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Superconductivity, Pair Density Wave, and Néel Order in Cuprates *

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We investigate in underdoped cuprates possible coexistence of the superconducting order at zero momentum and pair density wave (PDW) at momentum $\boldsymbol{Q} = (\pi, \pi)$ *in the presence of a Néel order. By symmetry, the d-wave uniform singlet pairing* dS_0 *can coexist with the d-wave triplet PDW* $dT_{\boldsymbol{Q}}$*, and the p-wave singlet PDW* $pS_{\boldsymbol{Q}}$ *can coexist with the p-wave uniform triplet* pT_0*. At half filling, we find that the novel* $pS_{\boldsymbol{Q}} + pT_0$ *state is energetically more favorable than the* $dS_0 + dT_{\boldsymbol{Q}}$ *state. At finite doping, however, the* $dS_0 + dT_{\boldsymbol{Q}}$ *state is more favorable. In both types of states, the variational triplet parameters* $dT_{\boldsymbol{Q}}$ *and* pT_0 *are of secondary significance. Our results point to a fully symmetric* Z_2 *quantum spin liquid with spinon Fermi surface in proximity to the Néel order at zero doping, which may not be adiabatically connected to the d-wave singlet superconductivity at finite doping with intertwining d-wave triplet PDW fluctuations and spin moment fluctuations. The results are obtained by variational quantum Monte Carlo simulations.*

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The mechanism of high-temperature superconductivity in cuprates remains to be an exciting research topic. One of the interesting proposals is Anderson's resonating valence bond (RVB) state.[1] A suitable Hamiltonian describing such a system is the one-band t–J model.[2] In this model, while the parent compound at half filling is automatically a Mott insulator for the charge degrees of freedom, the spin sector is much more intriguing. The RVB state may be viewed as a linear combination of configurations of the covering of spin singlets, a quantum spin liquid (QSL) with fractional spinon excitations. Chemical doping introduces mobile holes, leaving room for spin singlets to relocate and hence making the system a superconductor immediately.[3] Initially, an s-wave RVB state is proposed in view of the experimental robustness of superconductivity against impurity scattering,[4] whereas it is found later that the d-wave RVB state is energetically better,[5,6] and can actually be robust against impurities in doped Mott insulators because of charge renormalization.[7]

It should be pointed out that in the undoped limit, however, there is a local charge-$SU(2)$ symmetry following from the charge neutrality.[8] This symmetry relates various forms of RVB states. For example, the RVB state with a π-flux in the spinon hopping around a plaquette may be mapped to a state with d-wave pairing, etc. Such states are said to be gauge equivalent and describe the same spin liquid upon projection to the physical Hilbert space. The projective symmetry group (PSG) has been developed[9,10] to classify all possible and physically distinct spin liquids. It is then interesting to ask which type of spin liquid is adiabatically connected to the d-wave pairing at finite doping, although the d-wave RVB state is widely believed to be the candidate. This becomes even more challenging when the ground state develops Néel order so that spin-$\frac{1}{2}$ spinons would have been confined to form spin-1 magnons. However, inelastic neutron scattering experiments show that the spin excitations away from the Néel vector are broadened significantly, well beyond the linear spin-wave description.[11] A recent interesting proposal is that even in the presence of Néel order, the spinons may be deconfined in a partial region of the Brillouin zone,[12−14] although the same phenomenon could also be understood in terms of magnon-magnon scattering.[14−16] It is therefore interesting to consider spinon states on the background of the Néel order.

The charge-$SU(2)$ symmetry is broken at finite doping. The Néel order is weakened but persists at small finite doping. The effect of this order on superconductivity is also an intriguing topic. In this case, the spin-$SU(2)$ symmetry is broken down to $O(2)$. As a result, there is no longer sharp distinction between spin-singlet and spin-triplet Cooper pairings,

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and there is room for coexistence,[17,18] although the relative weight is not dictated by symmetry. There is a residual point group C_4 (with respect to a site), which leaves the spin moment invariant in the Néel state. This symmetry dictates what kinds of singlet and triplet could coexist. There are three irreducible representations for the C_4 group, namely, the 1D A and B representations, and the 2D E representation. Since the completely symmetric A representation is not a favorable pairing symmetry we will ignore it henceforth. The B representation transforms as d-wave. The doubly degenerate E representation transforms as p-wave. Therefore, the singlet and triplet should transform under C_4 identically either as d-wave or as p-wave. These possibilities are illustrated in Fig. 1. In the first case, the d-wave singlet Cooper pair at momentum $\mathbf{q} = 0$ (a), can coexist with a d-wave triplet at momentum $\boldsymbol{Q} \equiv (\pi, \pi)$ (b). The latter is a pair density wave (PDW), namely, the Cooper pairing at the center-of-mass momentum of $\boldsymbol{Q}$. In the second case, the $p + ip'$-wave singlet PDW at momentum $\mathbf{q} = \boldsymbol{Q}$ (c) can coexist with the same $p + ip'$-wave but triplet SC state (d). The four types of states are denoted in a self-explaining manner as dS_0, $dT_{\boldsymbol{Q}}$, $pS_{\boldsymbol{Q}}$ and pT_0. Interestingly, in the PDW states, an electron at momentum $\boldsymbol{k}_1 = \boldsymbol{k} + \boldsymbol{Q}$ pairs up with another at $\boldsymbol{k}_2 = -\boldsymbol{k}$. When $\boldsymbol{k}$ is on the so-called umklapp surface (US), so will be both $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$, related by mirror symmetry. The scattering of such a Cooper pair across the US was argued to be the key mechanism that could generate not only the single-particle gap but also two-particle gap near the antinodes and hence the pseudogap.[19] On the other hand, the chiral pT_0 state was recently proposed[17] to be present on the background of the Néel order, in an effort to explain in the underdoped regime the opening of a mini gap in the quasiparticle excitations along the otherwise gapless (nodal) direction of the dS_0 state.[20–24] Remarkably, if this were the case, the cuprate would be topologically nontrivial because of the chiral $p + ip'$ pairing.[25,26]

We are therefore motivated to investigate possible coexistence of superconducting (SC) order at zero momentum and PDW at momentum $\boldsymbol{Q}$ in the presence of a Néel order. We use the variational quantum Monte Carlo (VQMC) to treat the strong correlation effects. In the undoped case, we find that the novel $pS_{\boldsymbol{Q}} + pT_0$ state is energetically more favorable than the $dS_0 + dT_{\boldsymbol{Q}}$ state. On its own the $pS_{\boldsymbol{Q}}$ state is a fully symmetric Z_2 QSL with spinon Fermi surfaces, but it is unstable toward Néel ordering. At finite low doping, however, the $dS_0 + dT_{\boldsymbol{Q}}$ state is more favorable. Our results point to a novel QSL in proximity to the Néel order at zero doping, which is not adiabatically connected to the d-wave singlet SC at finite doping with intertwining d-wave triplet PDW fluctuations and spin moment fluctuations.

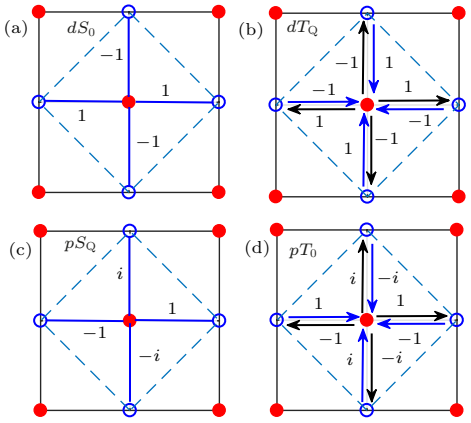

Fig. 1. Illustration of the pairing function Δ_{ij} on nearest-neighbor bonds, in the d-wave case [(a) and (b)], and the p-wave case [(c) and (d)], all on the background of a Néel order. The spin moments are opposite on the filled and open circles. For singlet pairing, Δ_{ij} is symmetric under exchange of i and j, hence its value is denoted on headless bonds in (a) and (c). For triplet pairing, the function is antisymmetric under the exchange of i and j, hence its value is denoted by colored text on colored arrows in (b) and (d). An arrow starts at i and points to j.

Model and method.—We begin with the t–J model on the square lattice, described by the Hamiltonian

$$H = -\sum_{n=1,2;\sigma}\sum_{\langle ij\rangle\in N_n} t_n P(c^{\dagger}_{i\sigma}c_{j\sigma} + \mathrm{H.c.})P + J\sum_{\langle ij\rangle\in N_1}\left(\boldsymbol{S}_i\cdot\boldsymbol{S}_j - \frac{1}{4}Pn_in_jP\right). \quad (1)$$

Here σ is the spin polarization, $N_{1,2}$ denote the first- and second-neighbor bonds with hopping integrals $t_1 = t$ and $t_2 = t'$, respectively; $c_{i\sigma}$ is the electron annihilation operator, J is the Heisenberg spin exchange, $\boldsymbol{S}_i$ is the local spin, and n_i is the local density. The operator $P = \Pi_i(1 - n_{i\uparrow}n_{i\downarrow})$ projects away any double occupancy. As typical parameters for cuprates, we take $(t, t', J) = (0.4, -0.12, 0.13)$ eV.[3] At half filling, the charge degrees of freedom are frozen, and the model reduces to the Heisenberg model. Upon hole doping, the doped holes may move without causing double occupancy, leading to metallicity and superconductivity.

The Hamiltonian includes infinitely strong correlations, due to the fact that no double-occupancy is allowed. In this work, we tackle the problem by variational quantum Monte Carlo (VQMC), which takes care of the no-double occupancy condition exactly. This method has been used extensively previously for the same system,[5,27–32] yielding considerable insights into the Néel state at half filling and the uniform d-wave SC state at finite doping. More recently, the VQMC has been extended to deal with essentially unlimited number of variational parameters.[33–35] Here we will go beyond the uniform d-wave ansatz yet limit

ourselves to a handful of motivated parameters as we now describe. For later convenience, we introduce the Nambu basis $\psi_i^\dagger = (c_{i\uparrow}^\dagger, c_{i\downarrow})$. The variational Hamiltonian can be written as

$$H_{\rm v} = \sum_{\langle ij\rangle\in N_1} (\psi_i^\dagger U_{ij}\psi_j + {\rm H.c.}) - \sum_i \psi_i^\dagger(\mu_z\eta_i + \mu\tau_3)\psi_i - \sum_{\langle ij\rangle\in N_2} (\psi_i^\dagger t_{2v}\tau_3\psi_j + {\rm H.c.}). \quad (2)$$

Henceforth $\tau_{1,2,3}$ are Pauli matrices in the Nambu (or particle-hole) basis. The second-neighbor hopping t_{2v}, the chemical potential μ and the exchange field μ_z are all variational parameters, and $\eta_i = \pm 1$ is a staggered sign for A/B sublattice. Furthermore, on the N_1 bonds,

$$U_{ij} = -t_{1v}\tau_3 + \Delta_{ij}\tau^+ + \Delta_{ji}^*\tau^-. \quad (3)$$

Henceforth we fix $t_{1v} = 1$ without loss of generality (since the only role of $H_{\rm v}$ is to construct the trial wavefunction, see below), $\tau^\pm = (\tau_1 \pm i\tau_2)/2$, and Δ_{ij} is the real-space pairing function on a directed bond $\langle ij\rangle$: For $\boldsymbol{b} = \boldsymbol{r}_j - \boldsymbol{r}_i = (b_x, b_y)$,

$$\Delta_{ij} = (b_x^2 - b_y^2)(S_0 + T_{\boldsymbol{Q}}\eta_i) + (b_x + ib_y)(S_{\boldsymbol{Q}}\eta_i + T_0),$$

where S_0 ($T_{\boldsymbol{Q}}$) is the singlet (triplet) part of the d-wave pairing, and $S_{\boldsymbol{Q}}$ (T_0) is the singlet (triplet) part of the chiral p-wave pairing. Note we assumed the triplets all have their d-vectors along z, the direction of the Néel moment. In this way, the total spin of a triplet Cooper pair is orthogonal to the Néel moment, a most favorable situation for triplets to develop on the Néel order induced by μ_z. On the other hand, the momentum $\boldsymbol{Q} = (\pi, \pi)$ in the PDW state is obvious from the staggered sign η_i over the lattice. The four cases of the pairing function Δ_{ij}, when only one of the four coefficients in it is nonzero, are illustrated in Fig. 1. To summarize, we consider the set of variational parameters

$$x = \{\mu, \mu_z, t_{2v}, S_0, T_{\boldsymbol{Q}}\}, \quad \text{d-wave case},$$
$$x = \{\mu, \mu_z, t_{2v}, S_{\boldsymbol{Q}}, T_0\}, \quad \text{p-wave case}. \quad (4)$$

Since the d-wave and p-wave states belong to different irreducible representations of the C_4 group, we consider them separately. We assume that all parameters in x are real, as this turns out to gain energy better.

The normalized trial ground state is constructed as $|G\rangle = P|\psi_0\rangle/\sqrt{\langle\psi_0|P|\psi_0\rangle}$, where $|\psi_0\rangle$ is the ground state of the free variational Hamiltonian $H_{\rm v}$ (which depends on the parameter set x), in the canonical ensemble with a definite total number of electrons, $N_{\rm e} = N(1-\delta)$, on the lattice. Here N is the number of sites and δ is the hole doping level. We use the standard Monte Carlo to calculate the average energy density E and the Néel order m, $E = \langle G|H|G\rangle/N$, $m = \sum_i \eta_i\langle GS_i^z|G\rangle/N$. We optimize the parameter set x automatically by adapting to our case the method proposed previously.[33–35]

Variational results at zero doping.—In Fig. 2 we show the energy (per site) in the two types of variational states, versus the inverse lattice size. In both the cases, the parameter μ_z is finite, leading to Néel order, see Fig. 4(b) at zero doping. The best energy of the d-wave state is about $E = -1.1645J$, or $\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j\rangle = -0.3322J$ on N_1 bonds, consistent with that reported in the literature.[3,5,32,36,37] Interestingly, the optimized energy is even lower in the p-wave state, $E = -1.1660J$. Translated as $\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j\rangle = -0.3330J$, the energy is so far the best using fermionic VQMC, and is fairly close to the results of bosonic VQMC ($-0.3344J$[38,39]), Green's function QMC ($-0.3346J$[40]) and stochastic series expansion QMC ($-0.3347J$[41]). The singlet part (dS_0 or $pS_{\boldsymbol{Q}}$) is essential to gain energy. It is surprising that the more favorable $pS_{\boldsymbol{Q}}$ singlet has a center-of-mass momentum $\boldsymbol{Q}$, in contrast to the usual dS_0 state widely assumed in previous fermionic VQMC. Therefore, our results point to a novel type of ground state with p-wave singlet PDW. An interesting question is whether such a pairing could persist at finite doping. We will come back to this point later.

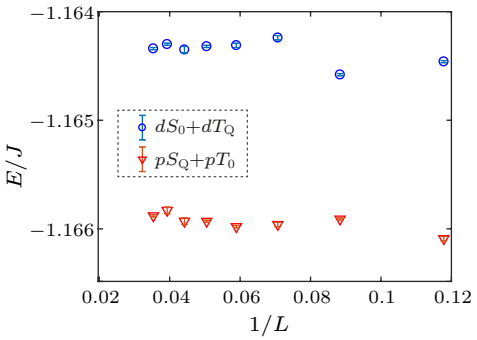

Fig. 2. Scaling of the optimized energy per site versus $1/L$, with $L = \sqrt{N}$ the linear lattice size, in the $dS_0 + dT_{\boldsymbol{Q}}$ state (open circles) and the $pS_{\boldsymbol{Q}} + pT_0$ state (open triangles). In both the cases the Néel order is included and optimized simultaneously.

A Z_2 QSL with nested spinon Fermi surfaces and a pair of Dirac nodes.—At zero doping, we find that if we switch off the Néel order (by setting $\mu_z = 0$), the optimized energy is $E = -1.1466J$ for the $pS_{\boldsymbol{Q}}$ state, and $E = -1.1406J$ for the dS_0 state. The $pS_{\boldsymbol{Q}}$ state is still better. The energy difference is far beyond statistical error. Furthermore, we can set $\mu = t_{2v} = 0$ without affecting the optimized energy. In this case the variational Hamiltonian is composed of the U_{ij} terms only, in Eq. (2). We take a closer look into such a $pS_{\boldsymbol{Q}}$ state to understand why it is better than dS_0, in terms of the PSG theory.[9,10]

There are staggered signs in Δ_{ij} or in U_{ij} in the $pS_{\boldsymbol{Q}}$ state, see Fig. 1(c). This can actually be gauged

away, given the exact charge neutrality (at zero doping) and hence local charge-$SU(2)$ gauge invariance in VQMC. After the gauge transformation,

$$U_{ij} \to V_i U_{ij} V_j^\dagger, \quad V_i = e^{i\boldsymbol{Q}\cdot\boldsymbol{r}_i\tau_3/2}, \tag{5}$$

where $\boldsymbol{r}_i = (x_i, y_i)$ is the coordinate vector, we obtain a uniform ansatz:

$$U_{i,i+\hat{x}} = -i\tau_0 + S_{\boldsymbol{Q}}\tau_2, \quad U_{i,i+\hat{y}} = -i\tau_0 + S_{\boldsymbol{Q}}\tau_1, \tag{6}$$

with $U_{ji} = U_{ij}^\dagger$. The invariant gauge group (IGG) of this state is Z_2, so all but Z_2 gauge fluctuations are gapped.[9,10] Moreover, the PSG leaving the ansatz invariant is composed of the following conventional symmetry group elements g and the associated local gauge transformation V_g:

$$\begin{array}{cccccc} g = & P_x, & P_y, & P_d, & T. \\ V_g = & (-)^{x_i} i\tau_1, & (-)^{y_i} i\tau_2, & \frac{i}{\sqrt{2}}(\tau_1+\tau_2), & (-)^{x_i+y_i}. \end{array} \tag{7}$$

In the above, $P_{\mathrm{x,y,d}}$ are mirrors sending $x \to -x$, $y \to -y$ and $x \leftrightarrow y$, respectively, and T is time reversal which acts on U_{ij} as $i\tau_2 U_{ij}^*(-i\tau_2) = -U_{ij}$.[9,10] This PSG can be labeled as Z2A$xy(12)n$,[9,10] not realized in previous VQMC or self-consistent MF studies.

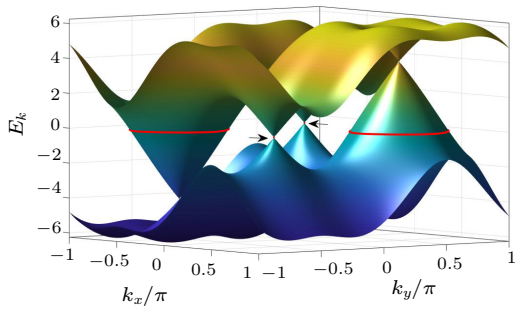

Fig. 3. Spinon dispersion, for $S_{\boldsymbol{Q}} = 1.7$ as an illustration. The thick red lines are the Fermi pockets and the arrows indicate a pair of Dirac nodes at the Fermi level.

Substituting the transformed ansatz Eq. (6) into Eq. (2), we obtain the spinon dispersion $E_{\boldsymbol{k}} = 2(\sin k_x + \sin k_y) \pm 2|S_{\boldsymbol{Q}}|\sqrt{\cos^2 k_x + \cos^2 k_y}$. This is gapless, and in fact there are two Fermi pockets enclosing $\pm\boldsymbol{Q}/2$, see Fig. 3. In addition, at the Fermi level there are two Dirac nodes at $\pm(\pi/2, -\pi/2)$. Importantly, the spinon Fermi surfaces and Dirac nodes are protected by the above PSG, and this serves as an indicator of the quantum order in such a gapless Z_2 QSL.[9,10]

We now discuss the fate of the above QSL. Even if we assume that the spinons are free from the coupling to the massive gauge fields, residual interactions between spinons can induce an instability toward Néel ordering in the presence of perfect nesting between the spinon Fermi surfaces, which eventually gaps out the spinons. We see an interesting example that the massiveness of gauge field fluctuations in a Z_2 QSL is insufficient to guarantee its stability against magnetic ordering.

Finite doping.—We have also performed systematic VQMC simulations at finite doping. In Fig. 4(a) we show the energy versus hole doping in the various variational states. We find that the energies in the dS_0 and $dS_0 + dT_{\boldsymbol{Q}}$ states are degenerate within statistical error, and so are the $pS_{\boldsymbol{Q}}$ and $pS_{\boldsymbol{Q}} + pT_0$ states. In contrast, the energy becomes poorer (not shown) if we get rid of the singlet components, in both the cases. This further enforces our view that the relevant pairing states are all singlets: dS_0 and $pS_{\boldsymbol{Q}}$. Moreover, while in the undoped case we find that the $pS_{\boldsymbol{Q}}$ (or $pS_{\boldsymbol{Q}} + pT_0$) state has lower energy, at finite doping we find that the dS_0 (or $dS_0 + dT_{\boldsymbol{Q}}$) state is systematically more favorable, down to the lowest nonzero doping we accessed. In fact, by linear interpolation the transition between these two types of states would be at a tiny hole doping level. We conjecture that the $pS_{\boldsymbol{Q}}$ state at zero doping is not adiabatically connected to the dS_0 state at finite doping.

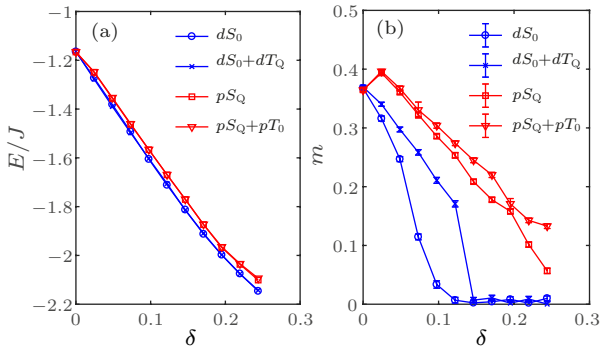

Fig. 4. The doping dependence of (a) energy per site and (b) the Néel moment, in the various types of variational states. The results are obtained in a lattice with $N = 82$, and are qualitatively robust in larger lattices.

Figure 4(b) shows the average Néel moment versus the hole doping. We find that the energy may be degenerate in the d-wave cases, or in the p-wave cases, as shown in (a), the moment will differ if the triplet component is included. For example, the Néel moment is larger in the $dS_0 + dT_{\boldsymbol{Q}}$ state than the dS_0 state, and similarly for $pS_{\boldsymbol{Q}} + pT_0$ versus $pS_{\boldsymbol{Q}}$. On the other hand, the Néel moment is suppressed more quickly in the d-wave cases. Taking the lower-energy $dS_0 + dT_{\boldsymbol{Q}}$ state at finite doping, we believe the harmlessness of the triplet component $dT_{\boldsymbol{Q}}$ to the optimized energy points to soft triplet PDW fluctuations and spin fluctuations in the underdoped regime. The dominant dS_0 is just the d-wave singlet pairing, well perceived in doped cuprates. The secondary $dT_{\boldsymbol{Q}}$ state may become important at higher energy scales (e.g., above the superconducting transition temperature), where the umklapp scattering of such pairs may be the key

process to generate the pseudogap near the antinodes in underdoped cuprates.[19] Unfortunately, this is already beyond the scope of VQMC for the ground state.

The numerical simulations were performed in High-Performance Computing Center of Collaborative Innovation Center of Advanced Microstructures, Nanjing University.

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