Charge pair hopping and Bose-Einstein condensation in underdoped Mott insulators

Sanjoy K. Sarker and Timothy Lovorn
Department of Physics and Astronomy
The University of Alabama, Tuscaloosa, AL 35487

Recently, a renormalized Hamiltonian has been derived by continuing spin states from the Mott limit of the t-J model to the underdoped region. We show that it naturally leads to a pairing mechanism in which the pair has a dual character. Its spin part is a spinon singlet which condenses at $T^*$. The charge part is a real-space holon pair formed at $T_p < T^*$, which undergoes Bose-Einstein condensation at $T_c < T_p$. While neither is observable separately, the combination is. The mechanism is consistent with the small superfluid density, the decline of $T_c$ at small doping, and the existence of pairs above $T_c$ in cuprates, indicated by the observation of diamagnetism and Nernst effect.

PACS numbers: 74.20.Mn, 74.72.-h, 71.27.+a, 74.20.-z.

In a conventional superconductor the metallic state is characterized by quasielectrons, which form spatially overlapping Cooper pairs. In contrast, the elementary excitations in the metal phase of a hole-doped cuprate superconductor are hidden, and their nature unknown, which raises the questions: what is paired and how? Here we address this issue using the t-J model, which describes electrons of concentration $1 - x$ hopping on a lattice such that no site is doubly occupied. At half filling ($x = 0$), the system is a Mott insulator, with electrons localized as moments (or spins) which interact antiferromagnetically. Unexpectedly, the metallic state in cuprates is two dimensional (2d), even though these are (layered) 3d materials. Anderson argued that in 2d electrons remain localized even for $x > 0$, metallic conduction results from the motion of positively charged spinless holons. It is then natural to invoke continuity and construct a theory of the doped region by continuing the spin states from the insulator [1]. Experimental support comes from the fact that the carrier concentration is $x$, not $1 - x$. Also, despite the reappearance of nodal quasielectrons below $T_c$, the $T = 0$ superfluid density $\rho_s(0) \sim x^2$ [2], which is suggestive of holon pairing.

In the t-J model, hopping by a localized electron can be described as an exchange of a spin-1/2 particle (spinon) and a spinless holon, and the AF interaction as an exchange of two spinons. This reflects the $U(1)$ gauge symmetry of the model. The electron field is then represented as $c^\dagger_{i\sigma} = b^\dagger_{i\sigma} h_i$, where $b^\dagger_{i\sigma}$ creates a spinon of spin $\sigma$ and $h_i$ destroys a holon, such that the particle number at each site $i$ is conserved and equals 1. The ground state at half filling is known: it is a mixture of the Arovas-Auerbach valence-bond (VB) state, in which bosonic spinons are paired into singlets, and the Neel state [3,4]. The VB state survives the destruction of AF order up to a temperature $T^*$. However, despite many attempts, the connection with the doped phase (with fermionic holons) has not been established theoretically.

Recently, we have solved the continuation problem and derived a renormalized Hamiltonian valid for small $x$, assuming that hole motion prevents long-range magnetic order beyond some small $x_c$ [5,6]. This implies that spin excitations are gapped since spinons are bosons. The new Hamiltonian has the symmetry of the original model, which allows us to determine its phases by continuing all the contiguous spin phases from half-filling. The approach is blind: no experimental inputs are introduced by hand. Thus, we avoid the universal practice of assuming two dimensionality of the normal state; instead it emerges as a consequence of the theory [6]. This is fundamental since 2d confinement is thought to be responsible for the unusual behavior. It provides a serious test since it is almost impossible to confine metallic conduction in the presence of 3d hopping; it has not been demonstrated in previous theories.

Initially no new order is introduced, so that the phases are fully constrained by the symmetries at half filling. Yet, we obtain exactly two normal phases for small $x$ as in cuprates [7], with properties that match experiments, but disagrees with other theories (see, [6] for details). (1) Recent experiments have shown that, for small $x$, the high-$T$ strange-metal phase actually behaves like an insulator, with no Drude peak and a resistivity exceeding the Mott maximum, consistent with dynamically localized charge carriers [8]. In our case, since the high-$T$ phase has the symmetry of the Hamiltonian, it is automatically an insulator, as holons are localized by gauge forces. (2) For $T < T^*$, the pseudogap temperature, holons form a spinless Fermi liquid of concentration $x$, in agreement with recent experiments [8,11]. (3) The renormalized Hamiltonian has a pair hopping term, which automatically leads to $d$-wave superconductivity. (4) There is strong evidence for a spin gap [2] which causes a downturn in the paramagnetic susceptibility below $T^0 > T^*$.

Here we take a closer look at pairing. We show that ours is necessarily a strong-coupling Hamiltonian which, together with low density, induces holons to form real space pairs below $T_c$. These undergo a Bose-Einstein condensation (BEC) [12] below $T_c < T_p$, which is qualitatively consistent with the observed downturn of $T_c$ at small $x$, and diamagnetism [13] and Nernst effect [14] above $T_c$. A key prediction is a composite gauge-invariant “Cooper pair”, which is observable above and below $T_c$. It is a well-defined excitation of momentum $\mathbf{q}$ and energy $\omega(\mathbf{q})$, whose charge is carried by a mobile (bound) holon pair, which picks up the local phase of spinon sin-
glets (representing localized electrons) already condensed below $T^*$. The $t$-$J$ Hamiltonian for a layered 3d system is given by

$$H = - \sum_{ij,\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} - 2 \sum_{ij} J_{ij} A_{ij}^\dagger A_{ij}. \quad (1)$$

The first term describes electron hopping from $j$ to $i$ such that no site is doubly occupied, the second is the exchange interaction between spins. Here, $A_{ij} = \frac{1}{2}[b_i b_j - b_i^\dagger b_j^\dagger]$ destroys a spinon singlet, and $J_{ij} = 4t_i^2/U$, where $U$ is the Hubbard repulsion. Minimally, we need nearest-neighbor ($t$) and next-nearest-neighbor hopping ($t'$) within each plane, and nearest-neighbor out-of-plane hopping $t_z << t$. For cuprates, $t'/t \sim 3 - 4$. In general, $t'/t$ is small, so is $J' = (t'/t)^2 J$, and will be neglected.

As described in [6], renormalization proceeds in two steps. Hole motion is violently opposed by AF correlations, which localizes the hole in a small region, within which it hops rapidly. The renormalized hole - bare hole plus surrounding spins - moves slowly through the lattice, preventing AF order. By continuity, the Hamiltonian has the same form as the original one, with renormalized parameters $t_{eff}, J_{eff}$ etc, except that spinons have a gap $\Delta_s$. The spin gap is the singlet breaking scale $\Omega \sim 2\Delta_s$. At small $x$, we expect $J_{eff}$, $\Omega$ to scale with $J$. One-hole calculations suggest that for $t > J$, in-plane hopping amplitude $t_{eff} < J$, and also scales with $J$. The spin gap allows us to decouple unpaired spinons from hopping, and derive a minimal Hamiltonian involving sublattice-preserving hopping of renormalized holons and singlets only. Now, due to the gap, singlets form within the plane since $J_z/J = (t_z/t)^2 << 1$ and a spinon belongs to one singlet at a time. As a renormalized hole hops it breaks a singlet. For hops within the plane, the excess energy $\Omega$ is removed if the hole makes a second hop, and the singlet is reconstructed. Eliminating the intermediate state, we get

$$H_2 = -t_s \sum_{ijml,z} C_{ijml}^\dagger(z)C_{ijml}(z) + h.c., \quad (3)$$

where $t_{sz} = t_z^2/\Omega$, and $C_{ij} = (c_{ij\uparrow}c_{ij\downarrow} - c_{ij\downarrow}c_{ij\uparrow})/2 = -A_{ij}F_{ij}^\dagger$ destroys a singlet made from physical electrons, which is equivalent to destroying a spinon singlet and creating a holon pair with $F_{ij} = h_i^\dagger h_j^\dagger$. The first term describes intraplane, and the second, interplane pair hopping. The full Hamiltonian is gauge invariant and has the symmetries of the original model, plus an additional symmetry: total number of holes in each sublattice is conserved.

Since the high-$T$ phase has full symmetry (nonordered) holons are localized by gauge forces. A pseudogap metal appears below $T^*$ as the spinon singlets condense, allowing holons to propagate coherently. It is connected to the VB state at $x = 0$, and is characterized by the order parameter

$$A_{ij} = << A_{ij} = Ae^{\pm iQ \cdot (\vec{r}_i - \vec{r}_j)}, \quad (4)$$

or, its gauge-related concepts, where $Q$ is the two-sublattice wave vector. This state has a quantum lattice order: on average singlets connect spinons on opposite sublattices. If we replace $A_{ij}$ by its average $A_{ij}$, the single-hole term [Eq. 2] describes free holon hopping within the sublattice, with a spectrum $c_h = -2t_h + 2t_h \sin k_x + \sin k_y)^2 + 4D_h t_s A^2 \sin k_x \sin k_y$, where $t_h = t_s A^2(1 - x)$. The last term is the Hartree contribution from the pair hopping term, and $D_h < x$ is the average hole hopping amplitude. At low-$T$, the pseudogap phase is thus a spinless Fermi liquid of concentration $x$. The corresponding small Fermi pockets are not gauge invariant, but have been seen indirectly in de Hass-van Alphen type experiments [11].

The condensate part of the pair-hopping term becomes

$$H_{2h} = -t_s A^2 \sum_{ijml,z} [F_{ij}(z)F_{ml}(z) + h.c.]$$

$$-t_{sz} A^2 \sum_{ij,z} [F_{ij}^\dagger(z)F_{ij}(z + 1) + h.c.]. \quad (5)$$

It clearly leads to 3d superconductivity via pair condensation, so that $F_{ij} = F_{ij}^\dagger = \neq 0$. The electron pair amplitude is then $< C_{ij} >= - < A_{ij} >= F_{ij}^\dagger \neq 0$. A $T = 0$ MF analysis showed that it is a robust $d$-wave, essentially due to the symmetry of the VB state at $x = 0$.

The MF approximation clearly would not work for $T > 0$ since the intraplane pair-hopping energy scale $t_s$ is essentially the same as that for single-hole hopping, $(t_s(1 - x))$ (Eqs. 2, 3) because they arise from the same singlet breaking mechanism. It would induce holons to form real-space pairs, particularly since (a) at small $x$, pairs would not overlap, and (b) they can reduce energy further by delocalizing in the $z$-direction. Here we show this by analyzing pair fluctuations using functional integral methods [12] and focusing on $T \geq T_c$. 

A holon pair field is written as a two-component vector field: $F_{ij}(\tau) = F_{ij,\eta}(\tau)$, where $\eta = (x,y)$. Let $F_{ij}(\tau) = \frac{i}{\sqrt{\eta^2}} \sum_{\rho} e^{i(k_{\rho} r_{\eta} + \eta / 2) - i\omega \tau} F_{ij}(k,\omega)$, where $\eta$ is the unit vector, and $p = (k,\omega)$. The Hamiltonian density is given by

$$H = \sum_p \xi(k) h_p^* h_p - \sum_{pq} E_{\eta}(k) F_{\eta}^*(p) F_{\eta}(p),$$

where $\xi(k) = \epsilon(k) - \mu_h$, and

$$E_{x,y}(k) = 2t_0 \cos k_{xy} + 2t_{z0} \cos k_z,$$

is the hopping energy for a holon pair. Here $t_0 = t_x A^2$, and $t_{z0} = t_z A^2$. We introduce an order-parameter field $\Delta_\eta(p)$, and do a Hubbard-Stratonovich transformation to obtain the action $S = S_0 - \sum_{\rho} [\Delta_\eta^*(p) \Delta_\eta(p) - \sqrt{E_{\eta}(k)} \Delta_\eta^*(k) F_{\eta}(p) + c.c.]$, where $S_0$ is the free (quadratic) holon part. Integrating out the holons and keeping terms to second order in $\Delta$ we obtain the effective action describing pairing fluctuations:

$$S_{fl} = -\sum_{\rho \rho'} [\delta_{\rho \rho'} - (E_{\eta}(k) F_{\eta}(k))^{1/2} \Pi_{\eta \rho}(\Delta_\eta(p) \Delta_\rho(p)),$$

where $\Pi_{\eta \rho}(\Delta_\eta(p) = \langle F_{\rho}^*(p) F_{\eta}(p) \rangle$ is the pair correlation function for noninteracting holons, which is given by

$$\Pi_{\eta \rho}(q, \omega) = -\frac{1}{N} \sum_k \frac{\sin k_x \sin k_y}{\sin \omega - \xi(k + q/2) - \xi(k - q/2)} [\tanh \frac{\xi(k + q/2)}{2T} + \tanh \frac{\xi(k - q/2)}{2T}].$$

The sum is over the 2d Brillouin zone and $k_B = 1$.

The thermodynamic potential is given by $\Omega = \Omega_0 + \Omega_\Delta$, where $\Omega_0$ is the free holon part, and $\Omega_\Delta = T \sum_{\rho} \ln(1 - \lambda_+(p)(1 - \lambda_-(p))]$, is the fluctuation contribution, with

$$\lambda_\pm = \frac{1}{2} [E_{xx} \Pi_{xx} + E_{yy} \Pi_{yy} \pm (E_{xx} \Pi_{xy} - E_{yy} \Pi_{yx})^2 + 4E_{xy} \Pi_{xy} \Pi_{yx}]^{1/2}.$$

Since $E_x(0) = E_y(0) = 2t_0 + 2t_{z0}$, we obtain, for $p = 0$ (noting that $\Pi_{xy} < 0$) $\lambda_\pm = E_x(0)[\Pi_{xx}(0) \mp \Pi_{yx}(0)]$. The order-parameter equation is given by $1 = \lambda_+(0)$, the larger eigenvalue, at $T_c$ it reads

$$\frac{1}{t_0 + t_{z0}} = \frac{1}{N} \sum_k (\sin k_x - \sin k_y)^2 \frac{\tanh(\xi(k)/2T_c)}{\xi(k)}.$$

This corresponds to a $d$ wave. The holon density is given by $x = x_1 + x_2$, where

$$x_1 = \frac{1}{2N} \sum_k [1 - \tanh(\xi(k)/2T)]$$

is the contribution from free holons, and

$$x_2 = -N^{-1} \partial \Omega_\Delta / \partial \mu_h$$

is the fluctuation contribution.

Solving Eqs. (11-13) we obtain $\mu_h$ and $T_c$ for small $x$. We take the bottom of the holon band to be at zero. In the MF (BCS) approximation fluctuations are neglected ($x = x_1$). The MF $T_c$, which we denote by $T_p$, is a measure of the pair binding energy, and remains finite as $x \to 0$, as shown in Fig. (1). When fluctuations are included, real-space pair states appear as poles of the two-holon Green’s function, i.e., as zeroes of $Re(1 - \lambda_\pm(q, \omega)$, which are at $\omega = \omega_\pm(q) = E_{pp}(q) - \mu_p$. (We have made an analytic continuation to real frequencies). Here, $E_{pp}$ is the energy of the pair, $\mu_p$ is the pair chemical potential, such that $\omega_\pm(q) \geq 0$. We need to consider only the lower pair band $E_{pp}(q)$ which has states below the free holon-holon ($h-h$) continuum, therefore are long lived. Once formed, the states near the bottom of the band (which is at $q = 0$), are quickly occupied by holons so that $x_1$ is negligible, and $x \approx x_2$. Then, $\mu_h \sim E_{pp}(0)/2$ is negative, and binding energy is $\sim 2|\mu_h|$. Bose condensation occurs for $\omega = \omega_+(q) = 0$, so that $\mu_p = E_{pp}(0) \sim 2|\mu_h|$. Numerical solution shows that, for small $x$, $|\mu_h| \sim t_s >> T_c$, so that pairs are strongly bound. Since pairs are bosons we need to consider only states near $q = 0$. It is easily shown that $\lambda_\pm$ is symmetric in the $q_x, q_y$ plane. Then for small $q$ the pair spectrum has the form

$$\omega_+(q) = a(q_x^2 + q_y^2) + b q_z^2,$$

which gives

$$T_c \approx 4.66(x_d^{2/3}(a b^2)^{1/3}.$$

The parameters $a, b$ also depend on $T_c$, and indirectly on $x$, and obviously $x_1 = x - x_2$ is not zero. However, these corrections arise from free holons, which require a finite energy $= 2|\mu_h|$ to produce. For small $T_c/|\mu_h|$, we obtain $x_1 \approx e^{-|\mu_h|/T_c} Z_1$, where $Z_1 = \sum_k e^{-\epsilon_h(k)/T_c}$. We find

$$T_c \approx 4.66(x_d^{2/3}(a b^2)^{1/3}$$

and

$$\omega_+(q) = a(q_x^2 + q_y^2) + b q_z^2,$$

which gives

$$T_c \approx 4.66(x_d^{2/3}(a b^2)^{1/3}.$$
from numerical calculations that $a \sim t_0$, and $b$ scales is $t_{3d} \ll t_0$. Since, $T_c$ also depends on $x_2 \sim x$, $T_c/\mu_n = (x^2 t_{3d}/t_0)^{1/3}$ is small, and $x_1$ vanishes exponentially. Fig. 1 shows $T_c$ and $T_p$ as a function of $x$.

The bound pairs continue to exist up to $T_p > T_c$. Holons are gapped below $T_p$, but there is no sharp transition. Now, a mobile holon pair is hidden, as it is not gauge invariant. However, in the present case, it becomes observable through the gauge-invariant physical electron (singlet) pair represented by: $G_{ij} = -A_{ij}F_{ij}$. Ordinarily the electron pair Green’s function $G_{\eta\eta'}^{\text{pair}}(p) = -\langle C_{\eta\eta'}(p)C^*_{\eta\eta'}(p) \rangle$ is incoherent since it is a convolution. However, a coherent part emerges as the spinon singlets condense below $T^*$. Replacing $A_{ij}$ by its mean-field value, we obtain $G_{\eta\eta'}^{\text{pair}}(p) = A^2G_{\eta\eta'}(-p)$, where $G_{\eta\eta'}^{\text{pair}}(p) = -\langle F_{\eta}(p)F^*_{\eta'}(p) \rangle$. To determine $G^F$ we use the exact relation $(E^*_{\eta}E_{\eta'})^{1/2} < F^*_{\eta}F_{\eta'} > = \delta_{\eta\eta'} + < \Delta^*_\eta\Delta_{\eta'} >$. This leads to

$$G_{\eta\eta'}^{\text{pair}}(p) = -A^2Z_{\eta\eta'}(-k) + \text{incoherent part}, \quad (14)$$

which is a key prediction. Here

$$Z_{\eta\eta'} = \left(\frac{u^*_{\eta}(\eta')}{E^*_{\eta}\omega_{\eta}}\right)^{1/2} \frac{\partial \lambda_+/\partial \omega}{\partial \lambda_+/\partial \omega},$$

evaluated at $\omega = \omega(k)$; $u_{\pm}(\eta)$ are the eigenvectors which diagonalize the effective action [Eq. 8], yielding eigenvalues $\lambda_{\pm}$. Eq. (14) describes a well-defined physical (gauge-invariant) pair excitation. However, unlike a Cooper pair, it is observable above and below $T_c$, and represents both electrons and holes. Since the pole is at negative energy $-\omega_+(\bar{k})$, the actual excitation represents a mobile hole pair of momentum $\mathbf{k}$ and energy $\omega_+(\mathbf{k})$, of charge $2e$. The spin part consists of a spinon singlet (representing a localized electron pair). These are already condensed below $T^*$, and tend to overlap because of high density $(1-x)$. With increasing $x$ and/or $T$, the excitation will be less sharp due to broadening by phase (gauge) fluctuations associated with $A_{ij}$, and the factor $A^2$, which vanishes for $T \geq T^*$.

These results resolve a number of puzzling issues. Holon pairing implies that $\rho_+(0) \sim x$. Superconductivity by BEC explains the decline of $T_c$ with decreasing $x$. That $T_c$ does not vanish at some $x_{SC} \sim x_c$ is not surprising since the renormalized Hamiltonian is not valid near $x_c$, where the spin gap is small, AF correlations are longer ranged and compete with superconductivity. Experimentally, $T_c \propto \rho_+(0)$, i.e., both vanish at same $x_{SC}$, as if the effective carrier density is $x - x_{SC}$. Early reports indicated that $\gamma = 1$[10]; however, recently $\gamma = 0.61$ has also been reported[17], which is not far form $2/3$. The existence of charged bound pairs above $T_c$ has been shown to account[18] for the observed diamagnetism[13]. On the other hand, since the spin part of the pair is condensed below $T^* > T_c$, and the associated phase (gauge) fluctuations couple to holon pairs, vortex type excitations are likely to exist. These may account for the observed Nernst effect[12], although more work would be needed to sort out these issues. Interestingly, in this theory, spinon singlet condensation is two-dimensional, so that the transition at $T^*$ is probably Kosterlitz-Thouless type, hence the lack of singularities. Then phase fluctuations are also two-dimensional. The theory is consistent with the observed decoupling between spin and charge responses. As $T$ decreases, the paramagnetic susceptibility decrease starting at the spin-gap temperature $T^0$, and seemingly unaffected by charge pairing below $T_p$ and superconductivity at $T_c$[11,12]. Finally, though not considered here, nodal electrons can appear as collective excitations[12], but would not directly take part in pairing.

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