

Superconducting charge-ordered states in cuprates

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Motivated by recent neutron scattering and scanning tunneling microscopy (STM) experiments on cuprate superconductors, we discuss charge-ordered states, in particular with two-dimensional charge modulation patterns, coexisting with superconductivity. We extend previous studies of a large- N mean-field formulation of the t - J model. In addition to bond-centered superconducting stripe states at low doping, we find checkerboard-modulated superconducting states which are favorable in an intermediate doping interval. We also analyze the energy dependence of the Fourier component of the local density of states at the ordering wave vector for several possible modulation patterns, and compare with STM results.

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I. INTRODUCTION

A series of recent experiments highlighted the importance of spin and charge ordering tendencies in the cuprate superconductors. Static stripe order has been established in Nd-doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$,¹ which appears to coexist with superconductivity at very low temperatures. Recently, superconducting samples of underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6.35}$ (Ref. 2) and nearly optimally doped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$,³ with T_c up to 90 K, have been found to show signatures of charge ordering. In addition, a variety of compounds display strong dynamic spin and charge fluctuations, which are enhanced by the application of moderate magnetic fields.^{4–6} This emphasizes that even materials where no static order can be observed in the absence of an external field are in close proximity to a critical point where spin and/or charge order is established in the superconducting state. Theories based on this assumption^{7–9} successfully explain a number of NMR (Ref. 10) and neutron scattering^{5,6,11} experiments.

In this paper, we will focus on superconducting states with static charge order, but dynamic spin fluctuations—this appears to be realized in the experiments of Ref. 2. Such states are found upon doping paramagnetic Mott insulators on a square lattice.^{12,13} The undoped quantum paramagnet has a broken translational symmetry associated with spontaneous bond charge (or spin-Peierls) order; at a small carrier concentration δ , this order persists and coexists with anisotropic superconductivity; a d -wave superconductor with a full square lattice symmetry appears above a critical δ . [“Charge order” is defined very generally as spatial modulation in any $\text{SU}(2)$ -invariant observables, such as the local density of states (LDOS) per *site*, or kinetic or exchange energy per lattice *bond*; the modulation in the total site charge density can be small due to long-range Coulomb interactions.] A particular feature of the superconducting charge-ordered states, found in Refs. 12 and 13, is that the modulation is bond centered and its real-space period p always takes even integer values (in units of the Cu lattice spacing); this is in contrast to the continuous doping evolution of the ordering wave vector $1/p$, usually assumed in the so-called “Yamada plot.”¹⁴ Interestingly, the experimental results of Refs. 2–4 appear to be remarkably well described by the states proposed in Refs. 12 and 13, as has also been

discussed in recent work^{15,16} which appeared while this paper was being completed: Neutron scattering on underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6.35}$ (Ref. 2) shows a charge order with a real-space period of eight lattice sites, whereas $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ close to optimal doping displays a $p=4$ modulation;^{3,4} furthermore the scanning tunneling microscopy (STM) data indicate an even-period modulation not only in the site charge density, but also in the bond kinetic energy and perhaps the bond pairing amplitude.¹⁵

We note that numerical studies of the t - J model¹⁷ have observed bulk charge order with a period $p=4$ at a doping level $1/8$; and paired hole states with different types of charge order in both insulators and superconductors have been discussed elsewhere.^{18–20} In the past, most theoretical work has been focused on states with one-dimensional (1D) charge modulation, often referred to as stripes. However, recent STM experiments^{3,4} indicate LDOS modulations in *both* x and y directions in a single CuO_2 plane (although a small anisotropy is observed). Theoretically, charge density wave (CDW) fluctuations are expected in both directions¹³ on the *disordered* side of the charge ordering transition (if the system has no intrinsic lattice anisotropy). Moving to the ordered side, it depends on microscopics whether CDW order in one or in two directions condenses, leading to stripelike or 2D modulations, respectively.

The purpose of this paper is twofold: In Sec. II, we reexamine the mean-field theory of Refs. 12 and 13, to investigate the possible existence of and the mechanism leading to superconducting charge-ordered states with 2D charge modulation. In Sec. III, we turn to a detailed discussion of the charge modulation pattern, by calculating the energy dependence of the LDOS Fourier component at the ordering wave vector and comparing it with the measurements of Ref. 3.

II. MEAN-FIELD THEORY

We start by reanalyzing the large- N theory of Refs. 12 and 13, which provides a microscopic description of doping mobile charge carriers into a paramagnetic Mott insulator.²¹ We consider an extended t - J Hamiltonian for fermions, $c_{i\alpha}$, on the sites i of a square lattice with spin $\alpha=1 \dots 2N$ ($N=1$ is the physical value):

$$\mathcal{H}_{tJV} = \sum_{i>j} \left[-\frac{t_{ij}}{N} c_{i\alpha}^\dagger c_{j\alpha} + \text{H.c.} + \frac{V_{ij}}{N} n_i n_j + \frac{J_{ij}}{N} \left(\mathbf{S}_i \cdot \mathbf{S}_j - \frac{n_i n_j}{4N} \right) \right]. \quad (1)$$

Here $n_i = c_{i\alpha}^\dagger c_{i\alpha}$ is the on-site charge density, and the spin operators \mathbf{S}_i are fermion bilinears times the traceless generators of $\text{Sp}(2N)$. For most of the following, the fermion hopping t_{ij} and exchange J_{ij} will be restricted to nearest-neighbor terms t and J ; for a detailed comparison with the Ref. 3 we will introduce a second neighbor hopping t' . The electronic Coulomb interaction is represented by the on-site constraint $n_i \leq N$ and the off-site repulsive interactions $V_{ij} = V/|r_{ij}|$. The average doping δ is fixed by $\sum_i \langle n_i \rangle = NN_s(1 - \delta)$, where N_s is the number of lattice sites. To proceed, we represent the spins by auxiliary fermions $f_{i\alpha}$ and the holes by spinless bosons b_i , such that the physical electrons $c_{i\alpha} = b_i^\dagger f_{i\alpha}$, and the necessary Hilbert space constraint is implemented by Lagrange multipliers λ_i . Via a Hubbard-Stratonovich decoupling of the antiferromagnetic interaction we introduce link fields Q_{ij} , defined on the bonds of the square lattice. After taking the limit $N \rightarrow \infty$, the slave bosons b_i condense, $\langle b_i \rangle = \sqrt{N} b_i$, Q_{ij} and λ_i take static saddle-point values, and we are left with a bilinear Hamiltonian which can be diagonalized by a Bogoliubov transformation. At the saddle point, the slave boson amplitudes fulfill $\sum_i b_i^2 = N_s \delta$, and the link fields are given by $N Q_{ij} = \langle \mathcal{J}^{\alpha\beta} f_{i\alpha}^\dagger f_{j\beta}^\dagger \rangle$, where $\mathcal{J}^{\alpha\beta}$ is the antisymmetric $\text{Sp}(2N)$ tensor; for further details, see Ref. 13.

Various ground states obtained from the numerical solution of the above mean-field equations have been discussed in Refs. 12 and 13. At $\delta=0$ the ground state is a fully dimerized, insulating spin-Peierls state, i.e., it has period-2 bond charge order. At low doping δ , the bare large- N t - J model tends to phase separate, and the inclusion of a moderate Coulomb interaction V leads to the formation of bond-centered, superconducting stripes with a 1D charge modulation. Large doping destroys charge order and leads to a pure d -wave superconducting ground state; in this regime the large- N approach reduces to the usual BCS mean-field theory with renormalized hopping matrix elements.

Motivated by the STM results of Refs. 3 and 4, we have searched for additional saddle-point solutions with 2D charge modulation; we have restricted our attention to states in which the charge distribution respects the 90° rotation symmetry of the lattice. Interestingly, there are several such saddle points which were overlooked in Ref. 13. In most of the low and intermediate doping regions the states with 1D and 2D modulations are close in energy; at small doping 1D stripelike states are preferred, whereas 2D checkerboardlike states are lower in energy in a certain interval of intermediate doping and Coulomb repulsion.

We have therefore concentrated on states with a real-space periodicity $p=4$, leading to a unit cell of 4×4 sites.³ (Recall that bond-centered stripes with the spatial period pinned to four sites were found over a rather large range of doping values in Refs. 12 and 13.) The most favorable mean-

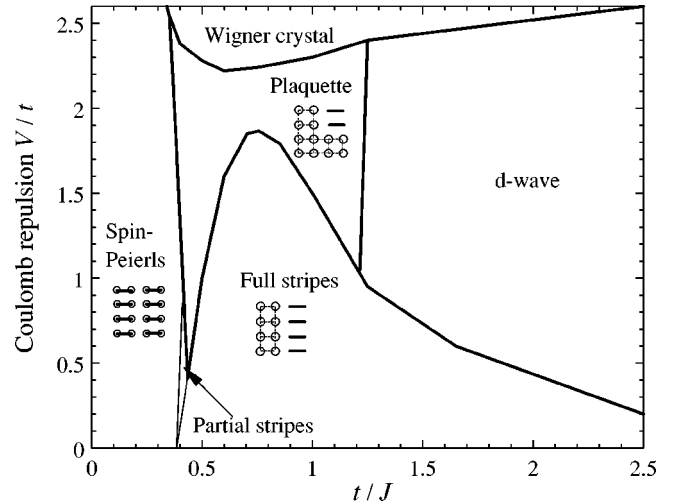


FIG. 1. Ground state phase diagram of the extended t - J model in the large- N limit at doping $\delta=20\%$. Thick (thin) lines indicate first- (second-) order transitions. All states except for the Wigner crystal have superconducting order. The stripe and spin-Peierls phases show 1D charge modulation; the plaquette phase has charge modulations with a real-space period 4 in both directions. The circles indicate the spatial distribution of hole density, the lines symbolize the strength of the bond variables Q_{ij} . “Full stripes” refers to states where the charge modulation in the large- N limit is maximal, i.e., the hole density is zero in the hole-poor regions, whereas the “partial stripe” states have a finite hole density there.

field states with 2D charge modulation are characterized by holes arranged in intersecting bond-centered stripes, i.e., the hole concentration is large on 12 sites and zero on four sites; see Fig. 1. The actual filling in the hole-rich regions is smaller than 1 and depends on microscopic parameters; due to the strong pairing correlations the 2D CDW states are good superconductors (with a d -wave-like pairing symmetry). Such 4×4 plaquette states occur as large- N ground states of Hamiltonian (1) for doping levels between approximately 13% and 25%. A sample phase diagram for doping $\delta=20\%$ is shown in Fig. 1 (also see Fig. 3 of Ref. 12). For small t/J phase separation tendencies do not occur, therefore the ground state is a doped spin-Peierls state with a homogeneous site charge distribution. At intermediate t/J , frustrated phase separation leads either to stripes or to plaquette states; at larger t/J the kinetic energy becomes dominant and weakens the phase separation tendencies, consequently a homogeneous d -wave state is reached. At very large Coulomb repulsion, the holes arrange into an insulating Wigner crystal.

The occurrence of plaquette-modulated CDW states in favor of stripes can be understood as an interplay of exchange, kinetic, and Coulomb energies as follows: The exchange term “prefers” dimerization between spins on neighboring sites and tends to expel holes—this produces the overall dimer structure of the Q_{ij} , and leads to a fraction of sites being undoped. The kinetic energy is lowered by possible hopping processes, i.e., by neighboring sites with a nonzero hole density. Clearly, the exchange term prefers stripes, whereas the kinetic term prefers plaquettes, where hopping in two directions is possible. Now the Coulomb energy of the 2D modulated state is significantly lower than that of a stripe

state, because the charge inhomogeneity is smaller in the plaquette state (which is also closer to a crystalline arrangement of charges). From this discussion it is clear that, at low doping, where the physics is dominated by the exchange term, stripe states are preferred. With increasing doping the kinetic energy becomes more and more important, which leads to 2D modulated CDW states at moderate values of V , before a homogeneous d -wave superconductor becomes the ground state.

In all CDW states superconductivity competes with charge order. In the stripe states superconductivity is very weak due to the strong anisotropy: bulk superconductivity is established only by Cooper pair tunneling between the stripes. In contrast, the plaquette CDW states are much better superconductors, due to the full 2D character of the charge distribution. This trend is consistent with the low T_c in Nd-doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (Ref. 1) compared to much higher T_c in apparently charge-ordered $\text{YBa}_2\text{Cu}_3\text{O}_{6.35}$ (Ref. 2) and $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ samples.³

III. MODULATION IN THE LOCAL DENSITY OF STATES

After having established the possible occurrence of plaquette CDW states in the large- N theory for the t - J model, we turn to a detailed analysis of the corresponding STM signal. Hoffman *et al.*⁴ introduced a STM technique of atomically resolved spectroscopic mapping, which allowed one to detect LDOS modulations around vortex cores with real-space period 4, i.e., at wave vectors $\mathbf{K}_x = (\pi/2, 0)$ and $\mathbf{K}_y = (0, \pi/2)$. Howald *et al.*³ used this technique to map the energy dependence $\rho_{\mathbf{K}}(\omega)$ of the spatial Fourier component of the LDOS at the ordering wave vectors $\mathbf{K}_{x,y}$. This energy dependence has recently been discussed within a model for the pinning of spin density wave (SDW)/CDW fluctuations by inhomogeneities,²² and in an analysis of different patterns of translational symmetry breaking in d -wave superconductors.¹⁵

For a comparison with the experimental situation³ we restrict ourselves again to states with period-4 modulation; we will employ hopping parameters t and t' that yield a realistic band structure. Furthermore we have to keep in mind the shortcomings of the large- N theory: the precise location of the phase boundaries is not reliable, and the theory underestimates fluctuations. Therefore we will work with superconducting gap values close to the experimentally observed ones, and discuss both self-consistent mean-field solutions as well as states where translational symmetry breaking is imposed by hand in the Hamiltonian.

To obtain initial information about the possible forms of $\rho_{\mathbf{K}}(\omega)$ we start by considering d -wave superconductors with additional modulation in one of the following quantities: site charge density, bond charge density (kinetic energy), and pairing amplitude. Such an analysis has also been independently performed by Podolsky *et al.*,¹⁵ but here we are interested in 2D modulations and furthermore diagonalize the mean-field Hamiltonian exactly for the 4×4 unit cell. The Hamiltonian thus has the form $\mathcal{H}_{\text{BCS}} + \mathcal{H}_{\text{mod}}$, where

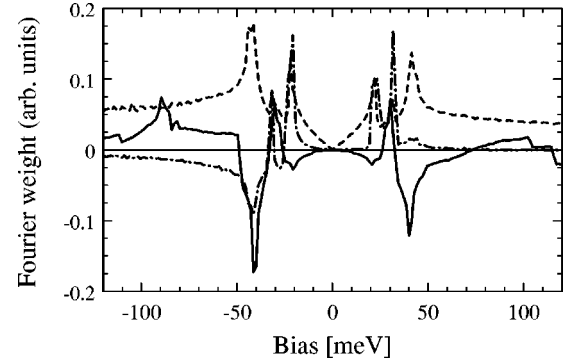


FIG. 2. Energy dependence $\rho_{\mathbf{K}}(\omega)$ of the Fourier component of the LDOS at $\mathbf{K} = (\pi/2, 0)$, obtained from diagonalizing $\mathcal{H}_{\text{BCS}} + \mathcal{H}_{\text{mod}}$ on a 4×4 unit cell. Bulk parameter values are $t = 0.15$ eV, $t' = -t/4$, doping $\delta = 17\%$, and a gap size $\Delta_0 = 40$ meV. The curves correspond to modulations as follows: solid—site charge density; dashed—bond charge density; dash-dotted—pairing amplitude. The amplitude of the LDOS modulation is proportional to V_0 in \mathcal{H}_{mod} ; here $V_0^{\text{site}} = V_0^{\text{kin}} = 10$ meV, $V_0^{\text{pair}} = 4$ meV.

$$\mathcal{H}_{\text{BCS}} = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} c_{\mathbf{k},\sigma}^{\dagger} c_{\mathbf{k},\sigma} + \sum_{\mathbf{k}} \Delta_{\mathbf{k}} (c_{\mathbf{k},\uparrow}^{\dagger} c_{-\mathbf{k},\downarrow} + \text{H.c.}) \quad (2)$$

in standard notation, with $\Delta_{\mathbf{k}} = \Delta_0 (\cos k_x - \cos k_y)/2$; \mathcal{H}_{BCS} is equivalent to the $\text{Sp}(2N)$ mean-field theory presented above in the region where the large- N ground state is a pure d -wave superconductor (with the correspondence $\Delta_{ij} = J_{ij} Q_{ij}$ where Δ_{ij} is the real-space Fourier transform of the energy gap $\Delta_{\mathbf{k}}$).

The modulation is introduced via \mathcal{H}_{mod} : for a site CDW we add $\mathcal{H}_{\text{mod}}^{\text{site}} = V_0^{\text{site}} \sum_i f(\mathbf{R}_i) c_{i,\sigma}^{\dagger} c_{i,\sigma}$; for a bond CDW we have $\mathcal{H}_{\text{mod}}^{\text{kin}} = V_0^{\text{kin}} \sum_{\langle ij \rangle} f[(\mathbf{R}_i + \mathbf{R}_j)/2] c_{i,\sigma}^{\dagger} c_{j,\sigma}$, and a pairing modulation is given by $\mathcal{H}_{\text{mod}}^{\text{pair}} = V_0^{\text{pair}} \sum_{\langle ij \rangle} f[(\mathbf{R}_i + \mathbf{R}_j)/2] (c_{i,\uparrow}^{\dagger} c_{j,\downarrow}^{\dagger} + \text{H.c.})$. The function $f(\mathbf{R})$ describes the modulation strength and pattern, and we will concentrate on 2D bond-centered period-4 modulations with $f(\mathbf{R}) = [\cos(\pi R_x/2 + \pi/4) + \cos(\pi R_y/2 + \pi/4)]/2$.

Diagonalization of $\mathcal{H}_{\text{BCS}} + \mathcal{H}_{\text{mod}}$ yields the local density of states for each site of the unit cells, from which we find $\rho_{\mathbf{K}}(\omega)$ by Fourier transformation [the real-space origin is chosen such that $\rho_{\mathbf{K}}(\omega)$ is real]; note that $\mathbf{K} = (\pi/2, 0)$ and $(0, \pi/2)$ are equivalent with the above choice of $f(\mathbf{R})$. Results for $\rho_{\mathbf{K}}(\omega)$ are shown in Fig. 2 for the three modulation cases listed above. If we compare the curves in Fig. 2 with the STM result in Fig. 3 of Ref. 3, which shows a peak in the magnitude of $\rho_{\mathbf{K}}(\omega)$ at subgap energies $|\omega|/\Delta_0 \approx 2/3$, it is clear that the experiments are not well described by a site charge modulation alone. In contrast, our result for a modulation in the pairing amplitude comes closest to the curves of Ref. 3. We note that our results in Fig. 2 are somewhat different from the ones of Ref. 15, this may be due to the 2D character of the modulation considered here and due to the approximations employed in Ref. 15.

It is clear that the experimentally realized CDW state will have modulations in all quantities invariant under spin rotation and time reversal. This can—at least in part—be cap-

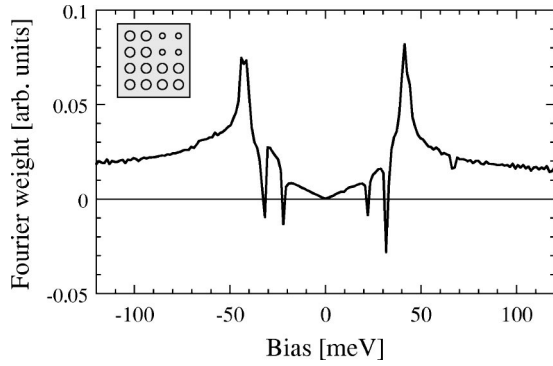


FIG. 3. As in Fig. 2, but for a state with plaquette site charge modulation as indicated in the inset ($V_0^{\text{site}}=5$ meV), and self-consistently determined pair fields Δ_{ij} , using $J=70$ meV. Note that LDOS modulations of similar magnitude occur at wave vectors $\pm \mathbf{K}_x \pm \mathbf{K}_y$.

tured by a self-consistent solution of the mean-field equations. A natural candidate is given by the plaquette state found as large- N ground state above (Fig. 1). Thus we employ the Hamiltonian $\mathcal{H}_{\text{BCS}} + \mathcal{H}_{\text{mod}}^{\text{site}}$, where the pair field Δ in \mathcal{H}_{BCS} [Eq. (2)] is determined self-consistently from $\Delta_{ij} = J_{ij} \langle \mathcal{J}^{\alpha\beta} c_{i\alpha}^\dagger c_{j\beta}^\dagger \rangle$, and $\mathcal{H}_{\text{mod}}^{\text{site}}$ imposes a weak modulation of the site charge density as shown in Fig. 3 (the strong modulation found in the large- N limit will certainly be weakened by fluctuation corrections beyond the large- N theory). Figure 3 displays a corresponding LDOS modulation $\rho_{\mathbf{K}}(\omega)$. The agreement with the available experimental data³ is not satisfying; in particular $\rho_{\mathbf{K}}(\omega)$ does not show a large peak at energies below the bulk superconducting gap.

This fact and the results in Fig. 2 led us to consider an additional effect not captured in the mean-field calculations: On general symmetry grounds, a static charge modulation will lead to a real-space modulation in the effective *pairing interaction*, because the CDW influences the local spin fluctuation spectrum. On the mean-field level, this can be phenomenologically accounted for by a modulation in the exchange interaction J . Therefore, we have studied self-consistent solutions of the mean-field theory, $\mathcal{H}_{\text{BCS}} + \mathcal{H}_{\text{mod}}^{\text{site}}$, as above, but in addition to a weak site charge modulation in $\mathcal{H}_{\text{mod}}^{\text{site}}$ we imposed a modulation of the J_{ij} exchange interaction, $J_{ij} = J_0 + V_0^J f[(\mathbf{R}_i + \mathbf{R}_j)/2]$ for nearest neighbor sites i

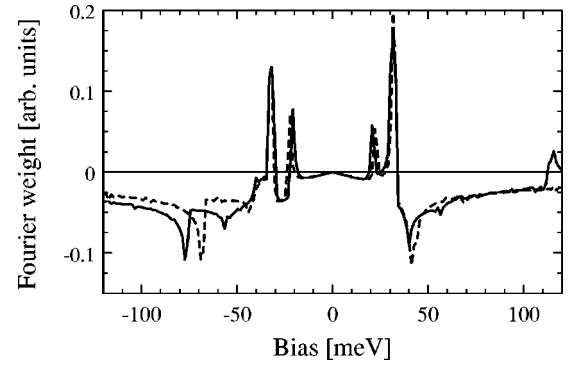


FIG. 4. As in Fig. 2, but for a model with plaquette modulation in both site charge density and exchange interaction, and self-consistently determined pair fields Δ_{ij} , using $V_0^{\text{site}}=10$ meV, $J_0=70$ meV, and $V_0^J=3.5$ meV. The dashed curve corresponds to a band structure with $t' = -t/3$ to demonstrate the robustness of the result.

and j , which leads to corresponding modulations in both the pair fields Δ_{ij} and the bond charge density (kinetic energy). Results for the LDOS Fourier component $\rho_{\mathbf{K}}(\omega)$ are shown in Fig. 4, with a rather good agreement with the experiments of Ref. 3.

IV. CONCLUSIONS

Summarizing, we have studied superconducting charge-ordered states of doped Mott insulators. Within a large- N theory we have established that ground states with 2D charge modulation can occur at intermediate doping where they are preferred over stripes. By analyzing the energy dependence of the LDOS modulation as observed in STM, we have found the data of Ref. 3 to be well described by combined modulations in charge density as well as exchange and pairing energy, caused by a modulation of the pairing interaction.

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