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. Other superconductors that 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 $\frac{1}{2}$-filled band triangular lattice Hubbard model with increasing frustration [7–16].

The frustrated Hubbard model has been applied most commonly to the CTS belonging to the $\kappa-(\mathrm{BEDT}-\mathrm{TTF})_2X$ ($\kappa-(\mathrm{ET})_2X$) family with the highest critical temperature (BEDT-TTF denotes bis(ethylenedithio) tetrathiafulvalene, and ET is its abbreviation). In these materials, the ET molecules form anisotropic triangular lattices of dimer pairs [5]. While $\kappa-(\mathrm{ET})_2X$ is formally $\frac{3}{4}$-electron filled ($\frac{1}{4}$-hole filled), considering the ET dimers as unit cells, the effective bandfilling becomes $\frac{1}{2}$. 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 [19] 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 [7–16].

We consider the Hamiltonian,

$$H = -t\sum_{\langle ij\rangle,\sigma}(c^{\dagger}_{i,\sigma}c_{j,\sigma} + H.c.) - t'\sum_{[kl],\sigma}(c^{\dagger}_{k,\sigma}c_{l,\sigma} + H.c.) + U\sum_{i} n_{i,\uparrow}n_{i,\downarrow}. \quad (1)$$

In Eq. (1), $c^{\dagger}_{i,\sigma}$ creates an electron with spin σ ($\uparrow$, $\downarrow$) on-site i, $n_{i,\sigma} = c^{\dagger}_{i,\sigma}c_{i,\sigma}$. 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]. Equation (1) and a variant with two diagonal hoppings have recently been investigated within a numerically more precise Path 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 $\frac{1}{2}$-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 where 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 $\frac{1}{2}$-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 $S_\sigma(\mathbf{q})$ and the bond order B_{ij} between sites i and j,

$$S_\sigma(\mathbf{q}) = \frac{1}{N}\sum_{\mathbf{r}\mathbf{r}'} e^{i\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}')}\langle(n_{\uparrow\mathbf{r}} - n_{\downarrow\mathbf{r}})(n_{\uparrow\mathbf{r}'} - n_{\downarrow\mathbf{r}'})\rangle \quad (2)$$

$$B_{ij} = \sum_\sigma \langle c^\dagger_{i,\sigma} c_{j,\sigma} + H.c.\rangle. \quad (3)$$

In Eq. (2), N is the number of lattice sites. We define the standard pair-creation operator $\Delta^\dagger_i$

$$\Delta^\dagger_i = \frac{1}{\sqrt{2}}\sum_\nu g(\nu)(c^\dagger_{i,\uparrow} c^\dagger_{i+\nu,\downarrow} - c^\dagger_{i,\downarrow} c^\dagger_{i+\nu,\uparrow}). \quad (4)$$

In Eq. (4), 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 sites $i + \hat{x}$, $i + \hat{y}$, $i - \hat{x}$, and $i - \hat{y}$; 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 $i + \hat{x} + \hat{y}$, $i + \hat{x} - \hat{y}$, $i - \hat{x} - \hat{y}$, $i - \hat{x} + \hat{y}$. We will calculate the pair-pair correlation function as a function of distance, $P(r) = \langle \Delta^\dagger_i \Delta_{i+\vec{r}}\rangle$.

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 $S_\sigma(\mathbf{q})$ versus $\mathbf{q}$, which exhibit strong peaks at $\mathbf{q} = (\pi, \pi)$. 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 the double occupancy $d = \langle n_{i,\uparrow} n_{i,\downarrow}\rangle$. In Fig. 1, we plot $S_\sigma(\pi, \pi)$ and B' for $t' = 0.5$. It is in this region of t' that the $d_{x^2-y^2}$ 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 (M-I) 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 all $t' < 1$. 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 (π, π) AFM order is not frustrated at $t' = 0$ (lattice 12(a) in Fig. 2 of [21]), and found comparable values of U_c.

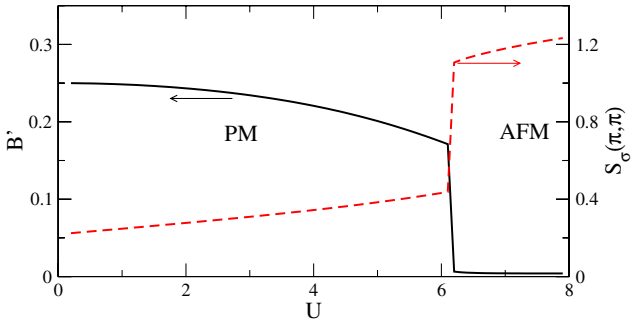

FIG. 1 (color online). Correlation functions as a function of U for $t' = 0.5$. Solid line: bond order B' along the diagonal bonds. Dashed line: $S_\sigma(\pi, \pi)$. Both are discontinuous at the AFM–PM transition at $U = 6.1$. Lines are guides to the eye based on calculations performed on a grid of size $\Delta U = 0.1$.

The magnetic ordering is qualitatively different for $t' > 1$, where $S_\sigma(\mathbf{q})$ peaks at $\mathbf{q} = (\pi, 0)$ instead of (π, π) [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 J_2/J_1 shifts to 1 when the number of diagonal bonds per site is one instead of two, corresponding to $t' = 1$ within the large U Hubbard model. This is exactly where we have found the transition in our numerical work.

In Fig. 2(a), we show B' and spin structure factor data for $t' = 1.1$ as a function of U. Unlike the discontinuous transition between the PM and AFM phases in Fig. 1, the correlation functions change continuously now. Importantly, the steepest decrease in B' and the changes in $S_\sigma(\mathbf{q})$, for both $\mathbf{q} = (\pi, \pi)$ and $\mathbf{q} = (\pi, 0)$, occur at different U, suggesting the existence of a third phase between the PM and the $(\pi, 0)$ magnetic order. In Fig. 2(b), we show the derivatives of B' and $S_\sigma(\pi, 0)$ with respect to U. At $U = 7.3$, a minimum occurs in dB'/dU, which is accompanied by a minimum in the derivative of the double occupancy d at nearly the same value of U (not shown). At $U = 13.8$, similar maxima occur in derivatives of B' and $S_\sigma(\pi, 0)$. We

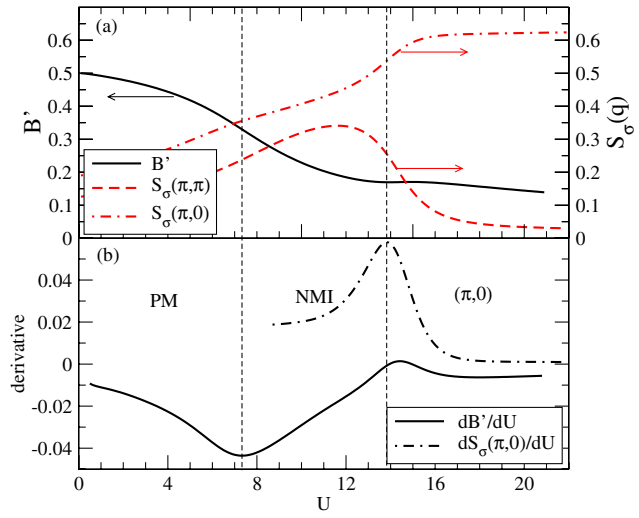

FIG. 2 (color online). Correlation functions as a function of U for $t' = 1.1$. (a) Diagonal bond order B' (solid line), $S_\sigma(\pi, \pi)$ (dashed line), and $S_\sigma(\pi, 0)$ (dot-dashed line). (b) Derivative of B' and $S_\sigma(\pi, 0)$ with respect to U. Vertical lines show estimated phase boundaries between PM, NMI, and $(\pi, 0)$ phases. Lines are guides to the eye and based on calculations performed on a grid of size $\Delta U = 0.1$; derivatives calculated with a three-point centered difference formula.

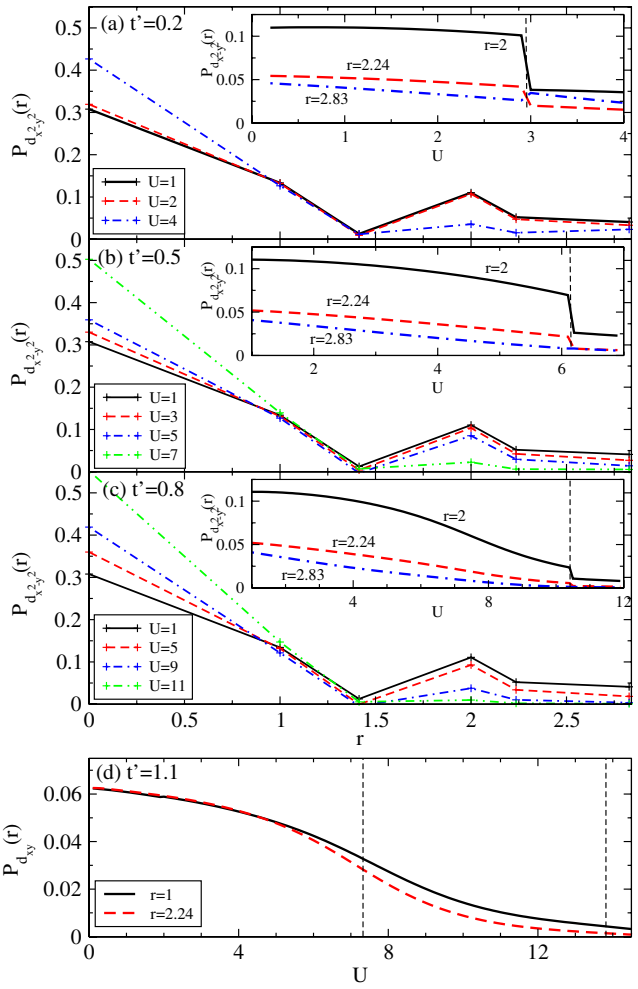

FIG. 3 (color online). (a)–(c) $d_{x^2-y^2}$ pair-pair correlation functions as a function of r for $t' = 0.2$, 0.5, and 0.8, respectively. Insets show U-dependence of $d_{x^2-y^2}$ pair-pair correlation functions for three different r. The PM–AFM transition is indicated by vertical lines. (d) U-dependence of the d_{xy} pair-pair correlations at two different r for $t' = 1.1$. Vertical lines mark PM–NMI and NMI–$(\pi, 0)$ transitions. Lines are guides to the eye and based on calculations performed on a grid of size $\Delta U = 0.1$.

therefore identify three distinct phases: a PM phase for $U \leq 7.3$, a nonmagnetic (NM) phase for $7.3 < U \leq 13.8$, and the magnetically ordered $(\pi, 0)$ phase for $U > 13.8$. Calculations for a 12 site 4×3 lattice (chosen to be compatible with columnar magnetic order) are also consistent with three distinct regions. In the PM phase, B' in the 4×4 lattice is larger than in the 4×3 lattice, as expected for a delocalized region at these system sizes. B' has nearly the same value between the two lattices in the NMI and $(\pi, 0)$ regions, hence indicating localization and the insulating character. Thus, the insulating character is reached at a U below the $(\pi, 0)$ magnetic transition.

Mean-field calculations finding SC generally agree that the pairing is of $d_{x^2-y^2}$ symmetry for $0 < t' < 1$. In Figs. 3(a)–3(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 $r = 2$, $\sqrt{5}$, 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 4×4 lattice for these r. For the 12 site cluster of reference [21] (see above), pair-pair correlations at fixed r also decrease monotonically with U, and the pair-pair correlation at the maximum r is smaller for 16 sites than 12 sites for all U.

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 $U = 4$ data in Fig. 3(a) and the $U = 7$ data in Fig. 3(b)]. This behavior of the short-range correlations

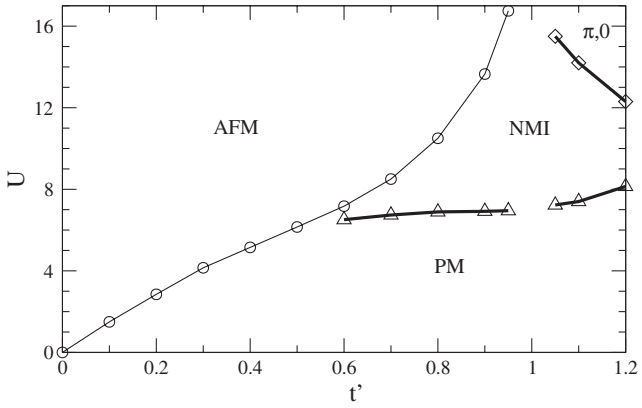

FIG. 4. Phase diagram as a function of t' and U. Lines are guides to the eye. The PM–AFM boundary is located to within $\Delta U = 0.1$ using the discontinuity of correlation functions. The PM–NMI and NMI–$(\pi, 0)$ phase boundaries are drawn from the inflection point in the correlation functions (see text.)

may be one reason embedded cluster methods using small cluster sizes find SC.

Based on the $\mathbf{q} = (\pi, 0)$ magnetically ordering for $t' > 1$, it has been proposed that the SC correlations have pairing symmetry d_{xy} or $s + d_{xy}$ [11,13,25]. 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 non-overlapping. 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' exactly 1 due to problems associated with degeneracy at this point. We have found that the NMI phase not only exists for $t' > 1$, but enters the phase diagram at approximately $t' \gtrsim 0.6$; for all $t' > 0.5$, inflection points as in Fig. 2 appear in B' and d. Our phase diagram for the insulating states and the boundaries between them are in good agreement with other numerical results [16,20,21,23,26]. We find, for example, that the minimum U necessary for the NMI phase is $U \sim 7$. This is very close to the boundary of $U \sim 6.7$ at $t' = 1$ found recently using a variational cluster approximation [26]. The data in Fig. 2 suggest that the PM–NMI transition is continuous [20], but confirmation of this would require finite-size scaling.

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]. Our theory can explain naturally the recently observed transition from a valence-bond solid to the SC state .

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