\( \omega > 2\Delta \)) are expected to undergo a hardening.

The phonon self energy, assuming a polarisation bubble with no vertex correction is given by,

\[
\sum_{\gamma}(q, i\nu_n) = \frac{T}{N} \sum_{k} \sum_{m} |\lambda_{\gamma}(k, k + q)|^2 \text{Tr}(\tau_3 G[k + q, i(\omega_n + \nu_n)]\tau_3 G(k, i\omega_n))
\]  

(5)

where \( \tau_3 \) is a pauli matrix, \( G(k, i\omega_n) \) is the fully interacting green’s function, \( \omega_n (\nu_n) \) is the fermion(boson) Matsubara frequencies, and \( \lambda_{\gamma}(k, k + q) \) is the electron phonon matrix element for scattering an electron of momentum \( k \) to \( k + q \) with momentum transfer \( q \) to or from a phonon with branch index \( \gamma \). The standard electron phonon interaction being,

\[
H_{e-ph} = \sum_{k, q, \gamma, \sigma} \lambda_{\gamma}^{k}(q) c_{k-q, \gamma, \sigma}^{\dagger} c_{k, \sigma} (a_{q, \gamma}^{\dagger} + a_{-q, \gamma})
\]  

(6)

In BCS theory the superconducting Green’s function is,

\[
G(k, i\omega_n) = -\frac{i\omega_n + \epsilon_k \tau_3 + \Delta_k \tau_1}{\epsilon_k^2 + \omega_n^2 + \Delta_k^2}
\]

Substituting this in the earlier equation and taking \( q = 0 \) we get,

\[
\sum_{\gamma}(i\nu_n) = -\frac{4}{N} \sum_{k} \tanh(\frac{E_k}{2T}) \frac{\Delta_k^2 |\lambda_{\gamma}^{k}|^2}{[2E_k(2E_k)^2 + \nu_n^2]}
\]

where \( E_k = (\epsilon_k^2 + \Delta_k^2)^{1/2} \) and \( \lambda_{\gamma}^{k} = \lambda_{\gamma}(k, k) \). Raman shift is the difference between the superconducting and normal state self energies, where normal state self energy can be extracted from the above equation by putting \( \Delta_k = 0 \). One still has to analytically continue \( (i\nu_n \rightarrow \nu + i\delta) \) and the real part describes the frequency shift and the imaginary part describes the change in phonon linewidth upon entering the superconducting state.

The simplest method of evaluating it, is to numerically evaluate it on the lattice after solving the self consistent gap equation. We have done so in this paper. We borrow the estimates of the electron phonon coupling function from the estimates by Normand et al.\cite{ref1}, i.e. \( t(u) = t(0)[1 - 2.03 \frac{u}{a}] \) where \( t \) is the inplane hopping matrix element, \( u \) is the small oscillation amplitude of the oxygen atoms out of the plane, and \( a \) is the lattice parameter. Also for the \( A_{1g} \) mode symmetry \( (u_x = -u_y) \) and for the \( B_{1g} \) mode \( (u_x = u_y) \). The electron phonon coupling function \( \lambda_{k,k} \) for \( A_{1g} \) and \( A_u \) phonons will be \( \lambda_{kk} = 4.06 \frac{t(0)}{a} \sqrt{\frac{1}{2m}} \gamma(k) \), where \( \gamma(k) = \cos k_x \pm \cos k_y \) for \( A_{1g} \) and \( B_{1g} \) phonon mode. The constants \( m \) and \( \omega \) are the mass of the oxygen atom and the bare phonon frequencies \( (q = 0) \) in the normal state for the \( A_{1g} \) and \( B_{1g} \) (340 cm\(^{-1}\) and 307 cm\(^{-1}\)) modes. This contribution to phonon frequency softening is due to the screening by the charge carriers in the planes. We have already calculated the phonon mode softening due to interlayer pair hopping process (additional screening due to pair tunneling process).

Both the contributions that shifts the phonon frequencies, the usual self energy contribution and the screening effect due to the pair propagation along the \( c \)-axis are shown as a function of temperature for the \( A_{1g} \) and \( B_{1g} \) phonon modes in Figs 3 and 4.

For the \( B_{1g} \) phonon the second contribution will be missing and is plotted in Fig. 4. We see that the pair tunneling contribution is much larger than that of screening by inplane charge carriers. Secindly from the form of electron phonon coupling function \( \lambda_{k,k} \) as well as the pair hopping contribution \( (k \) dependence of \( t_{\perp}(k) \)), it is clear that, the contributions to the frequency renormalisation of \( A_{1g} \) and \( B_{1g} \) phonons comes mostly from the \( M(0, \pi) \) points, and from the points along the diagonal respectively. This in the light of the WHA gap nature would mean that the change in frequency across the \( T_c \) will be more abrupt for the \( A_{1g} \) phonon, while \( B_{1g} \) frequency change will be gradual. At lower temperatures \( A_{1g} \) frequency will very sensitive to small doping variations near the optimal filling, decreasing as the fermi surface moves away from \( T_c \).

In conclusion, we find that, both scaling of \( T_c \) and the buckling angle with doping variation and large frequency softening of the buckling mode phonon compared to other optical modes of comparable frequencies, can be understood with the WHA mechanism.
is completely disordered on large length scales (i.e. a spatial variation of buckling angle unlike what we have considered). It is possible that this series of materials have a lot of unrelaxed structural disorder.

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Fig.1 Transition temperature versus doping concentration in the interlayer tunneling model.

Fig.2 Transition temperature versus doping concentration in the interlayer tunneling model.

Fig.3 Frequency versus Temperature of $A_{1g}$ phonon for three different chemical potentials($\mu = -457.0,-450.0,-440.0$ meV, i.e doping concentrations of 0.19, 0.18, 0.16). The corresponding $T_c$’s are 92.5, 88.0 and 85.0 degrees. The upper three curves shows the screening of inplane carriers(the bare polarisation bubble) , the middle ones are the contributions due to interlayer tunneling, and the bottom ones are the total contributions.

Fig.4 Frequency versus Temperature of $B_{1g}$ phonon for three different chemical potentials($\mu = -457.0,-450.0,-440.0$ meV, i.e doping concentrations of 0.19, 0.18, 0.16).
Buckling angle in degrees vs. Doping concentration
Phonon frequency (q=0) in meV vs Temperature in degree Kelvin