Absence of superconductivity in the half-filled band Hubbard model on the anisotropic triangular lattice

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We report exact calculations of magnetic and superconducting pair-pair correlations for the half-filled band Hubbard model on an anisotropic triangular lattice. Our results for the magnetic phases are similar to those obtained with other techniques. The superconducting pair-pair correlations at distances beyond nearest neighbor decrease monotonically with increasing Hubbard interaction $U$ for all anisotropy, indicating the absence of frustration-driven superconductivity within the model.

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The possibility of superconductivity (SC) in the doped square-lattice Hubbard model continues to be discussed in the context of cuprate superconductors. Theoretical results for the strength of superconducting pair correlations in this model are controversial, and arguments for [1, 2] and against [3, 4] long range order (LRO) both exist in the literature. Yet another class of superconductors which have been discussed using the Hubbard model are the organic charge-transfer solids (CTS) [5], which are known to have strong electron-electron interactions [6]. SC in the CTS is reached from proximate insulating states not by doping, but by application of pressure at constant carrier density. It is therefore natural to expect that a simple change of parameters within a model appropriate for the insulating states of the CTS can give SC. The proponents of spin fluctuation and resonating valence bond models for SC in the CTS suggest that precisely such an insulator-to-superconductor transition can occur within the 1/2-filled band triangular lattice Hubbard model with increasing frustration [7, 8, 9, 10, 11, 12, 13, 14, 15, 16].

The frustrated Hubbard model has been applied most commonly to the CTS belonging to the the $\kappa$-(BEDT-TTF)$_2$X ($\kappa$-(ET)$_2$X) family with the highest critical temperature. In these materials, the ET molecules form anisotropic triangular lattices of dimer pairs [3]. While $\kappa$-(ET)$_2$X is formally 3/4-electron filled (1/4-hole filled), considering the ET dimers as unit cells the effective band-filling becomes 1/2. It has been suggested that pressure modifies the interdimer electron hoppings and changes the degree of frustration [17]. Experimentally, both an antiferromagnetic (AFM) state [18] in systems with relatively large anisotropy, and a spin liquid state [10] for nearly isotropic triangular lattices, have been found as well as SC. Theoretically, strong enough frustration within the Hubbard model destroys the AFM order, giving a paramagnetic metal (PM). Within theories of SC mediated by AFM fluctuations, the SC occurs in a narrow region sandwiched between the AFM and PM. [7, 8, 10, 11, 12, 13, 14, 17, 16].

We consider the Hamiltonian,

$$H = -t \sum_{\langle ij \rangle, \sigma} (c_{i,\sigma}^\dagger c_{j,\sigma} + H.c.) - t' \sum_{\langle kl \rangle, \sigma} (c_{k,\sigma}^\dagger c_{l,\sigma} + H.c.) + U \sum_i n_{i,\uparrow} n_{i,\downarrow}$$

In Eq. 1, $c_{i,\sigma}^\dagger$ creates an electron with spin $\sigma$ ($\uparrow, \downarrow$) on site $i$, $n_{i,\sigma} = c_{i,\sigma}^\dagger c_{i,\sigma}$. $U$ is the on-site Hubbard interaction.

The lattice structure is the conventional square lattice (hopping integrals $t$) with additional hopping integrals $t'$ across the $x + y$ diagonals for a total of three bonds per site. The ratio $t'/t$ is 0 for the square lattice and 1 for the isotropic triangular lattice. In what follows, we express all quantities in units of $t$.

Accurate solutions to Eq. 1 are difficult to reach using existing analytical approaches. Many of the techniques that find SC are based on mean-field embedded cluster methods that use extremely small cluster sizes, including some as small as 4 or 8 sites [14, 16]. Eq. 1 and a variant with two diagonal hoppings have recently been investigated within a numerically more precise Path

[FIG. 1: (color online) Correlation functions as a function of $U$ for $t'=0.5$. Solid line: bond order $B'$ along the diagonal bonds. Dashed line: $S_x(\pi, \pi)$. Both are discontinuous at the AFM–PM transition at $U \approx 6.1$. Lines are guides to the eye based on calculations performed on a grid of size $\Delta U = 0.1$.]
Integral Renormalization Group (PIRG) technique [20]. The phase diagrams found by these authors contain PM, AFM, and nonmagnetic insulator (NMI) phases only and suggest implicitly the absence of SC along the AFM–PM boundary. The authors, however, did not calculate the SC pair-pair correlations. Recent exact diagonalization calculations also investigated the AFM–PM phase boundary, but did not calculate pair-pair correlations [21]. Specifically because of the difficulty of calculating SC pair-pair correlations, and the prediction of SC occurring only over a narrow region of the phase diagram, it is essential that SC correlations are calculated explicitly with high precision over the complete phase space.

We report precisely such calculations within Eq. 1 via exact diagonalization in the present Letter. We focus here solely on \(\frac{1}{4}\)-filling, numerical convergence for which is reached much faster than in doped systems. Although exact diagonalizations cannot necessarily establish the occurrence of SC, they can test precisely the necessary conditions for SC. First, if the model does indeed have a SC ground state with pair-pair correlations exhibiting LRO in the thermodynamic limit, finite clusters should necessarily exhibit short range order (SRO) in the pair-pair correlations. Second, if SC driven by the Hubbard \(U\) interaction is present, then there must exist some parameter region where \(U\) enhances the pair-pair correlations relative to \(U = 0\). Note that for the one strongly-correlated model in which it is known that SC exists, the negative-U Hubbard model, both the above conditions are met [22]. We show here that neither of the above criteria are met within Eq. 1 for repulsive \(U\) at \(\frac{1}{4}\)-filling. For all \(U\) and anisotropy, the SC pair correlations become monotonically weaker with increasing \(U\).

We solve exactly the ground state of Eq. 1 on a periodic 4×4 lattice. We calculate the spin structure factor \(S_\sigma(q)\) and the bond order \(B_{ij}\) between sites \(i\) and \(j\),

\[
S_\sigma(q) = \frac{1}{N} \sum_{r,r'} e^{iq(r-r')} \langle (n_{i\uparrow} - n_{i\downarrow})(n_{j\uparrow} - n_{j\downarrow}) \rangle
\]

\[
B_{ij} = \sum_\sigma \langle c_{i\sigma}^\dagger c_{j\sigma} + H.c. \rangle
\]

In Eq. 2 \(N\) is the number of lattice sites. We define the standard pair-creation operator \(\Delta^{\dagger}_i\)

\[
\Delta^{\dagger}_i = \frac{1}{\sqrt{2}} \sum_\nu g(\nu)(c_{i\uparrow\nu}^\dagger c_{i\downarrow\nu} - c_{i\downarrow\nu}^\dagger c_{i\uparrow\nu}^\dagger)
\]

In Eq. 3 the phase factor \(g(\nu)\) determines the symmetry of the superconducting pairs. For \(s\)-wave pairing \(g(\nu)\) is +1 for the four nearest-neighbor pairs \(i+x, i+y, i-x, i-y\) and \(i-x, i+y, i-x, i+y\); for \(d_{x^2-y^2}\) pairing \(g(\nu)\) alternates as +1,-1,+1,-1 for the same four sites; and for \(d_{xy}\) pairing \(g(\nu)\) alternates sign over the four sites \(i+x+y, i+x-y, i-x+y\). We will calculate the pair-pair correlation function as a function of distance, \(P(r) = \langle \Delta^{\dagger}_i \Delta_\nu \rangle\).

We first discuss the non-SC phases within the model. In the limit \(t' = 0\), the ground state of Eq. 1 has AFM LRO for any nonzero \(U\). This is usually determined from plots of \(S_\sigma(q)\) versus \(q\), which exhibit strong peaks at \(q = (\pi, \pi)\). For \(t' \neq 0\), the ground state is AFM only when \(U\) exceeds a critical value \(U_c(t')\) and remains a PM for smaller \(U\). We determine \(U_c\) from calculations of \(S_\sigma(\pi, \pi)\), the bond order corresponding to the diagonal \(t'\) bonds \((B')\), and the ground state expectation value of \(d_{x^2-y^2}\) SC phase is claimed to be broadest [16]. \(U_c\) is identified from a discontinuous decrease in \(B'\) accompanied by a simultaneous increase in \(S_\sigma(\pi, \pi)\).

The double occupancy \(d\) also decreases discontinuously at the same \(U_c\) (not shown). The large decrease of \(B'\) and \(d\) indicate loss of electron mobility associated with a metal-insulator (MI) transition, while the sharp increase in \(S_\sigma(\pi, \pi)\) indicates the AFM nature of the insulator. The simultaneous discontinuity of all observables at \(U_c\) is consistent with a first-order (discontinuous) quantum phase transition. The discontinuity continues to exist for \(t' = 1\). The size of the discontinuity in \(B'\) decreases as \(t'\) approaches 1; this is likely related to the appearance of the NMI phase as well as the change in the magnetic ordering at \(t' = 1\) we discuss below. We also performed calculations (not shown here) on a 12 site cluster, chosen so that \((\pi, \pi)\) AFM order is not frustrated at \(t' = 0\), and found comparable values of \(U_c\).

The magnetic ordering is qualitatively different for...
\( t' > 1 \), where \( S_\sigma(\mathbf{q}) \) peaks at \( \mathbf{q} = (\pi, 0) \) instead of \( (\pi, \pi) \) [20], with the amplitude of the peak increasing with \( U \). This transition is expected from the known results for the antiferromagnetic Heisenberg spin Hamiltonian with nearest \((J_1)\) and next-nearest \((J_2)\) neighbor exchange interactions (two diagonal bonds per site) [23]. Here \( J_2/J_1 = 0.5 \) is a quantum critical point, with \( \mathbf{q} = (\pi, \pi) \) dominating for \( J_2/J_1 < 0.5 \), and \( \mathbf{q} = (\pi, 0) \) for \( J_2/J_1 > 0.5 \) [24]. This value of \( J_2 \) corresponds to \( t' = 1/\sqrt{2} \) within the \( U \to \infty \) limit of the Hubbard model with two diagonal bonds, and this is where the transition to the \((\pi, 0)\) ordering is found [23]. From perturbation theory, the quantum critical transition to the \((\pi, 0)\) phase for \( U \to \infty \) at nearly the same value of \( t' \) (not shown). At \( U = 13.8 \) similar maxima occur in derivatives of \( B' \) and \( S_\sigma(\pi, 0) \). We therefore identify three distinct phases: a PM phase for \( U \leq 7.3 \), a nonmagnetic (NM) phase for \( 7.3 \lt U \leq 13.8 \), and the magnetically ordered \((\pi, 0)\) phase for \( U > 13.8 \). The transport behavior within the NM phase is difficult to determine from our finite-cluster calculations. The continuous decrease of \( B' \) and \( d \) in the intermediate \( U \) region is, however, consistent with an insulating character of this phase. We therefore identify the intermediate \( U \)-phase as the previously discussed NMI phase [20].

Mean field calculations finding SC generally agree that the pairing is of \( d_{x^2-y^2} \) symmetry for \( 0 < t' < 1 \). In Fig. [23(a)] - (c) we show the \( d_{x^2-y^2} \) singlet pair-pair correlation function \( P_{d_{x^2-y^2}}(r) \) as a function of distance \( r \) and Hubbard interaction \( U \), for \( t' = 0.2, 0.5 \) and \( 0.8 \), respectively. It is for the intermediate \( t' = 0.5 \) region that SC correlations have been claimed to be the strongest. The insets show the behavior of \( P_{d_{x^2-y^2}}(r) \) as a function of \( U \) for \( r = 2, \sqrt{3} \) and \( 2\sqrt{2} \). \( P_{d_{x^2-y^2}}(r) \) at the two larger values of \( r \) that we have chosen are least affected by the finite lattice size, since there is no overlap between pairs on the \( 4 \times 4 \) lattice for these \( r \).

Several features in the pair-pair correlations are immediately apparent. First, the pair-pair correlations decrease rapidly with distance, the values at the largest \( r \) being more than an order of magnitude smaller than those at \( r = 0 \). Second, like the other correlation functions (Fig. [1]), \( P_{d_{x^2-y^2}}(r) \) changes discontinuously at the PM–AFM transition. Finally, for \( r > 1 \), the pair-pair correlations are strongest at \( U = 0 \) and decrease monotonically with increasing \( U \). Pair-pair correlations at \( r = 0 \) are trivially enhanced with increasing \( U \). Nearest-neighbor correlations \( (r = 1) \) decrease with \( U \) for \( U < U_c \) for all \( t' \) but increase for \( U > U_c \) for large enough \( t' \). This increase is associated with the transition to AFM and occurs only on the insulating side of the PM–AFM boundary. Furthermore, the small increase in the \( r = 1 \) correlations is accompanied by a large simultaneous decrease in the long-range component, consistent with the entry into an insulating state (clearly seen in the \( U = 4 \) limit of the Hubbard model).
Based on the $q = (\pi, 0)$ magnetically ordering for $t' > 1$, it has been proposed that the SC correlations have pairing symmetry $d_{xy}$ or $s + d_{xy}$. In Fig. 3(d) we show the $U$-dependence for $d_{xy}$ pair-pair correlations at $t' = 1.1$, for the two separations where the pairs are nonoverlapping. Exactly as for $t' < 1$, we find no region of $U$ where the “long-range” $d_{xy}$ pair-pair correlations are enhanced compared to their $U = 0$ values. We have performed similar calculations for $s$ and $s + d_{xy}$ pairing symmetries, and find no enhancement for these either.

We give the phase diagram from our calculations in Fig. 4. We have not plotted data for $t' = 7$ data in Fig. 3(b). This behavior of the short-range correlations may be one reason embedded cluster methods using small cluster sizes find SC.

Our most important result is that SC pair-pair correlations do not exhibit even SRO within Eq. 1, in any channel and for any range of parameters. Theoretical models for SC in the CTS should either include electron-phonon interactions, or should be based on a different electronic model. We have recently suggested a correlated-electron theory of SC in the CTS that emphasizes their $\frac{1}{4}$-filled band nature, and the transition to the SC state from a density wave composed of local singlets [27]. Such a theory can explain naturally the recently observed transition from a valence-bond solid to the SC state [28].

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