Phase pinning and interlayer effects on competing orders in cuprates

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Over the past few years, several exciting experiments in the cuprates have seen evidence of a transient superconducting state upon optical excitation polarized along the c-axis [R. Mankowsky et al., Nature 516, 71 (2014)]. The competition between d-form-factor order and superconductivity in these materials has been proposed as an important factor in the observed enhancement of superconductivity. Central to this effect is the structure of the bond-density-wave along the c-axis, in particular, the c-axis component of the ordering vector \(Q_z\). Motivated by the fact that the bond-density-wave order empirically shows a broad peak in c-axis momentum, we consider a model of randomly oriented charge ordering domains and study how interlayer coupling affects the competition of this order with superconductivity.

I. INTRODUCTION

The cuprate superconductors have been a topic of active research since their discovery more than thirty years ago. The past several years have brought exciting new experimental works in underdoped cuprates on transient states showing signatures of electron-electron pairing\(^6,7\). In these experiments, the system is excited via mid-infrared laser pulses which drive phonon modes of the system and can lead to quasi-static changes of the lattice structure via non-linear phonon couplings\(^7\). For times close to the pump, features reminiscent of superconductivity can be seen in the optical conductivity \(\sigma(\omega)\), e.g. a \(1/\omega\) divergence in \(\text{Im} \sigma(\omega)\) and Josephson plasma resonances\(^7\).

Also in the last few years, there has been growing interest in charge ordering phases in several cuprate families\(^7,8,9\), which have been seen to compete and co-exist with superconductivity at low temperatures. One model for such order is a density-wave instability emerging from nesting of the Fermi surface\(^10,11\). Such a model predicts the experimentally observed d-wave form factor seen in experiments\(^12\), although it predicts a diagonal \((Q, Q)\) in-plane ordering vector instead of the observed axial \((Q, 0)\) order\(^13\).

The nature of the photo-excitation employed in experiments, as well as previous theoretical works, have suggested that it is important to understand the effect of interlayer coupling. Particular its role in the competition between charge order and superconductivity to have a full understanding of the effects seen under mid-infrared excitation. In particular, one scenario suggests melting of the competing charge order\(^14,15\) via modulation of the interlayer coupling as the underlying mechanism, motivated in particular by the suppression of charge ordering peaks in X-ray coinciding with the transient pairing states\(^6\). Additionally, coupling between the planes seems to play an important role in the experimental results\(^5,19\).

One consequence of driving the c-axis phonon modes is a transient quasistatic modification of the interlayer spacing\(^6\), leading to an enhancement of the hopping between the planes. In a previous work, we showed that an increase in interlayer coupling could lead to a melting of d-form-factor density wave order in a model of stacked planes\(^12\). Furthermore, within the model, the melting of the density wave order led to a corresponding enhancement of superconductivity. These results apply to the case of order which is constant along the c-axis, while charge order with c-axis momentum \(Q_z = \pi\) is robust to changes in the interlayer coupling. We, however, note that such a configuration is to be contrasted with the empirical observation that while the c-axis momentum seen in experiments is peaked about \(Q_z = \pi\), the feature is quite broad\(^8\). Along with scanning tunneling microscopy results\(^12\), this suggests a picture of patches of in-plane order which are only weakly correlated between planes. We consider here a model where the local phase and orientation of charge order are pinned by e.g. lattice impurities or distortions. Taking this phase and orientation to be random variables, we consider the Landau theory obtained by averaging over all such regions in the system. In general, we find that when in-plane pinning of the charge order is taken into account an increase of interlayer coupling leads to a melting of charge order and an enhancement of superconductivity.

The outline of the paper is as follows. In Section II we describe the \(t - J - V\) model\(^22,24\) of the planes and consider the non-interacting susceptibility in the d-form-factor density wave (dFF-DW) channel to find the wavevector of the strongest instability. In Section III we review the mechanism for enhancement of superconductivity in the case where dFF-DW is constant along the c-axis. Then, in Section IV we consider the averaged Landau free energy of competing superconductivity and dFF-DW order and study how interlayer coupling affects the competition between the two orders. Finally, in Section V we summarize and discuss our results.

II. MODEL

In order to explore the effects of the c-axis hopping on d-form-factor density wave (dFF-DW) order, we consider a minimal model of two planes. We model each Cu-O
plane as a $t - J - V$ model\textsuperscript{14,20–22} on a square lattice, setting the lattice constant $a$ to 1. Our model takes the form $H = H_0 + H_{\text{int}}$. The free part is given by

$$H_0 = \int_k \psi^\dagger_k \left[ \left( \xi_k \hat{\Sigma}_0 + t_k \hat{\Sigma}_1 \right) \otimes \delta_0 \psi_k \right]$$

$$t_k = t_z (\cos k_x - \cos k_y)^2 / 4$$

where $\hat{\Sigma}_i$ are Pauli matrices acting in the layer space and $\sigma_i$ act in the spin space. $\xi_k$ includes hopping up to third nearest neighbor\textsuperscript{23} and $t_k$ describes the hopping between layers\textsuperscript{24,25}. To this we add the layer-local interactions

$$H_{\text{int}} = \frac{1}{2} \sum_{(i,j)} \sum_L \left( V n_{i,L} n_{j,L} + J S_{i,L} \cdot S_{j,L} \right).$$

where $V$ and $J$ are nearest neighbor Coulomb repulsion and spin exchange, respectively, and $L$ is a layer index. The $V$ term suppresses $d$-wave superconductivity and functions as a way to tune the relative strength of the two instabilities.

The nearest neighbor form of the interaction allows us to decompose the potential into a sum of factorizable potentials

$$J_{\mathbf{k} - \mathbf{k}'} = \frac{1}{2} J \sum_l f_l^f(k) f_l^f(k')$$

$$V_{\mathbf{k} - \mathbf{k}'} = \frac{1}{2} V \sum_l f_l^f(k) f_l^f(k').$$

Here, the functions $f_l^f(k)$, listed in Tab. I, form a basis of nearest neighbor in-plane interaction vertices which transform as representations of $D_4$. Since we are interested in $d$-wave superconductivity (dSC) and dFF-DW we will be focusing on the terms containing $f_1^f(k) = \cos k_x - \cos k_y$, which correspond to a $d_{x^2-y^2}$-like form factor. In real space such a form factor corresponds to the case where $x$-links and $y$-links have opposite signs. Self-energy effects due to interactions in other channels will be assumed to have already been taken into account in the free dispersion.

We may then undertake a decoupling in the dFF-DW and superconducting channels. Due to the form of the interaction, we consider only layer-local order parameters. The superconducting order is taken to be $d$-wave and constant along the $c$-axis.

![Image](https://via.placeholder.com/150)

**FIG. 1.** Maximum eigenvalue of the matrix density wave susceptibility $\Pi_\phi$ as a function of in-plane ordering vector in the Brillouin zone. The strongest instability is generically at in-plane wavevector $(Q, Q)$ with $Q \sim 1.14$ and out of plane wavevector $Q_z = \pi$.

With these restrictions, at the mean-field level, we consider the order parameters

$$\phi_L(Q) = \frac{g_\phi}{2} \sum_{k,\sigma} f_1^f(k) \langle c_{k - Q/2, \sigma, L}^\dagger c_{k + Q/2, \sigma, L} \rangle$$

$$\Delta = \frac{g_\Delta}{4} \sum_{k, L, \sigma, \sigma'} f_1^f(k) \langle c_{-k, \sigma, L} ( -i \sigma_\sigma') c_{k, L, \sigma'} \rangle$$

where $g_{\phi, \Delta} = \frac{\Delta}{14} \pm V$ and $\langle \cdots \rangle$ indicates an ensemble average.

Having defined the order parameters we can also define associated normal state susceptibilities in these channels. In particular, we define the matrix dFF-DW susceptibility

$$\Pi(Q)_{ij} = - \sum_k [f_1^f(k)]^2 \times \text{tr}_{\sigma, \sigma'} \left[ \hat{G}_0(\epsilon_n, \mathbf{k} + \mathbf{Q}/2) \hat{v}_i \hat{G}_0(\epsilon_n, \mathbf{k} - \mathbf{Q}/2) \hat{v}_j \right].$$

where $\hat{G}_0$ is the non-interacting Green’s function and $\sum_k$ includes an integral over in-plane momentum and a sum of the Fermionic Matsubara frequency $\epsilon_n$.

In order to determine the in-plane charge-ordering wavevector, we calculated the susceptibility at various values of $Q$ and compared the maximum eigenvalues. An intensity plot of the strongest instability by wavevector is shown in Fig. I. For the in-plane component, we generically find the susceptibility to be greatest for a diagonal
The leading instability has an in-plane ordering momentum which connects the ‘hotspots’, the points where the Fermi surface intersects the magnetic Brillouin zone boundary, across the edge of the Brillouin zone, e.g. the hotspots labeled 1 and 2. The symmetry of the problem allows the mean-field Hamiltonian at only hot regions 1 and 2 to be considered.

FIG. 2. The leading instability has an in-plane ordering momentum which connects the ‘hotspots’, the points where the Fermi surface intersects the magnetic Brillouin zone boundary, across the edge of the Brillouin zone, e.g. the hotspots labeled 1 and 2. The symmetry of the problem allows the mean-field Hamiltonian at only hot regions 1 and 2 to be considered.

Due to the symmetries of the problem, it is then only necessary to consider one pair of hotspots with the mean-field Hamiltonian at only hot regions 1 and 2 to be considered.

FIG. 3. Schematic phase diagram of competing d-wave superconductivity and d-form-factor density wave. V, the nearest-neighbor Coulomb repulsion, acts as a tuning parameter for the relative strength of the two instabilities.

(Q, Q) nesting wavevector as is generally the case in such models.\textsuperscript{14,27,28}

III. HOTSPOT MODEL AND THE CHARGE ORDERING INSTABILITY

As was discussed in a previous work,\textsuperscript{19} interlayer hopping leads to a curvature effect that suppresses dFF-DW that is constant along the c-axis. This can be understood by looking at an effective low-energy model of ‘hotspots’ in the Brillouin zone. We begin by considering a single Cu-O plane. Noting the importance of anti-ferromagnetic fluctuations in the cuprates, we expand the Hamiltonian about ‘hot-spots’ where the Fermi surface is nested with anti-ferromagnetic wave-vector (π, π), i.e. where the Fermi surface intersects the magnetic Brillouin zone boundary as depicted in Fig. 2. This leads to a low-energy Hamiltonian

\[
H = \sum_{\vec{k},i} \xi_{i,\vec{k}} c_{i,\vec{k},\sigma}^{\dagger} c_{i,\vec{k},\sigma} + g_{abcd} \sum_{\vec{k},\vec{p}} \left[ c_{1,\vec{k},a}^{\dagger} c_{2,\vec{k},d}^{\dagger} \delta_{3,\vec{p},b} c_{4,\vec{p},c} - c_{1,\vec{k},a}^{\dagger} c_{2,\vec{k},d} c_{4,\vec{p},c} - c_{3,\vec{p},b} c_{1,\vec{k},a}^{\dagger} \right], \tag{7}
\]

where the interaction is

\[
g_{abcd} = -\frac{1}{4} J_{K} \bar{\sigma}_{ab} \cdot \bar{\sigma}_{cd} - V_{K} \delta_{ab} \delta_{cd}, \tag{8}\]

k and p are now the deviations from the hotspots, a − d are the electron spin indices, and i is now a hotspot index (e.g. as shown in Fig. 2). Inversion symmetry allows us to restrict attention to half of the hotspots in the Brillouin zone. We then undertake a simultaneous mean-field decoupling in the dSC and dFF-DW channels. Due to the symmetries of the problem, it is then only necessary to consider one pair of hotspots with the mean-field Hamiltonian

\[
\hat{H}_{MF}(\vec{k}) = \begin{pmatrix}
\xi_{1}(\vec{k}) & \bar{\phi} & \Delta & 0 \\
\bar{\phi} & \xi_{2}(\vec{k}) & 0 & \Delta \\
\bar{\Delta} & 0 & -\xi_{1}(\vec{k}) & -\bar{\phi} \\
0 & -\bar{\phi} & -\bar{\Delta} & -\xi_{2}(\vec{k})
\end{pmatrix}, \tag{9}
\]

where \(\Delta\) and \(\bar{\phi}\) are the dSC and dFF-DW order parameters respectively. From this mean-field Hamiltonian one can obtain a Landau free energy as a function of \(g_{\phi,\Delta} = \frac{3J}{4} \pm V\) and temperature:

\[
\mathcal{F} = \alpha_{\Delta} \Delta^{2} + \beta_{\Delta} \Delta^{4} + \alpha_{\phi} \phi^{2} + \beta_{\phi} \phi^{4} + \gamma_{\phi^{2}} \Delta^{2}. \tag{10}\]

For this model, one finds that \(\gamma > 0\), meaning that the orders \(\phi\) and \(\Delta\) compete. Nonetheless, there exists a parameter regime where the two coexist, consistent with experimental phase diagrams of the cuprates as schematically depicted in Fig. 3. If we now introduce a tunneling between planes along the c-axis, the picture is modified in two notable ways. The most obvious is that the free electron dispersion \(\xi\) changes. However, the dFF-DW order parameter no longer exactly connects hotspots 1 and 2 away from \(k_{z} = 0\). Since the Fermi surface changes shape with \(k_{z}\) while the ordering vector \(Q\) remains fixed, the hotspots cannot be nested at \(Q\) for all values of \(k_{z}\) as can be seen in Fig. 4. This can most readily be seen by observing the form of the dFF-DW susceptibility within the hotspot model. If we consider a point, \(\vec{k}\), the important quantities for the dFF-DW susceptibility are the energies \(\xi_{\vec{k}}\) and \(\xi_{\vec{k} + \vec{Q}}\). Let us define \(\xi_{\pm} = \frac{\xi_{\vec{k}} \pm \xi_{\vec{k} + \vec{Q}}}{2}\). We may then express the integrand of the susceptibility as

\[
\frac{\sinh \frac{\xi_{\pm}}{2T} \cosh \frac{\xi_{\pm}}{2T}}{2\xi_{\pm} \left( \sinh^{2} \frac{\xi_{\pm}}{2T} + \cosh^{2} \frac{\xi_{\pm}}{2T} \right)}. \tag{11}\]
stability is overwhelmingly of such form. Nevertheless, there are empirical reasons to believe that the experimental situation is a little more complicated. The main focus of this work is to address one such aspect.

IV. EFFECTS OF PHASE PINNING

Empirically, the c-axis ordering vector $Q_z$ of the density wave phase is broadly peaked around $\pi$, with a correlation length of approximately 0.6 lattice units. Motivated by this we consider a model in which the interlayer ordering is not defined by a single wavevector. Instead, we propose a model of the charge order wavevector $Q_L$, where the relative phase $\theta = \theta_1 - \theta_2$ in between the two layers and the relative orientation $Q_1 \cdot Q_2 \in \{0, Q^2\}$ of the ordering vector on the layers are taken to be random variables determined by disorder.

For the model under consideration, the Landau free energy generically takes the form

$$F_O[\theta] = \alpha_\Delta |\Delta|^2 + \beta_\Delta |\Delta|^4 + \alpha_{\phi,O}[\theta]|\phi|^2 + \beta_{\phi,O}[\theta]|\phi|^4 + \gamma_O[\theta]|\phi|^2|\Delta|^2,$$

where $\theta$ is as above, $O = ||, \perp$ is the relative orientation of the ordering vectors in the two planes, and $\Delta$ and $\phi$ are the superconducting and density wave order parameters, respectively. The coefficients may be calculated diagrammatically from the free particle action and depend parametrically on the interlayer couplings through the single-particle dispersion. The microscopic expressions for the Landau coefficients are given in the Appendix. We again find $\gamma > 0$, indicating competition between the two orders. For purposes of calculation it is useful to express the coefficients as a power series in $\cos(\theta)$

$$c_\parallel (\theta) = \sum_n c^{(n)} \cos^n \theta$$

$$c_\perp = \frac{1}{2\pi} \int_0^{2\pi} d\theta c_\parallel (\theta),$$

where $c \in \{ \alpha, \beta, \gamma \}$ and for a term including $\phi^m$ the coefficients $c^{(n)} = 0$ for $n > m/2$. This form allows moments of the terms to be calculated easily in terms of the circular moments $c^{(m\theta)}$.

The corresponding saddle-point equations admit three non-trivial solutions: a superconducting phase, a density wave phase, and a coexistent phase:

$$|\Delta| = \sqrt{\frac{\alpha_\Delta}{2\beta_\Delta}}, \phi = 0$$

$$\Delta = 0, |\phi| = \sqrt{-\frac{\alpha_\phi}{2\beta_\phi}}$$

$$|\Delta| = \sqrt{\frac{2\beta_\phi \alpha_\Delta - \gamma \alpha_\phi}{\gamma^2 - 4\beta_\Delta \beta_\phi}}, |\phi| = \sqrt{\frac{2\beta_\phi \alpha_\Delta - \gamma \alpha_\phi}{\gamma^2 - 4\beta_\Delta \beta_\phi}}.$$
Notably, there is little effect for \( \sigma \) in Fig. 5, increasing deviation \( \sigma \).}
FIG. 7. dFF-DW order parameter $\phi$ (top) and superconducting order parameter $\Delta$ (bottom) vs interlayer coupling for various values of $\sigma$. The coupling constants of the model have been normalized to keep the bare charge ordering temperature at $t_z = 30$ meV fixed. Increasing $t_z$ in general leads to a melting of dFF-DW and enhancement of SC with the effect becoming more pronounced as $\sigma$ is increased.

V. DISCUSSION AND CONCLUSION

In this work, we have shown that the broadly peaked nature of the $c$-axis structure for dFF-DW order in general means that an increase of interlayer tunneling leads to a melting of charge order and a corresponding enhancement of the competing superconductivity. Notably the presence of phase pinning or the dFF-DW is essential to this effect. This has implications for optical control experiments, which are believed to be inducing a transient superconducting state by coupling to interlayer degrees of freedom, and, in particular, to our previous work in which we investigated the effect of interlayer separation in cuprates on the competition between superconductivity and dFF-DW. In that work we considered specifically the case of an order which is constant along the $c$-axis. Here we have extended our analysis to consider the case where the interlayer phase difference for the dFF-DW order is a random variable to be averaged over.

One way to visualize our results is that domains with distinct phase differences form and these domains are susceptible to melting to different degrees. Such a picture is consistent with experiments where inhomogeneous enhancement of electron-electron pairing is observed, as one might expect from inhomogeneous melting of dFF-DW domains.

While the $c$-axis curvature effects seem to be too weak to explain the observed enhancement of superconducting correlations alone, there are still other theoretical and experimental reasons to believe that melting of charge order plays an important role. Other explanations have been considered for this effect such as redistribution of spectral weight, suppression of superconducting phase fluctuations, or other routes to melting of dFF-DW order. Most likely the complete explanation is some combination of factors, with a number of these frameworks forming complementary rather than competing mechanisms.

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Appendix: Microscopic expressions for Landau coefficients

As discussed in Sec. [V] the Landau theory for competing orders in this model takes the form

$$F_O[\theta] = \alpha_\Delta |\Delta|^2 + \beta_\Delta |\Delta|^2 + \alpha_{\phi,O} |\phi|^4 + \beta_{\phi,O} |\phi|^4 + \gamma_O |\phi|^2 |\Delta|^2, \quad (A.1)$$

The quadratic Landau coefficients are simply related to the susceptibilities in the corresponding channels

$$\alpha_\Delta = \frac{1}{g_\Delta} - \Pi_\Delta, \quad \alpha_{\phi} = \frac{1}{g_{\phi}} - \Pi_{\phi}. \quad (A.2)$$

The superconducting terms are simplest, with

$$\Pi_\Delta = \sum_k f^1(k)^2 \sum_{\pm} \frac{\tanh \frac{\epsilon_{k,\pm}}{2T}}{2\epsilon_{k,\pm}} \quad (A.3)$$

and

$$\beta_\Delta = \sum_k f^1(k)^2 \sum_{\pm} \frac{1}{2\epsilon_{k,\pm}} \times \left[ \frac{\tanh \frac{\epsilon_{k,\pm}}{2T}}{2\epsilon_{k,\pm}} + n_f(\epsilon_{k,\pm}) \right], \quad (A.4)$$
where $\epsilon_{k,\pm} = \xi_k \pm \xi_k$ are the eigenvalues of the free Hamiltonian.

As mentioned above the terms involving $\phi$ can be broken into coefficients of $\cos^n \theta$. Beginning with the dFF-

The quartic dFF-DW terms are

$$\rho^{(1)}_\phi = \sum_k \sum_{\lambda,\lambda'} (-1)^{\lambda-\lambda'} \left[ \frac{1}{2} f^1(k)^4 M_{\beta,1}(\epsilon_{\lambda}(k + Q2), \epsilon_{\lambda'}(k - Q2)) + f^1(k + Q)^2 f^1(k - Q)^2 M_{\beta,2}(\epsilon_{\lambda}(k), \epsilon_{\lambda'}(k - Q), \epsilon_{\lambda}(k + Q)) \right]$$

and

$$\rho^{(0,2)}_\phi = \sum_k \left\{ f^1(k)^4 \left[ \frac{1}{4} \sum_{\lambda,\lambda'} M_{\beta,1}(\epsilon_{\lambda}(k + Q2), \epsilon_{\lambda'}(k - Q2)) + f^1(k + Q)^2 f^1(k - Q)^2 \left[ \frac{1}{2} \sum_{\lambda_1,\lambda_2,\lambda_3} (-1)^{\lambda_2-\lambda_3} M_{\beta,2}(\epsilon_{\lambda_1}(k), \epsilon_{\lambda_2}(k - Q), \epsilon_{\lambda_3}(k + Q)) \right. \right. \right.$$ 

$$\left. \left. \left. + \sum_{\lambda,\lambda'} (-1)^{\lambda-\lambda'} M_{\beta,3}(\{\epsilon_{\lambda}(k + Q)\}_\lambda, \{\epsilon_{\lambda'}(k)\}_\lambda) \right] \right\},$$

where we have defined

$$M_{\beta,1}(x,y) = \frac{1}{(x-y)^2} \left( \frac{\tanh \frac{x}{2y} - \tanh \frac{y}{2x}}{x-y} + n'_f(x) + n'_f(y) \right)$$

$$M_{\beta,2}(x,y,z) = \frac{1}{z-y} \left( \frac{n_f(z)}{(x-z)^2} - \frac{n_f(y)}{(x-y)^2} \right) + \frac{1}{(x-z)(x-y)} \left[ n'_f(x) - n_f(x) \left( \frac{1}{x-z} + \frac{1}{x-y} \right) \right]$$

$$M_{\beta,3}({\{x_i\}_i}) = \sum_{j \neq i} n_f(x_i) \frac{x_i - x_j}{x_i - x_j}.$$ 

Finally for the competition term

$$\gamma^{(n)} = \sum_k f^1(k)^2 f^1(k_+) \sum_{\lambda,\lambda'} (-1)^{n(\lambda-\lambda')} \left[ f^1(k_+) M_{\gamma,2}(\epsilon_{\lambda}(k_+), \epsilon_{\lambda'}(k_-)) - f^1(k_-) M_{\gamma,1}(\epsilon_{\lambda}(k_+), \epsilon_{\lambda'}(k_-)) \right]$$

where $k_\pm = k \pm Q/2$ and we have defined

$$M_{\gamma,1}(x,y) = \frac{1}{(x-y)^2} \left( \frac{\tanh \frac{y}{2x} - \tanh \frac{x}{2y}}{2y} \right)$$

$$M_{\gamma,2}(x,y) = \frac{1}{2x} \left[ \frac{n'_f(x)}{x-y} - \frac{\tanh \frac{y}{2x}}{2x(x+y)} \right] + (x \leftrightarrow y).$$
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