Phase separation of the two-dimensional t-J model

C. T. Shih
Department of Physics, National Tsing Hua University, Hsinchu, Taiwan

Y. C. Chen
Department of Physics, Tunghai University, Taichung, Taiwan

T. K. Lee
Institute of Physics, Academia Sinica, Nankang, Taipei, Taiwan

(Received 14 May 1997)

The boundary of phase separation of the two-dimensional t-J model is investigated by the power-Lanczos method and Maxwell construction. The method is similar to a variational approach and it determines the lower bound of the phase-separation boundary with $J_c/t=0.6\pm0.1$ in the limit $n_e\sim1$. In the physically interesting regime of high-T_c superconductors where $0.3<J/t<0.5$ there is no phase separation. [S0163-1829(97)03441-3]

It is believed that the main physical properties of the high-temperature superconductors can be described by the two-dimensional (2D) t-J model on a square lattice. The Hamiltonian is

$$H=-t\sum_{\langle i,j\rangle\sigma}(\tilde{c}^{+}_{i\sigma}\tilde{c}_{j\sigma}+\text{H.c.})+J\sum_{\langle i,j\rangle}\left(\mathbf{S}_i\cdot\mathbf{S}_j-\frac{1}{4}n_in_j\right), \tag{1}$$

where $\langle i,j\rangle$ is the nearest-neighbor pairs and $\tilde{c}_{i\sigma}=c_{i\sigma}(1-n_{i,-\sigma})$. In this model the two terms compete with each other. The kinetic term favors the phase in which the electrons are homogeneously distributed in the plane to minimize the kinetic energy. While the exchange term attracts the electrons together to lower the magnetic energy. It is easy to see that for very large J/t the system will phase separate into a hole-rich region and a region without holes to maximize the magnetic energy gain.

There are experimental evidences as well as theoretical studies that indicate phase separation and superconductivity are closely related. It is even argued that the driving mechanism of superconductivity is the same as that of phase separation[1] or superconductivity comes from the frustrated phase separation.[2] Hence it is extremely important to determine the phase-separation boundary of the 2D t-J model to resolve these issues. This paper reports our findings of the phase-separation boundary.

Experimentally, phase separation of the superconducting $La_2CuO_{4+\delta}$ compound is observed by several measurements.[3–6] The compound phase separates for $0.01\leqslant\delta\leqslant0.06$ below $T_{ps}\approx300$ K into the nearly stoichiometric antiferromagnetic $La_2CuO_{4+\delta_1}$ with δ_1 less than 0.02 and Néel temperature $T_N\approx250$ K, and a metallic superconducting oxygen-rich phase $La_2CuO_{4+\delta_2}$ with $\delta_2\approx0.06$ with $T_c\approx34$ K. The Sr doped compound $La_{2-x}Sr_xCuO_{4+\delta}$ also phase separates for $x\leqslant0.03$ into superconducting $La_{2-x}Sr_xCuO_{4+\delta'}$ ($\delta'\approx0.08$) and nonsuperconducting $La_{2-x}Sr_xCuO_{4+\delta''}$ ($\delta''\approx0.00$) phases.[7] Recent muon spin-resonance and nuclear quadrupole-resonance experiments[8–10] on $La_{2-x}Sr_xCuO_4$ also indicate that the doped holes were inhomogeneously distributed mesoscopically and segregated into walls separating the hole-poor antiferromagnetic domains.

Theoretically, there are conflicting results. The first important paper on this issue is by Emery *et al.*[11] They used the exact diagonalization (ED) to study the 4×4 cluster. Using Maxwell construction they claimed that phase separation occurs for all values of J/t. This result is contradictory to the later calculations by using quantum Monte Carlo[12] (QMC) and ED (Ref. 13) on the Hubbard model, which should be consistent with the t-J model for small J/t. Putikka *et al.* studied this problem using the high-temperature series expansion and found phase separation at $T=0$ for J/t lying above a line extending from $J/t=3.8$ at zero filling to $J/t=1.2$ at half filling.[14] Prelovšek *et al.*[15] calculated the two-point and four-point density correlations using ED on clusters of size 18 and 20 sites. They found the two-hole bound state for $J/t>0.2$. For $J/t>1.5$ the holes form domain walls along the (1,0) or (0,1) direction, and phase separate into a hole-rich and a hole-free phase for even larger $J/t>2.5$. Hellberg *et al.* determined very accurately that the critical J/t for phase separation at low electron density limit is $J/t=3.4367$.[16] Poilblanc calculated the energy of two and four holes by ED on several clusters up to 26 sites. The phase diagram includes a liquid of d-wave hole pairs for $J/t\geqslant0.2$, a liquid of hole droplets (quartets) for larger $J/t\geqslant0.5$, and at even larger J/t, an instability towards phase separation.[17] Yokoyama *et al.* investigated the phase diagram by the variational Monte Carlo (VMC) method.[18] The critical J/t for phase separation at the high density limit they found is 1.5, which is consistent with Putikka *et al.*

Most recently Hellberg and Manousakis[19] investigated this problem by the Green's function Monte Carlo (GFMC) method and Maxwell construction for larger clusters. Their phase diagram is similar to Emery *et al.*[11] They conclude that the t-J model phase separates for all values of J/t in the low doping regime.

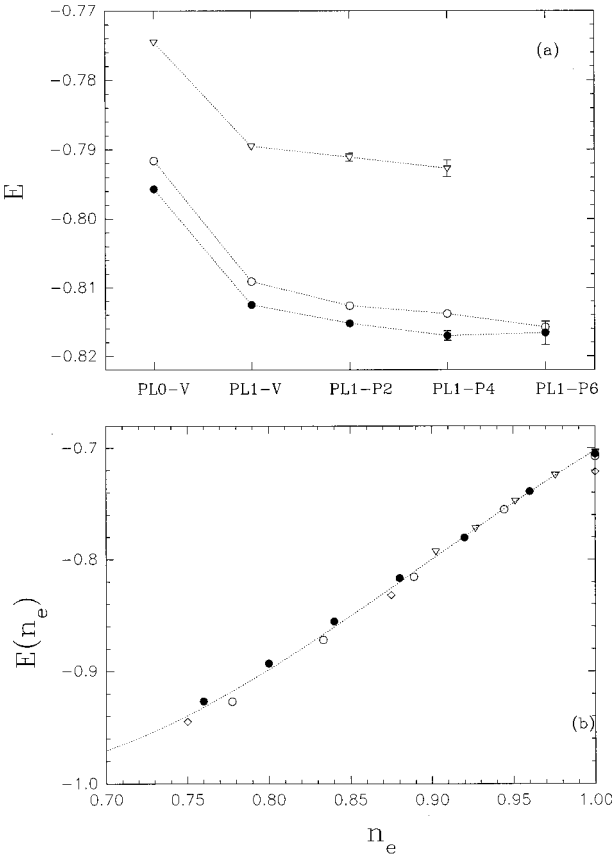

FIG. 1. (a): Typical plots of energy per site vs powers for $J/t=0.6$, $n_e=32/36$ (open circles), $n_e=44/50$ (full circles), and $n_e=74/82$ (open triangles); (b) energy per site as a function of electronic density for $J/t=0.6$ with different cluster sizes. Diamonds are the exact result of 16 sites. Open circles are for 36 sites and full circles are for 50 sites, both are obtained by $\mathrm{PL1}_{\mathrm{power=6}}$. Triangles are for 82 sites with $\mathrm{PL1}_{\mathrm{power=4}}$.

The theoretical results of different groups discussed above are consistent at the large J/t and low electron density region. But unfortunately, in the interesting physical regime of high-T_c superconductors, $0.3<J/t<0.5$ and high electron density $0.75<n_e<0.95$, they are in disagreement. We have used the power-Lanczos (PL) method[20,21] to obtain the best estimate of the ground state energy in this physical regime for the largest cluster (82 sites) that has been studied so far. Based on the variational argument we show that there is no phase separation in this physical regime.[22]

The ground state energy of the Hamiltonian of Eq. (1) is calculated by using the PL method. The PL method we used is similar to the GFMC method but without using importance sampling and the fixed node approximation. The method is essentially a variational approach. Applying more powers to a trial wave function implies a better approximation of the ground state wave function. Details of the method are discussed in Ref. 20. The trial wave functions we used are the optimized Gutzwiller wave functions, resonating valence bond state (RVB),[23] and RVB with antiferromagnetic long range order.[24] In Fig. 1(a) energy per site is plotted as a function of power for $J/t=0.6$ and three different densities: $n_e=32/36$ (open circles), $n_e=44/50$ (full circles), and $n_e=74/82$ (open triangles). An error bar is shown only when it is larger than the symbol. We also compared our energy of $J/t=1$ for 50/64 with the result of a high-temperature series expansion.[25] The best energy we get is $-1.183(2)$ while the high-temperature expansion result is $-1.20(2)$. They are well in agreement. In Fig. 1(b) we show the best energies we are able to obtain for clusters with 36, 50, and 82 sites as a function of electronic density. For comparison we also show the exact energies of 16 sites.[13] Energies are little lower for the smaller clusters. For 50 and 82 sites, there seems to be very little finite size effect. The energy per site is a fairly smooth function of density. We do not find large effect due to different Fermi surface topology in the physical regime.

To find the phase-separation boundary by using Maxwell construction we are interested in the variation of the slopes in figures like Fig. 1(b). In other words we are interested in the second derivative of energy with respect to the electronic density, or the inverse compressibility. It turns out that there is a systematic variation of this quantity as the energy approaches the ground state or as the power increases in our PL method. Although in the physical regime most of our best data have not yet converged to the exact ground state, this systematic variation is enough for us to determine the lower bound of the phase-separation boundary.

It is difficult to read out the slope variation from figures like Fig. 1(b), as the curve is almost a straight line for $n_e>0.85$. Therefore we shall follow Emery *et al.*[11] by examining another quantity. In the one-dimensional t-J model the phase-separated state contains electron-free and electron-rich phases. However, it phase separates into a hole-free phase, i.e., the antiferromagnetic Heisenberg island, and a hole-rich phase in the two-dimensional t-J model. Thus the energy of the phase separated state is in the form

$$E=(N_s-N)e_H+Ne_h, \tag{2}$$

where N_s is the total number of sites and N is the number of sites in the hole-rich phase. $e_H=1.169J$ denotes the Heisenberg energy per site.[26] And e_h is energy per site in the uniform hole-rich phase, which is a function of the hole density in this phase $x=N_h/N$. N_h is the number of holes. E can be rearranged into the form

$$E=N_s e_H+N_h e(x), \tag{3}$$

where

$$e(x)\equiv[-e_H+e_h(x)]/x. \tag{4}$$

If $e(x)$ of a particular J/t has a minimum at $x=x_m$ and the hole density of the total system is smaller than x_m, the system will adjust the size of the hole-rich phase N such that x_m is equal to N_h/N and it minimizes the total energy in Eq. (3). Since N_s, e_H, and N_h are all constants, the total energy is minimized as $e(x)$ is minimized. Thus x_m is the critical density for phase separation at this J/t.

We calculated $e(x)$ from the energy of the uniform states $e_h(x)$ by the PL method and found the minimum of $e(x)$ on 6×6, $\sqrt{50}\times\sqrt{50}$, and $\sqrt{82}\times\sqrt{82}$ clusters for several densities and J/t. It is very difficult to get the converged ground state energy in the physical regime due to the sign problem. After we have found the optimized wave function in the VMC calculation we used the PL method to project the trial

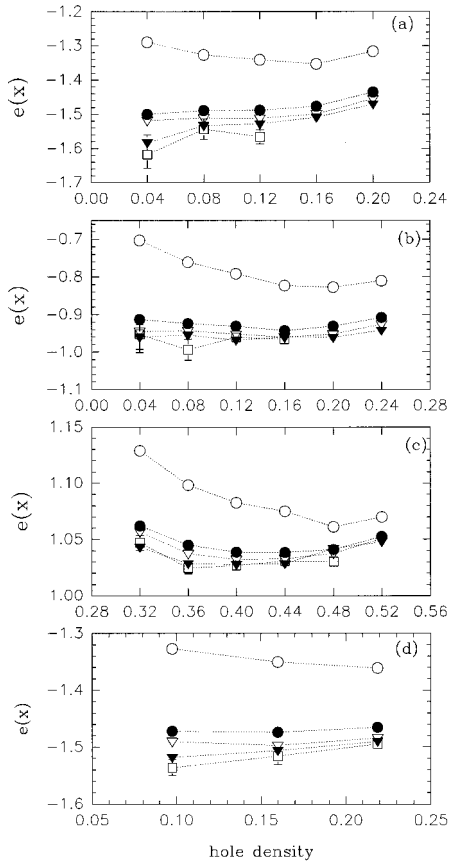

FIG. 2. $e(x)$ vs hole density x for (a) $J/t=0.4$, (b) $J/t=0.6$, and (c) $J/t=1.5$ for several powers: PL0-VMC (open circles), PL1-VMC (full circles), PL1-power=2 (open triangles), PL1-power =4 (full triangles), and PL1-power=6 (open squares). (d) $J/t=0.4$ for close shells for different size of lattices, 74/82, 42/50, and 50/64.

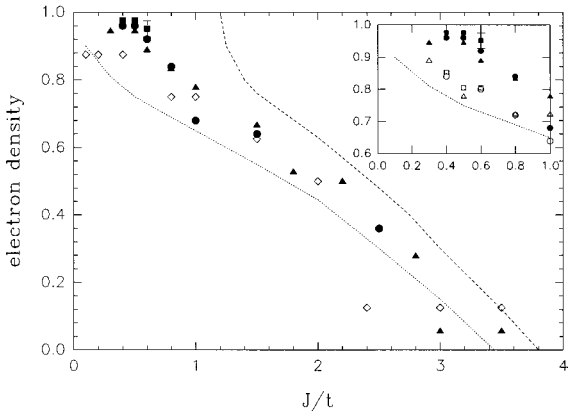

FIG. 3. Phase-separation boundary on the phase diagram of the two-dimensional t-J model evaluated by ED on the 4×4 lattice (Ref. 11) (open diamonds), by the high-temperature series expansion (Ref. 14) (dashed line), by the GFMC method (Ref. 19) (dotted line), and the PL method on 36 sites (full triangles), 50 sites (full circles), and 82 sites (full square). The phase boundary determined by the VMC method for 36 sites (open triangles) and 50 sites (open circles) is shown in the inset.

wave function onto the ground state systematically. The PL-1 power=4 (for 82 sites) or PL-1 power=6 (for 50 and 36 sites) energy is used here as the $e_h(x)$. It is about 2–4 % lower than the variational energy. We estimate the difference between the best PL energy is within 1 or 2 % of the true ground state energy.

$e(x)$ vs $x=1-n_e$ calculated on 50 sites for $J/t=0.4$, 0.6 and 1.5 is shown in Figs. 2(a)–2(c), respectively. It is interesting to note the trend of the shift of $e(x)$ with powers. For $J/t=0.4$ [Fig. 2(a)], at the VMC level, the minimum of $e(x)$ is at $x_m=0.16$. It shifts to $x=0.04$ (the minimum hole density we calculated for this cluster) immediately after the first-order Lanczos improvement (PL1-VMC) and stays at the density up to 6 powers. For $J/t=0.6$ [Fig. 2(b)], x_m shifts from $x=0.2$ (VMC) to $x=0.16$ (PL1-VMC) and to $x=0.08$ (PL1-power=6) at last. For $J/t=1.5$ [Fig. 2(c)], x_m shifts from $x=0.48$ (PL1-VMC) to $x=0.4$ (PL1-power=2) and to $x=0.36$ (PL1-power=6) at last. It is clear that x_m shifts monotonically toward a smaller value when the energy moves closer to the ground state.

The results presented in Figs. 2(a)–2(c) are calculated with a fixed lattice size and different electron numbers. Hence Fermi surfaces have different shapes and, in particular, there are open and closed shells. It has been argued[19] that comparing energies obtained for these different Fermi surfaces might be inaccurate. To examine this argument carefully, we have compared systems with closed shell Fermi surfaces only. In Fig. 2(d) $e(x)$ calculated from close shells of different size of lattices for $J/t=0.4$ shows similar behavior as Fig. 2(a). The minimum of $e(x)$ shifts toward smaller hole density. The trend of x_m moving with increasing power is the same for both close and open shells. Hence the shell effect is not important here.

In Fig. 3 we show the phase-separation boundary determined by the best x_m. The PL1-power=6 phase boundaries of 36 sites and 50 sites are shown as full triangles and full circles, respectively. Also some of the PL1-power=4 data of 82 sites are also shown as full squares. For $J/t=0.6$ the error bars of the $e(x)$ for $n_e=$ 80/82, 78/82, and 76/82 are larger than the difference of these three $e(x)$, thus error bars of x_m are shown in the figure near these electron densities.

The dashed line in Fig. 3 is the result of high temperature series expansion.[14] A similar result is obtained by the variational study.[18] They assumed the system separates into a hole-free Heisenberg antiferromagnet and an electron-free vacuum state. This overestimates the energy required for the phase-separated state, since electrons can ''evaporate'' from the Heisenberg island to gain energy. Their critical $J_c/t\approx1.2$ is larger than our $J_c/t\approx0.6$. Similar argument was also given by Hellberg and Manousakis.[19]

Our estimate of the $J_c/t=0.6\pm0.1$ is actually a lower bound. The exact phase-separation boundary should be to the right of our result in Fig. 3. When we use a much poorer estimate of the ground state energy as our VMC result, the phase boundary is shifted lower. This is shown in the inset of Fig. 3. The VMC results of 36 sites (open triangles) and 50 sites (open circles) show a much smaller J_c/t.

Another way to understand this argument of lower bound is to examine the variation of $e(x)$ with power. In Fig. 4 we show the change of $e(x)$ between PL1-power=6 and VMC

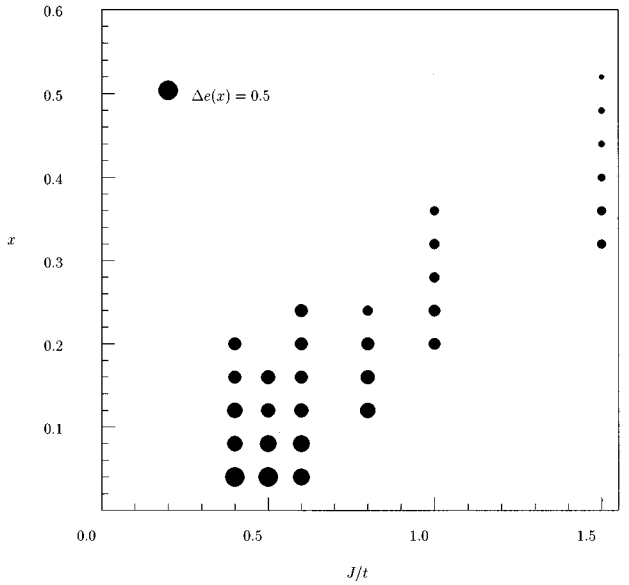

FIG. 4. The $e(x)$ difference between PL1-power$=6$ and PL0-VMC for 50 sites. The values are proportional to the area of the circles.

for 50 sites. The values are proportional to the area of the circles. Because of the $1/x$ factor in Eq. (4), the smaller the hole density the more improvement of $e(x)$ will likely occur. Because of the variational nature of the PL method, the larger the improvement observed between VMC and PL1-power$=6$ the larger the difference between the exact result and PL1-power$=6$ will be. Hence, once the minimum x_m is at the lowest hole density such as $J/t=0.4$ in Fig. 2, a better estimate of the ground state energy by applying more powers will not change the minimum to higher hole density. Based on this argument we are confident to conclude that there is no phase separation in the physical regime where $0.3<J/t<0.5$.

We have found that for $J/t\leqslant 0.5$, the minimum of $e(x)$ is always at two holes for clusters of different sizes (16, 36, 50, and 82). As argued by Dagotto *et al.*, this might indicate a two-hole bound state[13] but not phase separation. If there were phase separation, the x_m would be at the same (or nearby) density rather than the same number of holes.

It is also interesting to note that in Fig. 2(b), for $J/t=0.6$ the minimum x_m seems to be at four holes instead of two holes. This is observed for both 36 and 50 sites. It seems to be quite consistent with a recent claim by Poilblanc[17] that there is a phase with quartets for $0.5\leqslant J/t\leqslant 0.8$. But our data are not accurate enough for 82 sites to make a more definite conclusion.

Recently Hellberg and Manousakis[19] have used GFMC to determine the phase-separation boundary. The phase boundary they reported (dotted line in Fig. 3) is similar to our variational boundary (see the inset of Fig. 3). Without knowing details of their calculation we cannot completely understand this discrepancy. A possible clue is that they might not have obtained low enough energy in the high electron density regime. As shown in Fig. 2, in particular Fig. 2(d), until the energy is lower enough to be closer to the ground state, it is very easy to make the conclusion that there is a minimum of $e(x)$ at a finite hole density.

In summary, we determined the phase-separation boundary by the PL method and Maxwell construction. We have studied various size of clusters and densities of holes. The largest cluster studied is 2 holes in an 82-site lattice. Using the variational nature of the PL method and the systematic variation of the energy as a function of hole density we conclude that the critical J_c/t for phase separation in the low hole density limit is at least ≈ 0.6. There is no phase separation in the physical regime.

It should be pointed out that the results reported above are obtained by assuming the hole-rich region in the phase-separated state has a uniform hole density. We have not yet considered more exotic possibilities such as the stripe phase.[15,27,28]

We wish to thank C. S. Hellberg, H. Q. Lin, and W. O. Putikka for many useful discussions. This work was partially supported by the National Science Council of Republic of China, Grant Nos. NSC 86-2811-M-007-001R, 86-2112-M-001-042T, and 86-2112-M-029-001. Part of the computations were performed at the National Center for High-Performance Computing in Taiwan. We are grateful for their support.

[1] E. Dagotto *et al.*, Phys. Rev. B **49**, 3548 (1994).
[2] V. J. Emery and S. A. Kivelson, Physica C **209**, 597 (1993).
[3] J. D. Jorgensen *et al.*, Phys. Rev. B **38**, 11 337 (1988).
[4] P. C. Hammel *et al.*, Phys. Rev. B **42**, 6781 (1990).
[5] P. C. Hammel *et al.*, Physica C **185-189**, 1095 (1991).
[6] F. C. Chou *et al.*, Phys. Rev. B **54**, 572 (1996).
[7] J. H. Cho *et al.*, Phys. Rev. Lett. **70**, 222 (1993).
[8] J. H. Cho *et al.*, Phys. Rev. B **46**, 3179 (1992).
[9] F. Borsa *et al.*, Physica C **235-240**, 1713 (1994).
[10] F. Borsa *et al.*, Phys. Rev. B **52**, 7334 (1995).
[11] V. J. Emery, S. A. Kivelson, and H. Q. Lin, Phys. Rev. Lett. **64**, 475 (1990).
[12] A. Moreo *et al.*, Phys. Rev. B **43**, 11 442 (1991).
[13] E. Dagotto *et al.*, Phys. Rev. B **45**, 10 741 (1992).
[14] W. O. Putikka *et al.*, Phys. Rev. Lett. **68**, 538 (1992).
[15] P. Prelovšek and X. Zotos, Phys. Rev. B **47**, 5984 (1993).
[16] C. S. Hellberg and E. Manousakis, Phys. Rev. B **52**, 4639 (1995).
[17] D. Poilblanc, Phys. Rev. B **52**, 9201 (1995).
[18] H. Yokoyama and M. Ogata, cond-mat/9607050 (unpublished).
[19] C. S. Hellberg and E. Manousakis, Phys. Rev. Lett. **78**, 4609 (1997).
[20] Y. C. Chen and T. K. Lee, Phys. Rev. B **51**, 6723 (1995).
[21] E. S. Heeb and T. M. Rice, Z. Phys. B **90**, 73 (1993).
[22] A similar result has been reported by M. Khono, Phys. Rev. B **55**, 1435 (1997). Although he has also used the PL method, the largest cluster he used is 50 sites in the physical regime. Using the polynomial fitting of energy as a function of density, he determined the phase boundary with $J_c/t=0.75\pm 0.25$. This is

consistent with our result obtained with a much more careful analysis.
[23] C. Gros, Phys. Rev. B **38**, 931 (1988).
[24] T. K. Lee and C. T. Shih, Phys. Rev. B **55**, 5983 (1996).
[25] W. O. Putikka (private communication).
[26] E. Manousakis, Rev. Mod. Phys. **63**, 1 (1991).
[27] H. Tsunetsugu *et al.*, Phys. Rev. B **51**, 16 456 (1995).
[28] S. R. White and D. J. Scalapino, cond-mat/9610104 (unpublished).