Phase Separation in the t-J Model

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It is shown that, for the t-J model, dilute holes in an antiferromagnet are unstable against phase separation into a hole-rich and a no-hole phase. When the spin-exchange interaction J exceeds a critical value J_c, the hole-rich phase has no electrons. It is proposed that for J slightly less than J_c the hole-rich phase is a low-density superfluid of electron pairs. A brief discussion of phase separation in other related models is given.

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It has been clear for some time that a detailed understanding of the motion of "holes" in an antiferromagnet is of fundamental importance for the theory of high-temperature superconductors[1]—at least for those containing copper oxide planes. The parent compounds such as La_2CuO_4, $YBa_2Cu_3O_6$, or Nd_2CuO_4 are antiferromagnetic insulators but, when electrons or holes are introduced into the CuO_2 planes by doping, the magnetic order eventually gives way to superconductivity.[2] The precise manner in which this comes about surely is of importance for the electronic structure and for the nature of the quasiparticles in the metallic state, whatever the mechanism of superconductivity.

The simplest model for studying this problem is the t-J model.[3] It starts from the assumption that the parent compounds are well represented by the Heisenberg model with localized electrons of spin-$\frac{1}{2}$ occupying a square lattice and coupled by an exchange integral J. Doping is assumed to remove electrons thereby producing a "hole" or missing spin which is mobile because neighboring electrons can hop into its place with amplitude t. This is but one of several models that have been proposed for the study of correlation effects in high-temperature superconductors. Nevertheless, it is simple and has been widely studied, so it is certainly worthwhile to establish its phase diagram as a first step, and then to ask how more elaborate models may differ in their behavior.

The question to be addressed here is whether the holes have a uniform density (as is usually assumed) or separate into two phases of different density. The simplest possibility, and the one that turns out to be relevant, is that all of the holes are in one phase and the other phase is the undoped antiferromagnet. In that case, the problem may be reformulated by writing the energy in the form

$$E=(N_s-N)e_H+Ne_h\,. \tag{1}$$

Here N_s is the total number of sites in the system and N is the number of sites in the hole-rich phase. The energy per site in the Heisenberg (hole-free) phase is denoted by $e_H=-2BJ$, where BJ is the energy per bond: The best estimate of B by extrapolation from finite-size systems[4] is $B=0.584$. The energy per site in the hole-rich phase is denoted by e_h, which is a function of $x\equiv N_h/N$, where N_h is the number of holes. The system will separate into two phases if E has a minimum as a function of N at $N=N_m<N_s$. If E is rearranged into the form

$$E=N_s e_H+N_h e(x)\,, \tag{2}$$

where $e(x)\equiv[-e_H+e_h(x)]/x$, this will occur if $e(x)$ has a minimum at $x=x_m\equiv N_h/N_m$. Thus phase separation is equivalent to the hole's condensing into a self-bound system of concentration x_m. When x_m is close to 1, it may be simpler physically to think of the hole-rich phase as a low density of particles, but we shall continue to express the problem mathematically as a minimization of $e(x)$.

This discussion is obviously relevant, since either superconductivity or condensation into a liquid or both may be consequences of an attractive interaction. Indeed, liquid ^{3}He, a fermion superfluid, is self-bound in precisely the same sense as we are considering here. It may be objected that, unlike ^{3}He atoms, the holes in high-T_c superconductors are charged, and that Coulomb interactions will prevent condensation. Nevertheless, on the theoretical level, our results are significant because the t-J model as usually studied does not contain Coulomb interactions, and it is desirable to know what its phase diagram really is. Moreover, in the materials themselves, the holes are often donated by oxygen atoms which are quite mobile at temperatures below the consolute temperature of the holes, which we expect to be of order J/k_B. Hence the "background" O^{2-} atoms may also phase separate and compensate for any local charge imbalance. Precisely such phase separation occurs[5] in oxygen-doped La_2CuO_4. Evidently a complete picture would include the energies of the oxygen defects, but it is clear that the holes themselves may be a force for phase separation and that charge imbalance will not necessarily prevent it.

Another consideration is that liquid ^{3}He is a three-dimensional system, whereas holes in the CuO_2 planes are two dimensional. The distinction may be quite significant. Consider first the case of a purely attractive

interaction. It is well known[6] that the system will collapse to arbitrarily high density when the dimension $d > 2$. This may be shown by means of a variational argument, using a free Fermi-gas trial wave function. The attractive potential energy is proportional to the density n, whereas the kinetic energy varies as $n^{2/d}$. Thus, for large n, the potential energy dominates when $d > 2$: The Fermi pressure is incapable of sustaining the system against collapse. In practice, stabilization is provided by a short-range repulsion as, for example, in liquid ^{3}He or nuclear matter. But it is clear that in three dimensions the attractive part of the interaction does not have to be strong in order to produce a self-bound many-body system. In particular, it can certainly be much too weak to form a two-body bound state.

The situation is very different in two dimensions where an arbitrarily weak purely attractive potential will bind a pair,[7] but collapse is marginal and the formation of a many-body bound state is delicate. The instability is at long distances. *A priori*, it seems possible that a two-dimensional system may be able to form a low-density liquid of pairs, which should be a superfluid at low temperatures. To investigate this possibility for a potential with a short-range repulsion and a longer-range attraction of strength V, it would be necessary to determine the critical values of V for the formation of bound two-, three-, or four-particles bound states and for condensation into a liquid.

With this background, we now turn to the t-J model. The Hamiltonian is given by

$$H = J\sum_{i,k}(\mathbf{S}_i\cdot\mathbf{S}_j - \tfrac{1}{4}n_in_j) - t\sum_{i,j,s}[c^\dagger_{i,s}c_{j,s} + \text{H.c.}], \quad (3)$$

where $c^\dagger_{i,s}$ creates an electron of spin s on site i, and n_i,$\mathbf{S}_i$ are the electron number and spin operators, respectively. The Hamiltonian is supplemented by the constraint that there be no doubly occupied sites. Thus, there is an attractive potential $(-J)$ between electrons in singlet states on neighboring sites.

For perspective, it is interesting to consider generalizations of the model in which the spin coupling is anisotropic (Heisenberg-Ising model) and possibly ferromagnetic, and also to add an interaction V between electrons on neighboring sites. We have made a detailed study[8] of the phase diagrams of the extended model in the small-t limit and have found that phase separation is the rule rather than the exception. For the t-J model itself, we start out by considering the large-J and small-J limits.

When J is large, the hole-rich phase consists of all holes, $x = 1$. This may be understood in the following way. The energy to remove an electron from the purely antiferromagnetic phase ($x = 0$) is $2BJ = 1.168J$ and the gain in kinetic energy is $-4t$. Thus the fully phase-separated state is unstable to the transfer of single electrons when $J \le J_1 = 3.42t$. However, it may be shown that two electrons form a bound state when $J > 2t$. (The interaction is separable and the Schrödinger equation may be solved analytically for zero binding energy.) So, as J is decreased, there will be an instability to pairs before single particles; we have shown that it occurs at $J_2 = 3.828t$. It is also necessary to consider the possibility of three- or four-body bound states. We have not done this for an infinite system but, for a 4×4 lattice, we find that J must be considerably larger than J_2 in order to form three- or four-body bound states and that as J decreases, the first instability is to pairs at $J = 3.53t$. It seems likely that the first instability is to pairs in the infinite system also. If so, the hole-rich phase may be regarded as a dilute gas of pairs of electrons when J is slightly less than J_2. Since the pairs are bosons, they probably form a superfluid at low temperatures. As J is decreased still further, the density of pairs may build up sufficiently for the system to become a BCS superconductor but there should be a crossover to a new state at least by $J = 2t$ (where the pairs unbind) since a necessary and sufficient condition for BCS pairing *in a dilute gas* is that there is a two-body bound state.[7] But, for that value of J/t, the equilibrium density of electrons in the hole-rich phase is already quite high, and results for a low-density gas may no longer be relevant. In a BCS picture, a crossover to pairing in higher angular momentum states would occur as the density increases. We stress that we have not proved that x_m decreases continuously from 1 at large J; the system might, in principle, jump to a moderately high density of electrons, for which the preceding discussion would be inappropriate.

In the other limit, $J \ll t$, we follow an argument similar to that of Visscher[9] and of Ioffe and Larkin[9] and show that a very low density of holes is unstable to phase separation with the hole-rich phase ferromagnetic. By the variation principle, this argument is sufficient to demonstrate phase separation, whatever the character of the true ground state of the hole-rich phase. In a ferromagnetic region, the holes behave like spinless fermions with an energy spectrum

$$\epsilon_{\mathbf{k}} = -2t[\cos k_x + \cos k_y]. \quad (4)$$

For a low density of holes, the cosines may be expanded to second order in k_x and k_y and the total energy of the separated phase may be written in the form of Eq. (2) with

$$e(x) = 2BJ/x - 4t + 2\pi tx. \quad (5)$$

Minimizing with respect to x gives $x_m = (BJ/\pi t)^{1/2}$ and

$$e(x_m) = -4t + 4(BJ\pi t)^{1/2}. \quad (6)$$

This is to be compared with the energy per hole in a uniform phase which, in the low-density limit, is the same as that of a single hole in an antiferromagnet. If the hole does not create a ferromagnetic bubble around itself, its energy is[10] $-(12)^{1/2}t + f(J)$, where $f(J) > 0$. If it does create a ferromagnetic bubble, it is easy to show that the energy is given by Eq. (6) with the $\sqrt{tJ}$ term multiplied

by an extra factor of $2\sqrt{\pi}$. In either case, the energy of the phase-separated state is lower when J is sufficiently small.

The physical reason for phase separation in the large-J limit is that the gain in exchange energy by maximizing the number of antiferromagnetic bonds outweighs the cost in kinetic energy. In the small-J limit, phase separation occurs because the motion of a hole in an antiferromagnet is frustrated and it is better to put all of the holes into a region in which they have a lower kinetic energy. In one dimension, the same kind of argument goes through for large J but not for small J, because the kinetic energy is not frustrated.

Since there is phase separation for $J \ll t$ and $J \gg t$, it is reasonable to expect that the same qualitative behavior should be found for all values of J/t and that x_m increases as J/t increases. Physically, this is to be expected because if the background remains antiferromagnetically correlated there is an attractive interaction between holes on opposite sublattices. We have tested this idea by carrying out exact numerical diagonalization of the Hamiltonian on a 4×4 lattice[11] and interpreting the energy of N_h holes as the energy of a hole concentration $x = N_h/16$, which is reasonable when the number of *electrons* is not too small. For example, when $x = 0$, the energy per site differs from that of the infinite system by about 5%. Figure 1 shows $e(N_h/16)$ for a 4×4 lattice as a function of N_h, for $J/t = 0.1$, 0.4, and 1.0. The minima at $x = x_m(J)$ indicate phase separation for $x < x_m$. Figure 2 gives x_m as a function of J/t. The individual data points are for a 4×4 lattice and the curve illustrates the expected behavior for an infinite system, matching to $x_m = (BJ/\pi t)^{1/2}$ at small J/t and going to $x_m = 1$ when $J > 3.828t$. The points at $J/t = 2.5$, 3.0, and 3.5 all correspond to two-particle bound states, and surely are not representative of a finite concentration of holes. However, they do suggest that the hole-rich phase of an infinite system consists of a gas of pairs in this region.

An important implication of these results is that the transition from the ordered antiferromagnet to the doped state is first order. It is therefore not clear that the doped system can ever be regarded as a weakly disordered antiferromagnet.

Phase separation will occur in other proposed models of the copper oxide planes of high-temperature superconductors. The original arguments of Visscher[9] and of Ioffe and Larkin[9] were applied to the Hubbard model for large U/t. It can be seen that the discussion presented above relies on energies of the Heisenberg model, the ferromagnetic state, and the $U \to \infty$ limit, for all of which the t-J model and the Hubbard model are equivalent. To go further, it is necessary to resort numerical calculations since the small-U/t limit cannot be analyzed in a simple way. Riera and Young[12] studied the large-U form of the Hubbard model (t-J model plus pair hopping) on a 4×4 lattice and showed that $E_{B2} \equiv 2[e(\frac{2}{16}) - e(\frac{1}{16})]$ is negative for all relevant values of U/t but that $E_{B4} \equiv 4[e(\frac{4}{16}) - e(\frac{2}{16})]$ is negative only for $U \leq 5t$. These results are consistent with phase separation since $x_m \to 0$ as $U/t \to \infty$ and, if $e(x)$ is monotonic increasing for $x > x_m$, E_{B4} must be positive when $x_m < \frac{2}{16}$. It is evident that numerical calculations for much larger systems are required to study $e(x)$ for small values of x, but the possibility of phase separation

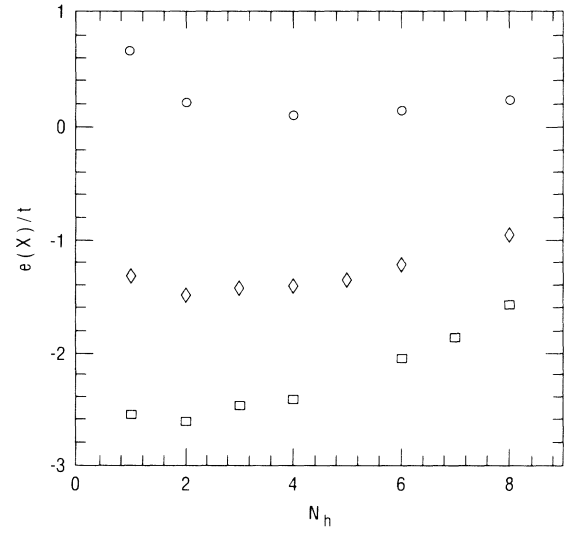

FIG. 1. $N_h e(x)$ is the energy to add N_h holes. $e(x)$ is shown as a function of N_h for $J/t = 0.1$ (squares), 0.4 (diamonds), and 1.0 (circles). Note that the states with odd or even N_h have different spin and that the oscillations in the energy are a finite-size effect.

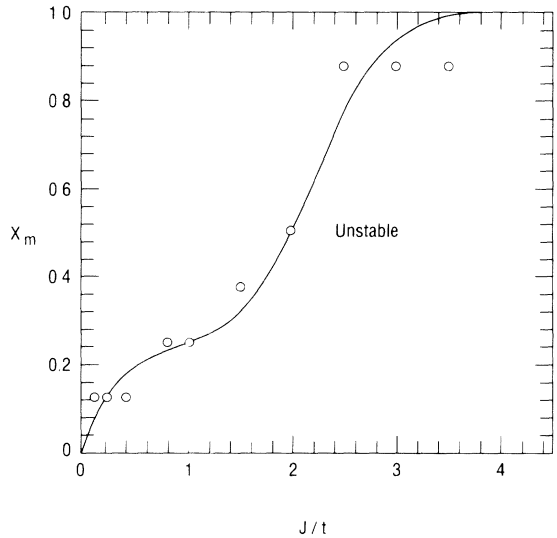

FIG. 2. $x_m = N_h/N_m$, the point at which $e(x)$ has minimum, is shown as a function of J/t. The data points are values for a 4×4 lattice and the curve illustrates the expected behavior for an infinite system (see the text).

should be investigated more thoroughly wherever it is found that two holes are bound. Recently, Dagatto *et al.*[13] have shown that $E_{B2} < 0$ for the Hubbard model on a 4×4 lattice with $U/t = 4$.

Phase separation may also occur in three-band models of the copper oxide planes,[14,15] but the physical reasons may depend on the particular range of parameters assumed. In some cases, the arguments are similar to those for the t-J model. But if the Coulomb interaction V between holes on neighboring sites is too large, that alone is sufficient to destabilize the system.[16] Materials aspects of phase separation have been discussed by Gor'kov and Sokol.[17]

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Note added.—After this paper had been submitted for publication, we received a report from Marder, Papanicolaou, and Psaltakis,[18] who show that there is phase separation in an N-component generalization of the t-J model when $N \to \infty$.

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