Superconductivity and Quantum Spin Disorder in Cuprates

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A fundamental connection between superconductivity and quantum spin fluctuations in underdoped cuprates is revealed. A variational calculation shows that *Cooper pair hopping* strongly reduces the local magnetization m_0. This effect pertains to recent neutron scattering and muon spin rotation measurements in which m_0 varies weakly with hole doping in the poorly conducting regime, but drops precipitously above the onset of superconductivity.

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When holes are introduced into the copper oxide planes of high T_c cuprates, spin and charge correlations change dramatically. The *local* magnetization m_0, measured by muon spin rotation [1] and elastic neutron scattering [2] on, e.g., $La_{2-x}Sr_xCuO_4$ reveals a qualitative difference between the insulating and superconducting phases: m_0 is rather insensitive to doping in the poorly conducting regime $0 \le x \le 0.06$, but drops precipitously above the onset of superconductivity at $x > 0.06$, becoming undetectable at optimal doping $x \approx 0.15$. Theoretically, holes can cause *dilution* and *frustration* [3] in the Heisenberg antiferromagnet, which create spin textures: either random ("spin glass") or with ordering wave vector away from (π, π) (sometimes called "stripes") [4]. However, the apparent reduction of local magnetization by the onset of superconductivity is a novel and poorly understood effect. Theory must go beyond purely magnetic models and involve the superconducting degrees of freedom.

We find that this problem is amenable to a variational approach, using hole-doped resonating valence bonds (RVB) states, originally suggested by Anderson for the spin correlations of high T_c cuprates [5–7].

These RVB states are excellent trial states for doped Mott insulators, with large Hubbard repulsion U: (i) Configurations with doubly occupied sites are excluded. (ii) Marshall's sign criterion for the magnetic energy [8] is satisfied, and Heisenberg antiferromagnetism at zero doping is accurately recovered. (iii) For doped systems, spin and charge correlations are parametrized independently, without explicit spin or gauge symmetry breaking.

These are important advantages over commonly used spin density wave, Hartree-Fock, and BCS wave functions for the antiferromagnet, metal, and superconducting phases, respectively. RVB states permit an *unbiased* determination of ground state spin and charge correlations appropriate for the cuprates.

A phenomenological low energy effective Hamiltonian is used, with two major components: Heisenberg interaction for spins, and single or Cooper pair hopping kinetic energy for fermion holes.

Our key results are as follows: (i) For the magnetic energy alone, the local magnetization m_0 is *weakly dependent* on doping concentration. This holds independently of interhole correlations for either randomly localized or extended states. (ii) In contrast to (i), m_0 is strongly reduced by the kinetic energy of *Cooper pair hopping*, which correlates the reduction of m_0 with the rise of superconducting stiffness, and hence [9] the transition temperature T_c.

Our results agree with the experimentally reported correlation between m_0 and T_c [1,2]. This relation appears to be *weakly* dependent on the precise hole density. A brief discussion concludes the paper.

Wave functions.—The hole-doped RVB states are compactly defined by

$$\Psi[u, v; x] = \mathcal{P}_G(x)\bar{\psi}[u, v],$$

$$\bar{\psi}[u, v] \equiv \exp\left(\sum_{ij} (v_{ij} f_i^\dagger f_j^\dagger + u_{ij}(a_i^\dagger b_j^\dagger - b_i^\dagger a_j^\dagger)\right)|0\rangle, \quad (1)$$

$$\mathcal{P}_G(x) \equiv \delta\left(\sum_i n_i^f - xL^2\right) \times \prod_i \delta(n_i^a + n_i^b + n_i^f - 1),$$

where $a_i^\dagger, b_i^\dagger$ and $f_i^\dagger$ are Schwinger bosons and hole fermions, respectively [10], $i = 1, \ldots, L^2$ is a site index on the square lattice, and $\mathcal{P}_G(x)$ is the Gutzwiller projection onto states with no double occupancies. As a result of the projection, Ψ can be written as a sum over bond configurations of singlets and hole pairs which cover the lattice as depicted in Fig. 1.

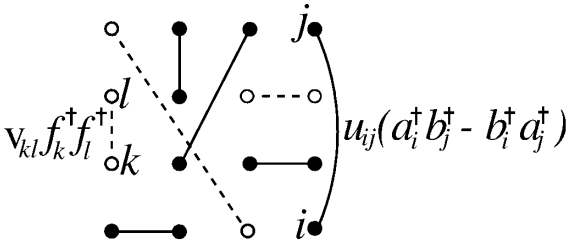

FIG. 1. A bond configuration in the doped RVB states $\Psi[u, v]$. Solid (empty) circles represent spins (holes) with bond correlations u_{ij} (v_{kl}).

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$u(\mathbf{r}_{ij})$ and $v(\mathbf{r}_{ij})$ are *independent* spin and hole bond parameters, respectively. $u_{ij} \geq 0$ connects i on sublattice A to $j \in B$, respectively, which ensures Marshall's sign.

The expectation value of an observable O is computed by a sum over loop coverings:

$$\langle O \rangle = \sum_{\gamma,\Lambda_\gamma} W_{\Lambda,\gamma} O_{\Lambda,\gamma}\,,$$
$$W_{\Lambda,\gamma} = \frac{1}{\langle \Psi[u,v]\,|\,\Psi[u,v]\rangle} \times \det_{ij\in\gamma}{}^{2}|v_{ij}| \prod_{\lambda\in\Lambda_\gamma}\left(2\prod_{(i,j)\in\lambda} u_{ij}\right). \quad (2)$$

Λ_γ denotes a list of directed loops $\{\lambda_\alpha\}$ which cover the lattice except for subset γ of xL^2 hole sites. W are positive Boltzmann weights, with which the Monte Carlo Metropolis step is defined following Refs. [6,7]. Ergodicity and convergence of our program was checked against precise transfer matrix results [11].

The Gutzwiller approximation (GA) amounts to dropping the projector $\mathcal{P}(x)$ in state (1) and setting $\Psi \to \bar{\psi}$, after adjusting the overall normalization of u and v to satisfy the global constraints $\langle n_i^a + n_i^b \rangle = (1-x)$, and $\langle n_i^f \rangle = x$. $\bar{\psi}$ is a Fock state of decoupled spins and holes, with easily computable correlations [12,13].

For the spin correlations we use power law decaying functions $u_p(r) \equiv 1/r^p$. The single variational parameter p determines the long range spin correlations and local magnetization [14]. [Other forms for $u(r)$, with qualitatively similar results, will be described elsewhere [13].]

We discuss four cases of interhole correlations:

$$v_{\rm ins}^{\gamma}(\mathbf{r}_{ij}) = \begin{cases} 1 & (i,j) \in \gamma\,, \\ 0 & (i,j) \notin \gamma\,, \end{cases}$$
$$v_{\rm met}(\mathbf{r}) = 1/L^2 \sum_{\mathbf{k}\in\Sigma} v_{\rm met}(\mathbf{k}) e^{-i\mathbf{k}\cdot\mathbf{r}}, \quad (3)$$
$$v_\alpha(\mathbf{r}) = \sum_{\hat{\eta}} c_\alpha(\hat{\eta})\delta_{\mathbf{r},\hat{\eta}}, \qquad \alpha = s,d\,,$$

where $|v_{\rm met}(\mathbf{k})| = 1$, $\hat{\eta}$ are nearest neighbor vectors on the square lattice, $c_s = 1$, and $c_d = \hat{\eta}_x^2 - \hat{\eta}_y^2$.

$v_{\rm ins}^{\gamma}$ puts the xL^2 holes on random sites. This state describes an insulator with disordered localized charges. $v_{\rm met}$ has filled Fermi pockets Σ, containing xL^2 occupied $\mathbf{k}$ states centered around $(\pm\pi/2, \pm\pi/2)$ in the Brillouin zone [15]. It describes weakly interacting holes in a "metallic" state. In real space, $v_{\rm met}(\mathbf{r}_{ij})$ decays slowly as $\sim r^{-3/2}$. Correlations in this state were previously computed by Bonesteel and Wilkins [7]. v_s and v_d describe tightly bound hole pairs in relative s- and d-wave symmetry, respectively.

Order parameters.—The local magnetization $m_0(L)$ is defined by $m_0^2 = 1/L^2 \sum_j \langle \mathbf{S}_i \mathbf{S}_j \rangle_p e^{-i(\pi,\pi)\mathbf{r}_{ij}}$. In Fig. 2(a), $m_0^2(p)$ for $\Psi[u_p, v; x = 0.1]$ is plotted for various choices of v. The GA (solid line) seems to work well for $m_0(p)$. Finite size scaling in Fig. 2(b) indicates

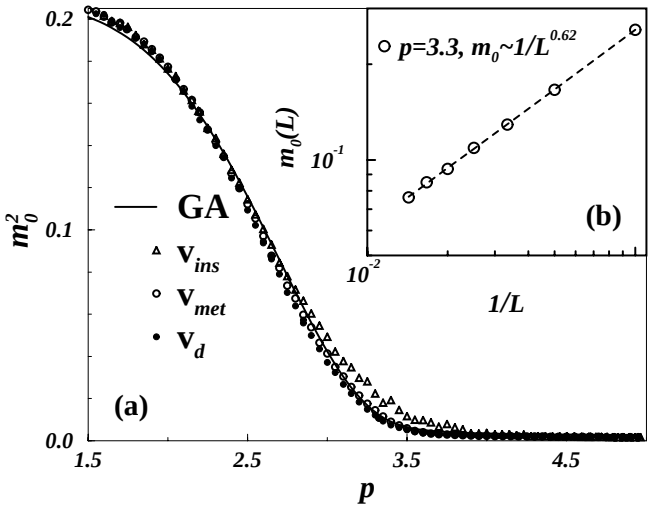

FIG. 2. (a) The local magnetization squared of doped RVB wave functions $\Psi[u_p, v]$ versus the variational power p, defined by the bond parameters $u_p(r) = 1/r^p$. Lattice size is 40×40, and hole concentration is 10%. Results agree well with the Gutzwiller approximation (solid line). The hole bond parameters v are defined in Eq. (3). Note that m_0^2 is weakly dependent on v; the data for $v = v_s$ overlap that of v_d. (b) Finite size scaling of $m_0(L)$ for $p = 3.3$ which indicates vanishing local magnetization at $L \to \infty$.

vanishing long range order $m_0 \to 0$ at $p_c = 3.3$, which lowers the bound given previously by Ref. [6]: at $p_c \leq 5$. The GA at $L \to \infty$ suggests that $p_c \leq 3$ [13].

The superconducting singlet order parameters are

$$\Delta_i^{s,d} = \sum_{\hat{\eta}} c_{s,d}(\hat{\eta}) \Delta_{i,i+\hat{\eta}}\,,$$
$$\Delta_{ij} = f_i^\dagger f_j^\dagger (a_i b_j - b_i a_j)/\sqrt{2}\,. \quad (4)$$

By gauge invariance imposed by the Gutzwiller projector, $\langle \Delta_{s,d} \rangle = 0$. However, $\Psi[u, v_{s(d)}; x > 0]$ describe true s- (d)-wave superconductors as seen by the (off-diagonal) long range order in $\Delta_{s,d}$ [13].

In contrast, the insulator states $\Psi[u, v_{\rm ins}, x]$ and the metallic states $\Psi[u, v_{\rm met}, x]$ have no long range superconducting order of either symmetry [13].

Effective Hamiltonians.—Magnetic order is driven by the diluted Heisenberg model [14],

$$\mathcal{H}^J = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j (1 - f_i^\dagger f_i)(1 - f_j^\dagger f_j)\,, \quad (5)$$

where, e.g., $S^x + iS^y \equiv a^\dagger b$. In Fig. 3 the expectation value $E_{\rm mag}(p) = \langle \mathcal{H}^J \rangle$ is plotted as a function of $m_0^2(p)$ for $x = 0.1$ and various choices of v from (3). Within numerical errors all states minimize $\mathcal{H}^J$ at around $p_{\rm min} \approx 2.7$, which by Fig. 2(a) yields local magnetization of $m_0^2 = 0.08 \approx m_0^2(0)(1 - 2x)$, where $m_0(0)$ agrees with the ground state local magnetization of the undoped Heisenberg model [16]. We have found that $p_{\rm min} \approx 2.7$ appears to be independent of x for $0 \leq x \leq 0.15$. Thus we conclude that, aside from the trivial kinematical

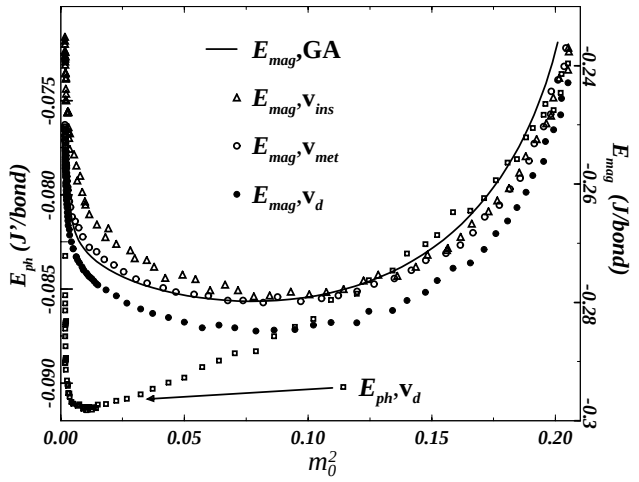

FIG. 3. The magnetic energy $E_{\rm mag}$ (5) and Cooper pair hopping energy $E_{\rm ph}$ (7) versus local magnetization squared m_0^2, used as a variational parameter. The density of holes is 0.1 and lattice size is $L = 40$. The magnetic energy is minimized at $m_0^2 \approx 0.08$, consistent with a diluted quantum Heisenberg antiferromagnet, and is weakly dependent on inter-hole correlations. The points of $E_{\rm mag}(v_s)$ overlaps $E_{\rm mag}(v_d)$. In contrast, Cooper pair hopping prefers *vanishing* m_0 at $L \to \infty$.

constraints, *the hole density and correlations have little effect on the magnetic energy at low doping.*

A single hole hopping in the antiferromagnetic background has been shown by semiclassical arguments [10,17] to be effectively restricted at low energies to hopping between sites on the same sublattice: Next we consider the single hole hopping process

$$\mathcal{H}^{t'} = \sum_{\langle ik\rangle \in A,B} t'_{ik} f_i^\dagger f_k (a_k^\dagger a_i + b_k^\dagger b_i), \qquad (6)$$

where i, k are removed by two adjacent lattice steps, and $t' > 0$. Unconstrained, the single hole ground state of $H^{t'}$ has momentum on the edge of the magnetic Brillouin zone, in agreement with exact diagonalization of $t - J$ clusters [18]. Previous investigations have found that *intersublattice* hopping (the t term in the $t - J$ model), is a high energy processes in the antiferromagnetic (AFM) correlated state [10,17]. We thus expect the same to hold even in RVB spin liquids with strong short range AFM correlations but no long range order. The primary effects at low doping may be to shift the ordering wave vector.

The single hole hopping (6) prefers the metallic state $v = v_{\rm met}$ over the superconductor $v = v_s, v_d$ [13]. It also prefers longer range $u(r)$ and thus actually *enhances* magnetic order at low doping. This is a type of a Nagaoka effect, where mobile holes separately polarize each of the sublattices ferromagnetically.

Now we consider Cooper pairs hopping terms

$$\mathcal{H}^{J'} = -J'\left(\sum_{ijk} \Delta_{ij}^\dagger \Delta_{ik} + \sum_{\langle ij\rangle,\langle \tilde{i}\tilde{j}\rangle} \Delta_{ij}^\dagger \Delta_{\tilde{i}\tilde{j}}\right). \qquad (7)$$

The first term is derived from the large U Hubbard model to order $J' = t^2/U$ [10]. It includes a *rotation* of the singlet pair, which prefers v_d over v_s. The second term is a parallel *translation* of singlets. It prefers superconductivity with $v = v_d$ over metallic states with $v = v_{\rm met}$ [19].

In Fig. 3 the ground state energy $E_{\rm ph}$ of (7) is plotted for $v = v_d$, $x = 0.1$, and $L = 40$. For $v = v_s$, $E_{\rm ph} > 0$. The variational energy is minimized at $p = 3.35$, which by the finite size scaling of Fig. 2(b) indicates vanishing m_0 at large L. Note the striking difference between the minima of $E_{\rm mag}$ and $E_{\rm ph}$. *Thus, Cooper pair hopping drives the ground state toward a spin liquid phase.*

A simple explanation is that pairs can hop with greater overlap when parallel bonds have maximum *singlet* components. When $u(r)$ is longer ranged, triplet contributions are larger, which inhibits pair delocalization. Incidentally, the Gutzwiller approximation fails to predict this effect since it decouples the local correlations between spins and hole pairs.

Since $\mathcal{H}^{J'}$ is the effective model which drives superconductivity it produces phase stiffness, which in the continuum approximation is given by

$$\mathcal{H}^{J'} \approx \frac{V_0}{2} \int d^2x (\nabla \phi_i)^2. \qquad (8)$$

The stiffness constant V_0 can be determined variationally from the RVB states by imposing a uniform gauge field twist on the bond parameters $v_{i,j} \to v_{i,j} \exp[i(x_i + x_j)\phi/2L]$ and measuring $E_{\rm ph}(\phi)$ to find $V_0 = d^2 E_{\rm ph}/d\phi^2$.

Following Ref. [9], at low doping for the square lattice V_0 is roughly equal to T_c.

In Fig. 4 we show our main result: The staggered magnetization m_0 for $\mathcal{H}^J + \mathcal{H}^{J'}$ is plotted against the superconducting to magnetic stiffness ratio V_0/J for different doping concentrations $x = 0.05, 0.1, 0.15$.

Two primary observations are made: (i) The local magnetization is sharply reduced at relatively low superconducting stiffness (and T_c/J). (ii) The relation between m_0 and V_0/J appears to be weakly dependent on the precise hole concentration.

Because of finite size uncertainty, m_0 in Fig. 4 is an *upper* bound on the thermodynamic local magnetization. The GA extrapolation suggests that m_0 may actually vanish already at $V_0/J \geq 0.2$. This is in qualitative agreement with the doping dependent of the local magnetization measured by Refs. [1,2], which diminishes rapidly above the onset of superconductivity.

In a quantized theory of stripes [20], mechanisms for diminishing m_0 assume anisotropic magnetic couplings, or fluctuating antiphase domain walls. A direct connection between superconductivity and m_0 is not obvious in these approaches.

In a recent *projected* SO(5) theory [21], spins and hole-pair dynamics have been considered with excluded

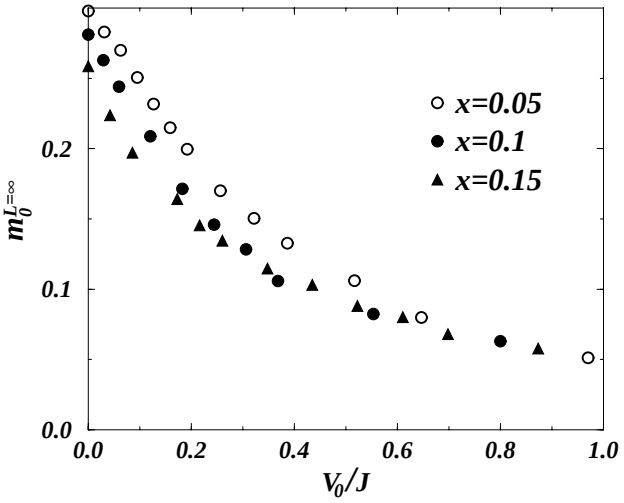

FIG. 4. The relation between thermodynamic local magnetization $m_0^{L=\infty}$ and superconducting phase stiffness V_0 (related to T_c; see text). J is the Heisenberg exchange energy. The points are considered *upper bounds* on m_0, which may even vanish for $V_0/J \geq 0.2$.

double occupancies. A variational relation is obtained between superconducting stiffness and the magnetic order parameter, which resembles the results of this paper.

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