Comment on "Superconductivity and Mott Physics in Organic Charge Transfer Materials"

Menke *et al.* [1] recently claimed that superconductivity (SC) in the κ-phase organic charge-transfer solids (CTS) can be understood within the two-dimensional half-filled anisotropic triangular-lattice Hubbard model. Experimentally, κ-CTS are mostly but not always antiferromagnetic (AFM) at ambient pressure and SC appears under pressure. In apparent agreement with this observation, Menke *et al.* found AFM ground states for small t/U and SC over a small region at the interface of AFM and Fermi liquid ground states with increasing t/U at fixed t'/t, where U is the Hubbard repulsion. Menke *et al's* computational results directly contradict those obtained using exact diagonalization [2] and Path Integral Renormalization Group (PIRG) [3] approaches. It is clearly of interest to determine the origin of this discrepancy, especially in view of the facts that (a) related arguments continue to persist in the context of cuprate SC superconductivity (which however involves doping), and (b) there exist CTS in which SC is not proximate to AFM, but is separated by an intermediate charge-disproportionated phase [4]. Here we show that Menke *et al's* conclusion regarding SC is incorrect and originates from a flawed assumption.

Menke *et al.* established the superconducting ground state using Cluster Dynamic Mean Field Theory (CDMFT), within which momentum-dependent susceptibilities are calculated by summing over all sites of a seven-site cluster. This approach does not distinguish between Cooper pairs separated by short versus long distances. We have performed computations on the triangular lattice Hubbard Hamiltonian for 4×4, 6×4, and 6×6 lattices using exact diagonalization and PIRG [5, 6] for the same $t' = 0.4t$, $t = 1$ as Menke *et al.* Fig. 1 (for the 6×4 lattice the ground state occurs in the total spin $S = 1$ subspace; our calculations are for the lowest $S = 0$ state). For these clusters PIRG is essentially exact [5, 6]. With our choice of axes, $d_{x^2-y^2}$ pairing corresponds to the "d_{xy}" pairing of reference 1. We calculate U- and distance r-dependent pair-pair correlations $P(r) = \frac{1}{2}\langle\Delta_i^\dagger\Delta_{i+\vec{r}} + \Delta_i\Delta_{i+\vec{r}}^\dagger\rangle$,

$\Delta_i^\dagger = 8^{-\frac{1}{2}}\sum_\nu g(\nu)(c_{i,\uparrow}^\dagger c_{i+\nu,\downarrow}^\dagger - c_{i,\downarrow}^\dagger c_{i+\nu,\uparrow}^\dagger)$, $c_{i,\sigma}^\dagger$ creates an electron on site i with spin $\sigma = \pm\frac{1}{2}$, and the phase factor $g(\nu)$ alternates as +1, -1, +1, -1 for the four sites $i+\hat{x}$, $i+\hat{y}$, $i-\hat{x}$ and $i-\hat{y}$. In addition to $P(r)$ we also calculated the spin-structure factor $S(\pi,\pi)$ to determine the occurrence of AFM.

One essential criterion for SC within the model Hamiltonian is simple: superconducting pair-pair correlations must be enhanced over the $U = 0$ values over a minimal range of U. In Fig. 1(a) - (d) we have plotted $P(0)$, $P(1)$, $P(r^*)$ and $\bar{P} = (1/N_c)\sum_{r>2} P(r)$ against U for all three lattices, where r^* is the next-to-furthest possible separation R between two lattice points on the finite lattice (r* = 2.24, and 3.16, and 3.61 in units of lattice constants for the 4×4, 6×4 and 6×6 lattices, respectively), and N_c is the total number of $r > 2$ correlations. Use of r* instead of the furthest distance avoids finite-size effects associated with the latter on periodic clusters.

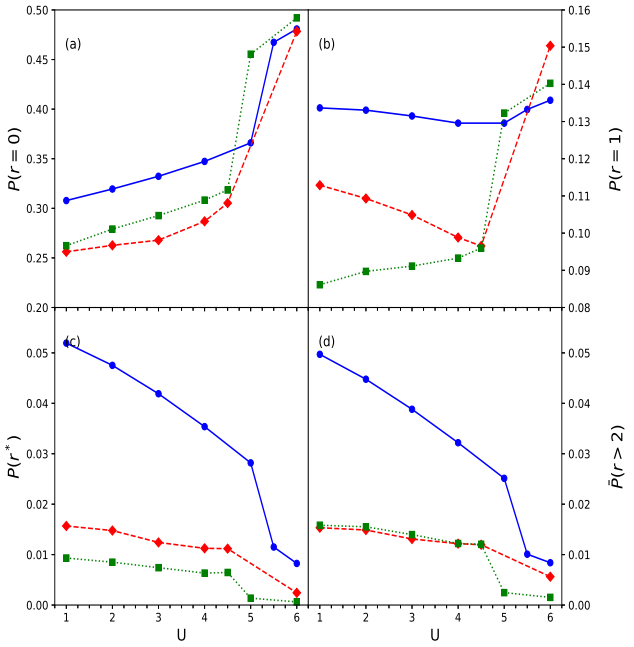

FIG. 1. $d_{x^2-y^2}$ pair-pair correlations for 4×4 (circles), 6×4 (diamonds), and 6×6 (squares) lattices. (a) r=0 (b) r=1 (c) r=r^*, and (d) $\bar{P}$ (see text).

Our results are identical for all three lattices. There is a sudden increase or decrease in $P(r)$ at a lattice-specific U_c, coincident with an increase in the spin structure factor indicating a transition to AFM. $P(0)$ increases with U in all cases while $P(1)$ increases with U for $U > U_c$. Both behaviors are directly determined by short-range antiferromagnetic spin correlations unrelated to SC. In contrast to these both $P(r^*)$ and $\bar{P}$ decrease continuously with U, starting from $U = 0$, and the behavior are qualitatively the same for both $U < U_c$ and $U > U_c$, clearly indicating absence of SC. Most importantly, the order(s) of magnitude larger magnitudes of $P(0)$ and $P(1)$ over the long-range counterparts explain why incorrect conclusions are reached within the CDMFT calculations: momentum-based calculations that place the same weight on short- versus long-range pair correlations *in a small cluster* are apt to give incorrect conclusions. Indeed, that going beyond the simple dimer Mott-insulator description is essential for understanding correlated-electron SC in the CTS has been clear since the discovery of pressure-driven AFM-to-charge disproportionation-to-SC transitions in β'-(BEDT-TTF)$_2$ICl$_2$, which can only be understood within a $\frac{3}{4}$-band filled description that takes into account of the charge degrees of freedom internal to the BEDT-TTF dimers. Interestingly, experiments on cuprates also

find charge-ordering in the pseudogapped state, perhaps also indicating that the explanation of SC requires going beyond the simplest Hubbard Hamiltonian.

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