

Limits on phase separation for two-dimensional strongly correlated electrons

W. O. Putikka*

*Theoretische Physik, ETH-Hönggerberg, CH-8093 Zürich, Switzerland
and Department of Physics, The Ohio State University, Mansfield, Ohio 44906*

M. U. Luchini

Department of Mathematics, Imperial College, London SW7 2BZ, United Kingdom

(Received 15 February 2000)

From calculations of the high-temperature series for the free energy of the two-dimensional t - J model we construct a series for ratios of the free energy per hole. The ratios can be extrapolated very accurately to low temperatures and used to investigate phase separation. Our results confirm that phase separation occurs only for $J/t \gtrsim 1.2$. Also, the phase transition into the phase-separated state has $T_c \approx 0.25J$ for large J/t .

The Hubbard and t - J models, though widely used to investigate high-temperature superconductors, remain controversial when doped away from one electron per site. The possibility that doped holes do not form a uniform phase but instead phase separate into distinct high- and low-density regions on the lattice is an important issue that has proved difficult to settle.¹⁻⁷ Phase separation for physical choices of model parameters would imply that more complicated models of two-dimensional (2D) strongly correlated electrons are required to describe high-temperature superconductors. Stability of a uniform density phase would leave open the possibility that simple models contain the relevant physics without additional terms.

While experiments have clearly observed phase separation in a few high- T_c systems, notably oxygen overdoped $\text{La}_2\text{CuO}_{4+\delta}$ with mobile interstitial oxygen atoms,⁸ phase separation does not seem to be a universal feature of the cuprates. However, the mechanism of phase separation causes holes to feel a net attraction, a possible precursor for the formation of stripe phases or superconductivity. Finding an attractive interaction for holes in models that have predominantly strong repulsive interactions is not easy, and all known possibilities deserve thorough investigation.

To investigate the properties of phase separation we have calculated the high-temperature series for the 2D t - J model free energy to 10th order in inverse temperature. The Hamiltonian for the t - J model is

$$H = -t \sum_{\langle ij \rangle, \sigma} (c_{i\sigma}^\dagger c_{j\sigma} + c_{j\sigma}^\dagger c_{i\sigma}) + J \sum_{\langle ij \rangle} \left(\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} n_i n_j \right), \quad (1)$$

where the sums are over pairs of nearest-neighbor sites and the Hilbert space is restricted to states with no doubly occupied sites. The series is generated for a 2D square lattice.

To determine the stability of the uniform phase we would like to investigate the ground-state energy per hole given by

$$e(\delta) = \frac{E_0(\delta) - E_0^{AF}}{\delta}, \quad (2)$$

introduced by Emery, Kivelson, and Lin.¹ Here $E_0(\delta)$ is the ground-state energy per site of the uniform phase for hole doping δ and $E_0^{AF} = -1.16944J$ is the ground-state energy per site for the Heisenberg model^{6,9} where $\delta=0$. If $e(\delta)$ is a monotonically increasing function of δ the uniform phase is

stable. If $e(\delta)$ is constant or decreasing for a range of dopings the uniform phase is unstable for those values of δ .

There are two main difficulties encountered in calculating $e(\delta)$ from numerical measurements (exact diagonalization, quantum Monte Carlo, or Green's-function Monte Carlo) of $E_0(\delta)$. The first is that $e(\delta)$ requires the subtraction of two large numbers $E_0(\delta)$ and E_0^{AF} to determine a small number which is then divided by δ , another small number. Given statistical uncertainty in numerically determining $E_0(\delta)$ (E_0^{AF} is essentially exact in comparison) this is a difficult task, especially for $\delta \ll 1$. The second difficulty is that numerical calculations are done on small clusters. Systematic errors in $E_0(\delta)$ are tough to estimate without knowing the finite-size scaling of the data and whether the cluster sizes considered are large enough to be in the scaling limit.^{6,7} In addition to these difficulties, phase separation is favored on small clusters for $\delta \ll 1$. The reduction in ground-state energy due to the kinetic energy of the holes, which disfavors phase separation, is not as large on a small cluster as it is for an infinite lattice. On a small cluster the electron system reduces its energy more through local interactions, which for the t - J model are attractive interactions for antiparallel spins due to the J term in the Hamiltonian.

High-temperature series provide a means to avoid these difficulties. We generalize $e(\delta)$ to $T > 0$ by

$$f(\delta, T) = \frac{F(\delta, T) - F^{AF}(T)}{\delta}, \quad (3)$$

where we have replaced the ground-state energy per site by the free energy per site and $\lim_{T \rightarrow 0} f(\delta, T) = e(\delta)$. This replaces the difficulties mentioned above by the need to analytically continue the series to low temperatures. For $J/t \lesssim 1$ and $\delta \ll 1$ we find ratios $f(\delta_2)/f(\delta_1)$ for two closely spaced dopings δ_1 and δ_2 are the best quantities to extrapolate. Series for ratios can be calculated exactly from the series for F , avoiding the need to subtract two large approximate numbers. The series coefficients are also exact for an infinite lattice so we have no explicit finite-size effects. The ratios are extrapolated using standard Padé approximants, but only *after* the exact series for a given ratio is calculated. The doping spacing we use is $\Delta\delta = 0.025$.

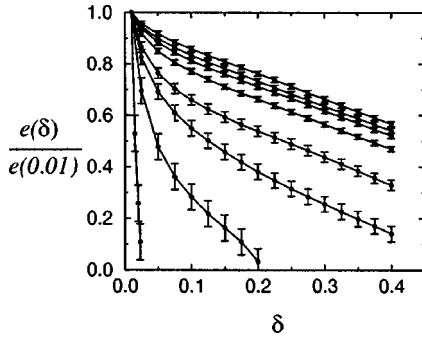


FIG. 1. Doping dependence of the energy per hole normalized to the energy per hole at $\delta=0.01$, plotted as a function of δ for different values of J/t . The J/t values from top to bottom are $J/t = 0.0, 0.2, 0.4, 0.6, 0.8, 1.0, 1.1$, and 1.2 . The monotonic decrease of the ratio with increasing δ indicates the uniform phase is stable for $J/t \leq 1.2$.

By extrapolating $f(\delta_2)/f(\delta_1)$ to $T=0$ we obtain estimates for $e(\delta_2)/e(\delta_1)$ in the uniform phase. Since high-temperature series start at infinite temperature and only have information for the phase above a nonzero T_c , all of our results are for the uniform phase. A description of what happens if we try to extrapolate below $T_c > 0$ is given below. Results for a range of dopings and J/t values are shown in Fig. 1. For the parameters considered here $e(\delta) < 0$ so that if $\delta_2 > \delta_1$ and the system phase separates we should find $e(\delta_2)/e(\delta_1) > 1$ if $T_c > 0$ or $e(\delta_2)/e(\delta_1) = 1$ if $T_c = 0$. If the uniform phase is stable we have $e(\delta_2)/e(\delta_1) < 1$. The 2D t - J model phase separates into a phase with $\delta=0$ and a doped phase with $\delta=\delta_0$. For phase separation we therefore expect $e(\delta)/e(0.01) \geq 1$ immediately upon doping.

In Fig. 1, $e(\delta)/e(0.01) < 1$ and falls monotonically with increasing δ for all J/t shown, indicating no instability towards phase separation in the 2D t - J model for $J/t \leq 1.2$.

In Ref. 1 a variational argument is used to support the presence of phase separation for $J/t \ll 1$. A variational phase-separated state was constructed from two pieces occupying different parts of the lattice: a Heisenberg antiferromagnet for the $\delta=0$ phase and a gas of spinless fermions for the $\delta=\delta_0$ phase. The energy of this state is then minimized with respect to δ , giving $E_0(\delta) = E_0^{AF} - 4t\delta(1 - \sqrt{B\pi J/t})$ for the phase-separated state and $\delta_0 = \sqrt{B\pi J/t}$, where $B = 1.16944/2 = 0.58472$. This energy was then compared to ground-state energy estimates for the uniform phase found by considering a single hole in an antiferromagnet. The energy for the phase-separated state was found to lie below the uniform state energy for small enough J/t , and since the variational energy lies above the true ground-state energy the conclusion of Ref. 1 was that the phase-separated state is stable. Extrapolating the result for a single hole to a finite density of holes assumes the energy bands remain rigid, a feature not obvious for a strongly correlated system.

In Fig. 2 we compare our estimates for the uniform ground-state energy to the phase-separated variational ground-state energy at $J/t=0.01$. We find that our energies lie below the variational energy for $\delta < \delta_0$. Note that from this result we cannot conclude that the uniform state is stable, but only that the variational state discussed in Ref. 1 is not sufficient to show phase separation at $J/t \leq 1$.

In Fig. 3 we show the temperature dependence of

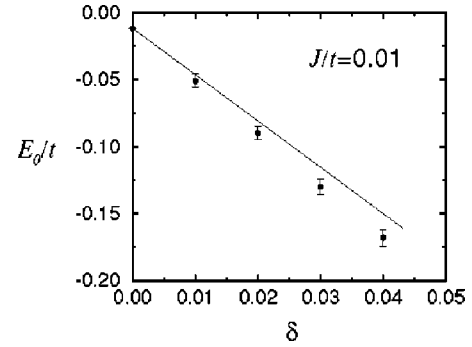


FIG. 2. Comparison of ground-state energy estimates at $J/t = 0.01$. The solid line is a variational phase-separated state from Ref. 1 which extends to $\delta_0 = 0.04314$. The data points are ground-state energy estimates for a uniform state calculated by extrapolating the high-temperature series for the free energy. The variational estimate lies above the uniform state estimate and is thus not sufficient to show phase separation at $J/t = 0.01$.

$f(0.02)/f(0.01)$ for a range of J/t values. Estimating the low- T behavior of this function is our only approximation. The weak temperature dependence for the ratio leads us to believe our results are reliable. The general trends of the data shown in Figs. 1 and 3 are due to the minimum in $E_0(\delta)$ moving to smaller δ as J/t is increased, causing $e(\delta)$ to decrease in magnitude faster than $e(0.01)$, though for the parameters shown $e(\delta)$ and $e(0.01)$ remain negative. For values of J/t larger than shown in Figs. 1 and 3 the ratio $f(\delta_2)/f(\delta_1)$ develops a spurious pole due to the crossing of F^{AF} and $F(\delta_1)$ at $T > 0$. This pole greatly degrades the accuracy of extrapolations of the ratios at lower temperatures. To investigate larger J/t we need another method.

The chemical potential $\mu = -\partial F/\partial \delta$ provides another means to investigate phase separation. We typically find μ is more difficult to extrapolate than $f(\delta_2)/f(\delta_1)$, with the error in the extrapolations for μ considerably larger than for the ratio. For $J/t \geq 1.2$ we do see $\mu(\delta)$ becoming quite flat for $\delta \ll 1$, as expected for a first-order phase transition into a phase-separated state. As the temperature is lowered, μ near the critical point (critical doping δ_c and temperature T_c) becomes flat, giving a diverging compressibility κ at the critical point. Results for μ are shown in Fig. 4. The flat region found in $\mu(\delta)$ can be used to estimate the boundary for phase separation. However, for larger δ distinguishing where

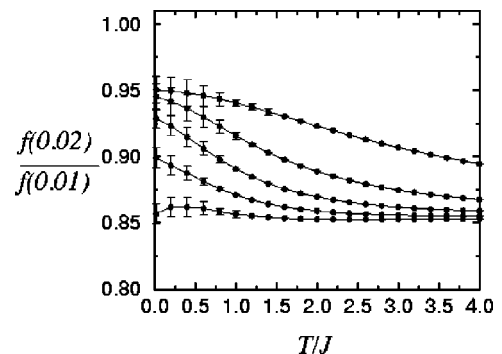


FIG. 3. Temperature dependence of $f(0.02)/f(0.01)$ for a range of J/t values. From top to bottom $J/t = 0.2, 0.4, 0.6, 0.8$, and 1.0 . The overall temperature dependence is fairly small, making the temperature extrapolations more reliable.

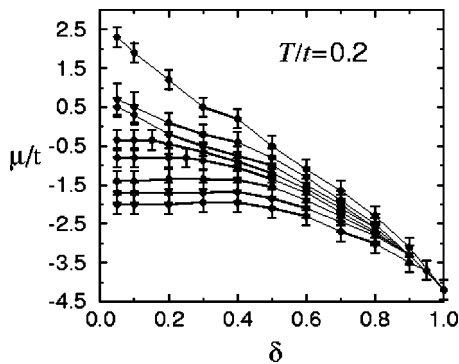


FIG. 4. Chemical potential at $T=0.2t$ as a function of doping for a range of J/t values. From top to bottom $J/t=0.4, 1.0, 1.2, 1.3, 1.4, 1.6, 1.8$ and 2.0 . For each $J/t \geq 1.3$ a range of dopings exists where the chemical potential is approximately constant, as would be expected near a critical point.

the flat region ends is difficult, leading to errors in the position of the phase-separation boundary.^{2,4}

Further evidence of phase separation at large J/t can be found by directly extrapolating $F(\delta, T)$ to estimate $E_0(\delta)$. Figure 5 shows results for $J/t=2.0$. The characteristic signature of phase separation is the reversed curvature observed from $\delta=0$ to $\delta \approx 0.45$. The reversed curvature of $E_0(\delta)$ (giving an unphysical negative compressibility) results from extrapolating the high-temperature uniform phase $F(\delta, T)$ through the $T_c > 0$ phase transition for phase separation. If $T_c=0$ we would find instead that $E_0(\delta)$ became linear in δ in the phase-separated region.

The reversed curvature shown in Fig. 5 indicates $T_c > 0$, but T_c is probably quite low. An indirect estimate of T_c can be made at large J/t , above $J/t=3.4367$ where the 2D t - J model phase separates at all densities¹⁰ into regions with $\delta=0$ and $\delta=1$. Here we know $E_0(\delta)$ for all δ , since $E_0(\delta)$ is the linear interpolation between $E_0(0)=-1.16944J$ and $E_0(1)=0$.

The ground-state chemical potential in this parameter range is the constant slope of $E_0(\delta)$ with the value $\mu/t = -1.16944J/t$. The chemical potential hits the bottom of the tight-binding band at $J/t=3.4367$ and as J/t is further reduced the gain in kinetic energy eventually limits the phase-

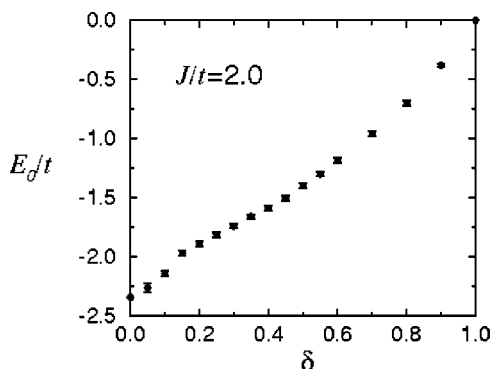


FIG. 5. Ground-state energy estimated by high-temperature series as a function of doping at $J/t=2.0$. The reverse curvature for $\delta \leq 0.45$ is due to extrapolating the high-temperature free energy through $T_c > 0$ into the low-temperature phase-separated state. The reverse curvature gives a negative compressibility, indicating the uniform phase is unstable towards phase separation.

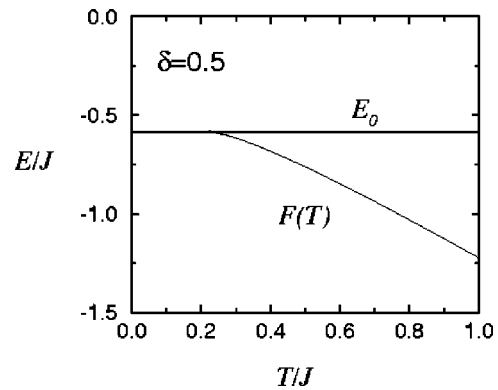


FIG. 6. Comparison of the free energy $F(T)$ as a function of temperature to the known ground-state energy E_0 for $\delta=0.5$, $J/t \rightarrow \infty$. The crossing of these two curves we interpret as a phase transition at $T_c \approx 0.25J$ into a phase-separated state.

separated state to $J/t \geq 1.2$. In Fig. 6 we compare E_0 to $F(T)$ in the limit $J/t \rightarrow \infty$ with $\delta=0.5$.

Comparing the extrapolated $F(T)$ to E_0 we see they tend to cross at $T \approx 0.25J$. Since $F(T)$ must be less than E_0 and a monotonic function of T this crossing cannot occur. We interpret the tendency to cross as a phase transition to phase separation with $T_c \approx 0.25J$.

Calculations for the 2D t - J model currently give a wide range of minimum J/t values for the presence of phase separation. Minimum J/t values reported in the literature are 0 (Refs. 1 and 4), 0.5–0.6 (Refs. 3, 5, and 6), and our result of 1.2.² The latter results are in qualitative agreement in that there is a minimum $J/t > 0$ for phase separation. The reasons for these differences are not clear at present. However, while statistical errors are well under control, systematic errors in ground-state energy calculations due to small cluster sizes are much more difficult to control. Calculations investigating phase separation in the 2D Hubbard model⁷ find $e(\delta)$ equal to a constant for a range of dopings near half filling for the $U=0$ tight-binding model. This spurious indication of phase separation is due to finite-size effects and is reduced for larger clusters. Resolving the different reported results for phase separation will probably require significantly larger cluster sizes.

In conclusion, by using an analysis of the high-temperature series for the free energy per hole $f(\delta)$ at different values of J/t we find that phase separation in the t - J model is limited to $J/t \geq 1.2$. In addition, we find by indirect arguments that $T_c \sim 0.25J$ for the first-order phase transition into the phase-separated state. Combining this with the demonstration that phase separation can only occur at $T=0$ for the 2D Hubbard model on a square lattice¹¹ supports the conjecture that the 2D Hubbard model does not phase separate for any positive U . Our results suggest phase separation in the 2D t - J model is a classical phase transition similar to a lattice gas with an attractive interaction¹² and that phase separation is not important for physical choices of the t - J model parameters.

This work was supported in part by a faculty travel grant from the Office of International Studies at The Ohio State University (W.O.P.), the Swiss National Science Foundation (W.O.P.), and by EPSRC Grant No. GR/L86852 (M.U.L.). W.O.P. thanks the ETH-Zürich for hospitality while part of this work was being completed.

*Permanent address: Department of Physics, The Ohio State University, Mansfield, Ohio 44906.

- ¹V. J. Emery, S. A. Kivelson, and H. Q. Lin, *Phys. Rev. Lett.* **64**, 475 (1990).
- ²W. O. Putikka, M. U. Luchini, and T. M. Rice, *Phys. Rev. Lett.* **68**, 538 (1992).
- ³M. Kohno, *Phys. Rev. B* **55**, 1435 (1997).
- ⁴C. S. Hellberg and E. Manousakis, *Phys. Rev. Lett.* **78**, 4609 (1997).
- ⁵C. T. Shih, Y. C. Chen, and T. K. Lee, *Phys. Rev. B* **57**, 627 (1998).
- ⁶M. Calandra, F. Becca, and S. Sorella, *Phys. Rev. Lett.* **81**, 5185 (1998).
- ⁷A. C. Cosentini, M. Capone, L. Guidoni, and G. B. Batchelet, *Phys. Rev. B* **58**, R14685 (1998); G. B. Batchelet and A. C. Cosentini, cond-mat/9810173 (unpublished); G. B. Batchelet (unpublished).
- ⁸J. D. Jorgensen *et al.*, *Phys. Rev. B* **38**, 11 337 (1988); P. C. Hammel *et al.*, *ibid.* **42**, 6781 (1990); F. C. Chou *et al.*, *ibid.* **54**, 572 (1996).
- ⁹A. Sandvik, *Phys. Rev. B* **56**, 11 678 (1997).
- ¹⁰C. S. Hellberg and E. Manousakis, *Phys. Rev. B* **52**, 4639 (1995).
- ¹¹G. Su, *Phys. Rev. B* **54**, R8281 (1996).
- ¹²See, for example, R. K. Pathria, *Statistical Mechanics*, 2nd ed. (Butterworth-Heinemann, Oxford, 1996), p. 319.