Competition between Superconductivity and Charge Stripe Order in High T_c Cuprates

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Members of the high T_c curapte family (with defect free CuO_2 planes) are suggested to have an '*Intrinsic single layer superconducting* T_c ', larger than the experimentally observed T_c , at a given hole concentration. This difference occurs to varying degrees among the different members, due to an anomalous response of the d-wave superconducting state to self generated charge perturbations. In particular, any quasi elastic charge stripe order, arising from electron-electron and electron- lattice coupling, can suppress the large intrinsic superconducting T_c . Existing experimental results on the wide T_c variation in the single layer cuprate families, such as the low T_c ($\approx 38K, 33K$) of LSCO and Bi-2201 compared to high T_c ($\approx 95K, 98K$) of Tl-2201 and Hg-1201 , the strain induced increase in T_c (25 to 50 K) observed in LSCO are qualitatively explained by our mechanism. We predict that under proper epitaxial strain the T_c of LSCO and Bi-2201 should increase from 30's all the way upto the intrinsic $T_c \sim 90$ K, at optimal doping..

I. INTRODUCTION

The superconducting T_c of cuprates[1,2] has a very wide variation from nearly zero all the way up to 165 K among the family members at optimal doping. With improved experiments it is becoming clear that most of these variations are intrinsic and are not driven by lattice defects or dopant induced disorder. From the materials science point of view, a thorough understanding of this phenomenon may provide suggestions for reaching even higher transition temperatures; from basic physics point of view this should help us sharpen our theoretical insights on the mechanism of cuprate superconductivity.

Strong electron correlations in the copper-oxygen based narrow d-band that is believed[3] to produces superconductivity is also capable of producing charge, spin and lattice related instabilities. In fact, charge stripe order seems generic to perovskite structures; any soft, rotational and distortional modes of the octahedra can enhance the charge localization (ordering) tendency. In particular, recent experiments[4−8] and theories[9−11] provide growing evidence for certain low energy dynamical charge ordering tendency called stripes. A natural question is *Do the charge stripe correlations, when they are dynamical with low frequency, provide a new mechanism for superconductivity or do they help or fight against an existing superconductivity mechanism.* A recent numerical analysis by White and Scalapino[11] already shows that charge stripes tend to suppress pairing correlations. Various mean field theories also bring out the presence of charge stripe correlations at low doping, before the emergence of superconductivity.

The aim of the present paper is to i) propose an 'Intrinsic single CuO_2 layer superconducting T_c ' hypothesis, ii) discuss the physics behind the charge stripe formation and iii) suggest that an anomalous response of the d-wave state to low energy or quasi static charge stripe fluctuations results in a large decrease of the intrinsic one-layer transition temperature T_c^{1*} .

We base our suggestions on a comparative study of the existing experimental results on the one layer family of cuprates, some existing numerical results and our own theoretical considerations. Our major emphasis is the observation that within the one layer family the superconducting T_c varies widely from 5 K to 98 K, even though the normal state remains reasonably unchanged. While impurities and doping induced disorder can cause part of this variation, we will argue that it is an anomalous response of the d-wave state to the development of low energy dynamical charge stripes, that causes the large suppression of an intrinsic superconducting T_c^{1*} of a single CuO_2 layer.

Some of the existing experimental results on the anomalous increase of T_c of LSCO under epitaxial strain as well as some pressure dependence of T_c can get qualitatively explained by our mechanism. One of our predictions is that under a proper epitaxial strain the T_c of optimal doped LSCO as well as Bi-2201 should increase all the way upto 95 K.

II. INTRINSIC SINGLE PLANE T_c HYPOTHESIS AND EXPERIMENTAL RESULTS

We hypothesize that an ideal rigid (with immobile atoms) single layer of CuO_2 has an intrinsic superconducting transition temperature T_c^{1*} for a given hole density x. The experimentally observed large variation in T_c among the families of cuprates must mean, by the above hypothesis, that there is some other mechanism or interaction that is either decreasing or increasing the intrinsic T_c at a given doping. From theory point of view an idea of the intrinsic T_c and its x-dependence can be obtained from the early RVB mean field theory of superconductivity[12] and their modern refined versions[13,14]

The maximum T_c of single layer materials LSCO , Bi-2201 , Tl-2201 and Hg-1201 are 38, 33, 95 and 98 K respectively. In spite of the varying T_c most of the ab plane properties including the T dependent resistivity ρ_{ab} at optimal doping are nearly the same in all the four materials. A major difference that distinguishes the low T_c LSCO and Bi-2201 from the high T_c Tl-2201 and Hg-1201 are the charge stripe tendencies in the former below the spin gap temperature scale. Neutron[4], X-ray[5] and NQR[6] studies show strong charge ordering tendency in LSCO . Hall angle studies in the ab-plane of LSCO and Bi-2201 show[8,15] a temperature dependence of the form $T^{2-\alpha}$ (with $\alpha \approx 0.3$) as opposed to T^2 in Tl-2201 [17] and most likely in Hg-1201 . The $T^{2-\alpha}$ dependence is easily interpreted as arising from the scattering of spinons by the incipient dynamical stripes[16].

We assume that the T_c of Tl-2201 is closest to the intrinsic T_c^{1*} . This is because experiments indicate that there is very little inter layer pair tunneling contribution to the superconducting condensation energy, suggesting that the mechanism of superconductivity that is operative is dominantly a single CuO_2 layer phenomenon. Thus we identify the maximum $T_c \approx 95$ with the intrinsic $T_c^{1*} = 95$ at optimal doping.

Another experimental input that we use and also explain is the doubling of critical temperature from 25 K to 50 K in $La_{1.9}Sr_{0.1}CuO_4$ under compressional epitaxial strain observed by Locquet et al.[18].

Numerical experiments on large U Hubbard model have brought out enhanced pair susceptibility with $d_{x^2-y^2}$ symmetry as well as charge stripe susceptibility. The work by White and Scalapino[11] indeed shows how in the presence of t' term the stripes are suppressed at the expense of superconductivity. The wave vector at which the charge susceptibility is largest in numerical studies[19] is close to the experimentally seen wave vectors, $4\pi(x,0)$ and $4\pi(0,x)$, where x is the hole density.

III. MECHANISM OF CHARGE ORDER

Central to stripe order at low doping is the interplay between one electron kinetic energy (t) and the super exchange energy (J), and to some extent short range repulsion energy. In the conducting state the super exchange term favours short range singlet bond formation and hole delocalization frustrates this tendency and vice versa. In the cuprates the parameters t and J have comparable values of $\sim$ 0.2 and 0.15eV. The doped 'holes' have an option to get delocalized isotropically in the plane and remain less coherent - some what like a collection of Brinkman-Rice holes with enhanced short time scale spin scrambling. This state will pave the way for the d-wave superconducting state at low temperatures. The other option is to delocalize slightly preferentially along a subset of parallel chains and become more coherent at least in one direction - this will be the charge stripe state. In the charge stripe state the enhanced quasi one dimensional motion of holes reduces the spin scrambling.

Very recent ARPES[7] brings out the high frequency (corresponding to a scale of about 0.5 eV) character of the stripe fluctuations. The stripe correlations quantum melt as we come to low frequencies and eventually disappear, leaving way for superconductivity. In some compounds perhaps it co-exists with superconductivity. We suggest that the stripe order that show up in mean field theories[20] should disappear or be strongly suppressed with proper inclusion of quantum fluctuations.

From many body theory point of view the high frequency insulating stripe correlations are analogue of *the strong short range crystalline order in liquid* He^3 *below the melting line at very low temperatures*. In liquid He^3 the crystalline correlation does not have a quasi one dimensional character. From the point of view of gaining delocalization energy the 'crystalline correlations' in the CuO_2 planes have quasi one dimensional character. These 'crystalline correlations' are the Mott insulating stripes that separate the more conducting 'chains'. In a sense the insulating stripes are short time scale memory of the parent Mott insulating state.

From the point of view of gain in kinetic energy the conducting stripe should be nearly 'quarter' filled, as also seen experimentally. The reason for this is that the holons along a conducting chain maximise their kinetic energy when they form a half filled band. This is easily seen in the limit $J \to 0$ where the spinless fermions maximise their kinetic energy gain in a chain only at half filling. Notice that in the one dimensional t-J model a quartered filled band of electrons corresponds to a half filled band of holons. Thus the major factor that determines the ordering wave vector is the mean distance between the nearly 'quarter' filled charged stripes at a given doping. Under this assumption the ordering wave vectors become $4\pi(x,0)$ and $4\pi(0,x)$.

Another important point we wish to make is that the high frequency stripe fluctuation can be frozen out if there is sufficient help from phonons. This is already visible in various numerical results where the boundary conditions stabilize stripes in the ground state. If we have a sufficiently soft phonon corresponding to the ordering wave vector and also strong coupling of the charge density fluctuations to this phonon, the lattice distortion and the stripe order can support each other self consistantly leading to quasi static or even true long range order.

We have done a simple RPA calculation coupling the hole density fluctuations to phonon and find that for the realistic parameters the freezing of the stripe order can occur. Our modelling involves coupling of the holon density of the spin gap phase to phonons. Assuming a model dispersion for the holons, the RPA expression for phonon frequency$\hbar\omega_Q$ at the nesting wave vector Q is given by the self consistent equation:

$$\hbar\omega_Q \approx \hbar\omega_0 - \frac{\lambda^2}{N}\sum \frac{1}{\hbar\omega_Q + 2\hbar v_c k} \quad (1)$$

where $\hbar\omega_0$ is the bare phonon frequency, λ is the holon-phonon coupling constant and v_c is the holon velocity close to the chemical potential. The second term in the above expression is the particle-hole susceptibility for the holons. In the spin gap phase the holons do not have an extended fermi surface, and hence the particle-hole susceptibility does not lead to a logarithmic divergence. However, the smallness of the holon velocity increases the pair susceptibility and equation (3) exhibits a zero frequency solution, and the onset of charge stripe instability, at a non zero critical value of $\lambda_c \sim \sqrt{\hbar\omega_0\epsilon_F \frac{v_c}{v_F}}$. This large value is only an upper bound, as we have not introduced important short range repulsion among the quasi particles that further encourage density wave instabilities. A modification of the susceptibility in the equation for the phonon frequency (equation 1) by a renormalized susceptibility $\chi(q.\omega) \rightarrow \frac{\chi(q,\omega)}{1-U\chi(q,\omega)}$ shows large reduction of the critical coupling constant. Here U is an effective short range repulsion among the quasi particles.

IV. HOW DOES CHARGE ORDER INSTABILITY SUPPRESS SUPERCONDUCTING T_c ?

In many body systems, when two or more instabilities occur simultaneously, they need not support each other. There may be some repulsive couplings between the two orders that will supress one in the presence of the other. This is a kind of level repulsion between the two different broken symmetry vacuua.

In this section we bring out a mechanism that brings out this repulsion quantitatively. It is well known that the response of d-wave superconducting state to static disorder is strong and anomalous, analogous to the response of s-wave superconductor magnetic impurities. Zn doping in cuprates illustrates this well. This makes the superconducting T_c very sensitive to development of low frequency periodic or random modulation of charges.

In the case of d-wave, unlike the s-wave, a finite density of scatterers leads to a finite quasi particle density of states[21,22] at the chemical potential. These low energy states emerge by depleting the d-wave condensate through a quantum interference effect arising from the changing sign of the d-wave amplitude in k-space. An approximate expression[23,24] for the resulting reduction in T_c is

$$k_B \Delta T_c \approx \frac{n_d}{4N(E_F)} \sin^2(\delta) \quad (2)$$

where n_d is the density of scatterers and δ is the phase shift arising from the individual scatterers. It is remarkable that the reduction in T_c is universal in the sense it is independent of the bulk T_c !

We use this formula to understand the sensitivity of T_c of the CuO_2 layer to the development of low frequency charge stripe correlations. *The low frequency charge stripe correlation provides a self consistent 'random' potential for the electrons.* The above formula is applicable as long as the charge stripe fluctuation frequency $\nu << \frac{\Delta(0)}{\hbar}$. The typical gap value $\Delta(0)$ is of the order of 20 meV in cuprates and the charge stripe correlation that has been seen by NQR, X-ray and neutron scattering are observed at much lower frequencies. Because of its dynamical character, as well as the non fermi liquid character of the normal state, the incipient holon density fluctuations affect the normal state transport less dramatically than they do T_c .

We can use the above formula to get an estimate of the reduction in T_c , and substitute in equation (2) for n_d the rms amplitude of the charge stripe order parameter, and for δ the phase shift experienced by the rest of the electrons due to the presence of a quasi static charge order over a length scale of the order of the coherence length of the cooper pair.

If we assume a phase shift of $\frac{\pi}{2}$, an rms amplitude of 1% leads to a reduction in T_c of about 50 K. The experimentally observed T_c in LSCO as well as Bi-2201 at optimal doping is reduced from our definition of intrinsic value of 95 K by about $60K$. The above is a very rough estimate, but tells us that T_c can get reduced considerably.

From experiments we can infer that the charge stripe order can not bring the T_c lower than 33 K at optimal doping, and perhas superconductivity can co-exist with charge stripe order at these low temperatures.

Our conclusion can be introduced as a phenomenological term in the Ginzburg Landau free energy as a coupling between the charge stripe order parameter $\phi(\mathbf{r})$ and the d-wave order parameter $\psi(\mathbf{r})$:

$$H_{int} \approx g \int d\mathbf{r} \phi^2(\mathbf{r})\psi^*(\mathbf{r})\psi(\mathbf{r}) \quad (3)$$

Since $g \approx \frac{\sin^2\delta}{4N(E_F)}$ is positive the charge stripe fluctuation always decreases T_c .

V. WHY DO LSCO AND Bi-2201 YIELD TO CHARGE STRIPE INSTABILITY IN CONTRAST TO Tl-2201 ?

We suggest that the enhanced charge stripe tendency (seen experimentally) in LSCO and Bi-2201 is responsible for their low transition temperatures. This means that phonons in LSCO and Bi-2201 corresponding to the nesting wave vector are softer and strongly coupled to the electrons in comparison with Tl-2201. We can justify this statement using a recent experiment on LSCO and in the process also explain the experimental result itself.

Locquet et al.[18] have found a remarkable result that under a compressive epitaxial strain the superconducting

Tc of $La_{1.9}Sr_{0.1}CuO_4$ is increased from 25 K to 50 K, while the ab plane resistivity remains unchanged above the spin gap phase temperature. We argue that the compressive epitaxial strain hardens the octahedral rotational and distortional phonon mode corresponding to the stripe order. In the process the charge stripe instability is removed and most of the latent superconducting order brought back.

Our explanation of the anomaly observed Locquet et al. also gives us a clue as to why Tl-2201 escapes the charge stripe instability. A closer look at the structure of Tl-2201 reveals that the 2d octahedral layer of CuO_4 is sandwiched between two layers of $(TlO)_2$. The natural lattice parameter of the $(TlO)_2$ is likely to be different from that of the 2d octahedral layer of CuO_2. Thus the CuO_2 layer is either under a compressive strain or expansional strain provided by the $(TlO)_2$ layers. This should harden the octahedral rotational and distortional phonon mode corresponding to the stripe order, there by escaping the charge stripe instability. Indeed, the ab plane lattice parameter of Tl-2201 and LSCO are 3.864 Au and 3.787 Au respectively, indicating the relative stretch and the consequent stiffness of the CuO_2 layer in Tl-2201.

In the same fashion in the one layer Hg-1201 dumb bell co-ordination of Hg and the intervening oxygen of the HgO plane seems to stiffen the CuO_2 planes there by discouraging charge stripe order. In Hg-1201 in the presence of Hg deficiency one has already seen[25] evidence of stripe tendency, in the sense of decrease of T_c around the magic hole density of $x = \frac{1}{8}$. This result can be interpretted as the beginning of the stripe tendency arising from Hg deficiency and the consequent lattice softening.

Our observation also qualitatively explains the mystery of the strong increase in T_c of oxygenated LSCO in pure state as well as in thin film epitaxy observed[26] in many experiments. Several intriguing pressure dependences of T_c [27] in a family of cuprates can be perhaps explained by our mechanism.

VI. CONCLUSIONS

In general we can say that *any lattice strain arising from sandwiching layers or from ionic radii mismatch of cation, that hardens the lattice mode corresponding to the charge stripe ordering wave vector, will disfavor charge stripe order and encourage superconductivity.* This is consistent with the finding of Attfield et al.[28] where they find a systematic increase of the optimal T_c with cation radius.

Based on our theory we also predict that one can increase superconducting Tc of LSCO and Bi-2201 from 35 K to nearly 95 K by producing appropriate epitaxial strain at optimal doping.

Ong[29] also suspects exclusion of stripe fluctuations in the superconducting state that are stabilized by epitaxial strains in some of the samples studied by transport measurements. It will be important to study systematically the correlation between the development of stripe order and the reduction in T_c as predicted by us, in Tl-2201 , Hg-1201 and Bi-2201 systems.

We will discuss the case of bilayer and tri layer in a separate paper[30], where there are interesting competition between the single layer superconductivity, interlayer pair tunneling and the charge stripe order.

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