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Article

The Origin of T_c Enhancement in Heterostructure Cuprate Superconductors

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Abstract: Recent experiments on heterostructures composed of two or more films of cuprate superconductors of different oxygen doping levels have shown a remarkable T_c enhancement (up to 50%) relative to single compound films. We provide a simple explanation of the enhancement which arises naturally from a collection of experimental works. We show that the enhancement could be caused by a structural change in the lattice, namely an increase in the distance of the apical oxygen from the copper-oxygen plane. This increase modifies the effective off-site interaction in the plane which in turn enhances the d-wave superconductivity order parameter. To illustrate this point we study the extended Hubbard model using the fluctuation exchange approximation.

Keywords: high T_c superconductivity; heterostructures; cuprates

1. Introduction

The effort to increase the transition temperature of superconductors by chemical and structural manipulations has been a major driving force in condensed matter research for quite some time. While most research in cuprate superconductors has focused on chemical doping, more recently the study of cuprate heterostructures has gained attention [1,2]. Early studies of cuprate heterostructures by Li *et al.* [3] have shown that heterostructure manipulation is a promising route to try and increase

T_c, and more recent experiments [4] shed remarkable light on the microscopic mechanisms at play in these systems.

In 2008 Yuli *et al.* [1] presented a novel technique to enhance the superconducting transition temperature (T_c) of LSCO films. The enhancement was achieved in a heterostructure of two thin layers of the same cuprate superconductor with different carrier density in the two layers. The experimental setup used $La_{2-x}Sr_xCuO_4$ (LSCO) structures where the top layer had $x = 0.35$ (overdoped) and in the bottom layer x ranged from the underdoped to the overdoped regime. In all measured heterostructures T_c was higher in the heterostructure than that of a single compound LSCO film with the same doping x as the bottom layer. The largest enhancement occurred around $x = 0.12$ where T_c of the heterostructure was 32 K, about 50% larger than the T_c of the single compound film. Similar behavior was observed in another experiment where Gozar *et al.* [2] created heterostructures from layers of metallic LSCO (overdoped) and either insulating LCO (the parent compound) or superconducting LCO (in this experiment the compound was $LaCuO_{4+\delta}$ with $\delta = 0$ for the insulator).

2. The Phenomenology of Cuprate Heterostructures

These intriguing results present a possible new direction in the exploration of high temperature superconductors. At the moment, the phenomenon of T_c enhancement is far from understood and seems to depend crucially on materials and growth method [5]. A few ideas have been put forth regarding the mechanism by which the enhancement occurs. These ideas are closely related to the different points of view on the origin of the pseudogap and its relation to the superconducting phase.

For example, if one takes the point of view that the pseudogap is related to an order parameter which competes with superconductivity then the tendency to develop such an order reduces T_c. Therefore, if in the heterostructure this tendency is suppressed superconductivity will prevail to higher temperatures. Some support for this point of view is provided by the experiment [1] in the following way. The largest enhancement occurs close to 1/8 filling. At this filling, the single compound films have a dip in the T_c *vs.* x curve. The dip is believed to be due to charge stripes which are favorable at this filling. In the heterostructures no such dip is observed.

Another possible origin of the pseudogap regime is that in the underdoped side of the superconducting dome, T_c is restricted by phase fluctuations while pairing persists up to a higher energy scale, possibly of the order of the pseudogap temperature, T^*. Taking this point of view, it has been suggested by Yuli *et al.* [1] and theoretically explored by Berg *et al.* [6] and by Goren and Altman [7], that in the heterostructure, enhancement may occur if phase fluctuations are suppressed compared to the single compound film. To suppress the fluctuations the superfluid density should be increased, making vortices more costly in energy. Higher superfluid density can be achieved by higher density of states at the Fermi level which can be provided by a metallic layer. This view is supported by the fact that the enhancement only occurs on the underdoped side of the superconducting dome.

3. The Role of Apical Oxygen

Both of the directions above are interesting and deserve further investigation. In this work, however, we point at a different direction which does not necessarily discriminate between the two scenarios

above but may lead the way to a microscopic description of the phenomenon. We are guided by both experimental evidence and relevant numerical findings. We first assume that the cause for the T_c enhancement resides *within* the copper-oxygen plane. This starting point should be contrasted with approaches that include a bilayer interface of the two materials and rely on strong hopping between the layers [6,7]. However, the inter-layer hopping strength in the cuprate materials is too weak to explain such strong variations in T_c. A quantitative measure of the interlayer hopping strength has been seen, for example, in transport properties [8,9] and optical conductivity [10]. There is no evidence to suggest that the heterostructures have stronger inter-plane coupling than that of the single compound film.

What is then the difference between the single compound film and the heterostructure? One difference may be the electronic density. The amount of doping x in the bottom layer of the heterostructure may be different from that of the starting material due to charge migration between the two layers. This leads to a "self-doping" effect which may change T_c. This effect alone, however, cannot account for the large T_c enhancement in the experiment. Doping alone can not yield a transition temperature that is significantly higher than that of the single compound film at optimal doping.

Another difference is structural; atoms move and bonds stretch/shrink to relieve strain resulting from the two layer mismatch. A recent experiment by Zhou *et al.* [11] provides evidence that structural changes are intimately related to the T_c enhancement. In these experiments heterostructures of the parent compound (LCO) and overdoped metallic LSCO are fabricated. These heterostructures also display superconductivity with an enhanced T_c relative to the single film. In order to detect *where* in the heterostructure superconductivity occurs, the experimental group introduced zinc impurities to the sample, layer by layer [12]. In-plane zinc impurities are known to suppress T_c in the cuprates by a factor of about 2. In the heterostructure their effect was significant only when introduced to one specific layer which resides one layer above the interface, on the insulating/superconducting LCO side. This leads to the conclusion that only one layer is responsible for the heterostructure's high T_c. In addition, the apical oxygen distance from each layer (d) was measured by X-ray scattering [11] and was found to vary significantly in the heterostructure, between $d \approx 2.3\mathring{A}$ and $d \approx 2.75\mathring{A}$. In contrast, both the insulator (LCO) and metal (LSCO) have a bulk apical distance of $d \approx 2.4\mathring{A}$ (as pointed out in Reference [11] and measured in Reference [13]). Therefore, the heterostructure achieves apical oxygen distances in doped LSCO, that are not reached in bulk samples. We believe that this lattice rearrangement is responsible for the T_c enhancement and motivate this point of view using a microscopic model below.

4. Model and methods

In order to discuss superconductivity in the cuprates without speculating on the pairing mechanism and the origin of the pseudogap we explore the extended Hubbard model. This model includes the hopping parameters that are relevant to the cuprates and can be fit to the observed band structure (e.g., ARPES [14]) and the strongly correlated nature of this electronic system. The Hamiltonian reads

$$\mathcal{H} = -\sum_{ij,\sigma} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + U_0 \sum_i n_{i\uparrow} n_{i\downarrow} + \frac{1}{2} V_0 \sum_{\langle ij \rangle} n_i n_j \tag{1}$$

where the indices i, j go over the lattice sites with angular brackets denoting nearest neighbors and $\sigma = \uparrow\downarrow$ is the spin label. The number of electrons at site i is denoted by $n_i = n_{i\uparrow} + n_{i\downarrow} = \sum_\sigma c^{\dagger}_{i\sigma} c_{i\sigma}$.

The numerical analysis of this model leads to d-wave superconductivity [15]. In order to see how this comes about one has to go beyond simple mean field and include spin and charge fluctuations. In this model the on-site repulsion U_0 promotes d-wave superconductivity while the off-site repulsion V_0 suppresses it. This can be shown, in the fluctuation exchange (FLEX) approximation [15]. FLEX is essentially a perturbative method where we dress the interaction vertex by a subset of diagrams representing spin and charge fluctuations, summing to all orders in perturbation theory. The renormalized interaction vertex together with the electronic Green's function, is used in the Eliashberg [16] theory to determine the strength of pairing. The analysis shows that when the off-site interaction V_0 is increased, d-wave pairing is suppressed. If V_0 is larger than some critical value a charge density wave becomes more energetically favorable than superconductivity.

Specifically, the FLEX approximation, applied to an extended Hubbard Hamiltonian [15,17,18] yields the effective interaction vertex in the singlet pairing channel

$$\begin{aligned}\Gamma_s(k,k') = \sum_{\mathbf{r},\mathbf{r}'} \Big[&\frac{3}{2}\left[V^m \cdot \chi_{sp} \cdot V^m\right]_{\mathbf{r},\mathbf{r}'}(k-k')e^{i(\mathbf{k}\cdot\mathbf{r}+\mathbf{k}'\cdot\mathbf{r}')} \\ &- \frac{1}{2}\left[V^d \cdot \chi_{ch} \cdot V^d\right]_{\mathbf{r},\mathbf{r}'}(k-k')e^{i(\mathbf{k}\cdot\mathbf{r}+\mathbf{k}'\cdot\mathbf{r}')} + \frac{1}{2}V^s_{\mathbf{r},\mathbf{r}'}e^{i(\mathbf{k}\cdot\mathbf{r}'-\mathbf{k}'\cdot\mathbf{r})}\Big]\end{aligned} \tag{2}$$

where the sum is over $\mathbf{r},\mathbf{r}' = 0, \pm\hat{x}, \pm\hat{y}$, and the momenta k, k' represent both momentum and (fermionic) Matsubara frequency $k = (i\omega_n, \mathbf{k})$. Furthermore, the spin χ_{sp} and charge χ_{ch} susceptibilities are given by

$$\begin{aligned}\chi_{sp} &= \frac{1}{\chi^{-1} + V_m} \\ \chi_{ch} &= \frac{1}{\chi^{-1} + V_d}\end{aligned} \tag{3}$$

where the bare susceptibility is

$$\chi_{\mathbf{r},\mathbf{r}'}(\mathbf{q}, i\nu_m) = -T\sum_n \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}G(k+q)G(k) \tag{4}$$

where $i\nu_m$ are bosonic Matsubara frequencies, and $G(k) = G(\mathbf{k}, i\omega_n)$ is the bare single particle Green's function. Finally, the various potential matrices are

$$V^d_{\mathbf{r},\mathbf{r}'}(q) = \begin{cases} U_0 + 4V_0\left(\cos(q_x) + \cos(q_y)\right) & ,\mathbf{r} = \mathbf{r}' = 0 \\ -V_0 & ,\mathbf{r} = \mathbf{r}' \in \{\pm\hat{x}, \pm\hat{y}\} \end{cases} \tag{5}$$

and

$$V^m_{\mathbf{r},\mathbf{r}'}(q) = \begin{cases} -U_0 & ,\mathbf{r} = \mathbf{r}' = 0 \\ -V_0 & ,\mathbf{r} = \mathbf{r}' \in \{\pm\hat{x}, \pm\hat{y}\} \end{cases} \tag{6}$$

in addition to

$$V^s_{\mathbf{r},\mathbf{r}'} = \begin{cases} 2U_0 & ,\mathbf{r} = \mathbf{r}' = 0 \\ V_0 & ,\mathbf{r} = \mathbf{r}' \in \{\pm\hat{x}, \pm\hat{y}\} \\ V_0 & ,\mathbf{r} = -\mathbf{r}' \in \{\pm\hat{x}, \pm\hat{y}\} \end{cases} \tag{7}$$

The effective singlet pairing interaction term in the action is then

$$S_{int} = -\sum_{k,k'} \Gamma_s(\mathbf{k}, i\omega_n; \mathbf{k}', i\omega_n') \left(c_{k,\uparrow} c_{-k,\downarrow}\right)^\dagger \left(c_{k',\uparrow} c_{-k',\downarrow}\right) \tag{8}$$

from which we can extract the effective U, V parameters of our model

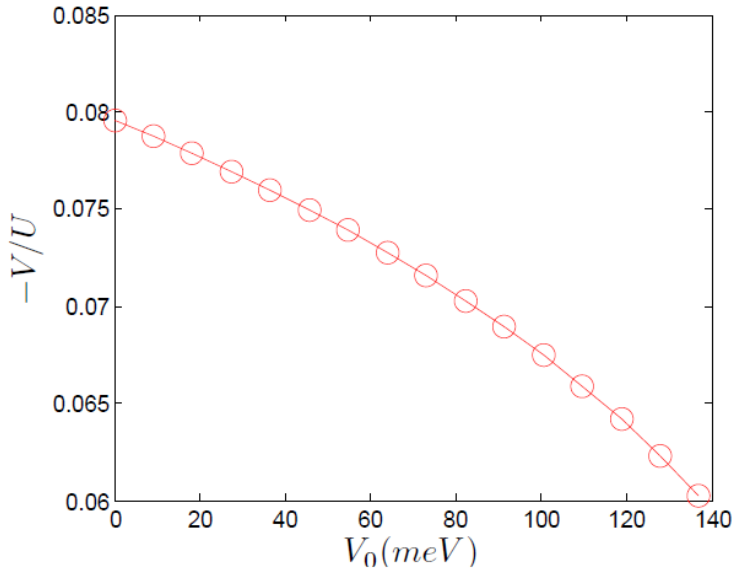
$$\begin{aligned} U &= \int_{\mathbf{q},\mathbf{p}} \Gamma_s(\mathbf{p}+\mathbf{q}/2, 0; \mathbf{p}-\mathbf{q}/2, 0) \\ V &= \int_{\mathbf{q},\mathbf{p}} e^{i\hat{x}\cdot\mathbf{q}} \Gamma_s(\mathbf{p}+\mathbf{q}/2, 0; \mathbf{p}-\mathbf{q}/2, 0) \end{aligned} \tag{9}$$

such that $\mathbf{q} = \mathbf{k} - \mathbf{k}'$ is the relative momentum, and $\mathbf{p} = \frac{\mathbf{k}+\mathbf{k}'}{2}$ is the average momentum, and we have taken all frequencies to be zero. These effective parameters can now be used in an effective Hubbard model

$$\mathcal{H} = -\sum_{ij,\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} + \frac{1}{2} V \sum_{\langle ij \rangle} n_{i\uparrow} n_{j\downarrow} \tag{10}$$

The FLEX method is perturbative and therefore is not guaranteed to work in a strongly correlated system. In addition, it takes into account only a subset of the diagrams in perturbation theory, making even the perturbative treatment approximate. The diagrams that are taken into account are those thought most significant for the pairing channels in the interaction vertex. One can hope that a model with relatively moderate interactions would be continuously connected to the strong interaction case, with the same qualitative features, even though it would be surprising if the FLEX approach would also get quantitative details right. Since our intent is to find a qualitative trend of stronger superconductivity with increasing apical distance, this limited approach should suffice.

Figure 1. The effective attractive off-site interaction V as a function of the bare repulsive, off-site, interaction V in units of the on-site repulsion U. In the calculation the values $t = 488\ meV$ and $U = 2.56\ eV$ were used. The (bare) repulsive V_0 axis is in units of meV. The values taken for the bare repulsive V_0 range from $V = 0$ to its expected value for bulk LCO as found in Reference [19].



5. Results and Discussion

The connection between the off-site interaction term and the apical oxygen distance from the Cu-O plane has been recently explored from first principles by Yin and Ku [19] and by Weber *et al.* [20]. The conclusion of these studies is that the distance of the apical oxygen from the layer is inversely related to the bare V_0. For materials with larger apical distance the off-site repulsion is smaller. Other terms in the Hamiltonian, however, are rather insensitive to this distance. The result is a d-wave superconductor with larger amplitude. This is the main point of our paper—the apical oxygen distances in the heterostructure are significantly larger than in bulk LSCO, in particular at the superconducting layer. This causes a reduction of the off-site repulsion V_0 and superconductivity is therefore enhanced.

Starting with both U_0, V_0 repulsive we calculate the effective interactions U, V as described above. We vary the strength of V_0 to capture the effect of the apical oxygen distance. The calculated renormalized interactions U and V are presented in Figure 1. Note that the effective off-site interaction is attractive. Using renormalized interaction coefficients is crucial, since superconductivity requires a pairing channel.

An intuitive understanding of the mechanism by which off-site interaction is related to superconductivity emerges from the FLEX approximation. We start with a repulsive on-site and off-site interaction in the extended Hubbard model, as appropriate for this system. However, when considering both interactions together in the vertex function the total off-site channel is attractive. This can be seen even at the level of the standard Hubbard model (without any bare off-site interaction). The Hubbard model close to half filling was studied by Scalapino in Reference [21] where it was found that the on-site repulsive interaction U_0 leads to an effective off-site attraction. This is the result of proximity to the antiferromagnetic phase at half filling which is due to double hopping processes of the order of t^2/U_0. Away from half filling there is no long range Neél order but the susceptibility is still peaked around (π, π). The contribution of such a structure to the interaction vertex is, again, strongest at this point in momentum space which leads to a large attraction on nearest neighbor sites. This can be viewed as large Friedel oscillations. Electrons on nearest neighbor sites take advantage of these oscillations through pairing in the d-wave channel.

In order to determine T_c realistically one should use the renormalized vertex in a full Eliashberg calculation as was done by Onari *et al.* [15] In the present work we calculate effective interaction parameters using the FLEX approximation, and turn to mean field theory in order to sketch the qualitative effect of the interaction on the order parameter. In the effective model the on-site interaction U is repulsive while the off-site interaction V is attractive. The bare repulsion, and the induced effective attraction combine to give an effective *attractive* off-site interaction and a renormalized on-site repulsion. When the apical oxygen distance is increased, the bare repulsion is decreased, and the overall effective interaction is more strongly attractive. We show the mean-field result for the above effective Hubbard model in Figure 2. In this figure, as the ratio between the magnitude of the attractive V and the on-site repulsion U is increased d-wave superconductivity is enhanced and the Neél order (or spin density wave) is suppressed. This result is obtained by the usual mean field decomposition of the interaction terms in

the superconducting and antiferromagnetic channels. The order parameters are found by minimizing the mean-field free energy.

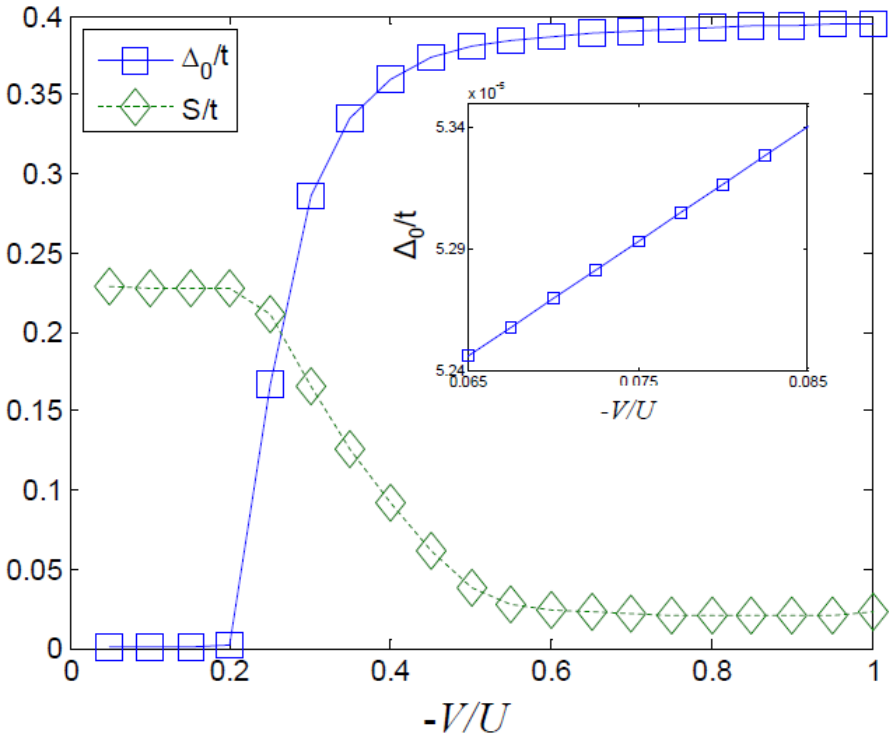
$$\begin{aligned}
\mathcal{F} &= -\sum_{\mathbf{k}\in RBZ}[E_1+E_2] - NU(\bar{n}^2 - S^2) - NV\Delta_0^2 \\
E_1 &= \sqrt{(US)^2 + (V\Delta_{\mathbf{k}})^2 + \frac{\epsilon_{\mathbf{k}}^2 + \epsilon_{\mathbf{k}+\mathbf{Q}}^2}{2} + (\epsilon_{\mathbf{k}} + \epsilon_{\mathbf{k}+\mathbf{Q}})\xi} \\
E_2 &= \sqrt{(US)^2 + (V\Delta_{\mathbf{k}})^2 + \frac{\epsilon_{\mathbf{k}}^2 + \epsilon_{\mathbf{k}+\mathbf{Q}}^2}{2} - (\epsilon_{\mathbf{k}} + \epsilon_{\mathbf{k}+\mathbf{Q}})\xi} \\
\xi &= \sqrt{(US)^2 + \left(\frac{\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}+\mathbf{Q}}}{2}\right)^2}
\end{aligned} \tag{11}$$

where RBZ stands for the sum over the reduced Brillouin zone, $\epsilon_{\mathbf{k}} = -2t(\cos(k_x)+\cos(k_y)) - \mu + U\bar{n}$ is the band dispersion which includes a chemical potential shift $U\bar{n}$, $\Delta_{\mathbf{k}} = \Delta_0\chi_{\mathbf{k}}$ with $\chi_{\mathbf{k}} = 2(\cos(k_x) - \cos(k_y))$ is the gap function, S is the Neél order parameter which folds the Brillouin zone with the wavevector $\mathbf{Q} = (\pi, \pi)$ and N is the number of sites. We have used an attractive potential to derive these equations and write $|V|$ to emphasize this point. The mean field equations read:

$$\begin{aligned}
&\frac{U}{2N}\sum_{\mathbf{k}\in RBZ}\left[\frac{1+(\epsilon_{\mathbf{k}}+\epsilon_{\mathbf{k}+\mathbf{Q}})/2\xi}{E_1} + \frac{1-(\epsilon_{\mathbf{k}}+\epsilon_{\mathbf{k}+\mathbf{Q}})/2\xi}{E_2}\right] = 1 \\
&-\frac{|V|}{2N}\sum_{\mathbf{k}\in RBZ}\chi_{\mathbf{k}}^2\left[\frac{1}{E_1} + \frac{1}{E_2}\right] = 1 \\
&-\frac{1}{2N}\sum_{\mathbf{k}\in RBZ}\left[\frac{\frac{\epsilon_{\mathbf{k}}+\epsilon_{\mathbf{k}+\mathbf{Q}}}{2}+\xi}{E_1} + \frac{\frac{\epsilon_{\mathbf{k}}+\epsilon_{\mathbf{k}+\mathbf{Q}}}{2}-\xi}{E_2}\right] = \bar{n}
\end{aligned} \tag{12}$$

where at half filling the third equation has only a trivial solution ($\bar{n} = 0$). We point out that the numbers we find from our simple mean field treatment in Figure 2, suggest that the superconductor is significant only if $\frac{|V|}{U} > 0.2$, while Figure 1 shows a value no higher than 0.08 for this ratio. However, in a full Eliashberg treatment even a small attractive off-site potential (like that of Figure 1) can cause the superconducting T_c to be non-zero [15]. We therefore conclude that our simplistic analysis gives the correct qualitative results, while leaving something to be desired in terms of quantitative results.

Figure 2. The mean field result for d-wave superconductivity order parameter (squares) and Neél order (diamonds) as a function of the ratio between the off-site interaction V and the on-site interaction U. The inset shows a zoom-in on the relevant interaction ratio for the superconducting order parameter Δ_0.



6. Suggested Experiments

Let us briefly discuss the two possible determining factors of T_c in the underdoped side, namely phase fluctuations and a competing order. In the model above only the amplitude of the d-wave pairing order parameter is considered. However, there is a lot of experimental evidence that phase fluctuations are important in the pseudogap phase. Naturally, the question arises whether phase fluctuations are reducing T_c from the temperature in which pairs are formed (which should be associated with a higher energy scale, possibly as high as T^*) or pairing and phase coherence appear at the same temperature (T_c) while the higher energy pseudogap is due to a competing order parameter. To shed more light on this issue it would be helpful to study the pseudogap regime of the heterostructure and measure T^*. We propose to perform the following experiments. Though experimentally challenging, it may be possible to measure the density of states (DOS) in the heterostructure and determine its pseudogap temperature T^*. If we adopt the point of view that T^* is due to a competing order, unrelated to superconductivity we expect T^* to be lower in the heterostructure than in the single compound films. This will reflect the competition; when the competing order is suppressed by a larger apical oxygen distance, d-wave superconductivity is enhanced. If, on the other hand T^* is the temperature at which pairing begins it should be higher in the heterostructure since T_c and T^* are related. Another important measurement is the Nernst effect which has provided evidence for phase fluctuations in the past [22] together with a

diamagnetism probe [23]. It is important to determine whether the phase fluctuations are enhanced or suppressed in the heterostructure relative to the single compound film.

In addition, we would like to propose a third experiment which would directly address the role of the apical oxygen in the T_c enhancement. Applying tensile stress along the c-axis will act to reduce the apical oxygen distance. This, according to the scenario we present here, should increase the bare off-site repulsion and reduce T_c.

7. Summary

To summarize, we provide a simple explanation of the enhancement of the superconducting transition temperature, T_c, in LSCO heterostructures compared with homogeneous, single compound films, as seen in recent experiments [1,2]. We have analyzed the extended Hubbard model in the FLEX approximation and used a heuristic mean-field analysis to assess the effect of the apical oxygen distance on the order parameter magnitude. We conclude that the off-site nearest neighbor interaction has a tremendous effect on T_c in d-wave superconductors. In particular, when this interaction becomes more negative (in other words when its bare repulsive component is reduced) the transition temperature increases. This interaction has been found to be inversely related to the apical oxygen distance from the copper oxygen planes [19,20]. It has also been seen that the apical oxygen distance is larger in the heterostructure, compared to the single compound film [11]. We conclude that it is this structural difference between the heterostructure and the single compound film that is responsible for the enhancement of T_c.

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