Phase Separation of Holes in Antiferromagnets

BNL--43373

DE90 003255

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Received by

NOV 29 1989

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ABSTRACT

It is shown that dilute holes in an antiferromagnet are unstable against phase separation into a hole-rich phase and a no-hole phase. When the spin exchange interaction J exceeds a critical value J_c , one phase consists of all holes, the other all electrons. The argument is presented in detail for the $t-J$ model but evidence of phase separation in other models is also mentioned.

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A good deal of the theoretical work on high-temperature superconductors¹ has been concerned with the properties of diluted antiferromagnets. By now, it is well-established experimentally that parent compounds such as La_2CuO_4 , $YBa_2Cu_3O_6$ or Nd_2CuO_4 are antiferromagnetic insulators², and that superconductivity is induced by doping electrons or holes into CuO_2 planes. It is therefore natural to suppose that the correlations which are responsible for antiferromagnetism are of importance for the electronic structure in the metallic state, and possibly for the mechanism of superconductivity.

It has been proposed³ that the essential features of the problem are expressed in the t - J model, which starts from the assumption that the parent compounds are well-represented by the Heisenberg model, with localized electrons of spin-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 models³⁻⁵ 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.

Most studies of the t - J model have assumed that the density of the holes is uniform and have investigated the stability of various magnetic phases (such as spiral states or flux phases) and their relationship to superconductivity⁶. The question to be addressed here is whether the holes do indeed have a uniform density 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:

$$E = -2BJN_s + N_h e(x) \quad (1)$$

Here N_s is the total number of sites in the system and BJ is the energy per bond in the Heisenberg antiferromagnet: the best estimate of BJ by extrapolation from finite size systems⁷ is $B = 0.584$. The number of holes is N_h and the number of sites in the hole rich phase is N , with $x \equiv N_h/N$. Thus $N_h e(x)$ is the energy to add N_h holes to an antiferromagnet at concentration x . Now E will have a minimum at $N = N_m < N_s$ if $e(x)$ has a minimum at $x = x_m \equiv N_h/N_m$. Thus phase separation is equivalent to the holes' forming a self-bound system of concentration x_m : if the average concentration is less than x_m , the holes will condense into a liquid or solid state with 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 $e(x)$. Holes in the copper oxide planes are a two-dimensional system. In our investigation of the conditions for and the properties of a self-bound state, we have found that there are interesting differences between two- and three-dimensions⁸.

These are obviously relevant considerations since either superconductivity or condensation into a liquid or both may be consequences of an attractive interaction. Indeed, liquid He^3 , a fermion superfluid, is self-bound in precisely the same sense as we are considering here. It may be objected that, unlike He^3 atoms, the holes in high T_c superconductors are charged, so Coulomb interactions will prevent condensation. Nevertheless, on the theoretical level, our results are

relevant 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 occurs⁹ 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.

The Hamiltonian of the $t-J$ model is given by:

$$H = J \sum_{i,j} (\vec{S}_i \cdot \vec{S}_j - \frac{1}{4} n_i n_j) - t \sum_{i,j,s} [C_{i,s}^\dagger C_{j,s} + H.c.] \quad (2)$$

where $C_{i,s}^\dagger$ creates an electron of spin s on site i ,

$$n_i = \sum_s C_{i,s}^\dagger C_{i,s} \quad (3)$$

and

$$S_i^m = \frac{1}{2} \sum_{s,s'} C_{i,s}^\dagger \sigma_{ss'}^m C_{i,s'} \quad (4)$$

is the m th component of the spin operator and $\sigma_{ss'}^m$ are elements of Pauli matrices. 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.

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 $J \leq J_1 = 3.42t$. However, it may be shown that two electrons form a bound state when $J > 2t$.

So, as J is decreased, there will be an instability to pairs before single particles at $J = J_2 > J_1$. 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. It seems likely that the same is true for an infinite system. Note that the binding energy on a 4×4 lattice provides a variational estimate of the binding energy for an infinite system.

If the transition is continuous then for $J < J_2$, the hole-rich phase may be regarded as a dilute gas of pairs of electrons. Since the pairs are bosons, they should be 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 $J = 1.69t$ (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 state⁸. 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 the transi-

tion is continuous and that 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 have followed an argument similar to that of Ioffe and Larkin¹⁰ and show that a very low density of holes is unstable to phase separation with the hole-rich phase ferromagnetic and

$$x_m = \sqrt{BJ/\pi t} \quad (5)$$

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.

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 , since 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 even if the background remains antiferromagnetically correlated there is an attractive interaction between holes on opposite sublattices. We have tested this idea by carrying out numerical calculations on a 4×4 lattice⁹ and interpreting the energy of N_h holes as the energy for $x = N_h/16$, which is reasonable when number of

electrons is not too small.

There is evidence⁸ of phase separation in the Hubbard model and in other models¹¹ of the copper oxide planes. It is evident that analytical studies should explore the possibility of phase separation in order to ensure that proposed phases do not occur in the unstable region. If it is argued that phase separation is unphysical and, in practice, would be removed by Coulomb or other interactions, then those interactions should be included in the model, as studied. Certainly Coulomb interactions will inhibit pairing and consequently conventional superconductivity. Moreover, the present calculations imply 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.

As we have argued above, the holes maybe a force for phase separation in the real materials. Their consolute temperature is of order J/k_B which is in the range 1500-2000K. Thus the holes tend to phase separate at typical annealing temperatures, where the oxygen ions are mobile and therefore are able to compensate for any charge imbalance.

Acknowledgements This work was supported in part by NSF grants DMR 87-06250 (at SUNY), PHY 82-17853 (at UCSB) supplemented by funds from NASA at UCSB and by the U.S. Department of Energy under Contract No. DE-AC02-76CH00016.

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