Phase separation and the existence of superconductivity in a one-dimensional copper-oxygen model

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Abstract

The phase separation instability occurring with increasing nearest-neighbor repulsion \( V \) in a two-band Hubbard model (CuO chain) is discussed. Quantum Monte Carlo simulations indicate that this transition is associated with a level-crossing if the filling fraction \( \langle n \rangle \) is close to 1 (half-filled lower band). Spin-density-wave fluctuations then dominate before phase separation. Superconducting fluctuations dominate only at considerably higher doping levels.

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It has been suggested that superconductivity in strongly correlated electron systems is favored close to regions in parameter space where phase separation occurs. This hypothesis is based on the notion that the fluctuations that cause the system to separate into high- and low-density regimes can lead to pairing if they are not quite strong enough to lead to phase separation. An explicit example of this mechanism is observed in the one-dimensional (1D) $t$-$J$ model. For small ratios $J/t$ this system is a Luttinger liquid, characterized by a single exponent $K_\rho$ which governs the decay of the spin and charge correlations, as well as the singlet and triplet pair correlations. A system with $K_\rho > 1$ has dominant singlet pairing fluctuations, whereas spin-density-wave fluctuations dominate if $K_\rho < 1$. The exponent is related to the compressibility $\kappa$ and the Drude weight $D$ according to:

$$K_\rho^2 = \pi \kappa D/4.$$  

For the 1D $t$-$J$ model, the compressibility increases smoothly with increasing $J/t$ and diverges as the system phase separates for $J/t \approx 3$. The Drude weight remains finite across the phase separation boundary, vanishing only for slightly larger $J/t$ (the high-density phase is initially conducting). $K_\rho$ hence diverges at the phase separation boundary, and there is always a regime with dominant pair correlations prior to phase separation. Emery, Kivelson and Lin have proposed that the same fluctuations that lead to phase separation may induce pairing also in the 2D $t$-$J$ model. Dagotto et al. have recently conjectured that the existence of superconductivity in the neighborhood of phase separation is a generic feature of strongly correlated electron systems.

Here we study a 1D two-band version of the 2D three-band Hubbard model suggested as a model of the CuO$_2$ planes of the high-$T_c$ cuprate superconductors. We fix the single-particle potentials and the on-site Cu repulsion at values appropriate for the high-$T_c$ cuprates, and study the behavior versus the near-neighbor repulsion $V$. We consider fillings $\langle n \rangle$ ranging from 1 (half-filling) to 2 (completely filled lower band). Using a quantum Monte Carlo (QMC) technique, we find that phase separation close to half-filling is a result of a level crossing: A band of already phase separated states cross the Luttinger liquid ground
state as $V$ is increased. The system has dominant spin-density-wave fluctuations before phase separation. On the other hand, for $\langle n \rangle$ close to 2, fluctuations in the Luttinger liquid state do lead to dominant pair correlations, and also appear to cause the phase separation. Hence, we have an example of a system where the interactions causing phase separation do or do not also induce superconductivity, depending on the filling.

The Cu-O chain model is defined by the Hamiltonian

$$\hat{H} = -t \sum_{\sigma} \sum_{i=1}^{2L} [c_{i+1,\sigma}^+ c_{i,\sigma} + c_{i,\sigma}^+ c_{i+1,\sigma}]$$

$$+ \sum_{i=1}^{2L} [\Delta_i n_i + U_i n_{i,\uparrow} n_{i,\downarrow} + V n_i n_{i+1}].$$

(2)

Here $c_{i,\sigma}^+$ is a creation operator for a spin-$\sigma$ particle on site $i$, with odd and even $i$ corresponding to copper $d$ and oxygen $p$ orbitals, respectively, on a lattice with $L$ unit cells ($N = 2L$ sites), and $n_i = n_{i,\uparrow} + n_{i,\downarrow}$. $\Delta_i$ and $U_i$ denote the single-particle potential and Hubbard repulsion, with $\Delta_i = 0, U_i = U_d$ for odd $i$ and $\Delta_i = \Delta, U_i = U_p$ for even $i$. We work in the hole representation, i.e. increasing the filling above $\langle n \rangle = 1$ corresponds to hole doping the insulating cuprate parent compounds. Typical estimates for the parameters appropriate for a CuO$_2$ plane are, in units of $t$, $\Delta = 2.5 - 3, U_d = 6 - 10, U_p = 2 - 4$. $V$ is difficult to estimate. For the 1D Cu-O model studied here we choose $U_d = 6, U_p = 0$, and $\Delta = 2$, which gives a robust charge-transfer insulator at half-filling, and regard $V$ as a variable.

We use a QMC method based on stochastic series expansion (a generalization of Handscomb’s technique). This method is free from errors associated with the Trotter decomposition used in standard methods. We have calculated the spin and charge structure factors

$$S_{\rho,\sigma}(q) = \frac{1}{L} \sum_{k,j} e^{i(q-k)j} \langle N_{\rho,\sigma}(k)N_{\rho,\sigma}(j) \rangle,$$

(3)

and the corresponding static susceptibilities $\chi_{\rho,\sigma}(q)$. Here, $N_{\rho,\sigma}(j)$ is the charge ($\rho$) or spin ($\sigma$) in unit cell $j$. We have also calculated current-current correlation functions, from which the Drude weight can be obtained. Using these quantities we have extracted the $V$-dependence of the low-energy parameters, $K_{\rho}$ in particular, at several fillings. Details of
the computations are discussed in Ref. 15. Here we focus on the mechanism causing phase separation in this model, and the issue of whether the system possesses dominant pairing fluctuations in the neighborhood of phase separation.

For fillings $1 < \langle n \rangle < 2$ a large $V$ causes the system to separate into high-density (HD) and low-density (LD) phases. This can be easily seen in the limit $V \to \infty$, where nearest-neighbor sites cannot be simultaneously occupied. At half-filling all $d$ sites are then singly occupied, and the $p$ sites are all empty. Adding particles to this state, the $V$ repulsion is minimized by the formation of a HD regime with doubly occupied $p$ sites and empty $d$ sites, and an LD regime which remains as the half-filled system with zero $p$ occupation. Upon lowering $V$, the particles acquire some mobility, but the HD and LD phases initially remain at insulating densities $\langle n \rangle_{HD} = 2$ and $\langle n \rangle_{LD} = 1$, respectively. For even lower $V$, our results discussed in Ref. 15 show that particles from the phase boundary first evaporate into the LD phase. The system is still phase separated for some range of $V > V_{PS}$, but the LD phase is conducting, with $\langle n \rangle_{LD} > 1$.

To shed further light on the details of the phase-separation instability, and in what $(V, \langle n \rangle)$ regime it occurs, it is useful to study local quantities that can be expected to change rapidly as $V$ approaches $V_{PS}$, such as the kinetic energy, the occupation of $p$ sites, and the number of doubly occupied $p$ sites. These are shown versus $V$ for a filling $\langle n \rangle = 1.25$ in Fig. 1. The simulations were carried out at temperatures low enough to give essentially ground state results (inverse temperatures $\beta = t/T$ up to 128 were used). For $V \lesssim 2.5$ there is little dependence on $L$. We have verified that the system is a Luttinger liquid in this regime, by checking the consistency of relations among the low-energy parameters such as Eq. (1). As $V$ is increased, there is a size dependent range where there is a rapid change in all the quantities shown. It is natural to associate this with phase separation. Indeed, the behavior of the charge structure factor also changes qualitatively in the same range. For example for $L = 32$, $S_\rho(q)$ at $V = 2.6$ decreases to zero as $q \to 0$, whereas it is strongly peaked at $q = q_1 = 2\pi/L$ for $V = 2.7$. The latter behavior is a clear sign of phase separation. The size dependence of $V_{PS}$ is probably related to the existence of finite boundaries between
the LD and HD phases, where the $V$ interactions are not avoided as efficiently as in the bulk of the phases, and which are relatively large in small systems.

A striking feature is that all quantities appear to change discontinuously at $V_{PS}$ for the larger systems. We propose that this behavior is associated with a level spectrum schematically outlined in Fig. 2: In addition to the Luttinger liquid and its standard spin and charge excitations, there is a band of states corresponding to a phase separated system. These states are present even for $V$ significantly lower than $V_{PS}$, but are then high in energy. As $V$ approaches $V_{PS}$ they approach, and cross, the Luttinger liquid ground state.

Assume that there are indeed two sets of low-energy states $\{LL\}$ and $\{PS\}$ corresponding to a Luttinger liquid and a phase separated system, respectively, as illustrated in Fig. 2. For a large system, states belonging to different sets will in general be very different, in the sense that their expansions in terms of eigenstates of the $r$-space number operators are dominated by different states, i.e. if a state belonging to $\{LL\}$ has a large expansion coefficient $a_i$ for a state $\psi_i$, all states belonging to $\{PS\}$ have small coefficients $a_i$. This can be expected to have consequences for a simulation carried out in the $r$-space basis. Typical low-temperature configurations can then be uniquely associated either with $\{LL\}$ or $\{PS\}$. A configuration belonging to $\{LL\}$ will require a large modification in order to “tunnel” into $\{PS\}$, and vice versa. Since the simulation proceeds in steps of small modifications of the configurations, one can expect tunneling between the sets to be rare. Consider the “partial partition functions”

$$Z_{LL} = \sum_{i \in \{LL\}} e^{-\beta E_i}, \quad Z_{PS} = \sum_{i \in \{PS\}} e^{-\beta E_i},$$

and the corresponding free energies $F_{LL} = -\ln(Z_{LL})/\beta$ and $F_{PS} = -\ln(Z_{PS})/\beta$. For an infinitely long simulation both sets will be sampled, the time spent in each of them being proportional to the corresponding partial partition function. However, since there are large barriers between the two types of configurations, a simulation of practical duration might sample just one of the sets. If the free energies $F_{LL}$ and $F_{PS}$ are sufficiently different, one might expect that the simulation after an initial equilibration settles within the set
with the lowest free energy. Hence, for a large system, a simulation carried out at a very low temperature would ideally show a discontinuous change in all calculated quantities at $V = V_{PS}$. However, in a small system there will always be some tunneling between \( \{LL\} \) and \( \{PS\} \) at any finite temperature if $V \approx V_{PS}$, and the transition will therefore appear smoothed.

The evolution of the kinetic energy and the $p$ occupation with Monte Carlo "time" is illustrated in Fig. 3 for a system with $V$ larger than but close to $V_{PS}$. Each point represents an average over a “bin” consisting of a large number of Monte Carlo configurations. Initially they fluctuate around values corresponding to the Luttinger liquid close to phase separation (see Fig. 1), but then change to values typical after phase separation. Smaller fluctuations towards the phase separated regime can also be seen, and after the transition there are small fluctuations back towards the Luttinger liquid values. Typically, low-temperature simulations rapidly equilibrate to either the LL or PS regime, depending on $V$, and behavior such as that in Fig. 3 is seen only very close to $V_{PS}$.

The energy versus $V$ is shown in Fig. 4, and has a behavior typical for a level crossing. There are two approximately linear regimes, with different slopes. The intersection of lines fitted to the two sets of points indicate that $V_{PS} \approx 2.63$. The point for $V = 2.675$ was actually obtained from the part of the simulation illustrated in Fig. 3 before the system equilibrated into the phase separated regime. Hence, it represents the energy of a meta-stable Luttinger liquid. Fig. 4 indicates that also $V = 2.65$ is on the separated side of the transition, and hence that this simulation remained in the meta-stable state, which is not surprising this close to the transition.

The scenario outlined above also implies an interesting finite-temperature behavior. For $V$ close to but smaller than $V_{PS}$ there should be a regime at elevated temperatures where $F_{PS} \ll F_{LL}$, and the system behaves as if phase separated. Lowering the temperature below the lowest PS state, $F_{PS}$ increases and the system crosses over into a Luttinger liquid regime where it remains down to $T = 0$. This is indeed observed. Fig. 5 shows results for the charge structure factor and the spin susceptibility for a system with $V < V_{PS}$ at
two temperatures. At the higher temperature $S_\rho(q)$ is sharply peaked at long wavelengths, which is a clear indication of phase separation. At the lower temperature the behavior is drastically different, and corresponds to a uniform system. The spin susceptibility is peaked around $q = \pi$ at the higher temperature, reflecting strong antiferromagnetic fluctuations in the LD phase of a phase separated system. At the lower temperature there is a clear peak at $q = 2k_F$, corresponding to the behavior of a Luttinger liquid with $K_\rho < 1$. The tendency to phase separation at elevated temperatures and the subsequent stabilization of the Luttinger liquid as the temperature is lowered seem counterintuitive, but follow naturally from the level spectrum we propose.

At higher doping levels the discontinuous behavior observed for the local quantities in Fig. 1 is much less prominent, but indications of a discontinuous phase separation transition persist even at $\langle n \rangle = 1.50$. At $\langle n \rangle = 1.75$ the transition appears completely smooth. We believe that this change in behavior with increasing doping is due to increasing density fluctuations in the Luttinger liquid state, which eventually, close to $\langle n \rangle = 2$ cause the system to phase separate without a level crossing. This has implications for the existence of dominant superconducting fluctuations in the neighborhood of phase separation. At $\langle n \rangle = 1.25$ our estimate of $K_\rho$ close to phase separation is $K_\rho \approx 0.8$, i.e. the system has dominant spin-density-wave fluctuations. This is also consistent with a spin susceptibility strongly peaked at $q = 2k_F$ as shown in Fig. 3. At $\langle n \rangle = 1.50$ the fluctuations appear to become large enough before phase separation to result in a regime with dominant pairing fluctuations. This could be a finite size effect, however, since for $L = 32$ the regime is very narrow. For $\langle n \rangle = 1.75$ the charge fluctuations appear to increase steadily before phase separation, and $K_\rho > 1$ for $1.4 \lesssim V \lesssim 1.7$. In this regime we also find indications of a gap in the spin excitation spectrum.

It can be noted that Dagotto et al. found that the pair correlations in small systems ($L = 6$) are considerably stronger at $\langle n \rangle = 1.67$ than at $\langle n \rangle = 1.33$. This is consistent with the scenario we have outlined above.

We conclude that the mechanism causing phase separation in the Cu-O chain model
depends on the filling. Close to half-filling it is a result of a level crossing. There is then no associated increase in the long-wavelength density fluctuations before phase separation, and therefore no dominant pairing fluctuations. Further away from half-filling the density fluctuations of the Luttinger liquid can increase enough to produce pairing before phase separation. At fillings close to two particles per unit cell these fluctuations also appear to drive the phase separation. This intricate behavior is clearly related to the two-band nature of the model. It would be interesting to explore this issue in 2D as well.

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FIGURES

FIG. 1. The kinetic energy $\langle \hat{k} \rangle$, the $p$ site occupation $\langle n^p \rangle$, and the density of doubly occupied $p$ sites $\langle d^p \rangle$ vs. $V$ for systems with $L = 8 - 32$ unit cells at filling $\langle n \rangle = 1.25$.

FIG. 2. Schematic outline of the level spectrum suggested to account for the observed phase separation behavior. The dashed curves represent the Luttinger liquid and its spin and charge excitations. The solid curves represent a band of states corresponding to a phase separated system, which crosses the Luttinger liquid ground state at the point marked PS.

FIG. 3. The kinetic energy and the $p$ site occupation versus Monte Carlo time. Each point represents an average of measurements performed on a large number of configurations.

FIG. 4. The energy per site vs. $V$ for a system with 32 unit cells at $\langle n \rangle = 1.25$. Solid and open circles are for the Luttinger liquid and phase separated regime, respectively. The intersection of lines fitted to the points gives $V_{PS} \approx 2.63$. Solid circles above this point indicate metastable states.

FIG. 5. The spin susceptibility (top panel) and charge structure factor (bottom panel) for an $L = 32$ system with $\langle n \rangle = 1.25$ and $V = 2.6$ at two different inverse temperatures.
FIG. 1, A. W. Sandvik and A. Sudbø
FIG. 2, A. W. Sandvik and A. Sudbø
FIG. 3, A. W. Sandvik and A. Sudbø
FIG. 4, A. W. Sandvik and A. Sudbø
FIG. 5, A. W. Sandvik and A. Sudbø