

## Doping asymmetry in the three-band Hamiltonian for cuprate ladders: Failure of the standard model of superconductivity in cuprates

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The relevance of the single-band two-dimensional Hubbard model to superconductivity in the doped cuprates has recently been questioned, based on density matrix renormalization group (DMRG) computations that found superconductivity over an unrealistically broad doping region upon electron-doping, yet a complete absence of superconductivity for hole-doping. We report very similar results from DMRG calculations on a Cu<sub>2</sub>O<sub>3</sub> two-leg ladder within the parent three-band correlated-electron Hamiltonian. The strong asymmetry found in our calculations are in contradiction to the deep and profound symmetry in the experimental phase diagrams of electron- and hole-doped cuprate superconductors, as seen from the occurrence of quantum critical points within the superconducting domes in both cases that are characterized by Fermi surface reconstruction, large jumps in carrier density, and strange metal behavior.

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The mechanism of unconventional superconductivity (SC) found in the high- $T_c$  cuprates and other strongly correlated materials remains an outstanding problem in condensed matter physics, more than three decades after its discovery. At the heart of the problem is the choice of the minimal model for the CuO<sub>2</sub> planes that can account for SC. Since the work of Zhang and Rice, who showed that under certain limits the three-band model of the CuO<sub>2</sub> planes could be reduced to a simpler one-band Hubbard model [1], the majority of theoretical work has focused on the single-band Hubbard model, as well as even simpler approximations such as the  $t$ - $J$  model. While cluster variants of dynamical mean-field theory find SC in the doped single-band model on a square lattice [2–9], density matrix renormalization group (DMRG) and quantum Monte Carlo (QMC) calculations have detected the absence of long-range superconducting order [10–12].

An accurate description of the band structure of the cuprates within a one-band correlated-electron Hamiltonian requires inclusion of second neighbor hopping  $t'$  [13–16]. DMRG calculations have therefore been performed on quasi-one-dimensional cylinders for the  $t$ - $t'$ - $J$  model, where  $t'/t$  negative (positive) corresponds to hole (electron)-doped regimes. No signature of pairing is found in the negative  $t'/t$  region [17]. Surprisingly, the strong signature of dominant superconducting pair-pair correlations is found in the positive  $t'/t$  region, over a very broad range of electron-doping [17]. Enhanced pairing correlations in the electron-doped region have been confirmed from DMRG calculations on related extended  $t$ - $J$  models on one-band six-leg cylinders [18,19]. These results are exactly opposite to the experimental observation in real cuprates, where significantly higher  $T_c$  over a much broader doping region is found with hole doping. The

authors of [17] have subsequently extended their calculations to the parameter region with nonzero third neighbor hopping  $t''$  [19]. The absence of pairing in the hole-doped region and strong pairing tendency over very broad region of electron-doping persist within the  $t$ - $t'$ - $t''$ - $J$  model [20]. Quantum Monte Carlo calculations have claimed long-range superconducting correlations for both electron and hole doping at finite  $U$  in the  $U$ - $t$ - $t'$  Hamiltonian, with *stronger* pairing on the hole-doped side [21]. DMRG calculations for the same model contradict these results, however, and only find pairing on the electron-doped side [22]. The origin of the differences in these numerical results and the more serious discrepancy from experimental observations remain not understood.

The single-band model calculations suggest that there are potential problems with reducing the electronic structure of the CuO<sub>2</sub> planes to Cu-site-based effective models. Clearly a comparison of hole- versus electron-doped pairing tendencies within the full three-band correlated-electron Hamiltonian for the cuprates will be more useful in this context. We report here the results of high-precision DMRG computations on the three-band two-leg cuprate ladder over a wide range of hole- and electron-doping. The corresponding single-band Hubbard ladder has been widely investigated in the past [23–27]. The undoped (half-filled) single-band two-leg Hubbard ladder has spin-gapped ground state, with spins on the ladder rungs paired into singlets [28]. Doped holes or electrons (equivalent since the single-band Hubbard ladder has particle-hole symmetry) occupy ladder rungs in pairs, which is favored over unpaired charge carriers that would destroy two singlets instead of one. The ground state of the single-band ladder for weak to moderate doping consequently has a spin gap and exhibits singlet superconducting correlations with

quasi-long-range order [23–27,29]. The above result breaks down for the hole-doped three-band ladder, where a recent DMRG study showed that, even though a spin gap persists in the undoped state, superconducting correlations in the hole-doped decay *faster* than  $1/r$ , indicating dominance of charge over pairing correlations at long distances [30]. The decay of pair correlations in this case is caused by pair-breaking hole hopping between the O ions, and is strongest when both Coulomb interactions between holes on the same O and O-O hopping are included [30]. The doped holes in hole-doped cuprates primarily reside on oxygen sites; the results for the hole-doped ladder indicate a breakdown of the Zhang-Rice theory [1]. In what follows we compare hole- versus electron-doped three-band two-leg ladder within high precision calculations.

We consider the  $\text{Cu}_2\text{O}_3$  two-leg ladder Hamiltonian

$$\begin{aligned}
 H = & \Delta_{dp} \sum_{i\sigma} p_{i,\sigma}^\dagger p_{i,\sigma} + \sum_{\langle ij \rangle, \lambda, \sigma} t_{dp}^\perp (d_{i,\lambda,\sigma}^\dagger p_{j,\sigma} + \text{H.c.}) \\
 & + \sum_{\langle ij \rangle, \lambda, \sigma} t_{dp} (d_{i,\lambda,\sigma}^\dagger p_{j,\sigma} + \text{H.c.}) \\
 & + \sum_{\langle ij \rangle, \sigma} t_{pp} (p_{i,\sigma}^\dagger p_{j,\sigma} + \text{H.c.}) \\
 & + U_d \sum_{i,\lambda} d_{i,\lambda,\uparrow}^\dagger d_{i,\lambda,\uparrow} d_{i,\lambda,\downarrow}^\dagger d_{i,\lambda,\downarrow} \\
 & + U_p \sum_j p_{j,\uparrow}^\dagger p_{j,\uparrow} p_{j,\downarrow}^\dagger p_{j,\downarrow}. \quad (1)
 \end{aligned}$$

In Eq. (1)  $d_{i,\lambda,\sigma}^\dagger$  creates a hole with spin  $\sigma$  on the  $i$ th Cu-site on the  $\lambda$ th leg ( $\lambda = 1, 2$ ) of the ladder and  $p_{j,\sigma}^\dagger$  creates a hole of spin  $\sigma$  on the  $j$ th O  $p$  orbital. The O-ion can be located on a rung or either leg of the ladder. Parameters  $t_{dp}^\perp$  and  $t_{dp}$  are the nearest-neighbor (n.n.) Cu-O rung and leg hopping integrals, respectively, while  $t_{pp}$  is the n.n. O-O hopping integral. The phase relations between the orbitals (see Fig. S1 in the Supplemental Material [31]) determine the sign convention for the hopping integrals. We have taken all  $t_{dp}^\perp$  as negative, while  $t_{dp}$  and  $t_{pp}$  alternate signs along the length of the ladder.  $U_d$  ( $U_p$ ) is the Hubbard repulsion between hole pairs on Cu- $d$  (O- $p$ ) orbitals, and  $\Delta_{dp} = \epsilon_p - \epsilon_d$  is the site-energy difference between Cu- $d$  and O- $p$  orbitals. We consider ladders with  $L$  rungs and an open boundary condition, with rungs at both terminal ends. Calculations are for ladders up to  $L = 96$  (192 Cu and 286 O sites) and  $N$  holes, with the undoped state corresponding to one hole per Cu site ( $N = 2L$ ). For hole (electron) doping we add (remove) particles and define the hole (electron) doping fraction as  $\delta_h$  ( $\delta_e$ ) =  $N/(2L) - 1$  [ $1 - N/(2L)$ ]. In the following we make comparisons of three-band results with those obtained from single-band Hubbard ladders. The single-band Hubbard repulsion and the rung and leg hopping parameters are written as  $U$ ,  $t$ , and  $t^\perp$ , respectively. The single-band doping fraction is written as  $\delta$ .

We set  $|t_{dp}| = 1$  ( $t_{dp}^\perp = -1$ ) and take other Hamiltonian parameters from recent first-principles calculations,  $\Delta_{dp} = 3$ ,  $U_d = 8$ ,  $U_p = \{3, 4\}$ , and  $t_{pp} = \{0.5, 0.6\}$  [15,16]. These parameters are similar to commonly accepted values [32–34]. We employed an  $S_z$ -conserving DMRG algorithm using the

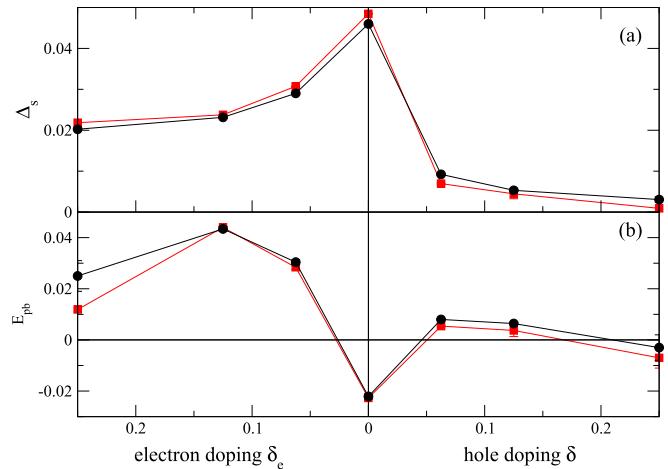


FIG. 1. (a) The doping dependence of the extrapolated spin gaps  $\Delta_s$  in the infinite-length limit ( $L \rightarrow \infty$ ). (b) Pair-binding energy  $E_{pb}$  as a function of doping (see text). Circles and squares are for  $(U_p, t_{pp}) = (3, 0.5)$  and  $(4, 0.6)$ , respectively. A transition to a band state with near-equal populations of charge carriers on Cu- and O-sites occurs at  $\delta_e$  larger than that shown here. Lines are guides to the eye.

ITENSOR library [35] with real-space parallelization [36]. We used a maximal bond dimension of up to 19 000, giving a truncation error of less than  $1 \times 10^{-7}$ . All results were extrapolated to the limit of zero truncation error (see [30] for examples of extrapolation).

The characteristic behavior of the two-leg ladder is determined by its spin gap  $\Delta_s$ . SC can occur only if the spin gap found in the undoped ladder persists under doping [26,29,37]. We calculated  $\Delta_s$  using finite-size extrapolation from ladders of lengths up to  $L = 64$ . Figure 1(a) shows the doping dependence of the  $L \rightarrow \infty$  extrapolated  $\Delta_s$ . For the undoped ladder, the behavior of  $\Delta_s$  against  $U_d/|t_{pd}|$  is very similar to that of the spin gap versus  $U/t$  in the single-band Hubbard ladder [23], with a maximum in  $\Delta_s$  for  $U_d/|t_{pd}| \approx 8$  [30]. However,  $\Delta_s$  behaves *qualitatively differently* for the electron versus hole-doped ladders within Eq. (1). For electron-doped ladders  $\Delta_s$  remains large over a wide doping range, while for hole doping  $\Delta_s$  decreases rapidly with doping. The normalized spin gap  $\tilde{\Delta}_s \equiv \Delta_s(\delta_e)/\Delta_s(\delta_e = 0)$  for the electron-doped ladder is comparable to  $\tilde{\Delta}_s$  for the single-band Hubbard ladder with  $U = 8$  and  $t^\perp = t$  [38]. For the single-band ladder,  $\tilde{\Delta}_s(\delta = 0.125) \approx 0.42$ , and is only slightly smaller at  $\delta = 0.25$  [38]; in comparison, for the electron-doped cuprate ladder with  $U_d = 8$ ,  $U_p = 3$ , and  $t_{pp} = 0.5$ ,  $\tilde{\Delta}_s(\delta_e = 0.125) = 0.49$ , and  $\tilde{\Delta}_s(\delta_e = 0.25) = 0.45$ . However, for hole doping,  $\tilde{\Delta}_s(\delta_h = 0.125) = 0.14$  and  $\tilde{\Delta}_s(\delta_h = 0.25) = 0.02$ .  $\Delta_s$  increases with increasing  $t_{pp}$  in the undoped three-band model [33]. This effect can be explained in the undoped case from perturbative calculations of the effective exchange  $J$  between n.n. Cu spins. About two-thirds of the contribution to  $J$  involves  $t_{pp}$ , demonstrating the critical role that the oxygen sublattice plays even in undoped cuprates [39]. Our DMRG results show that while  $\Delta_s$  increases with  $t_{pp}$  with electron doping,  $\Delta_s$  *decreases* with  $t_{pp}$  for hole doping. We also calculated the finite-size scaled pair-binding energy  $E_{pb}$  for both hole- and electron-doping,

defined as in [40],

$$E_{\text{pb}} = 2E(N_{\uparrow} - 1, N_{\downarrow}) - E(N_{\uparrow} - 1, N_{\downarrow} - 1) - E(N_{\uparrow}, N_{\downarrow}). \quad (2)$$

The calculated pair-binding energies, shown in Fig. 1(b), are consistent with the calculated  $\Delta_s$ .

The doped single-band two-leg ladder belongs to the Luther-Emery universality class, with gapped spin degrees of freedom and a single gapless charge mode [25,26,29,41]. For the three-band cuprate ladder we define the local charge density operator  $n_j$  for the  $j$ th unit cell as the sum of the charge density operators for the two Cu sites on a rung, the rung O, and two leg O sites. The charge correlation function is defined as  $C(r) = \langle n_i n_j - \langle n_i \rangle \langle n_j \rangle \rangle$ , where  $r \equiv |i - j|$  is the rung-rung distance. We define the superconducting pair-pair correlation function  $P(r) = \frac{1}{2}(\langle \Delta_i^\dagger \Delta_j \rangle + \langle \Delta_i \Delta_j^\dagger \rangle)$ , where  $\Delta_i^\dagger = \frac{1}{\sqrt{2}}(d_{i,1,\uparrow}^\dagger d_{i,2,\downarrow}^\dagger - d_{i,1,\downarrow}^\dagger d_{i,2,\uparrow}^\dagger)$  creates a spin singlet pair between Cu sites on the  $i$ th rung. In the Luther-Emery universality class, charge and pairing correlations decay as power laws in the long distance limit, with asymptotic behavior  $C(r) \sim r^{-K_\rho}$  and  $P(r) \sim r^{-1/K_\rho}$ , respectively. While true long-range superconducting order is absent in a one-dimensional system, for  $K_\rho > 1$  pair correlation decay with distance is slower than that of charge correlation and there is quasi-long-range superconducting order. Conversely, for  $K_\rho < 1$  charge correlations dominate over superconducting quasi-long-range order.

The direct approach to determine if superconducting correlations follow a power-law decay with distance involves fitting  $P(r)$  against  $r$ . To reduce finite-size effects caused by the open boundary conditions of our ladders [26,30], we calculate  $P(r)$  from an average of  $N_{\text{avg}}$  correlations of the same distance  $r$ , centered about the midpoint of the ladder. The results shown here use  $N_{\text{avg}} = 10$  ( $N_{\text{avg}} = 11$ ) for even (odd)  $r$ . In Fig. 2, we show the normalized pair-pair correlation function [ $P(r)/P(r = 1)$ ] for 96-rung ladders with  $U_d = 8$ ,  $U_p = 3$ ,  $t_{\text{pp}} = 0.5$ , and a range of dopings. We find that  $P(r)$  is well fit by a power law  $P(r) \sim r^{-\alpha}$  over a range of electron and hole dopings. As can be seen in Fig. 2, there is a very clear difference in the power-law exponent for hole versus electron doping, with a noticeably faster decay with distance for hole-doped ladders. For electron doping,  $\alpha < 1$  over a large range of doping, corresponding to a correlation exponent  $K_\rho > 1$ , which indicates quasi-long-range superconducting order. In contrast,  $K_\rho < 1$  for hole doping [30]. With increased hole doping, pair correlation decays *faster* with distance [30].

A more accurate approach to determining the correlation exponent  $K_\rho$  in DMRG calculations is to fit the charge density (Friedel) oscillations caused by the open boundaries of the ladder [26,42]. This method also permits more accurate extrapolation of  $K_\rho$  to the  $L \rightarrow \infty$  limit [26]. We use the following fitting function for the charge density  $n_k$  [26,30,42]:

$$n_k = n_0 + A \frac{\cos(N\pi k/L_{\text{eff}} + \phi)}{\sin(\pi k/L_{\text{eff}})^{K_\rho/2}}. \quad (3)$$

In Eq. (3)  $n_0$  is the background charge density,  $A$  the Friedel oscillation amplitude,  $\phi$  a phase shift, and  $L_{\text{eff}}$  an effective length. Typically  $L_{\text{eff}}$  is smaller than  $L$  to account for end effects [26]. The amplitude of the charge density oscillations at

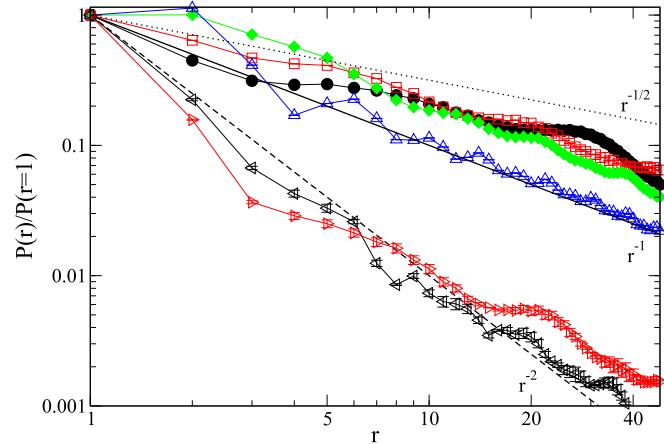


FIG. 2. Normalized pair-pair correlation function  $P(r)$  as a function of the rung-rung distance  $r$  for 96-rung ladders with  $U_p = 3$  and  $t_{\text{pp}} = 0.5$  for several electron dopings  $\delta_e$  and hole dopings  $\delta_h$ . Solid, dashed, and dotted lines are power laws  $r^{-1}$ ,  $r^{-2}$ , and  $r^{-1/2}$ , respectively. Circles, squares, diamonds, and up triangles correspond to electron dopings  $\delta_e = 0.0625, 0.0833, 0.125$ , and  $0.25$ , respectively. Right and left triangles are for the hole-doped ladder with  $\delta_h = 0.0625$  and  $0.125$ , respectively [30]. Lines are guides to the eye.

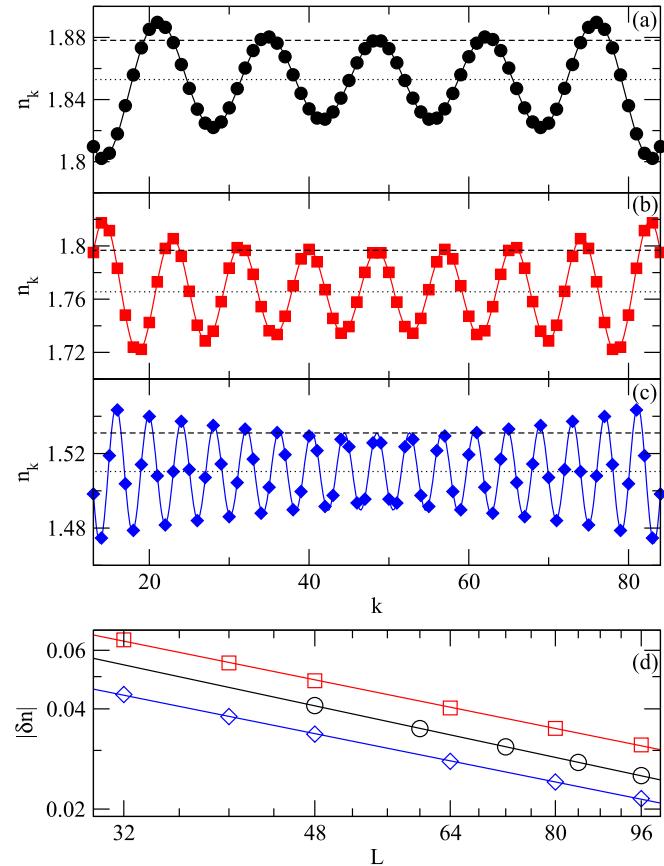
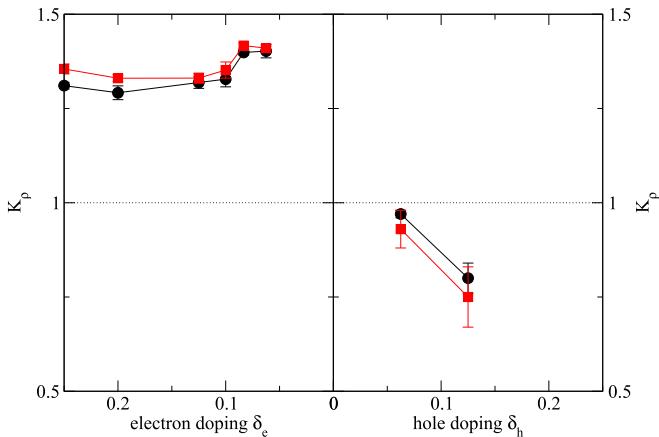


FIG. 3. The local charge density profile on a 96-rung ladder with  $U_p = 3$  and  $t_{\text{pp}} = 0.5$  for electron dopings  $\delta_e =$  (a) 0.0833, (b) 0.125, and (c) 0.25. The curves are fits to Eq. (3). Dotted and dashed lines represent  $n_0$  and  $n(L/2)$ . (d) Amplitude of Friedel oscillations at  $L/2$ ,  $\delta n$  (see text), as a function of ladder length  $L$ . The lines are linear fits. Circles, squares, and diamonds correspond to  $\delta_e = 0.0833, 0.125$ , and  $0.25$ , respectively.



relationship between the quantum criticality and superconductivity. Very recent research indicates that charge carriers in the strange metallic state of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  may be charge  $2e$  bosons [52]. All the above continue to be challenging within standard models of cuprate SC.

We end this Letter by pointing out that the quantum criticality and associated phenomena can be qualitatively understood within a valence transition theory of cuprates we recently proposed [53–55]. Within this theory, the Fermi surface reconstruction in both hole- and electron-doped compounds is due to the dopant-induced transition from positive to negative charge transfer gap state. The transition involves change in Cu-ion ionicity from  $\text{Cu}^{2+}$  to  $\text{Cu}^{1+}$ , resulting in the transfer of nearly all Cu-ion  $d_{x^2-y^2}$  holes to the O-ions. Similar quantum critical transitions between different ionicities have been widely discussed over four decades in the context of neutral-ionic transition in organic donor-acceptor charge-transfer solids [56] and heavy fermion systems [57]. Carrier densities of  $1 + p$  and  $1 - p$  holes are naturally expected within this approach following the valence transition. Transport in the normal and superconducting states with both hole and electron doping then involve the nearly 3/4-filled strongly correlated O-band alone, explaining the mysterious symmetry between the two cases. Previous

calculations on the single-band 2D 3/4-filled Hubbard Hamiltonian showed that (i) precisely at this carrier concentration there is a strong tendency to transition to a paired-electron crystal (PEC), which is a charge-ordered state of spin-singlet electron pairs [58,59], and (ii) very close to this concentration there occurs an enhancement of superconducting pair correlations by the Hubbard  $U$  [60,61]. In the absence of phase coherence the spin-coupled electron pairs can conceivably be the bosonic charge carriers in the strange metallic state. Importantly, the occurrence of the strange metallic state under pressure in the organic superconductor  $(\text{TMTSF})_2\text{PF}_6$  [48], known to possess a 1/4-filled hole band (3/4-filled electron band) is indirect confirmation of this approach. These and related topics are currently under investigation.

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