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Electronic Phase Separation and High Temperature Superconductors

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Abstract

We review the extensive evidence from model calculations that neutral holes in an antiferromagnet separate into hole-rich and hole-poor phases. All known solvable limits of models of holes in a Heisenberg antiferromagnet exhibit this behavior. We show that when the phase separation is frustrated by the introduction of long-range Coulomb interactions, the typical consequence is either a modulated (charge density wave) state or a superconducting phase. We then review some of the strong experimental evidence supporting an electronically-driven phase separation of the holes in the cuprate superconductors and the related Ni oxides. Finally we argue that frustrated phase separation in these materials can account for many of the anomalous normal state properties of the high temperature superconductors and provide the mechanism of superconductivity. In particular, we show that the T-linear resistivity of the normal state is a paraconductivity associated with a novel composite pairing, although the ordered superconducting state is more conventional.

I. INTRODUCTION:

Much of the theory of correlated electron systems and high temperature superconductors [1] is concerned with stable electronic phases of uniform density, although the actual materials are not always so well-behaved. High temperature superconductors typically are metastable compounds [2] in which electronic inhomogeneity is more the rule than the exception, [3,6] although high temperature superconductivity itself is a robust and obvious phenomenon. At the same time, numerical experiments [7] on a variety of models of correlated electron systems have so far failed to produce any indication of a significantly enhanced superconducting susceptibility in models that incorporate the most important short-range interactions in the copper oxide planes [8]. Moreover these calculations usually omit the long range part of the Coulomb interaction, which in any BCS-like theory would further suppress superconductivity, the more so since retardation is relatively ineffective, especially in materials with the highest transition temperatures. All in all, it is difficult to escape the feeling that an essential piece of physics is missing.

Our candidate for the missing ingredient is the competition between two features of correlated electron systems: the tendency of doped holes in a commensurate insulator [9–11] to separate into hole-rich and hole-free phases, and the long-range part of the Coulomb interaction which prevents macroscopic phase separation from actually taking place. From this perspective, high temperature superconductivity is a consequence of the conversion of a global instability (phase separation) into a local instability (superconductivity) by recruiting the long range part of the Coulomb interaction instead of fighting it. In this review we shall show that *electronic* phase separation is an established feature of idealized models *and of the real materials*, and that it may present itself as either a macroscopic effect or a local and dynamical phenomenon. We then go on to describe our progress in constructing a theory of the anomalous normal state properties of high temperature superconductors and the inevitable evolution into a superconducting state as the temperature is lowered. This part of the story is based on exact or controlled solutions of several models but, so far,

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the relationship between the various models has not been established to our satisfaction. In this sense, the *existence* of electronic phase separation is theoretically and experimentally more secure than our understanding of its consequences. Nevertheless, following this route, we have been led to a compelling interpretation of the well-known T-linear resistivity $\rho(T)$ of high temperature superconductors as a *paraconductivity* arising from composite (or equivalently odd-frequency) pairing fluctuations, with a high temperature scale of order the antiferromagnetic exchange integral J. This approach provides a natural explanation of the hitherto mysterious insensitivity of $\frac{d\rho}{dT}$ to phonons and impurities, [12] which is an important requirement of any theory. In a companion paper, [13] we discuss some of the implications of these ideas for the ordered (superconducting) state, and show in particular how odd-frequency fluctuations may evolve into even-frequency order.

The remaining sections of this paper are intended to be self-contained reviews of the various aspects of phase separation in high temperature superconductors: theoretical arguments (Section II), the experimental basis (Section III) and the consequences for the normal state properties and superconductivity (Section IV).

A central feature of high temperature superconductors is that they are doped insulators [8], obtained by chemically adding charge carriers to a highly-correlated antiferromagnetic insulating state. By now there is a good deal of theoretical evidence [9,10], reviewed in Section IIA, that, in the absence of long-range Coulomb interactions (*i.e.* for neutral holes), a low concentration of holes is typically unstable to phase separation into a hole-rich "metallic" phase and a hole-deficient antiferromagnetic phase. In one sense phase separation certainly can be regarded as a strong attractive interaction between holes, and hence it seems natural that it should be related to the mechanism of superconductivity. But the situation is somewhat more complicated for high temperature superconductors since the mechanism of phase separation is more properly regarded as the ejection of holes from the antiferromagnet. Nonetheless a variety of model calculations show that the interactions that produce phase separation can also lead to superconductivity. Section IIB discusses the appearance of superconductivity near the phase boundary for phase separation, and reviews the rea-

sons for believing that this mechanism can only lead to low temperature superconductivity. Section IIC describes examples of an alternative scenario in which superconductivity (and possibly high temperature superconductivity) arises when macroscopic phase separation is "frustrated" by a long-range repulsive interaction. Finally Section IID elaborates the nature of the phase diagram when the frustrating interaction is a $1/r$ Coulomb repulsion between holes. In particular, it is shown that the Coulomb interactions do *not* generally favor a uniform density phase but rather produce charge-modulated structures, with periods that are unrelated to nesting wave vectors of the Fermi surface. These structures may be static, as in a charge density wave or a "cluster spin-glass" phase, or dynamic with a finite length scale, especially when the system is sufficiently quantum in character. We shall refer to the latter situation as "fluctuating phase separation."

In Section III, we turn to experimental evidence that holes in the cuprate superconductors tend to phase separate. Section IIIA considers materials in which the dopant atoms are mobile on laboratory timescales, so that actual macroscopic phase separation is possible. Early on we predicted [9] that this effect is electronic in origin, and is not driven by elastic interactions between the excess oxygen ions. The persuasive evidence that this is the case is discussed in some detail for the best studied example, $La_{2-x}Sr_xCuO_{4+\delta}$. Section IIIB discusses the more usual situation of dopants that are immobile on laboratory timescales and presents evidence that fluctuating phase separation occurs in a large and relevant region of the phase diagram. Recent experiments showing (i) that the spin glass phase observed at low doping is, in fact, a cluster spin glass, [14] and (ii) that the dynamical spin structure factor $S_{AF}(\vec{q},\omega)$, where $\vec{q}$ is the departure of the wave vector from (π,π), takes the form of a product $F(\vec{q})\tilde{S}(\omega,T)$: [15] both confirm predictions made some time ago [16–18] on the basis of frustrated electronic phase separation. These experiments provide quantitative confirmation of the idea that there is substantial charge inhomogeneity, with locally Néel ordered spins in the hole deficient regions.

Section IV summarises the status of our efforts to understand the anomalous normal state properties and the existence of high superconducting transition temperatures in the cuprates

as consequences of fluctuating phase separation. In the most general terms, it is widely recognized that the normal state properties of high temperature superconductors require that, for an extended range of doping and temperature, there is a class of low frequency (quasi-classical) collective excitations which strongly scatter the conduction electrons. Typically, such excitations are on the verge of condensation into an ordered state. Moreover, the fact [19] that superconductivity in purposely disordered materials is only destroyed when $k_F l = 1$ (where a superconductor to insulator transition occurs) implies that, whatever the mechanism of superconductivity, it *cannot* rely on any pronounced structure in k-space. The only known ordered state which overlaps the metallic phases of the cuprate superconductors is the cluster spin glass. In our view, the slow density fluctuations in the metal and the existence of the cluster spin glass are closely related consequences of frustrated phase separation. Given the glassy nature of the nearby ordered phase, and the fact that frustrated phase separation is not related to a Fermi surface instability, it is natural to ignore the k-dependence of the scattering of the conduction electrons. Thus the important processes that determine the low-energy and low-temperature behavior of high temperature superconductors are essentially local in space. We show that they are governed by the same fixed point as the two-channel Kondo problem and that they provide a natural explanation of much of the normal state phenomenology of high temperature superconductors. In particular, we find a compelling interpretation of (a) the well known T-linear resistivity in terms of a paraconductivity arising from composite (or equivalently odd-frequency) pairing fluctuations with a high temperature scale J; and (b) the specific frequency and temperature dependence of the dynamical spin structure factor, as measured by neutron scattering and NMR.

There are several extant reviews dealing with various aspects of phase separation in high temperature superconductors. The present review is, in many ways, an update of a previous paper by the two of us, [18]. A review of numerical approaches to the problem can be found in [20]. The proceedings of two recent conferences [4,5] on the subject are excellent sources of both experimental and theoretical overviews. The reader is particularly referred to a paper by Di Castro and Grilli [21] in which a rather different perspective on electronic

phase separation and high temperature superconductivity is discussed. [22] In this context, it is important to stress that a wide variety of models and materials exhibit phase separation, and that this can occur for reasons having nothing to do with antiferromagnetism. For instance, in various three band models of the copper oxide planes, phase separation occurs [21] when the system is close to a charge-transfer instability. It also is possible that non-electronic effects could cause phase separation in some range of doping. While these other interactions could augment the tendency toward phase separation in high temperature superconductors, we focus on the role of antiferromagnetism because the undoped system is an antiferromagnetic insulator, and related spin fluctuations remain a prominent feature of the doped system. Moreover, as we shall see, experiments indicate that one of the two coexisting phases is essentially hole-free. This favors a magnetic mechanism [9] rather than a charge-transfer instability, [21] which would lead to hole-rich and a hole-poor phases.

Finally, it is often useful for perspective to view a given phenomenon in a wider context. In this case there are two other systems to which comparison may be made. The first is ^{3}He - ^{4}He mixtures whose phase diagram, shown in Fig. 1, displays a two-phase region extending at $T = 0$ from $0.06 < x < 1$, where x is the concentration of ^{3}He. Not only does this system phase separate but, as in the case of holes in an antiferromagnet, the mechanism is the expulsion of defects from the ordered state (the superfluid phase of ^{4}He), not a trivial classical interaction. Indeed ^{3}He and ^{4}He differ only in their masses and their statistics, both of which are irrelevant in classical statistical mechanics. Thus, there is an analogy between the superfluidity of the ^{4}He and antiferromagnetism, and between the ^{3}He concentration and the hole concentration. The second system worth mentioning is $La_2NiO_{4+\delta}$, which is identical in structure to $La_2CuO_{4+\delta}$ with Ni replacing the Cu. When undoped ($\delta = 0$), this system is a spin 1 antiferromagnet, so it is expected that quantum fluctuations will be considerably less important than in the cuprates. When doped, $La_2NiO_{4+\delta}$ is known to form a variety of modulated phases, [23] although the complete phase diagram has not yet been fully worked out.

II. PHASE SEPARATION IN MODEL PROBLEMS AND ITS IMPLICATIONS

This section gives a critical discussion of theoretical arguments and numerical studies of phase separation. The reader who is willing to accept our conclusion that in models of neutral holes in an antiferromagnet there is phase separation into a hole-rich phase and a hole-poor phase could omit this section and proceed to the discussion of experiments on the cuprates (Section III), and the consequences of frustrated phase separation (Section IV).

A. Phase Separation in Models of Holes in an Antiferromagnet

There is a general tendency for mixtures of substantially different entities to phase separate at low temperatures; a doped antiferromagnet is such a system. The antiferromagnet may be represented as a hard core Bose liquid in which a site is occupied by a boson when the spin is up and unoccupied when it is down. The bosons have a heavy mass corresponding to a bandwidth of order J. The holes can be represented as a gas of spinless fermions with a light mass. (Note the analogy with ^{3}He - ^{4}He mixtures.) Thus, it is hard to imagine a mixture of two more dissimilar objects. Moreover, it is easy to see that two holes in an antiferromagnet attract each other. At the shortest distances, this attraction arises from the fact that the number of antiferromagnetic bonds removed by two isolated holes is reduced by one if the holes are on nearest-neighbor sites. At larger distances, two holes attract each other through the exchange of magnons.

Stripped to its bare essentials, the tendency to phase separation is a reflection of the electrons' attempt to minimize their zero-point kinetic energy. The problem of simultaneously maintaining local antiferromagnetic order, which optimizes the zero-point energy for the half-filled band, and allowing itineracy for the holes away from half-filling is very difficult because these two requirements are intrinsically contradictory. Phase separation is the simplest solution to such problems of frustration, and typically it has the lowest energy. At zero temperature, there are two contributions to the energy of the holes which tend to

oppose phase separation: the Fermi pressure and the long-range Coulomb interaction. In this section, we consider neutral holes, that is to say holes with no long-range $1/r$ Coulomb repulsion.

As far as we know, all solvable models, or solvable limits of models of holes in a Heisenberg antiferromagnet show phase separation. That is to say, at zero temperature, there is a miscibility gap below a critical hole concentration x_c which can depend on the details of the model and the values of coupling constants. Based on this observation, we have conjectured that

> *In dimensions $d \geq 2$, there always exists a miscibility gap for neutral holes in a Heisenberg antiferromagnet at sufficiently low temperature and hole concentration.*

It is still not known whether this general result is true. As discussed below, we believe that evidence to the contrary obtained from numerical studies is quite inconclusive. At the same time, the Heisenberg symmetry is essential, as may be seen from the solution of the $t-(J_z,J_\perp)-V$ model which we introduced some time ago: [10]

$$H = -t \sum_{<i,j>,\alpha} [c^\dagger_{i,\alpha} c_{j,\alpha} + H.c.] + \sum_{<i,j>} [J_z S^z_i S^z_j + J_\perp \vec{S}^\perp_i \cdot \vec{S}^\perp_j - V \rho_i \rho_j] \qquad (1)$$

where $c^\dagger_{i,\alpha}$ creates an electron with z component of spin α on site i, $\vec{S}_i = \sum_{\alpha,\alpha'} c^\dagger_{i,\alpha} \vec{\sigma}_{\alpha,\alpha'} c_{i,\alpha'}$ and $\rho_i = \sum_\alpha [c^\dagger_{i,\alpha} c_{i,\alpha} - 1/2]$ are respectively the spin and density operators on site i, $< i,j >$ designates pairs of nearest-neighbor sites, $J_z/J_\perp \neq 1$ is a Ising-Heisenberg anisotropy, and there is an implied constraint of no double occupancy, $\rho_i = \pm 1/2$. For $J_z = J_\perp = J$ and $V = J/4$, this model reduces to the usual $t-J$ model. [24] We addressed the issue of phase separation by studying the model in the solvable small-t limit, where phase separation is the general rule. However, it has recently been shown [25] that for $J_z \gg t \gg \sqrt{|J_z - V| J_z}$ and in the extreme Ising limit, $t \gg J_\perp$ there exists a narrow region of parameter space in which phase separation does not occur. This is an interesting result, but it does not address the fundamental issue, since in the extreme Ising limit, the induced interactions between holes

are extremely short-ranged. In the Heisenberg limit, by contrast, the induced interactions have infinite range.

While this general question is of considerable accademic interest, it is of little practical importance. All that matters is that phase separation is a possible consequence of doping holes into an antiferromagnet. It is then to be determined, *from experiment*, whether this behavior is exhibited in the actual materials in the relevant range of concentrations and temperatures. This is particularly true since phase separation is a first order transition and may therefore depend on microscopic details which can never be incorporated accurately in any simple model. It will be shown in Section III that experiment indicates that the doped holes in high temperature superconductors tend to phase separate at all experimentally relevant temperatures and, as a minimum, for those dopant concentrations for which superconductivity is observed.

1. Phase Separation in Solvable Models of Holes in an Antiferromagnet

The most extensive analytical studies of phase separation have been carried out in the context of the $t-J$ model. It is evident that phase separation occurs at large J/t, where the attractive exchange interaction between electrons exceeds the Fermi energy. [9] Here, at zero temperature, the critical hole density x_c is given by:

$$x_c(J/t) = 1, \quad \text{for} \quad J/t > 3.828, \tag{2}$$

i.e. the system phase-separates into a no-hole antiferromagnetic phase ($x = 0$) and a no-electron phase ($x = 1$). As J/t decreases below 3.828, the critical concentration drops continuously from 1, i.e.

$$x_c(J/t) < 1, \quad \text{for} \quad J/t < 3.828. \tag{3}$$

At *very* small J/t the physics is quite different. However, in this limit, it is easy to show [26,9] that the holes are strongly dressed; they become large ferromagnetic polarons with essentially

infinite effective mass and hence vanishing Fermi pressure. The small residual attraction then leads to phase separation. While this conclusion is not rigorous, it is supported by a strong variational calculation, [9] and there seems to be little doubt that it is correct. Here, the critical concentration is quite small and in fact it vanishes as $J/t \to 0$. Specifically the variational calculation [9] gives:

$$x_c(J/t) \sim 0.43\sqrt{J/t}, \quad \text{for} \quad J/t \ll 1. \tag{4}$$

These results are exhibited as the solid lines in the zero temperature phase diagram in Fig. 2. At intermediate J/t, there are only numerical results, discussed in Subsection 3.

The phase diagram of holes in an antiferromagnet has also been obtained by generalizing the $t-J$ model so as to produce a small parameter. Thus, both the large N [27–29] and large S [30,18] generalizations of the $t-J$ model have been shown to phase separate. The behavior in the large N limit is dominated by Nagaoka physics, as for very small J/t. The large S limit is somewhat more revealing. It was shown by Auerbach and Larson [30] that for $J/t < 1$ the solution of the single hole problem in the limit $S \to \infty$ is a small polaron (whose size and shape depends on J/t) with a ferromagnetic core and undisturbed antiferromagnetic order beyond some finite radius. They found that coherent motion of these polarons does not occur to any finite order in 1/S, but rather there is an effective hopping matrix element $t^{eff} \sim \exp[-Sf(t/J)]$, where the function f is of order 1. On the other hand, we have shown [18] that in order 1/S the exchange of magnons induces long-range interactions between holes. Thus phase separation into some appropriate hole crystal phase is always favored in the large S limit. In a sense this shows that the system renormalizes to the large J/t limit, even if the bare J/t is small. In this context it is worth pointing out that the best current numerical estimate [31] of the renormalized bandwidth of one hole in the $t-J$ model (exact diagonalization up to 26 sites) gives $W^{eff} \approx 2.3J$ for small J/t, or, in other words, $J/t^{eff} \approx 4$.

2. Phase Separation and the Hartree-Fock Approximation

There have been several variational calculations of the ground state of the lightly doped Hubbard model [32]- [39] or a three-band model of the CuO_2 planes of high temperature superconductors, [40] mostly using the Hartree-Fock approximation. As with the $t-J$ model, we consider the Hubbard model on a square lattice (or, when we consider dimensions other than $d=2$, a hpercubic lattice) where t again denotes the nearest-niehgbor hopping matrix, and U is the repulsion between two electrons on the same site.

Two-sublattice Hartree-Fock: The simplest trial wave function [39] allows for the possibility of broken spin-rotational symmetry and broken translational symmetry with a two-site supercell. This permits Néel phase, but none of the variety of helical or striped phases that have been considered for the doped system; we will discuss these more complicated states below. For small Hubbard U, and on an arbitrary dimensional hypercubic lattice (where nesting is always complete in the half-filled band) it is easy to solve the $T=0$ Hartree-Fock equations analytically.

For dimensions $d \neq 2$, the density of states at the Fermi energy per site per spin polarization, ρ_0, is constant, and one finds the well known result that a spin-density wave with Néel symmetry has the lowest energy for the half-filled band (*i.e.* for chemical potential $\mu = 0$), with a resulting gap in the charge-exciation spectrum,

$$\Delta_0 \sim \exp\{-1/\rho_0 U\}[1+\mathcal{O}(U/t)]. \tag{5}$$

This state has condensation energy (relative to the metallic state) $E_A = -\frac{1}{2}\rho_0\Delta_0^2[1+\mathcal{O}(U/t)]$, and it remains the lowest energy state for a finite range of μ, reflecting the existence of a charge gap. However the zero temeperature free energy of a metallic state with no broken symmetry is $E_M = -\rho_0\mu^2[1+\mathcal{O}(U/t)]$, and it has the lowest free energy for $\mu > \Delta_0/\sqrt{2}$. In other words, there is a two-phase region with a critical concentration,

$$x_c = 2\rho_0\mu = \sqrt{2}\Delta_0[1+\mathcal{O}(U/t)], \tag{6}$$

and the energy per hole of the two-phase state is

$$\epsilon = \mu = \Delta_0/\sqrt{2}. \tag{7}$$

Dimension $d = 2$ is special, even at this crude level of approximation, because of the Van Hove singularity in the density of states at the Fermi energy ($\epsilon = 0$) of the half-filled band, $\rho(\epsilon) \sim \rho_0 \log\{1/\rho_0|\epsilon|\}$ and $\rho_0 = 1/8\pi^2 t$. Nonetheless, the above analysis goes through with little change. The charge gap at half-filling is [41]

$$\Delta_0 \sim \exp\{-\sqrt{1/2\rho_0 U}\}[1 + \mathcal{O}(\sqrt{U/t})], \tag{8}$$

with corresponding condensation energy $E_A = -\frac{1}{2}\rho_0\Delta_0^2 \log(1/\rho_0\Delta_0)[1 + \mathcal{O}(\sqrt{U/t})]$. The metallic state has zero temperature free energy $F_M = -\rho_0\mu^2[\log(1/\rho_0\mu) + 3/2]$, which means that a first order transition to the metallic state occurs as a function of μ at $\mu^2[\log(1/\rho_0\mu) + 3/2] = \frac{1}{2}\Delta_0^2 \log(1/\rho_0\Delta_0)[1 + \mathcal{O}(\sqrt{U/t})]$, or in other words, $\mu \approx \Delta_0/\sqrt{2}$. This implies that the two-phase region extends to

$$x_c = 2\rho_0\mu[\log(1/\rho_0|\mu|) + 1] = \Delta_0/\sqrt{2\log(1/\rho_0\Delta_0)}[1 + \mathcal{O}(\sqrt{U/t})], \tag{9}$$

and that the energy per hole in the two phase region is

$$\epsilon = \mu = \Delta_0/\sqrt{2}[1 + \mathcal{O}(\sqrt{U/t})]. \tag{10}$$

Even the simple two sublattice Hartree-Fock equations cannot be readily solved analytically at finite temperature, but a numerical solution is not difficult. [39] However, independent of details, it is clear that Δ_0 sets the temperature scale for phase separation and that, in the Hartree-Fock approximation, phase separation can never occur at temperatures in excess of the Néel temperature. (Fluctuation effects, of course, can cause serious violations of this bound.)

Incommensurate magnetic structures and stripes: Several studies [32] - [39] have included more complex Hartree-F ck states, particularly ones with large supercells. These are found by minimizing the Hartree-Fock energy numerically for arbitrary broken symmetry within a specified supercell. In general there are many local minima with different broken symmetries but similar energies, [35] depending on the value of U/t, the hole concentration x,

and the nature of the unit cell, *i.e.* the nature of the boundary conditions. The most widely discussed solutions of this sort are ones in which the magnetic order is incommensurate and collinear (Ising-like). In the dilute hole limit, these incommensurate states become increasingly anharmonic and are best described as striped phases in which the holes are localized in domain walls (discommensurations) of width $\xi_0 = \hbar v_F/\Delta_0$, in which the hole density is 1 per site. In $d = 1$, the domain walls are simply the Hartree-Fock versions of the charge e and spin 0 solitons created upon doping the $d = 1$ Hubbard model; the soliton state has Hartree-Fock energy [39] $\epsilon \leq 2\Delta_0/\pi = 0.64\Delta_0$ per hole, and hence has lower energy than the phase-separated state. In $d = 2$ stripe states are local minima of the energy for small enough x. [32] The stripes are vertical (along a lattice direction) for small U while, for larger U, diagonal stripes are thought to have lower energy. There is no analytic estimate of the energy of these striped phases. Schultz [32] obtained an estimate $\epsilon = 0.66\Delta_0$ for a vertical stripe by fitting a numerical solution of the Hartree-Fock equations on a 96×24 lattice. Since he worked with $U/t \sim 1$, this energy is equal to the energy of the phase separated state within the uncertainty of the calculation. It is important to stress that there is no doubt that, whatever its precise nature, the Hartree-Fock ground-state involves substantial clustering of charge. For our purposes, this is all that is important, since in the presence of long-range Coulomb interactions, it is only a local tendency to clumping of charge that survives.

Finite Size Effects as Geometric Frustration: It is important to realize that studies on finite-size periodic lattices, in effect, frustrate phase separation. For example the evidence for a striped phase in a mean-field approximation for the Hubbard model with 7 holes on a 7×7 lattice obtained by Poilblanc and Rice [34] is consistent with phase separation for the thermodynamic limit. If a finite number of holes tend to cluster, they will form domains whose shape is partly determined by the dynamics of the holes and partly by the boundary conditions. [33,35] It is reasonable to suppose that the hole kinetic energy prefers elongated domains, and then the spin boundary conditions would force the domain to lie on the diagonal of the 7×7 supercell, as found by Poilblanc and Rice. [34]

The stripe solution does not minimize the variational energy for any other hole density; in effect the stripe curls up on itself within the unit cell or the holes cluster together and surround themselves with a vortex spin texture. [35] Some studies have *fine-tuned* the hole density to favor a striped phase for a particular supercell. [34] This is a completely reasonable way to study the properties of the putative striped phase, but it is clearly not the optimal way to determine whether the striped phase has the lowest energy in the thermodynamic limit. In effect, boundary conditions can be chosen for finite size systems either to favor or to frustrate particular states. The only way to find the true mean-field ground state is to compare energies extrapolated to the thermodynamic limit.

Fluctuations about the Hartree-Fock state: The occurrence of a striped phase is a consequence of the competition between commensurability effects (which favor regions of Néel ordered antiferromagnet) and the nesting of the Fermi surface. In this sense, they reflect similar physics to phase separation. When the hole concentration is so low that the separation L between domain walls is large compared to $\xi_0 = \hbar v_F/\Delta_0$, the Hartree-Fock interaction between domain walls is repulsive but exponentially small: $V_{int} \sim exp(-L/\xi_0)$. [32] Two sorts of quantum fluctuations can make qualitative changes in the physics of the striped phase. 1) The domain walls themselves can fluctuate. So long as these fluctuations are not too great, they renormalize the width ξ_0 of the domain wall but do not destroy the integrity of the stripes. If, on the other hand, fluctuations are so severe that the system is on the rough side of the quantum roughening transition, then the striped phase is completely eliminated. 2) Assume, for present purposes, that the domain walls do not roughen, and focus on the effect of long-range (power-law) interactions induced by *magnetic* fluctuations, [18] which dominate the (exponentially small) mean-field repulsion between the domain walls. If the domain walls are ordered on alternate sublattices, (*i.e.* for a diagonal striped phase) the long-range interaction between neighboring domain walls is strictly attractive. Since, by assumption, the system is on the smooth side of the quantum roughening transition, there is no analog of the Fermi pressure to prevent bunching of the domain walls. Thus,

a low density diagonal striped phase is unstable to quantum fluctuations;

the domain walls themselves will phase separate, forming a high density striped phase coexisting with an undoped antiferromagnetic phase. It is at present unclear whether a uniform density vertical striped phase is similarly unstable against quantum fluctuations.

3: Phase Separation or not in Numerical Studies of Holes in an Antiferromagnet

There have been a fairly large number of numerical studies of phase separation for particular models of holes in an antiferromagnet. They are of two types: high temperature series expansions which address the thermodynamic limit, and exact diagonalization, finite temperature Monte Carlo or projection Monte Carlo studies, which deal with finite-size systems. There are two basic problems with the finite size studies: 1) The systems are all too small to permit a convincing finite size scaling analysis, so there is no systematic way to extract the thermodynamic limit. 2) When the critical hole concentration is small, it may not be possible to access the relevant regime of concentrations at all. The finite temperature studies are, of course, reliable only at high enough temperatures. This is particularly troublesome since phase separation is a first order transition so there need not be any precursor effects above the critical temperature. To illustrate these points, we give a (non-exhaustive) discussion of some representative numerical calculations.

Exact Diagonalization of the $t-J$ Model: There have been several such studies, [9,42] which agree on the facts, but disagree on interpretation. We considered a 4×4 system with periodic boundary conditions and an arbitrary number of holes, [9] as a supplement to analytical arguments, in order to obtain the boundary for phase separation. For this purpose we interpreted the results as an approximation for the thermodynamic limit of the ground-state energy as a function of the corresponding concentration of holes. Then the results imply phase separation at all values of J/t, with a phase boundary that interpolates smoothly between the large and small J/t limits. This is shown as the crosses in Fig. 2. In the later calculations of Dagotto and coworkers, [42] on a 4×4 lattice, the same results

were interpreted as evidence of pair binding rather than phase separation. (The same was true of earlier calculations on small Hubbard model systems by Riera and Young. [43])

All groups agree on the nature of the phase diagram at large J/t, and none of the numerical calculations purports to address the nature of the phase diagram in the regime of Nagaoka physics, $J/t \ll 1$. However, in the interesting range [24] $0.1 \leq J/t \leq 0.5$, the difference in interpretation rests on the following point. Let $E_N(n)$ represent the ground-state energy of an N site system with n holes. (Of course, n and N are, necessarily, both integers.) Then, for $0.1 \leq J/t \leq 0.5$, it is found that $[2E_{16}(1) - E_{16}(2) - E_{16}(0)] > 0$, but $[2E_{16}(2) - E_{16}(4) - E_{16}(0)] < 0$ and $[E_{16}(3) - E_{16}(2) - E_{16}(1) + E_{16}(0)] < 0$. Dagotto *et al* [42] concluded that there is pair binding but not phase separation. That is to say, they made the identification $\lim_{N\to\infty}[E_N(n) - E_N(0)] \approx [E_{16}(n) - E_{16}(0)]$. We on the other hand [9] used the same results to conclude that $x_c(J/t) < 3/16$ in this regime of parameters, i.e. we made the identification $\lim_{N\to\infty}[E_N(xN) - E_N(0)]/N \approx [E_{16}(x16) - E_{16}(0)]/16$ for $x = n/16$. The only way to use numerical calculations to decide between the two interpretations is to work with larger systems containing either two holes or a fixed hole concentration, and to extrapolate to the thermodynamic limit. Boninsegni and Manousakis [44] have carried out such a study, using the Greens function Monte Carlo technique to find the energy of a two-hole d-wave bound state for periodic lattices up to 10×10. Their results show that the interpretation of the results on a 4×4 lattice in terms of pair-binding is problematic; for instance, for $J/t = 0.4$, they find that the pair-binding energy on an 8×8 lattice, $[2E_{64}(1) - E_{64}(2) - E_{64}(0)] \approx 0.1t$, is approximately one third of its value on a 4×4 lattice, $[2E_{16}(1) - E_{16}(2) - E_{16}(0)] \approx 0.35t$. Extrapolating to the thermodynamic limit, they conclude that there is no pair binding for $J < J_c \sim 0.3t$. At the present time, it is not feasible to carry out the same program for a fixed concentration and thus, the question of the phase boundary at intermediate J/t remains open. In this regard, it is worth commenting that pair binding (or indeed, the binding of any finite number of particles) is neither necessary nor sufficient for phase separation. The two effects are entirely unrelated: phase separation is a global instability whereas pair-binding is a local property.

Projection Monte Carlo on the Hubbard Model: Recently Furukawa and Imada [45] used projection Monte Carlo on the Hubbard model to conclude that the compressibility vanishes as the hole concentration approaches zero, but that there is no phase separation. These results, which are primarily for 8×8 periodic lattices, but include some results for 12×12 systems, are the largest size calculations of the ground state energy of the Hubbard model. Nevertheless, there is still a substantial finite size effect. For example, in the best studied case (an 8×8 system with the Hubbard $U = 4t$) the chemical potential as a function of hole concentration has a discontinuity at $x = 0$, corresponding to the charge gap Δ_0 at half filling, and then is constant within numerical error over the range $0 < x \leq 7/32$. If these results were representative of the thermodynamic limit, they would imply phase separation with a critical concentration $x_c = 7/32$. An alternative explanation, adopted by Furukawa and Imada, relies on shell structure. In the noninteracting system, the momentum eigenstates occur in degenerate bands, and for the 8×8 Hubbard model the chemical potential would be constant for $-7/32 \leq x \leq 7/32$. Furukawa and Imada then assert that the flatness of the chemical potential indicates simply that the shell structure survives rather strong interactions. Therefore they ignore all results save those at hole concentrations corresponding to filled shells. For a $2n \times 2n$ system, the minimum hole concentration corresponding to a filled shell is $x_{min} = (n-1)/4n^2$ which, even for a 12×12 system is $x_{min} = 0.15$. Thus, *if this interpretation is adopted*, these calculations teach us nothing about phase separation with a critical concentration $x_c < x_{min}$, and it is unlikely that the results will ever shed much light on the issue if the Hartree-Fock value $x_c \sim \rho_0 \Delta_0$, discussed above, is correct.

High Temperature Series: The phase diagram of the $t-J$ model has also been studied by high temperature series and resummation by Padé approxmiants. The different approximants begin to diverge from each other for temperatures below about $J/2$, [46] and indeed this problem becomes worse as J/t decreases, as might be expected for a system with more than one characteristic energy scale. Thus, the conservative approach is to trust these results only for $T > J/2$, which may exceed the highest temperature at which phase separation

occurs. Singh and Gleinster [46] have computed the uniform spin susceptibility and the spin structure factor $S(\vec{k})$ for $T > J/2$ and $J/t = 1/2$. They find that $S(\vec{k})$ is peaked at $\vec{k} = (\pi, \pi)$ for $x = 0$ and 0.2, whereas it is strongly incommensurate, $\vec{k} = (\pi(1-q), \pi)$, with $q \approx 0.6$ when $x = 0.4$. This is suggestive of phase separation into antiferromagnetic droplets with a critical concentration $0.2 < x_c < 0.4$ for $J/t = 1/2$, consistent with our estimates based on finite size studies [9,24] of $1/8 < x_c < 1/4$. On the other hand, Putikka *et al* [47] have studied the high temperature series for the chemical potential, μ, and have argued that it can be more readily extrapolated to low temperature than the series for the uniform spin susceptibility studied by Singh and Gleinster. Thus, Putikka *et al* have attempted to use high temperature series to compute $\mu(x, T)$ in the limit $T \to 0$; in this way, they concluded that for $J/t < 1.2$, the system does not phase separate. The phase boundary deduced by Putikka *et al* is shown as the dashed line in Fig. 2. It is not clear to us why the series for $\mu(x, T)$ should be reliable to so much lower temperatures than $T \sim J/2$ at which the different Padé approximants for the spin susceptibility diverge. At present, details of the analysis of Putikka *et al* have not been published. It should also be noted that Putikka *et al* actually look for the position of the pseudospinodal (where the compressibility diverges) which, a priori, need not be close to the boundary of the two-phase region.

Monte Carlo Calculations on the Hubbard Model: Moreo and coworkers [48] have performed quantum Monte Carlo calculations on the Hubbard model. In addition to the troubles with finite size effects discussed above, these calculations are typically carried out at too high temperatures to address the issue of phase separation. Again, from the Hartree-Fock results, we expect to see phase separation only at temperatures low compared to Δ_0.

Variational Monte Carlo Calculations: Extensive variational Monte Carlo calculations of the ground-state energy of the $t - J$ model have been carried out by Trivedi and coworkers based on various Gutzwiller projected RVB wave-functions. [49] Although the authors did not interpret their results in this way, a simple Maxwell construction applied to their numerical results leads to the conclusion that there exists a miscibility gap at low hole concentration of about the same magnitude as we ourselves have found.

B. Superconductivity at the Border of Phase Separation

Since phase separation can, in some sense, be thought of as a strong attractive interaction between holes, it seems reasonable that, just outside the two-phase region of the phase diagram, there might enough residual attraction to produce superconductivity but not phase separation. We have demonstrated analytically that this is the case for large J in the $t-J$ model. [9] Specifically, as discussed above and as shown in Fig. 1 for zero temperature, we found that $x_c = 1$ for $J/t > 3.828$, whereas for J/t slightly less than 3.828, $x_c(J/t) \propto [3.828 - J/t]$. Thus, for $x_c(J/t) < x < 1$, there is a stable, dilute electron phase. We also found that two electrons form a bound-state in the $t-J$ model for all $J/t > 2$. Since for the dilute electron gas in two dimensions it has been shown [50] that pair binding is a necessary and sufficient condition for superconductivity, this implies that the hole rich state is indeed superconducting. Moreover, since the two-particle bound state is in the s-wave channel, it follows that the superconducting phase has spin singlet, s-wave symmetry. (As an aside, it was shown by Lin [51], that four electron bound states form only for $J/t > 4.85 \pm 0.05$.)

This idea was extended by Dagotto and Riera [52] by numerical diagonalization of the $t-J$ model on small periodic lattices. They found that for smaller values of J/t, both d-wave and s-wave correlations are strong, and that the d-wave seems to be dominant at somewhat smaller values of $J/t \sim 3$ and $x \sim 1/2$. This suggests the possibility that a transition from s-wave to d-wave pairing might occur in the $t-J$ model near the boundary of the two-phase region. While these results are inconclusive due to the small systems sizes, it is worth noting that a transition from s-wave to d-wave pairing is not unprecedented; it is known to occur as a function of density in a dilute gas with a short range repulsive interaction and a long range attraction. [53]

Another route to superconductivity at the border of phase separation is pairing via the exchange of density fluctuations, which may be soft because of the proximity of the pseudospinodal (where the compressibility vanishes). [54] However it is not easy for this mechanism to give rise to high temperature superconductivity because it would require that the

pseudospinodal be close to the phase boundary. In general this is not the case. For first order transitions the fluctuation region is even smaller than for a higher order phase transition.

As an illustration of these physical arguments, consider the case of ^{3}He - ^{4}He mixtures. At low enough temperatures, there is a possiblility that the border of the two-phase coexistence region may have a "double superfluid" phase in which both the ^{3}He and ^{4}He components are superfluid. (See Fig. 1) To date, no experimental evidence of fermion superfluidity in the minority ^{3}He fraction has been detected. Theoretical estimates [53,55] show that the superfluid transition temperature has a maximum value $T_c \sim 1\mu K$, for $x \sim 1.3\%$. This is not on the phase boundary and, since the Fermi energy of the minority ^{3}He fraction is $E_F \sim 0.14K$ it certainly is not an example of high temperature superconductors.

C. Superconductivity and Frustrated Phase Separation in Solvable Models

Another route to superconductivity is to "frustrate" the phase separation by introducing a longer range repulsion which prevents the system from actually decomposing. We addressed [10] this question through an analytic solution of the $t-J-V$ model [24] defined in Eq. (1) (with $J_z = J_\perp$) in the small t limit. Of course, for $V = 0$, the system phase separates with $x_c = 1$. The zero temperature phase diagram of this model is shown in Fig. 3, where it is evident that, for $1/2 < x < 1$, charge 4e or 2e superconducting phases are produced when V is increased above a critical value. The superconducting transition temperatures in these phases are, respectively, $T_c \sim t^4/J^3$ and $T_c \sim t^2/J$, and in both cases are peaked in the vicinity of the upper phase boundary (where the maximum T_c is, respectively, $T_c^{max} \sim t^2/J$ and $T_c^{max} \sim t$.) At large V the phase marked FL is a Fermi liquid at least down to very low temperatures. Numerical calculations on this model were later carried out by Dagotto and Riera [56] for large but finite J/t and $x = 1/2$. Even for a 4×4 system they obtained evidence of superconducting correlations which are strongly *enhanced* as a function of an increasingly repulsive interaction V.

Of course, both of these studies deal with a rather unphysical limit of the model, and so

strictly do no more than answer the question of principle. However, an important perspective is suggested by these calculations. Numerical studies of small systems may not be able to detect superconductivity for any realistic model since superconducting correlation lengths may be as large or larger than the systems that are amenable to numerical solution. However, if we truly understand the mechanism of high temperature superconductivity, then we should be able to change the parameters to reduce the superconducting correlation length to an accessible value. It is *only* on the border of phase separation and especially in models in which phase separation has been supressed by the inclusion of additional long-range repulsive interactions that *clear* evidence of superconductivity has been obtained in any numerical study. [52,56] This observation is a strong piece of evidence that frustrated phase separation is a viable mechanism for high temperature superconductivity.

Having established the principle that frustrating phase separation can give rise to superconductivity, we must add that we do not believe that these model calculations truly represent what is going on in high temperature superconductors which are, in fact, much more interesting. First of all, in the $t-J-V$ model, the value of V was "tuned" to produce finite clusters (pairs or quartets). Since the finite clusters are bosons they condense into a superfluid state. But this route will not necessarily give rise to high temperature superconductivity because the clusters may be rather heavy. Nor will it produce certain anomalous normal state properties of high temperature superconductors such as the T-linear resistivity. Secondly, phase separation in high temperature superconductors arises from "bare" repulsive interactions, and the residue of frustration is not clustering but strong nonlinear local density fluctuations, as described in Section IV. Our idea is that the scattering of the mobile holes from these local density fluctuations is responsible for the anomalous normal state properties and for high temperature superconductivity itself. In order for phase separation to be manifested locally, it clearly is necessary to be between the pseudospinodals of the neutral system. (Phase separation is a global instability whereas spinodal decomposition is, of course, local.) In this way it is guaranteed that the effects of the collective modes are strong, in contrast to the situation on the border of phase separation.

It is worth pointing out that some of the physics in which we are interested may be studied with the aid of numerical calculations on finite size systems since, as pointed out above, the boundary conditions can introduce sufficient geometric frustration to effectively suppress static phase separation.

D. Effect of Coulomb Interactions

It has always been clear that macroscopic phase separation is suppressed by the long-range Coulomb interaction. [9] However this does *not* mean that the Coulomb interactions stablize uniform density states of the neutral system. Indeed, for jellium, the Coulomb interaction favors *local* charge-inhomogeneity (a Wigner crystal) whereas it is the *kinetic energy* that inclines the system to be uniform. As we have shown, the situation is entirely different for a correlated electron systems for which minimization of the zero-point kinetic energy is achieved by separation into hole-rich and hole-free regions: all energies conspire to produce a state that is inhomogeneous on some length scale and time scale, although of course macroscopically it must be uniform. The dynamical character of this state of frustrated phase separation typically stems from the *quantum* nature of the problem and is not easily displayed in a simple model. As a warmup problem, we have studied a *classical* Ising pseudospin model [57] of the effects of Coulomb interactions on a system with a local tendency to phase separation.

The Hamiltonian for the model is given by:

$$H = K \sum_j S_j^2 - L \sum_{<ij>} S_i S_j + \frac{Q}{2} \sum_{i \neq j} \frac{S_i S_j}{r_{ij}} \tag{11}$$

Here $S_j = \pm 1, 0$ is a coarse-grained variable, representing the local density of mobile holes. [18] Each site j lies on a two-dimensional square lattice and represents a small region of space in which $S_j = +1$ and $S_j = -1$ correspond to hole-rich and hole-poor phases respectively, whereas $S_j = 0$ indicates that the local density is equal to the average value. The fully phase separated state has $S_j^2 = 1$ and is ferromagnetically ordered, with $S_j = +1$ in one half of the volume, and $S_j = -1$ in the other, so as to maintain overall charge neutrality.

The zero temperature phase diagram was determined for the complete range of parameters by using a combination of numerical and analytical techniques. [57] It was found that the pure Coulomb interaction favors a Néel state (equivalent to a Wigner crystal) but, as Q decreases, the system crosses over to a ferromagnetic (phase-separated) state via a rich structure of highly symmetric striped and checkerboard phases. Regions with uniform charge density, corresponding to sites with $S_j = 0$ do not occur unless K is positive and sufficiently large.

The solution of this simple model confirms our intuition that local inhomogeneity is the expected consequence of frustrated phase separation and that it should be a characteristic behavior of metallic correlated electron systems. But it also indicates that *ordered* charge-modulated states are a likely outcome if the system is sufficiently classical. [58] As we shall see in the next section, such charge-modulated states occur in $La_{2-x}Sr_xNiO_{4+\delta}$, and indeed some aspects of the complex phase diagram of this material have been explained by a variant of the model of Eq. (11). [59] It is evident that high temperature superconductivity can only occur as a consequence of frustrated phase separation if the charge ordering is suppressed by quantum fluctuations. This is the major difference between $La_{2-x}Sr_xNiO_{4+\delta}$ and $La_{2-x}Sr_xCuO_{4+\delta}$. The fact that Ni is spin-one whereas Cu is spin-half clearly is a significant factor, but the different atomic states of the doped holes may also play a role.

III. ELECTRONIC PHASE SEPARATION IN CUPRATE SUPERCONDUCTORS AND RELATED MATERIALS

The existence of phase separation in model calculations does not, of course, prove that it occurs in real materials. In this section we discuss some of the unambiguous experimental evidence that an electronic tendency of the holes to phase separate is an important feature of the cuprates. It is necessary to distinguish two limiting situations. If the charge donors are mobile on laboratory timescales (i.e. if the dopant distribution does not fall out of equilibrium in the relevant temperature regime) they can be dragged along by the holes and compensate the long-range part of the Coulomb interactions; in this case, the holes

can macrocopically phase separate. On the other hand, if the dopant atoms are absolutely immobile then clearly phase separation can only occur as a short distance, fluctuation effect. The latter is the situation in the majority of the cuprate perovskites in the experimentally relevant temperature regime.

Figure 4 shows a schematic phase diagram for a representative cuprate superconductor, (solid lines) using $La_{2-x}Sr_xCuO_{4-\delta}$ as the canonical example. Dashed lines show the putative boundary of the two-phase region for neutral holes, and dotted lines the corresponding pseudospinodals. Note that the boundary of the two-phase region is drawn in such a way that most of the interesting physics, except for antiferromagnetism, occurs within the unstable region of the neutral hole phase diagram. In the present section, we will analyze some of the experimental evidence which supports the validity of both this picture and the implication that high temperature superconductivity occurs in a region of frustrated phase separation. By extension, it is therefore reasonable (although, of course, not inevitable) to conclude that the interesting physics of these materials in this regime of doping is a consequence of frustrated phase separation.

A. True Phase Separation in the Case of Mobile Dopants

As far as we know,

> *in all cases in which the dopant species are mobile on laboratory time scales, there exists a miscibility gap at small dopant concentrations.*

In particular, in photo-doped materials, in oxygen-doped $La_2CuO_{4+\delta}$, and in H-doped $YBa_2Cu_3O_{7-\delta}H_x$, there is strong evidence of electronic phase separation associated with the rise of antiferromagnetic correlations. We feel that the fact that phase separation occurs in all known cases where the dopants are mobile provides strong experimental support for an electronic mechanism for phase separation whenever the long-range Coulomb interactions do not prohibit it.

Some may find it strange that the mobile holes can actually drive phase separation of the dopants; the tail seems to be wagging the dog. It is easy to see that this feeling is unwarranted for the examples discussed above for two reasons: 1) The energy associated with electronic phase separation is of order $J \sim 0.1$eV, which is larger than in most other magnetic materials. 2) The very fact that, at room temperature, the dopants are mobile on laboratory timescales means that the characteristic energy differerences between different dopant configurations cannot be very much larger than ~ 0.025eV.

This is not to say that dopant interactions play no role in determining the *location* of the boundaries of the two phase region. Tendencies of the dopants to form preferred structures (staging) will certainly affect the equilibrium hole density in the hole-rich phase. Even in the extreme case in which the configuational energy of the dopants is completely negligible, we surely can never ignore the configurational entropy, and this alone can cause a significant depression of T_{PS}, the critical temperature for phase separation. [18]

1. O doped $La_2CuO_{4+\delta}$

The additional oxygen in $La_2CuO_{4+\delta}$ is mobile on laboratory timescales down to temperatures of order $T = 200$K, and the dopant distribution reflects the equilibrium thermodynamics of the system. Following the landmark work of Hundley *et al*, [60] Hammel *et al* [61] have obtained evidence from the La NMR spectrum that phase separation occurs at $\sim 230K$, and they obtained the phase diagram shown in Fig. 5. NMR is a local probe, and both the Knight shift and the relaxation rate $1/T_1$ are extremely sensitive to static differences in the local environment. Above the Néel temperature T_N, Hammel *et al* observe a single NMR line, which implies that the material is homogeneous, at least on the long times characteristic of NMR. Below T_N, the NMR spectrum splits into two distinct pieces, one similar to that of metallic $La_{2-x}Sr_xCuO_{4-\delta}$, and the other similar to that of the undoped antiferromagnetic insulator La_2CuO_4. This behavior is completely reversible; the system becomes single-phase again once the temperature is raised above T_N, implying that phase

separation in these materials *is* an equilibrium phenomenon. Similarly, Jorgensen *et al* [62] have shown by means of neutron diffraction that oxygen-doped $La_2CuO_{4+\delta}$ consists of two phases at low temperature: one nearly stoichiometric $La_2CuO_{4+\delta'}$ with $\delta' \approx 1\%$ and the other oxygen-rich $La_2CuO_{4+\delta''}$ ($\delta'' \sim 8\%$). Moreover, at lower temperatures, this material has a superconducting $T_c = 38K$, independent of δ, which strongly supports the existence of phase separation and implies [63,64] that the metallic phase has a hole concentration between 0.13 and 0.20.

We now examine the evidence that the observed phase separation is driven by the electronic physics of the CuO_2 planes, not by superfluous oxygen chemistry. In the first place, the concentrations of the two coexisting phases have special significance for the electronic physics; the hole-rich phase is an optimally doped superconductor and the hole-deficient phase lies on the boundary for antiferromagnetism. It would be a remarkable coincidence if oxygen chemistry chose two such special concentrations. However, the most direct proof of an electronic mechanism comes from the careful studies by Cho *et al* [64] of O-doped $La_{2-x}Sr_xCuO_{4+\delta}$, with x and δ in the range 0 to 0.03. It is well known that for $\delta = 0$ (no mobile O), $T_N(x)$ is a rapidly decreasing function of x and reaches zero at $x = x_N \approx 0.02$. Cho *et al* observed that, in the O-doped material ($\delta > 0$), $T_N(x,\delta)$, the superconducting T_c, and the Meissner fraction are all diminished by Sr doping. In fact for $\delta > 0$, $T_N(x,\delta)$ still vanishes at about the same $x = x_N$, the Meissner fraction vanishes continuously at $x = x_S \approx 0.03$, but T_c remains finite for all $x < x_S$. For x just greater than x_S, apparently there is no antiferromagnetic order, no static phase separation (as inferred from the vanishing Meissner fraction), and no superconductivity. This behavior is precisely what is expected for electronically-driven phase separation. The presence of immobile Sr dopants implies a minimum hole concentration in the hole-poor phase. This clearly reduces the energetic advantage of phase separation and hence reduces $T_{PS}(x,\delta)$ ultimately to a point where the oxygen as well as the Sr dopants cease to be mobile on laboratory timescales. Small hole concentrations substantially disrupt the antiferromagnetic correlations in the CuO_2 planes, which are the primary driving force for both phase separation and Néel ordering. Thus,

T_N is rapidly driven to zero and $T_{PS}(x,\delta)$ is rapidly depressed by Sr doping. In the limit of small interplane coupling, longer inplane magnetic correlations are required for Néel ordering than for phase separation. Consequently we expect that $x_N < x_c = x_S$, where x_c is the critical concentration for phase separation at $T \approx 200$K (the temperature at which the system falls out of equilibrium). This is in agreement with experiment. On the other hand this whole scenario would be entirely unexpected if the driving force for phase separation were the interaction between the oxygen ions.

The behavior of this system is strongly dependent on thermal history, [65,66] and the observed phase diagram does not reflect the equilibrium physics of the oxygen defects below about $T = 200$K where the oxygen mobility decreases rapidly. It is likely, for instance, that the boundary of the two phase region is not as vertical as in Fig. 5, but rather continues to move to higher hole concentration as the temperature is lowered. Indirect evidence of this can be derived from experiments of Ahrens *et al*, [66] who find that in samples of $La_2CuO_{4+\delta}$ annealed at a given temperature, T_{aneal}, and then rapidly quenched to low temperatures, T_c is a function of T_{aneal}. This is true even for annealing temperatures as low as 150K and probably implies that, at least on some rather short length scale, it is possible for the oxygen distribution to equilibrate on the timescale of the aneal. The fact that T_c continues to depend on T_{aneal} implies that the hole concentration of the hole-rich phase increases with decreasing temperature between 200K and 150K, although the slow diffusion rate of the added oxygen masks this phenomenon on a macroscopic scale.

2. $YBa_2Cu_3O_{7-\delta}$

The oxygen atoms in the chains of $YBa_2Cu_3O_{6+\delta}$ with $0 \leq \delta \leq 1$ are somewhat mobile [67,68] even at room temperature, although not as much as in $La_2CuO_{4+\delta}$. (Presumably, this is due to the covalent bonding of the chain oxygen to the Cu.) Nonetheless, there appears to be a miscibility gap at low hole concentrations in these materials as well. Specifically, in well-anealed samples, there is a first order transition [67] from an antiferromagnetic insulating

phase for $\delta < 0.45$ to a superconducting phase with $T_c \sim 60$K for $0.45 < \delta < 0.7$. However, we do not consider this to be strong evidence of electronically-driven phase separation, since models of oxygen chain-ordering, based on oxygen-oxygen interactions, account for this behavior in considerable detail. [69] Thus, while these experiments are consistent with the general observation of a miscibility gap between the doped and undoped phases of the cuprates, they do not directly lend support to the notion that there is an intrinsic tendency of holes in the Cu-O planes to phase separate.

3. H doped $YBa_2Cu_3O_{7-\delta}$

The hydrogen in $YBa_2Cu_3O_7H_x$ acts as an electron donor, or, in other words, acts to undope the Cu-O planes. It was determined in recent ^{1}H and ^{63}Cu-NMR studies [70] of $YBa_2Cu_3O_{6.95}H_x$ ($0 \leq x \leq 0.6$) that the hydrogen phase separates, producing a two-phase material. The hydrogen-rich phase exhibits the same NMR spectrum, the same Néel temperature ($T_N \approx 410$K) and the same temperature dependence of the order parameter as the parent insulating compound, $YBa_2Cu_3O_6$. The other phase is hydrogen free, and hence is superconducting below the same transition temperature ($T_c \approx 90$K) as $YBa_2Cu_3O_{6.95}$. Phase separation occurs in these samples regardless of thermal history, which implies that the critical temperature for phase separation is greater than room temperature (i.e. the temperature at which the materials are protonated). Again, the fact that the material phase separates into the same two electronically special phases as $La_2CuO_{4+\delta}$ is highly suggestive that the driving force for phase separation is dominantly electronic.

4. Photodoped cuprates

In the extensive studies of photodoped cuprate perovskites, [71–73], the insulating (antiferromagnetic) parent material is exposed to high intensity light with photon energies in excess of the insulating gap, and the photo-induced changes in the optical absorption spectrum and the photo-conductivity are monitored. The holes rapidly find their way to the

CuO_2 planes, while the electrons play a role analogous to the dopant atoms in chemically doped materials. Generally, one interprets these experiments as reflecting the properties of lightly photo-doped materials, but the electrons are far more mobile than chemical dopants, and so can respond on laboratory timescales to the putative clumping tendency of the holes in the CuO_2 planes. There is strong experimental evidence of clumping of the holes in photodoped materials. This evidence has been sumarized in several articles, for example in Refs. [18] and [74]

B. Experimental Evidence for Frustated Phase Separation

We now consider the case in which the dopant atom distribution is quenched. Here, the intrinsic tendency of the holes to phase separate is forbidden by the long-range Coulomb interactions and it can only manifest itself as large amplitude charge and spin density inhomogeneities. *Static* inhomogeneities are either charge density wave states with period unrelated to a nesting vector of the Fermi surface or, in the presence of disorder, a cluster spin glass phase in which the density modulations are randomly pinned. On the other hand, in sufficiently quantum systems, the modulated structures intrinsically dynamical and we shall refer them as "fluctuating phase separation".

In the interest of space, we focus on a few of the experiments which most clearly show evidence of fluctuating phase separation. Basically, there are two classes of phenomena that we discuss here: 1) The thermodynamic definition of two-phase equilibrium is the existence of a range of density for which the chemical potential is constant. Since fluctuating phase separation implies the coexistence of small regions of the metallic phase and the hole-poor insulating phase, the change in the chemical potential (or Fermi level) should be anomalously small as the dopant concentration is varied. [75] 2) The classic way to probe the proximity of a system to any thermodynamic instability is to study the response to an applied field which couples to the putative order parameter of the ordered phase. In the case of frustrated phase separation, this means the response of the system to weak, local electric potentials; *i.e.*

small variations in the local potential will lead to large amplitude electronic inhomogeneities. By contrast, in a stable metal, due to screening, small inhomogeneities in the structure or composition of the material produce very little in the way of electronic inhomogeneity. Thus the second direct experimentally observable manifestation of frustrated phase separation is a tendency of a system to strongly overscreen any material defect, producing pinned, local regions of one or the other of the two stable phases.

1. Pinning of the Chemical Potential

Direct evidence of fluctuating phase separation comes from photoemission studies of $Nd_{2-x}Ce_xCuO_4$. [76] Here, it is found that, as the dopant concentration varies, the chemical potential remains within the insulating gap, while the density of states in the gap grows roughly in proportion to the electron concentration. Similar evidence was obtained for hole doped $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_8$ by Watanabe *et al.* [77] This is precisely the expected behavior for an inhomogeneous system: The density of states in the gap arises from the metallic regions, whereas the gap features are associated with the insulating regions. The conditions of chemical equilibrium require that the chemical potential be equal in the two regions, and hence within the insulating gap. This is quite different from the behavior of a semiconductor or the *one-dimensional* Hubbard model [78], for which the chemical potential moves discontinuously from the middle of the Mott-Hubbard gap into the lower or upper Hubbard band. Of course, photoemission is a volume averaged probe, and has limited spatial and temporal resolution, so these results need not imply static and macroscopic phase separation. Nonetheless, the photoemission results strongly suggest that in doped cuprates, segregation of the added charges into heavily doped (hole-rich or electron-rich, depending on the type of doping) and lightly doped (hole or electron-poor) regions must occur on some time and length scale.

2. The Cluster Spin Glass

Sample inhomogeneity (and a consequent electronic inhomogeneity) is known to plague studies of all of the high temperature superconductors, especially when they are not "optimally doped'. [6,67,68,63] For the reasons noted above, we feel this, in itself, is evidence of an intrinsic electronic tendency to phase separation.

As shown in Fig. 4, when the insulating "parent" materials of the high temperature superconductors are lightly doped with immobile dopants, the antiferromagnetic long-range order gives way to a "spin glass" phase, in which there are frozen local magnetic moments but no magnetic long-range order. [79–82] According to μSR, NMR and Mössbauer studies, the spin-freezing temperature goes to zero at about optimal doping. Some time ago, we suggested [16,17] that frustrated phase separation implied that this phase is actually a "cluster spin glass", in which there are regions of nearly perfect antiferromagnetic insulator separated by rivers of metal. Because the metallic regions cause the magnetic order to suffer a more or less random phase shift, the axis of quantization of the Néel order varies randomly from region to region. Recent experiments on $La_{2-x}Sr_xCuO_{4-\delta}$ strongly support this point of view. [14] A similar picture rationalizes some striking results obtained by Keimer *et al* [83] from neutron scattering studies of $La_{2-x}Sr_xCuO_{4-\delta}$, with x between 2% and 5%. They found that the dynamical spin structure factor, $S(\vec{q},\omega)$, is peaked at $\vec{q} = (\pi,\pi)$, with an inverse correlation length $\kappa(x,T)$ well-approximated by the simple empirical relation

$$\kappa(x,T) = \kappa_o(x) + \kappa(0,T) \tag{12}$$

where κ_o is the zero temperature value of the inverse correlation length, a monotonically increasing function of the dopant concentration, and $\kappa(0,T)$ is the inverse correlation length of the undoped material. [84] This is precisely the expected form for a cluster spin glass so long as $\kappa_o^{-1}(x)$ is interpreted as the typical size of an antiferromagnetic region: for x=2%, $\kappa_o^{-1} = 200\mathring{A}$ and, for x=4%, $\kappa_o^{-1}(x) = 40\mathring{A}$. To further corroborate this picture, one would like to observe accompanying charge density fluctuations with characteristic length scale $\kappa_o^{-1}(x)$, for instance by diffuse x-ray scattering or NQR.

The appearance of a cluster spin-glass phase, which even coexists with the superconducting phase over a substantial region of the phase diagram, is probably the single strongest piece of evidence of the persistence of the physics of frustrated phase separation in the presence of quenched dopants. While there is probably a fair amount of dopant disorder, especially in underdoped materials, it is a characteristic of the high temperature superconductors that the dopant atoms are outside the conducting planes, as in modulation-doped semiconductor devices. Thus, the potential produced by the disordered dopants is actually rather gentle, and is smaller than the Fermi energy at moderate doping. That frozen moments with substantial local antiferromagnetic correlations can be induced in a fairly good metal by the introduction of such mild disorder is a striking demonstration of the intrinsic tendency of these materials to develop local electronic inhomogeneities. Of course, introduction of strong disorder potentials, as when the system is doped by Cu substitution, should also induce the formation of a pinned local region of one of the stable phases.

3. Modulated Structures in $La_2NiO_{4+\delta}$

$La_2NiO_{4+\delta}$ is a material with the same structure as $La_2CuO_{4+\delta}$. It is an antiferromagnetic insulator for $\delta = 0$ but does not become a superconductor on doping with Sr or oxygen. A major difference between the two materials is that Ni is spin-one, whereas Cu has spin-half. $La_2NiO_{4+\delta}$ has a wonderfully complex phase diagram, which is just beginning to be explored. [23] It displays phase separation, oxygen staging, charge density wave order, striped phases, and modulated magnetic structures. Once again, the systematics of the phase diagram, in particular the interplay between charge order and antiferromagnetism, supports the notion that the phase separation is driven by the dynamics of the doped holes. In our view, there is static order rather than superconductivity in the nickelates because kinetic effects, especially spin fluctuations for this spin-one system, are less important than in the cuprates, so the system is more classical. In this sense, $La_2NiO_{4+\delta}$ provides an excellent counterpoint for theories of hole dynamics in the cuprates, and gives independent evidence for electronic

phase separation of holes doped in an antiferromagnet.

4. Dynamical Spin Structure Factor in Superconducting Materials

Direct evidence of *fluctuating* phase separation has recently been obtained in neutron scattering experiments on superconducting $YBa_2Cu_3O_{6.6}$, with $T_c = 46K$. [15] It was observed that, within experimental error, the dynamical spin structure factor is a product of two terms: $S_{AF}(\vec{q},\omega) = F(\vec{q})\tilde{S}(\omega,T)$, where $\vec{q}$ is the departure of the wave vector from (π,π), and $F(\vec{q})$ is independent of temperature and frequency. $F(\vec{q})$ can then be viewed as a snapshot of a fluctuating antiferromagnetic domain; its diameter is found to be of order eight lattice constants. In order to understand this experiment in any detail, it is first necessary to study the dynamics of the fluctuating phase separation, which we do in the following section.

IV. CONSEQUENCES OF FRUSTRATED PHASE SEPARATION IN THE HIGH TEMPERATURE SUPERCONDUCTORS

In general it is difficult to obtain a unified understanding of any system in which distinct physics occurs at several different energy scales. Frustrated phase separation in high temperature superconductors is no exception: the "high energy physics" is the competition between the local tendency to phase separation and the long range Coulomb interaction, while the low energy physics is determined by the scattering of the mobile holes from the ensuing large amplitude density fluctuations.

A detailed description of the crossover between the various scales often is finessed by solving such a problem in stages, using an appropriate effective Hamiltonian for each energy scale at which distinct physics occurs. Following the same logic, we have constructed a simple model of the scattering of an electron gas by the low-energy collective modes in order to compute the consequences of fluctuating phase separation and to compare these results with experiments in the high temperature superconductors. While our ultimate goal

is to *derive* the effective low energy theory from a microscopic Hamiltonian, it is our hope (and belief) that the present version captures the essential features of the full problem. This hope is bolstered by the fact that the low energy properties of the low energy model are governed by a critical fixed-point Hamiltonian, so that predictions of the universal low energy behavior are representative of the original, more complicated problem provided only that we have identified the correct fixed point.

The salient features of fluctuating phase separation are: i) the existence of large amplitude local charge and spin density fluctuations, *i.e.* the medium is locally highly polarizable; ii) characteristic wave vectors reflecting the structure of the local antiferromagnetic correlations and any local charge modulations, but *not* particularly nesting the Fermi surface. Both imply that the k-dependence of the important scattering processes is unimportant, and we neglect it in the simplified model. This is equivalent to the assumption that the collective mode is local. Indeed, as mentioned in the introduction, experimental evidence of the extreme insensitivity of T_c to disorder [19] (other than Cu substitution) implies that no particular structure in k-space can be important; this observation lends support to the notion that any important collective mode is quasi-local. Charge conservation implies that the mode must be neutral, but our physical picture suggests that it has a dipole moment, *i.e.* it is a "local dipolar mode." [18] In addition to these assumptions, we ignore spin-orbit coupling and also assume that the strongest coupling between the collective mode and the conduction electrons is a nucleation process involving the local current density. Spin-orbit coupling is a relevant perturbation in the renormalization group sense, and neglecting it relies on the fact that the bare coupling is small, so that it may not become effective in the interesting temperature range. The assumption of dominant current-current coupling is related to the frustration of the motion of a hole in an antiferromagnet; the hole *must* polarize the medium in order to move. On the other hand, a static hole induces a much milder rearrangement of charge and spin in its neighborhood. We do not, at this point, know which results, if any, are sensitive to this assumption; it does not affect fixed point properties but it could influence the amplitude and relative importance of various processes. We have, however,

proven [39] that there exists a duality tranformation which relates the model at strong current coupling to the same model with weak current coupling, so any dependence on the assumed dominance of the current coupling must be subtle indeed.

We are thus led to model the collective modes as an array of local dipolar modes. The array may be ordered, anealed, or quenched-disordered. To be concrete, we consider a one-dimensional lattice realization of this model which can be solved exactly. [85,13] The electronic degrees of freedom are represented by a simple tight binding Hamiltonian, H_0, in which $c^\dagger_{n,\sigma}$ creates an electron with z component of spin $\sigma = \pm 1$ on site n. The collective modes are represented by pseudospin 1/2 operators $\vec{\tau}_{\mathbf{n}}$, where $\mathbf{n}$ labels the positions of the subset of *bonds* on which the local dipolar modes are situated. (Bond $\mathbf{n}$ connects site n and site $n+1$.) In terms of these variables the Hamiltonian is:

$$H = H_0 + \sum_{\{\mathbf{n}\}} \{K_x S^x_{\mathbf{n}} \tau^x_{\mathbf{n}} + K_z S^y_{\mathbf{n}} \tau^y_{\mathbf{n}} + K_z S^z_{\mathbf{n}} \tau^z_{\mathbf{n}}\} \tag{13}$$

where and $S^a_{\mathbf{n}}$, like $\tau^a_{\mathbf{n}}$, is a pseudospin 1/2 operator such that

$$S^x_{\mathbf{n}} = \frac{1}{2} \sum_\sigma \left[c^\dagger_{n+1,\sigma} c_{n+1,\sigma} - c^\dagger_{n,\sigma} c_{n,\sigma} \right] \tag{14}$$

is the local electronic dipole density,

$$S^y_{\mathbf{n}} = \frac{1}{2} \sum_\sigma \left[c^\dagger_{n+1,\sigma} c_{n,\sigma} + \text{H.c.} \right] \tag{15}$$

is the local electronic kinetic energy density, and

$$S^z_{\mathbf{n}} = -\frac{i}{2} \sum_\sigma \left[c^\dagger_{n+1,\sigma} c_{n,\sigma} - \text{H.c.} \right] \tag{16}$$

is the local electronic current density. As mentioned above, we assume that the current coupling is the strongest, $K_z > K_x, K_y$. One can also consider the effects on the properies of the model of a local pseudomagnetic field, $\vec{h}_{\mathbf{n}}$, which couples to the pseudospin $\vec{\tau}_{\mathbf{n}}$. Physically, $\vec{h}_{\mathbf{n}}$ is some sort of local disorder potential since $\vec{\tau}_{\mathbf{n}}$ is itself a charge-density mode. It is important to reiterate that other microscopic realizations of the same physical processes yield identical results in the long wavelength limit.

We have solved this model for certain values of the coupling constants in the continuum (field theory) limit. [86,85,13] In particular, we find that there is a dilute limit when the concentration c of local dipolar modes is sufficiently small, and the results in this limit are universal and dimension independent. The method of solution, and some of the basic results are discussed in the following companion paper. [13] Here we list some of the salient features of the solution, and discuss their physical implications.

A. The Existence of a Dilute Limit and the Two-channel Kondo Fixed Point

We have shown [85] that, for sufficiently small c, the physics of the single dipolar mode is in the same universality class as the two-channel Kondo problem, a critical system [87] for which many the properties are universal for temperatures and frequencies small compared to the Kondo scale Γ. (Γ is a function of the parameters which enter the Hamiltonian.) However, it is important to note that the proper identification of various scaling operators with measurable physical observables is dependent on the particular realization. Thus, for the two-channel Kondo *problem* itself, the variable $\vec{\tau}$ is a spin and only s-wave scattering of the electrons is considered. Then, since there are no vertex corrections for s-wave scattering, the dc conductivity σ_{dc} can be computed from the electron self-energy, and it is found [88] to be inversely proportional to c and directly proportional to $\sqrt{T}$, reflecting the *non-Fermi liquid* $\Sigma \propto \sqrt{\omega}$ behavior of the electron self-energy Σ. By contrast, in our model (which has the same *fixed point* but not the same *bare* Hamiltonian) the fact that $\vec{\tau}$ is an orbital variable implies that the scattering is *not* simply s-wave, and that the conductivity is *not* simply related to the electron self-energy. In fact, as discussed below, we calculate the conductivity *exactly* from the Kubo formula and find that σ_{dc} is proportional to c and inversely proportional to T at zero frequency. [86]

From our solution of the one dimensional array we have found that there is a crossover concentration of dipolar modes, c_0, such that for $c < c_0 < 1$, there exists a regime of temperatures $T < \Gamma$ in which the properties of the array are well approximated by properties

of an isolated dipolar mode. Coherent behavior, involving the coordinated dynamics of many dipolar modes, sets in below a temperature, $T_{coh} < \Gamma$, where T_{coh} vanishes as either Γ or c tend to 0. (c_0 is also a function of Γ and $c_0 \to 0$ as $\Gamma \to 0$.) For $c > c_0$, $T_{coh} > \Gamma$, so there is no range of temperatures where the universal properties of the scattering from a single dipolar mode are observable. Specifically, for the one-dimensional array we found that $c_0 \sim \Gamma/W$, where W is the bandwidth. We believe, but have not proven, that the same qualiative physics applies in higher dimensions, although the dependence of c_0 on Γ/W is likely to be dimension dependent. An approximate expression for c_0 may be obtained in arbitrary dimension by using a generalization of the Doniach criterion [89]; $c_0 \sim (\Gamma/W)^{1/d}$. This expression agrees with the exact result in one dimension and implies that $c_0 \to 1$ as $d \to \infty$, consistent with other results. [90]

B. Dilute Dipolar Modes and the Normal State: $T_{coh} < T < \Gamma$

Wherever condensation into a charge-modulated structure is suppressed by quantum fluctuations, the dipolar modes will be rather dilute; we imagine this is the case in the high temperature superconductors. Thus, to understand the properties of the normal state of these materials we study the problem of dilute dipolar modes for $T_{coh} < T < \Gamma$, where the properties of the system are governed by the two-channel Kondo fixed point. One of the notable features of this fixed point is a resonance between the dipolar mode and a pair of mobile electrons, [86] characterized by an enhancement of correlations involving a non-standard "composite" pairing operator. The latter involves a pseudospin operator as well as the creation operators of a pair of mobile electrons. An alternative, but equivalent representation of this resonance is odd-frequency pairing [86,91]. The composite pairing correlations are significant up to temperatures of order Γ.

The imaginary part of the pseudospin susceptibility for $h = 0$ has the form [92,86,93]

$$\chi''(\omega, T) = \frac{1}{2}\, tanh(\omega/kT)\, \frac{\Gamma}{\omega^2 + \Gamma^2}. \tag{17}$$

It has previously been pointed out by Cox [94] that, for $\omega \ll \Gamma$, there is no external energy scale governing the frequency and temperature dependence of χ'', and that this behavior is similar to that of the normal state of high temperature superconductors. [95] While the pseudomagnetic field h is a relevant perturbation, it is only weakly so and has little effect so long as $T > T_h$ where $T_h \sim h^2/\Gamma$. Bearing in mind that in our realization of the model the local dipolar modes couple directly to an external electric field and also have internal antiferromagnetic structure, we have found that the frequency and temperature dependence of χ'' is impressed on many physical properties of the system. Two important examples are the optical conductivity and the spin susceptibility.

The optical conductivity [85] has the form $\sigma(\omega, T) = \sigma_0(\omega, T) + \delta\sigma(\omega, T)$, where σ_0 is the conductivity in the absence of the dipolar modes and $\delta\sigma(\omega, T)$ contains two anomalous contributions from the impurity scattering;

$$\delta\sigma(\omega, T) = \sigma_{bound}(\omega, T) + \sigma_{mobile}(\omega, T). \tag{18}$$

Here $\sigma_{bound}(\omega, T)$ comes from the direct coupling of the external electromagnetic field to the local dipolar mode, and is thus analagous to a bound-charge absorption:

$$\sigma_{bound} \propto c\ (\omega/\Gamma^2)\ \chi''(\omega), \tag{19}$$

which vanishes as $\omega \to 0$ and is peaked at $\omega \sim \Gamma$. The second contribution $\sigma_{mobile}(\omega, T)$ comes from the mixing between the mobile holes and the Kondo pseudospin:

$$\sigma_{mobile}(\omega, T) \propto c\ \frac{\chi''(\omega, T)}{\omega}. \tag{20}$$

and has the property that for $T \gg \omega$, $\sigma_{mobile} \propto c/T$, while for $T \ll \omega$, $\sigma_{mobile} \propto c/\omega$, as required to explain the T-linear and ω-linear resistivity of high temperature superconductors. The fact that σ_{mobile} is proportional to c, rather than inversely proportional to c as in the usual case of impurity scattering, identifies it as a paraconductivity, associated with the composite pairing fluctuations. As discussed in the companion paper [13], this goes a long way to explaining one of the most mysterious features of the observed T-linear resistivity

($\rho = 1/\sigma$) in high temperature superconductors, the insensitivity to other scattering processes. For instance, scattering from optical phonons should become significant above room temperature, yet for $La_{2-x}Sr_xCuO_{4-\delta}$ with $x = 0.15$, the T-linear behavior persists [12] to at least $T = 1000$K, with no change in slope.

For the CuO_2 planes of high temperature superconductors, the dynamical spin structure factor $S_{AF}(\vec{q},\omega)$ (where $\vec{q}$ is the departure from the antiferromagnetic wavevector (π,π)) determines the relaxation rate $^{63}T_1^{-1}$ of Cu spins (as measured by an NMR experiment [96]) and the intensity of spin-flip neutron scattering. It is natural to suppose that the dominant contribution to $S_{AF}(\vec{q},\omega)$ for $YBa_2Cu_3O_{7-\delta}$ comes from the locally-antiferromagnetic hole-deficient regions, since (π,π) is not a special nesting vector of the Fermi surface. [97] Then, as a function of space and time, S_{AF} may be written as a product of two structure factors, one representing an antiferromagnetic hole poor region and the other the dynamics of the collective mode. Equivalently, the Fourier transform of S_{AF} is a convolution of the two factors. In this way the charge dynamics of frustrated phase separation is impressed on the spin dynamics, and experiments which are sensitive to the spin excitations with wavevector near (π,π) give important information concerning the internal structure of the collective mode.

To obtain an explicit expression for $S_{AF}(\vec{q},\omega)$, we imagine that the hole-free regions are a few lattice spacings in size, so it is sufficient to use the single-mode approximation to evaluate their contribution to the structure factor, *i.e.* to retain only the lowest spin excitation, the triplet with wave vector (π,π). This approximation has been shown to be very good by numerical calculations on small clusters. [98] In this way we obtain:

$$S_{AF}(\vec{q},\omega) = F(\vec{q}) \sum_{\sigma} \int_{-\infty}^{\infty} d\omega' S_{\sigma}(\omega')\ S_I(\omega-\omega'), \tag{21}$$

where

$$\sum_{\sigma} S_{\sigma}(\omega') = 3\ A(\beta\omega_g)\ \frac{\delta(\omega-\omega_g)-\delta(\omega+\omega_g)}{1+e^{-\beta\omega}} \tag{22}$$

is the dynamical spin structure factor for the interior of a region, with

$$A(x) = \frac{e^x + 1}{e^x + 3}. \tag{23}$$

Here we have assumed that the spin excitations within a hole-free region are infinitely long-lived, although of course in reality they will be broadened by the interaction with their surroundings. The contribution from charge fluctuations is given by:

$$S_I(\omega) = \frac{1}{2(1 - e^{-\beta\omega})} \frac{\Gamma}{\omega^2 + \Gamma^2}, \tag{24}$$

which is equivalent to Eq. (17). (Note that, for some models of fluctuating phase separation, a $\delta(\omega)$ contribution is added to the Lorentzian factor in this equation.) Finally, $F(\vec{q})$ is the Fourier transform of the equal time spin-spin correlation function, within the hole-free regions. From these equations it follows that: [101]

$$S_{AF}(\vec{q}, \omega) = \frac{F(\vec{q})}{4} \tilde{S}(\omega, T) \tag{25}$$

where

$$\tilde{S}(\omega, T) = \frac{3A(\beta\omega_g)}{\Gamma} \left(\frac{tanh\frac{1}{2}\beta(\omega - \omega_g) + tanh\frac{1}{2}\beta(\omega + \omega_g)}{1 - e^{-\beta\omega}} \right) \tag{26}$$

It can be seen that the effect of the charge dynamics is to move spectral weight to low frequencies. The NMR relaxation rate $^{63}T_1$, which is sensitive to spin fluctuations at (π, π), is obtained by setting $\omega = 0$ and integrating $S_{AF}(q, \omega)$ over q, to find:

$$^{63}T_1 = B(4 + e^{\beta\omega_g} + 3e^{-\beta\omega_g}) \tag{27}$$

Equations (25) - (27) were given in reference [18].

Striking confirmation of the predicted form of $S_{AF}(\vec{q}, \omega)$ in Eq. (25) has been obtained in recent neutron scattering experiments by Sternlieb *et al.*. [15] They found that a separable form $\tilde{S}(\omega)F(\vec{q})$ of the dynamical structure factor $S_{AF}(\vec{q}, \omega)$ for $YBa_2Cu_3O_{6.6}$, with $T_c = 46K$, described the data for all energies between $2meV \leq \hbar\omega \leq 40meV$ and that the "shape" function $F(\vec{q})$ is the same in both the superconducting and normal states, *i.e.* $F(\vec{q})$ is independent of temperature. The Fourier transform of $F(\vec{q})$ gives a characteristic size of $4a$,

where a is the Cu-Cu distance. In our picture, this is a measure of the radius of the hole-free regions. The frequency dependence of the scattering is well-described by Eq. (26), with a sample-dependent broadening of the spin excitations within a hole-free region, showing that the regions are indeed fluctuating.

Further evidence for the fluctuation of the domains comes from NMR, which measures the spectral weight transferred to zero frequency. Mehring and coworkers have analyzed NMR experiments on a variety of high temperature superconductors and concluded that Eq. (27) gives an excellent fit to the data. [99]

Finally, it is important to have some estimate of the magnitude of the Kondo scale, Γ. Since Γ characterizes the strength of the interaction between the mobile holes and the local dipolar modes, we expect that $\Gamma \sim J$. This is consistent with the observation that the bound-state contribution to the optical absorption in the high temperature superconductors is typically found to peak at $\omega \sim 0.1$eV. [100] (σ_{bound} in Eq. (19) is peaked at $\omega = \Gamma$.)

C. Coherence Effects and the Superconducting State: $T < T_{coh}$

In the one dimensional model we have studied, there cannot be long-range order but we have found that the susceptibilities corresponding to three different order parameters diverge like T^{-1} below T_{coh}: 1) Composite odd-parity spin-singlet η pairing, 2) Ordinary, even parity, spin-singlet pairing, and 3) Transverse pseudospin ordering. Higher dimensional couplings, *e.g.* weak coupling to other one-dimensional chains, favors ordering of 2) and 3). As a result, we find that, despite the fact that the normal state properties are dominated by composite superconducting fluctuations, the ordered state into which the system ultimately condenses is likely to be either an ordinary s-wave superconducting state or a charge-modulated state, in which the local dipolar modes order. In either case the composite pairing resonance of the isolated dipolar mode should affect the properties of the condensed state at frequencies greater than T_{coh}, even if the ordering itself does not.

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VI. FIGURE CAPTIONS

Figure 1: Phase diagram of ^{3}He - ^{4}He mixtures. x is the concentration of ^{3}He and T is the temperature. The solid lines show the ^{4}He lambda line marking the transition from the normal liquid to the superfluid state, and the edges of the two-phase region. The point marked "A" is the ^{3}He superfluid phase; the transition temperature, $T_c \approx 2$mK, is too low to be visible in the figure. The line segment marked "B" is where the possibility of a superfluid state at the border of phase separation occurs; no transition has been detected down to the lowest temperatures obtainable in the laboratory.

Figure 2: Phase separation in the $t-J$ model at $T = 0$. The solid line segments mark the boundary of the two-phase region obtained from the analytic results of Ref. [9]. The stars are the inferred extension of these results from the finite size numerical diagonalization studies of the same authors. The dashed curve marks the phase boundary obtained in Ref. [47] by extrapolating the high temperature series results to $T = 0$. The region marked "s-wave" has been shown analytically to be superconducting in Ref. [9]. The region marked "A" is where numerical studies of Ref. [52] reveal substantial d-wave, as well as s-wave superconducting correlations.

Figure 3: Phase diagram in the small t limit and for $T = 0$ of the $t-J-V$ model defined in Eq. (1), reproduced (with minor modifications) from Ref. [10]. Here, x denotes the hole concentration. The region marked "two-phase" is the region of two-phase coexistence; the regions marked "2e" and "4e" are superconducting phases with charge 2e and charge 4e order parameters respectively. The phase marked "FL" is a Fermi liquid, at least down to very low temperatures. There are unlabelled line phases corresponding to hole-crystals with various lattice structures.

Figure 4: Schematic phase diagram for a model cuprate perovskite. The variables are temperature (T) and hole concentration (x). The dashed lines give the phase boundaries for neutral holes. The solid lines indicate phase boundaries for charged holes and a quenched uniform distribution of dopants. The regions enclosed by solid lines and marked AF, SG,

and SC have antiferromagnetic, spin glass, or superconducting order respectively.

Figure 5: Phase diagram of $La_2CuO_{4+\delta}$from Ref. [61]. More recent work has indicated that the high temperature boundary of the 2-phase region of the phase diagram may be more complicated than shown.

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[101] We take the opportunity to note that, due to a typographical error, two pairs of parentheses were omitted from Eq. (4) of reference [18], which is properly identicle to Eqs. (25) and (26) of the present paper.

Fig 1

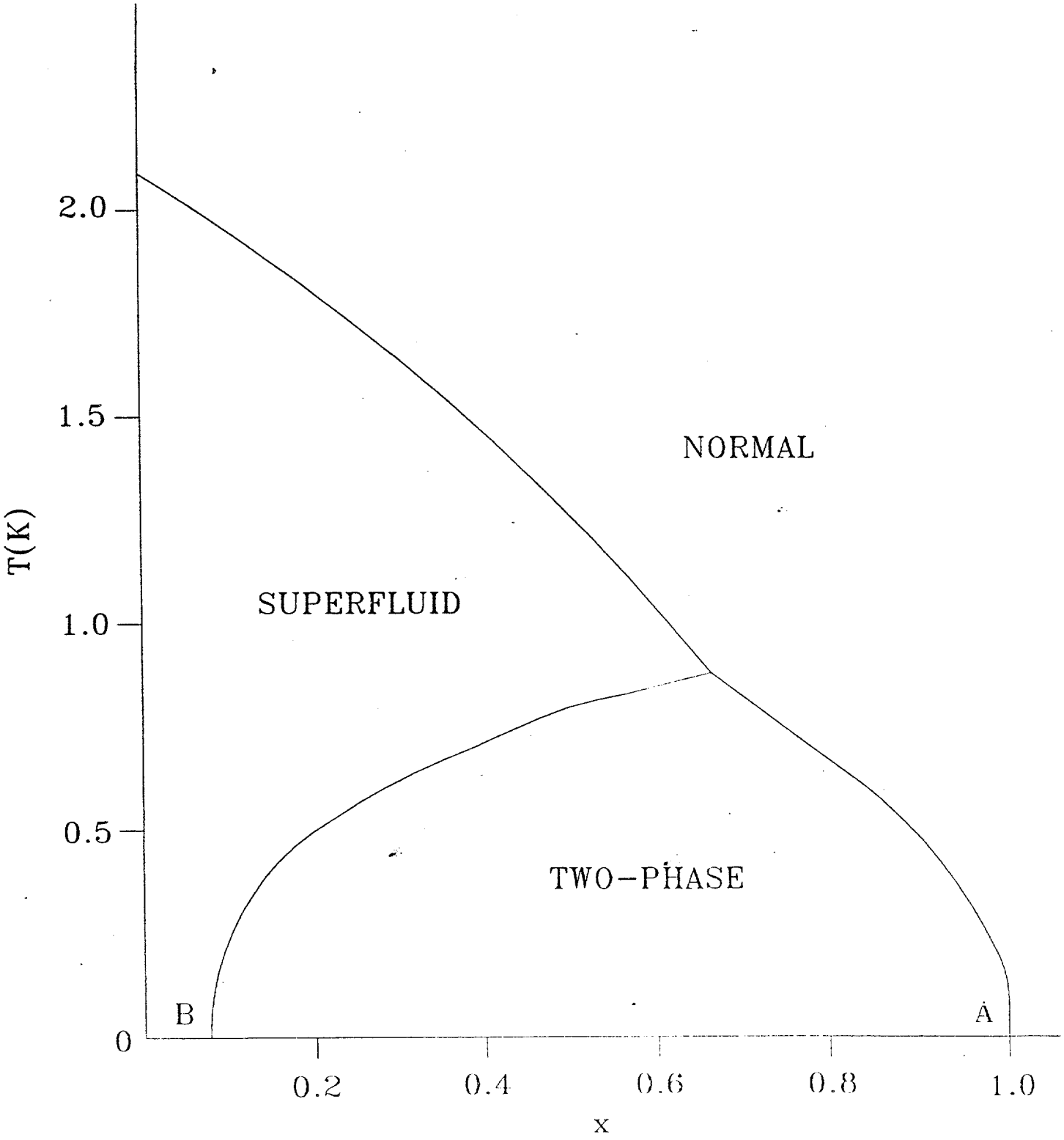


Fig 2

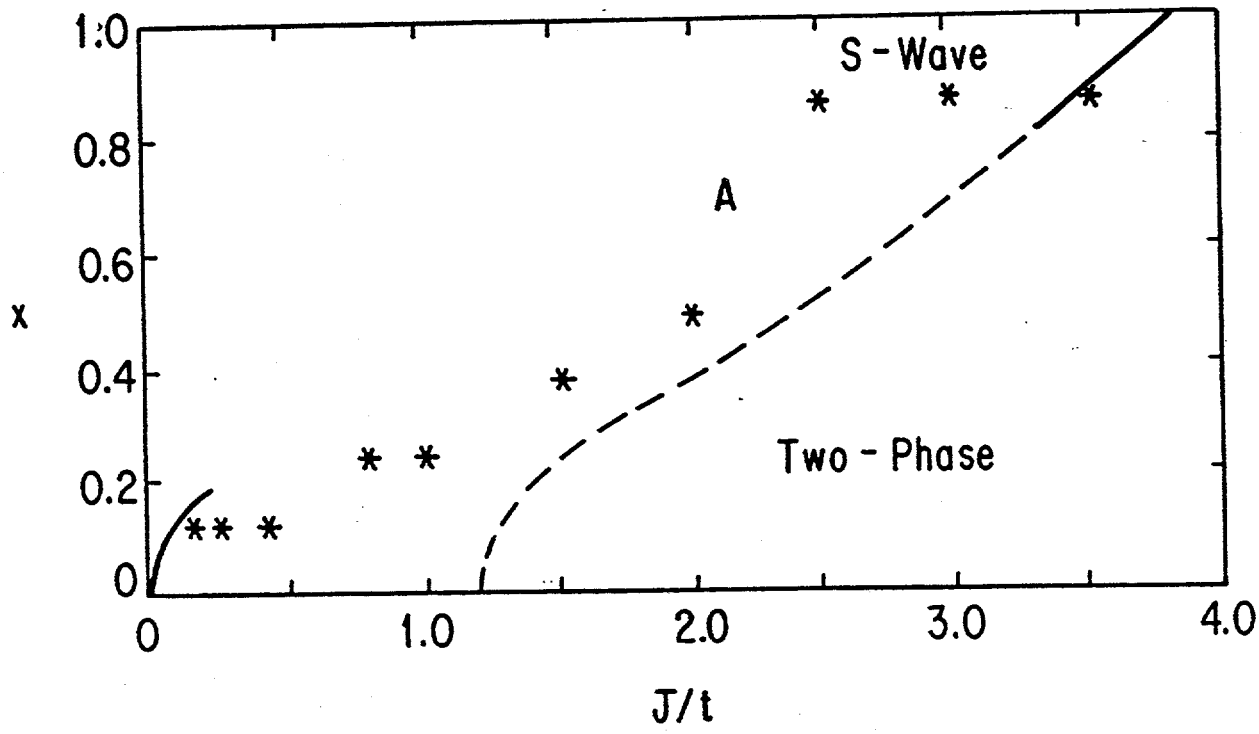


Fig 3

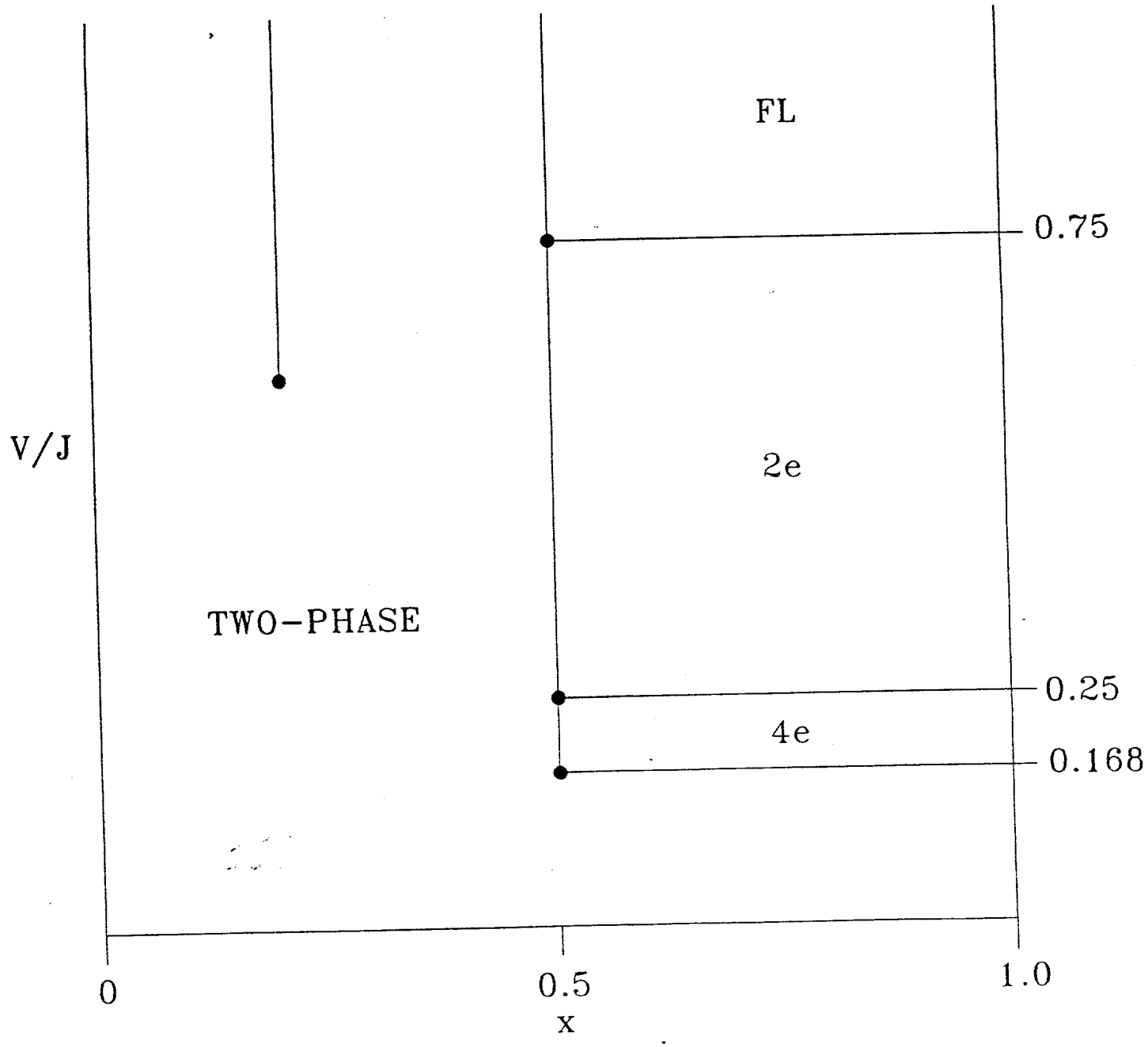


Fig 4

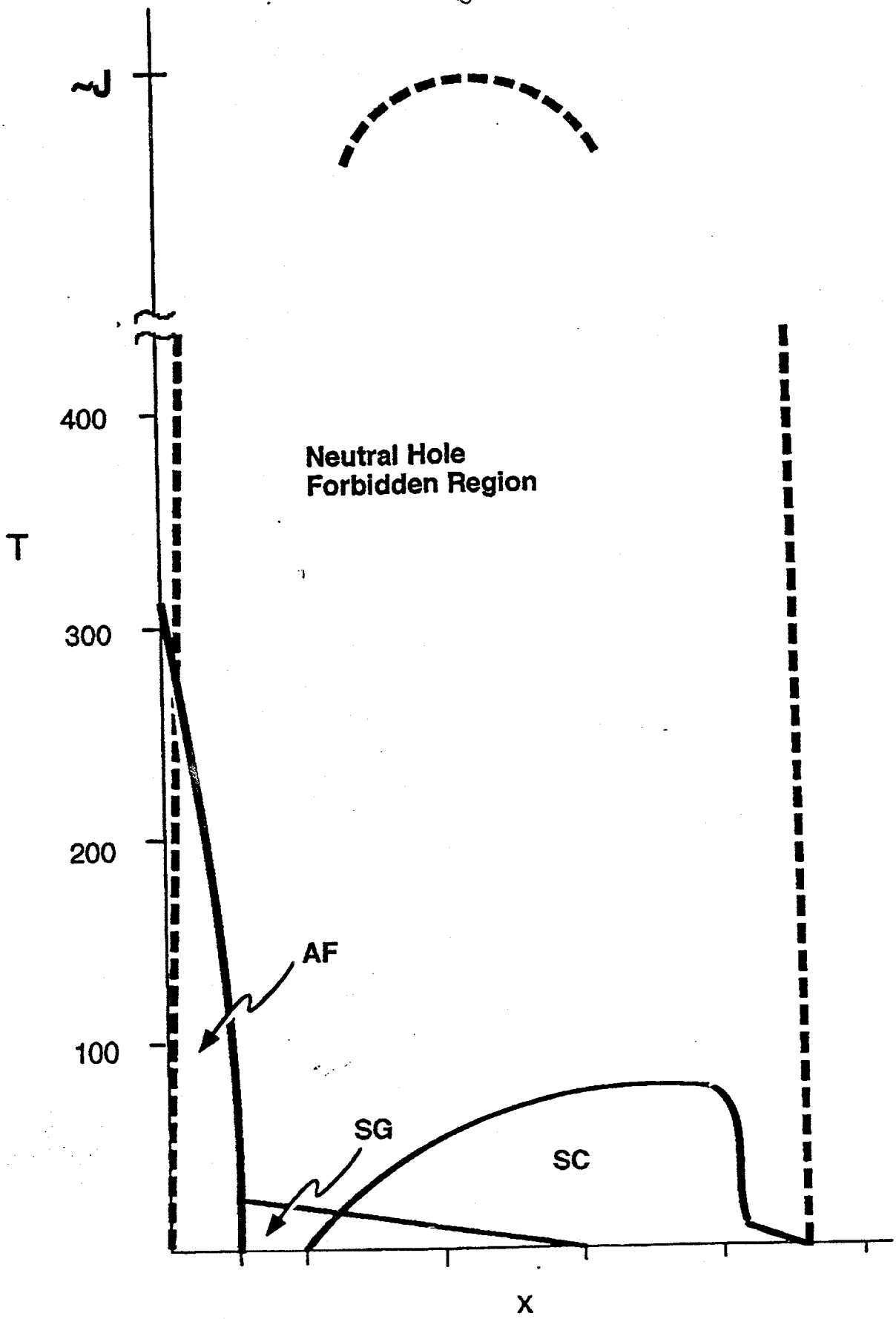
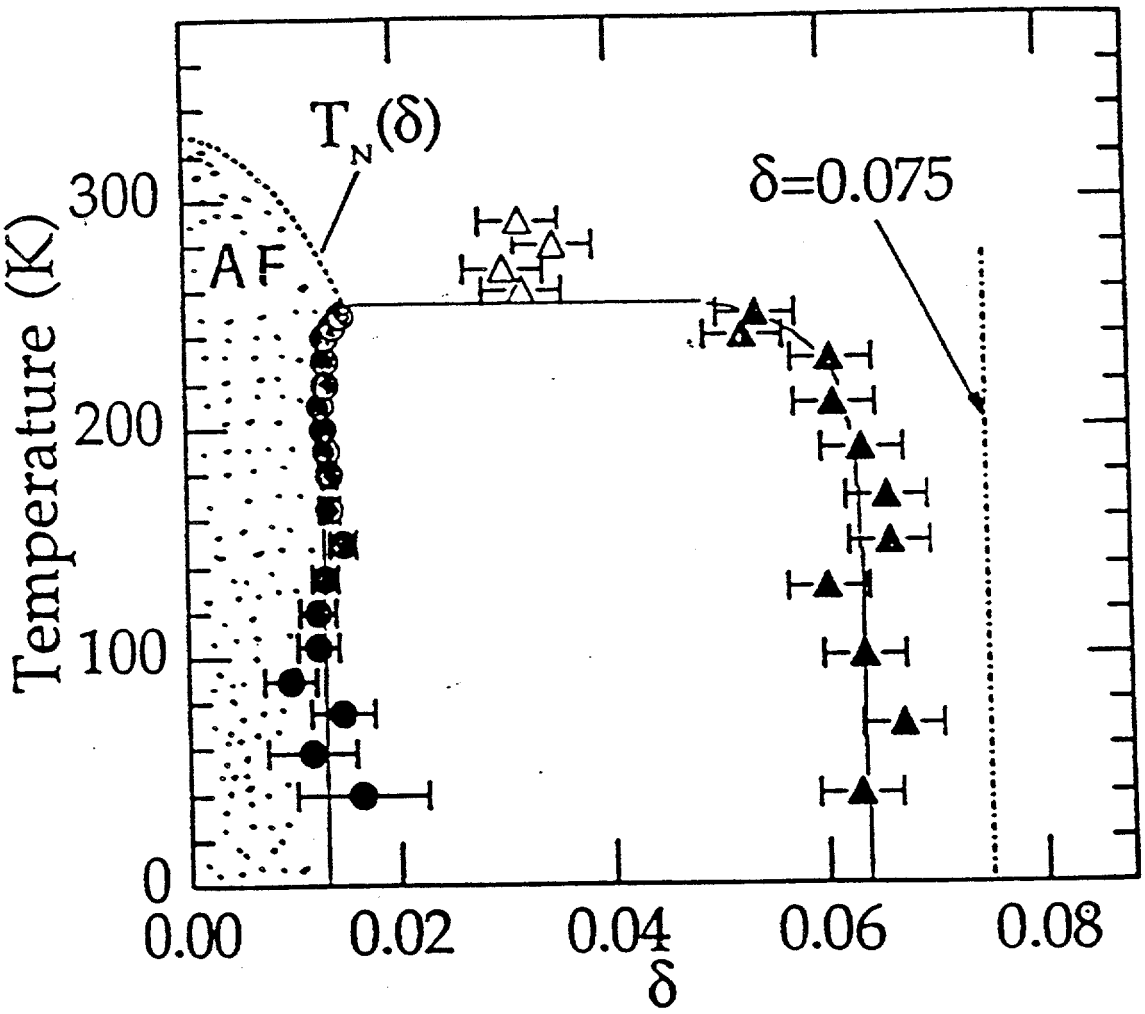


Fig 5


$T_N(\delta)$
δ=0.075
AF
Temperature (K)
300
200
100
0
0.00
0.02
0.04
0.06
0.08
δ