Conf 9205231--1

BNL--47796

DE92 019249

Talk Presented at the Workshop on
"Phase Separation in Cuprate Superconductors"
Erice, Sicily May 6 - 12, 1992

Frustrated Phase Separation and High Temperature Superconductivity

V.J. Emery
Dept. of Physics Brookhaven National Laboratory, Upton, NY 11973

S. A. Kivelson
Dept. of Physics UCLA, Los Angeles, CA 90024

[illegible]
AUG 1 [illegible] 1992

ABSTRACT

A dilute system of neutral holes in an antiferromagnet separates into a hole-rich and a hole-poor phase. The phase separation is frustrated by long-range Coulomb interactions but, provided the dielectric constant is sufficiently large, there remain large-amplitude low-energy fluctuations in the hole density at intermediate length scales. The extensive experimental evidence showing that this behavior gives a reasonable picture of high temperature superconductors is surveyed. Further, it is shown that the scattering of mobile holes from the local density fluctuations may account for the anomalous normal-state properties of high temperature superconductors and also provide the mechanism of pairing.

1. Introduction

On the experimental level, high temperature superconductivity[1] is a robust phenomenon, occurring in a wide variety of materials containing CuO_2 planes. However, despite various analytical arguments suggesting that there is an attractive interaction between charge carriers, numerical experiments[2] on Hubbard, extended Hubbard, $t-J$, and related models have so far failed to produce any indication of a significantly enhanced pairing susceptibility, although the models are believed to incorporate, at least crudely, the most important physical interactions.[3] It is conceivable that more extensive numerical experiments may still succeed in finding evidence for superconductivity; nevertheless it is striking that such a gross property has failed to show up so far, and it seems likely that something is missing. We shall argue that the long-range Coulomb interaction is the essential piece of physics that has been ignored.

The central feature of high temperature superconductors is that they are doped insulators,[3] obtained by chemically adding charge carriers to a highly-correlated antiferromagnetic insulating state. We have argued previously,[4,5] on the basis of analytical and numerical studies of the two dimensional $t-J$ model that a low concentration of holes in an antiferromagnet is unstable to phase separation into a hole-rich and a hole-deficient phase. The $t-J$ model describes neutral holes (no long-range Coulomb

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED interaction) with hopping amplitude t, and local moments with exchange interaction J. Phase separation certainly can be thought of as a strong attractive interaction between holes, although in a real sense the mechanism is more properly regarded as the ejection of holes from the antiferromagnet. The characteristic energy scale of this interaction is set by magnetic energies, so one expects to see phase separation only below temperatures of order the exchange energy J. In the presence of the long-range Coulomb repulsion between holes, the system cannot macroscopically phase separate unless the counterions are mobile. Thus, the system is frustrated. However, so long as the Coulomb interactions are not too strong, i.e. if the background dielectric constant is large enough, the local tendency toward phase separation will still have important consequences. Specifically, there will be large local density fluctuations so that a snapshot of the system at moderate doping will show metallic regions, in which the hole density is greater than average, and antiferromagnetic droplets, in which it is smaller than average. This structure may be frozen, as in a pinned charge density wave state, or fluctuating with very slow dynamics due to the metastability implied by the frustration.

In this paper, we propose to see what we can learn about the the properties of high temperature superconductors by looking at the materials from this new perspective. In particular, it will be shown that there is considerable evidence of a purely electronic tendency towards phase separation of the holes, and that the consequent large-amplitude local density fluctuations may provide an understanding of low-energy long-wavelength phenomena, such as the anomalous normal-state properties[6,7] and high temperature superconductivity itself. Indeed, it is widely recognized that the normal-state properties of high temperature superconductors imply the existence of a class of low-frequency collective excitations which strongly scatter the conduction electrons. Here we have, ready made, a rather obvious candidate. The presence of low-frequency collective modes typically indicates a nearby phase in which these fluctuations condense into a new ordered state. The only known ordered state which overlaps the metallic phases of the cuprate superconductors is the so-called spin-glass phase, in which there are frozen local magnetic moments but no long-range magnetic order.[8,9] We suggest that this phase should be regarded as a "cluster spin-glass", in which there is substantial local charge inhomogeneity, and the spins in the hole-deficient regions are locally Neel ordered, but with a random direction of the staggered magnetization. Thus the slow density fluctuations in the metal are related to the existence of the nearby cluster spin-glass phase.

Frustrated phase separation may also give rise to superconductivity, possibly with a high transition temperature. Indeed there are known examples of such behavior. One is the $t-J-V$ model in the weak hopping limit. Here, V is a repulsive interaction between holes on neighboring lattice sites. It has been shown[5,10] that a finite value of V frustrates the phase separation that occurs for $V = 0$ and that, for $0.168 < V/J < 0.75$, the system becomes a superconducting square liquid or dimer liquid. A second example, which is more physical but comes from another area of physics is provided by atomic nuclei, which have a gap in their energy spectrum analogous to that of a BCS superconductor.[11] Nuclei are droplets formed because the Coulomb interaction between protons frustrates the condensation of nuclear matter into a liquid state. The energy gap is quite small in nuclear matter[12] but relatively large in a finite nucleus. The reason is that pairing takes place in the low-density surface region of a nucleus, and the energy gap increases as the density decreases.[12] Thus the first effect of the nuclear force is condensation into a liquid state (phase separation), and the secondary effect is low temperature superconductivity. However the long-range Coulomb interaction frustrates the phase separation and leads to droplets (nuclei), which are high T_c superconductors. Part of this story is relevant for the cuprates, but, in that case, high temperature superconductivity is more subtle because, unlike nucleons, electrons have a short-range interaction which is repulsive.

This paper is organized as follows: In Sec. 2, we summarize the theoretical arguments leading to the conclusion that dilute holes in an antiferromagnet are unstable to phase separation. These arguments are discussed in greater detail in our previously published work on the subject.[4,5] In Sec. 3, we summarize arguments showing that collective modes with slow dynamics are a consequence of frustrated phase separation. We also introduce an effective model from which low-energy properties may be calculated. An important feature of this model is that the behavior is governed by a fixed point, which is consistent with the robustness and universality of the behavior of high temperature superconductors. In Sec. 4 we review experiments that demonstrate the existence of frustrated phase separation in the high temperature superconductors. First, we emphasize the fact, discussed extensively at this conference, that phase separation on laboratory time scales occurs in oxygen-doped La_2CuO_4 and in photo-doped materials. In both cases, the negative counter charges are mobile enough to allow phase separation to take place. We then consider the more usual situation in which the dopants are not mobile, and describe some of the experimental evidence to support the notion that the doped state is inhomogeneous at intermediate distances and times. We argue that the fact that it is difficult to produce electronically homogeneous materials is a reflection of the tendency of the system to phase separate locally in response to a fluctuation in the local potential. Whereas a normal metal would screen small local variations in the concentration of dopant ions, the doped antiferromagnet tends to overscreen, thus amplifying the effect of any small inhomogeneity. We also show that large-amplitude density fluctuations provide a natural explanation for a number of experimental anomalies in these materials and outline our understanding of the implications for the mechanism of high temperature superconductivity. Finally, in Sec. 5 summarize our results. Brief accounts of some of our ideas have previously been given in conference reports.[13] More detailed and extended accounts will be presented in papers currently under preparation.[14]

2. Neutral Holes in an Antiferromagnet

We have previously reported[4,5] on the results of extensive analytical and numerical studies of the $t-J$ model, which led us to suggest that phase separation always occurs at zero temperature, provided the hole concentrations x is less than a critical value $x_c(J/t)$. 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 two holes on nearest-neighbor sites break one less antiferromagnetic bond than two far-separated holes. At larger distances, two holes attract each other through the exchange of magnon pairs. In essence, phase separation is the best way to minimize the zero-point kinetic energy of the electrons and to reconcile the mobility of doped holes with the maintenance of local antiferromagnetic order (which optimizes the zero-point energy for the half-filled band).

A wide variety of models and materials exhibit phase separation, although the reasons may have nothing to do with antiferromagnetism. For instance, it has been shown in various three-band models of the copper oxide planes that, near to the charge-transfer instability, phase separation occurs on doping.[15] It also is possible that non-electronic effects, such as chemical interactions between the oxygen dopant atoms, could cause phase-separation in some range of doping. While these other interactions might augment the tendency toward phase separation in high temperature superconductors, we focus on the role of the antiferromagnetism because it is a prominent feature of the doped and undoped systems. Moreover, as we shall see, experiments indicate that one of the phases is essentially hole-free, suggesting a magnetic mechanism, rather than a charge-transfer instability, which would lead to hole-rich and hole-poor phases.[15]

As outlined above, studies of various models in various limits have *established* that phase separation is a common behavior of dilute holes in an antiferromagnet. We *believe*, but have not yet proven, that it is generic, essentially model-independent, behavior. However, if it were shown that a given model of holes in an antiferromagnet did not exhibit phase separation for a physically reasonable choice of parameters, we would turn to the experimental evidence (discussed below) that there is a strong local tendency toward phase separation in the high temperature superconductors, and conclude that that there is an important piece of physics missing from that model.

3. Implications of Frustrated Phase Separation

It is easy to demonstrate that a model with a short-range tendency for phase separation together with long-range Coulomb interactions is highly frustrated. In a coarse-grained sense, we can represent the hole-rich, hole-poor, and average-density phases as different orientations of a local "block spin". The local tendency toward phase separation is modelled as a short-range ferromagnetic interaction between spins while, the long-range Coulomb interaction corresponds to a long-range antiferromagnetic interaction. The latter is known to be highly frustrating; at the classical level it produces Devil's staircases[16] and a large degree of metastability. At the quantum level, the frustration is reflected in the existence of spatially-localized, large-amplitude density fluctuations, or collective modes, in which local regions of the hole-rich and hole-poor phases nucleate and disappear.[14] The large amplitude of the distortion implies that the collective modes are very "heavy" in the sense that they have little dispersion; even a modest disorder will localize them. Once again there is a large degree of metastability and, associated with it, extremely slow "glass-like" dynamics,[14] which is manifested via the spin fluctuations as spin freezing. The mobile holes create, annihilate and scatter from the collective modes. These processes, together with the distribution of energies of the collective modes determine the low-energy properties of the system. At present, it is not possible to give a fully deductive theory of all of these phenomena and their consequences, starting from e.g. an extended Hubbard model with long-range Coulomb interactions. Therefore we proceed in stages, first constructing a lower-level model from which the low-energy physics may be calculated. Such a theory will be described in detail in a future publication:[14] for the present, we describe the essential ideas.

Our approach is to calculate directly the consequences of the interactions between the mobile holes and the collective modes, which dominate the low-energy behavior of the system and determine the temperature-dependence of physical quantities. Because the collective modes correspond to local phase separation, they have significant internal structure; in particular, where the hole density is low, we expect behavior characteristic of the antiferromagnetic insulating state. For the internal degrees of freedom, we shall use the single-mode approximation, in which we ignore all internal excited states save one. This approximation is valid both for small clusters[17] and in the thermodynamic limit.[18] The single internal mode is a spin-1 excitation with momentum (π,π) and energy ω_g. An NMR experiment, or a neutron scattering experiment which measures the q-integrated intensity of the (π,π) peak, does not probe the spatial structure of the collective modes. Consequently, for many purposes, we may regard them as point-like objects and introduce operators $b_0^\dagger(\vec{r})$ and $b_{1,\sigma}^\dagger(\vec{r})$, which create a collective mode at position $\vec{r}$ in the spin-0 ground-state or in the spin-1, $S_z = \sigma = (-1, 0, +1)$ excited state respectively. These operators are well-defined in the adiabatic limit, where the collective-mode fluctuations are either frozen (due to the disorder) or slow compared to the equilibration times of the microscopic modes. Since phase separation is driven by the tendency of the antiferromagnetic ground-state to exclude holes, it follows that the existence of large-amplitude density fluctuations requires that the system be reasonably close to the adiabatic limit. By the same token, the maximum energy scale (ultraviolet cutoff) of the fluctuations must be of the order of the exchange integral, J. In general, we expect this approximation to be particularly good in low- or moderately-doped materials whereas, in sufficiently heavily-doped materials, corrections to the adiabatic approximation may be more significant. (A more complete discussion will be presented in a forthcoming publication[14]).

In order to calculate correlation functions, we note that the collective modes are local in space and that there cannot be two at the same point. Moreover, the fact the collective modes correspond to a local charge inhomogeneity implies that they have a dipolar character and that there is a local flow of current as they are created. Using these properties, we have shown[14] that the scattering of the mobile holes from the collective modes is equivalent to a two-channel Kondo problem. It has already been noted by Cox[19] that the behavior of the latter has much in common with the phenomenology of the normal state of high-temperature superconductors.[6,7,20] When the excitation energies of the collective modes may be neglected, the imaginary part of the susceptibility for $b_0^\dagger(\vec{r})$ has the form[21,22]:

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

where Γ is the Kondo energy scale.

We have found that this structure is impressed on many of the observed properties of high-temperature superconductors, in particular the optical conductivity[22] and spin fluctuations. As a specific example, we shall consider the dynamical structure factor $S(\omega, T)$ of the operator $b_{1,\sigma}^\dagger(\vec{r}) b_0(\vec{r})$. For a material such as $YBa_2Cu_3O_{7-\delta}$ this function determines the relaxation rate T_1 of Cu spins measured by an NMR experiment,[23] and the q-integrated intensity of the (π, π) peak in neutron scattering. The point is that (π, π) is not a special nesting vector of the Fermi surface[24] in $YBa_2Cu_3O_{7-\delta}$ and thus the amplitude for spin excitations at this wave vector is significant only within the hole-poor part of a collective mode. We have shown[14] that, when ω is small compared to Γ, $S(\omega, T)$ is given by

$$S(\omega, T) = A(\frac{\omega_g}{kT})[e^{\frac{\omega_g}{kT}} F(\omega - \omega_g, T) + F(\omega + \omega_g, T)] \tag{2}$$

where

$$F(\omega, T) = \frac{\pi}{2} \delta(\omega) + \frac{1}{\Gamma(1 + e^{-\frac{\omega}{kT}})} \tag{3}$$

and

$$A(x) = \frac{const.}{e^x + 3} \tag{4}$$

If the excitation energy of the collective modes had been included it would have broadened the δ-function in Eq. (3). A comparison of these results with NMR and neutron scattering experiments will be made in Sec. 4.

An additional feature of the fixed point of the two-channel Kondo problem is a resonance between the "impurity" and a pair of conduction electrons,[22,25] which may be responsible for high-temperature superconductivity. It is important to note that this is a fixed point theory and hence the conclusions are insensitive to the detailed structure of the materials. The dipolar nature of the collective modes is essential, otherwise we would have obtained a single-channel Kondo problem which does not have the same low-energy behavior as the normal state of high temperature superconductors.

The spatial structure of a collective mode may be probed by a q-resolved neutron scattering experiment. The major effect is a cutoff in the q-width of the (π, π) peak when the correlation length reaches the size of the cluster. The latter may be quite large for lightly-doped materials but is a few lattice spacings for superconducting materials, close to optimum doping, as we shall see later.

4. Experimental Evidence for Frustrated Phase Separation

The existence of phase separation in model calculations suggests an examination of experiments that might show if it is a feature of real materials. There are two quite different situations. If the dopant atoms are absolutely frozen, then clearly phase separation can occur only as a short-distance, fluctuation effect. This true of the majority of the cuprate perovskites. On the other hand, if the charge donors are mobile on laboratory time scales, they can be dragged along by the holes and compensate the long-range part of the Coulomb interactions; in this case, the holes can actually phase separate. This occurs in two special cases: In photo-doped materials and in oxygen-doped $La_2CuO_{4-\delta}$ where the oxygen ions remain fairly mobile to low temperatures. The evidence for true phase separation is discussed in other lectures at this workshop.[26]

Of course, there is an issue as to whether the observed phase separation is driven by the physics of the CuO_2 planes, or is induced by extraneous factors such as the oxygen chemistry. However we feel that the fact that phase separation occurs in *both* of the two known cases where the dopants are mobile provides strong experimental support for an electronically driven tendency for phase separation whenever the long-range Coulomb interactions do not prohibit it.

We shall now go on to consider the evidence for frustrated phase separation in materials in which the countercharges are immobile.

4.1 Lightly Hole-Doped Antiferromagnets

When the antiferromagnetic insulating "parent" materials of the high temperature superconductors are lightly doped with immobile charge-donors such as Sr, the antiferromagnetic long-range order is rapidly destroyed (at about 2% in Sr doped La_2CuO_4), although superconductivity requires somewhat larger dopant concentrations (of order 5% in Sr doped La_2CuO_4). We imagine that, in this entire range, if the dopant atoms were mobile, the system would phase separate below a characteristic temperature which is some fraction of the antiferromagnetic exchange energy J. Consequently, chemical or other kinds of inhomogeneity would be enhanced rather than screened, and would have an unusual effect on the electronic properties. Indeed, worries about sample homogeneity plague all studies of lightly doped materials.

4.1.1 Neutron Scattering

Some of the most interesting recent data on the lightly-doped material comes from neutron scattering studies of $La_{2-x}Sr_xCuO_{4-\delta}$ with x between 2% and 5%, where there is neither Neel order nor superconductivity. Recent data by Keimer et al[27] show a peak in the dynamical spin structure factor, $S(\vec{q},\omega)$, at $\vec{q} = (\pi,\pi)$, with an inverse correlation length $\kappa(x,T)$ which they fitted by the simple empirical relation

$$\kappa(x,T) = \kappa_o(x) + \kappa(0,T) \qquad (5)$$

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.

There are other ways to fit this data, especially considering the large error bars.[28] Nevertheless, it is worth noting that Eq. (5) displays precisely the form we expect on the basis of frustrated phase separation and that we may deduce the size of the antiferromagnetic regions from the data: for x=2%, $\kappa_o^{-1} = 200\mathring{A}$ and, for x=4%, $\kappa_o^{-1}(x) = 40\mathring{A}$.

4.1.2 Optical Absorption

It has been found by Thomas et al[29] that the electronic contribution to the optical absorption is quite similar in lightly hole-doped and electron-doped materials. The salient features are two very broad characteristic absorption peaks in the insulating gap, one with an energy of order 500 meV, and the other with an energy of order 100 meV. They seem to be present in all the cuprate superconductors, although they may be more or less well-resolved, depending on their widths in the different materials. The transitions are certainly associated with bound charges, since in all cases the optical absorption vanishes as T and ω tend to 0. Later, when we consider more heavily-doped materials, it will be shown that there is a natural interpretation of the lower energy peak in terms of fluctuating phase separation.

4.2 Local Droplet Fluctuations in Superconducting Materials

We now examine the evidence that fluctuating local phase separation occurs in more heavily hole-doped, superconducting materials.

4.2.1 Photoemission

Thermodynamically, the *definition* of phase separation is that, in a region of the phase diagram, the chemical potential remains constant over a range of concentrations. From this perspective, the most direct evidence of phase separation, at least at short length scales, comes from photoemission studies[30] of both $La_{2-x}Sr_xCuO_{4-\delta}$ and $Nd_{2-x}Ce_xCuO_4$, which show that, as the hole concentration varies, the chemical potential remains within the insulating gap, while the density of states in the gap grows in proportion to the hole concentration. This is precisely the expected behavior for an inhomogeneous system in which insulating and metallic regions coexist in chemical equilibrium: The density of states in the gap arises from the metallic region, 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 it must lie within the insulating gap. This is quite different from the behavior of a semiconducting model or the *one-dimensional* Hubbard model.[31] Of course, photoemission is basically a high-energy, short-wavelength probe, so it is sufficient to invoke *dynamical* phase separation in order to account for these experiments.

4.2.2 Motion of the Apex Oxygens

Phase separation of the holes also has structural implications. The apical oxygens, which sit directly above or below the Cu ions, are a likely probe.[32] In the first place, it is generally true that only cuprates possessing apical oxygens can be hole-doped; the so called electron-doped materials, which do not have apical oxygens, cannot be hole doped.[33] A possible exception is the infinite-layer compound[34] $(Sr_{1-x}Ca_x)_{1-y}CuO_2$. Nevertheless, it is clear that the apical oxygens may play a significant role in compensating the charge of the holes in the CuO_2 planes. Moreover, measurements of the crystal structure as a function of hole concentration[35] in $YBa_2Cu_3O_{7-\delta}$ show that the average distance of an apical oxygen from the CuO_2 planes increases by about $0.15\AA$ as δ increases from 0 to 1. We therefore conclude that there must be a significant coupling between the *local* hole concentration and the apical oxygen distance. In effect the oxygen motion modulates the local chemical potential of the holes and compensates the local charge (i.e. it reduces the effective strength of the long-range Coulomb repulsion between holes). Thus, the polarizability of the apical oxygens enhances phase separation and, at the same time, implies slow (ionic) dynamics for large amplitude density fluctuations. However this is not the only way in which the Coulomb interaction may be compensated: the essential ingredient is a large dielectric constant, which is a common feature of oxide superconductors. We emphasize that the important role of the apex oxygens is to contribute to the dielectric constant, *not* to provide an electron-phonon interaction.

Evidence for these effects has been obtained by Egami et al[36] in neutron scattering experiments on $Tl_2Ba_2CaCu_2O_8$. They found that there are dynamical fluctuations of $0.3\AA$ in the position of the apical oxygens. This is a rather large displacement and it signifies large local differences in the local hole concentration.

4.2.3 Frozen Moments

Muon spin rotation experiments (μSR) on $La_{2-x}Sr_xCuO_{4-\delta}$ observe a so-called "spin-glass" phase, characterized by frozen magnetic moments, extending from the point at which long-range antiferromagnetic order is destroyed to higher Sr concentrations where the low temperature phase is superconducting.[8] The fraction of frozen moments and the spin freezing temperature are found to be continuously decreasing functions of the Sr concentration and to extrapolate to zero at x=15%. Similarly, in $YBa_2Cu_3O_{7-\delta}$ the fraction of frozen moments and the spin-freezing temperature extrapolate to zero for $\delta \approx 0.3$. At higher doping the hole-deficient droplets are fluctuating more rapidly than μSR time scales, which are quite long (of the order of a microsecond). No information about the droplet sizes or shapes can be extracted from the μSR data. Measurements using the Mossbauer effect give a similar picture and are reviewed by P. Imbert at this workshop.[9]

In the context of frustrated phase separation, the existence of frozen moments is quite natural. The system should be regarded as a cluster spin glass, in which the charge is inhomogeneous and the spins in the hole-poor regions locally Neel ordered, but with a random orientation of the staggered magnetization.

4.2.4 Magnetic Correlations

Nuclear magnetic resonance experiments on $YBa_2Cu_3O_{7-\delta}$ show that the relaxation rate of the nuclear spins is an order of magnitude larger on Cu than on oxygen[37]. The current interpretation is that the Cu nuclear spin is relaxed by antiferromagnetic fluctuations, which are strongly suppressed at an oxygen nucleus by geometric form factors[23]. The latter reflect the position of an oxygen atom between two Cu atoms and are given by $(1 + cosq_a)$ or $(1 + cosq_b)$, both of which vanish when $(q_a, q_b) = (\pi, \pi)$, the wave vector for antiferromagnetic order in the insulating phase[23]. Thus the experiments require that the wave vector at which spin fluctuations peak in the metallic phase is not too different from (π, π) and, indeed, this is confirmed by neutron scattering experiments[38,39] on $YBa_2Cu_3O_{7-\delta}$. From a microscopic point of view, this behavior is not easy to understand. The wave vector (π, π) does not appear to be special for the Fermi surface[24] of $YBa_2Cu_3O_{7-\delta}$ and indeed nesting vectors are, if anything, along the (1,0) and (0,1) directions. However, it is quite natural to have spin fluctuations at (π, π) if hole-poor regions occur as low-energy fluctuations. In fact the neutron scattering experiments provide our best estimate of the size of the hole-poor domains (about four lattice spacings for $YBa_2Cu_3O_{7-\delta}$ with $\delta = 0$).

According to the discussion of Sec. **3**, the relaxation rate of a Cu nucleus $^{63}T_1^{-1}$ is proportional to $S(\omega = 0, T)$, where $S(\omega, T)$ is given by Eqs. (2)-(5). It follows that:

$$^{63}T_1 = B(4 + e^{\frac{\omega_g}{kT}} + 3e^{-\frac{\omega_g}{kT}}) \tag{6}$$

where B is a constant. This expression provides a quite good description of the temperature-dependence of $^{63}T_1$ in $YBa_2Cu_3O_{6.63}$ (ref.40), with $\omega_g = 240K$, and in $YBa_2Cu_4O_8$ (ref.41), with $\omega_g = 285K$. The calculation of the Knight shift and T_1 for other nuclei will be presented in a future publication.[14]

Recent neutron-scattering experiments[42] on $YBa_2Cu_3O_{7-\delta}$ for $\delta = 0.4$ and $T_c = 53K$, were fitted to a phenomenological expression for $S(\omega, T)$, similar to that quoted in Eqs. (2) and (3), except that the $\delta(\omega)$ term in Eq. (3) was omitted. It was found that $\omega_g = 9meV$, which is about half as large as as the value required to fit the NMR experiments. As pointed out by Tranquada et al,[42] this may not be a serious discrepancy, since ω_g varies rapidly with small changes in oxygen content. Moreover, the value of ω_g was determined above T_c by NMR but below T_c by neutron scattering experiments which, at least initially, were designed to find the superconducting gap. Clearly it is desirable to fit the the results of neutron scattering experiments carried out above T_c to the expressions given in Sec. **3**.

$La_{2-x}Sr_xCuO_{4-\delta}$ is different from $YBa_2Cu_3O_{7-\delta}$ because there are incommensurate peaks near to (π, π) which are related to a Fermi surface instability.[43] Consequently, NMR and neutron scattering experiments on this material do not give *direct* information about the "doped-insulator" effects that we have explored here and it is inappropriate to invoke a spin gap. In our picture, any specific frequency- and temperature dependence is obtained because the dynamics of the collective modes are impressed on the motion of the mobile holes.[14] Experimentally,[44,45] the frequency-dependence of the q-integrated susceptibility was found to be consistent with $arctan(\omega/2kT)$. However, within experimental error, an equally good fit can be obtained using $tanh(\omega/2kT)$, as suggested by Eq. (1).

4.2.5 Optical Absorption Spectrum

While there is by now considerable agreement on the nature of the optical absorption spectrum in the cuprate superconductors from a few hundreths of an eV to a few eV, the interpretation of this data is still very controversial. There are two schools of thought: the one-component, or Drude school,[46] infers a single species of (mobile) charge carrier for which the scattering rate and effective mass depend strongly on frequency. On the other hand, the two-component or Drude-Lorentz school[29,47-50] proposes that there are two types of charge carrier: mobile charges which dominate the absorption at low frequency and which, at low temperatures, form the superconducting condensate; and bound charges which dominate the mid-infrared absorption but depend weakly on temperature.

There is no doubt that a two-component theory is required by the data in reduced T_c materials, where a well-defined finite-frequency peak in the absorption is observed in the normal state, as we saw earlier. However, in the highest T_c materials, the normal-state absorption is monotonic and is dominated by its Drude part (regardless of interpretation). It is not at all clear that the current data are sufficient to distinguish these two models unambiguously; the whole controversy turns on how seriously one is to take the continuity of behavior as a function of doping. There is, microscopically, only one type of charge carrier, hence the appeal of the one-component theory. On the other hand, continuity of phenomena at finite frequency is expected on general grounds, as stressed by the two-component enthusiasts. From the point of view of frustrated phase separation, it is natural to expect the spontaneous generation of a two component absorption spectrum, one associated with the metallic regions, the other with the insulating (hole-poor) regions. Moreover, it is clear that the separation into two components should begin to break down when the Drude response of the metallic regions extends out to frequencies characteristic of the density fluctuations. This interpretation should be testable, especially in reduced T_c materials.

Following the argument of Sec. **3**, we may obtain an expression for the contribution $\sigma_L(\omega)$ to the optical conductivity from the local collective modes. It is found that:[14]

$$\sigma_L(\omega) = const.\omega\chi(\omega) \tag{7}$$

where $\chi(\omega)$ is given in Eq. (1). This expression has a peak at Γ and, as argued in Sec. **3**, Γ should be of the order of the antiferromagnetic exchange integral J. It has been emphasized by Thomas[29] that there is a mid-infrared peak at this energy in the cuprates at all levels of doping. We have also found that, as a consequence of the fluctuating trapping of charge in the local collective modes, the major contribution to the optical conductivity at low frequency has the form:

$$\sigma_c(\omega) = const.Re\frac{\chi(\omega)}{i\omega} \tag{8}$$

where the imaginary part of $\chi(\omega)$ is given in Eq. (1). This expression is proportional to ω^{-1} when ω is larger than T, and to T^{-1} when T is larger than ω, as observed in optimally-doped materials[46,51]. Note that the frequency- and temperature dependence is attributed to the $tanh(\omega/2kT)$ prefactor in Eq. (1), and not to the scattering rate Γ.

5. Summary

We conclude by summarizing the gist of our approach and our most important conclusions.

1) There is a strong tendency for an antiferromagnet to expel holes, which leads to phase separation in a wide class of models of dilute, neutral holes in an antiferromagnet.

2) This tendency is present in the high temperature superconductors, as evidenced by the fact that whenever the countercharges are mobile on laboratory time scales, the holes do, in fact, phase separate into a hole-rich metallic phase and a hole-poor antiferromagnetic phase.

3) If the background charge is fixed, the Coulomb interactions between the holes prohibit true phase separation but, so long as the dielectric constant is large enough, the frustrated tendency toward phase separation is still important, and gives rise to large-amplitude, collective density fluctuations which have little dispersion and are readily pinned by even moderate disorder, so in practice they are spatially localized.

4) There are strong indications of local, fluctuating phase separation in all the superconducting cuprates. Specifically, the most striking evidence is: a) The chemical potential changes relatively little as a function of doping concentration, suggesting a tendency toward local phase separation. b) In most of the high temperature superconductors the dopant concentration is relatively high ($\sim 0.1 - 0.3$); and in the case of $YBa_2Cu_3O_{7-\delta}$ the Fermi surface is rotated by 90° relative to that of a simple tight-binding model on a square lattice. As a result, $\vec{Q} = (\pi, \pi)$ is not close to a nesting vector, and may not even be a spanning vector of the Fermi surface. Nevertheless, the dominant structure in the spin susceptibility is peaked in the neighborhood of $\vec{Q}$, as evidenced by neutron scattering and the difference between the O and Cu NMR. This surely implies that memory of the antiferromagnetic insulating state is retained even in the heavily doped metallic regime. Frustrated phase separation naturally accounts for the persistence of pronounced structure at $\vec{Q}$, whereas theories based on homogeneous states have greater difficulty. c) Less direct, but still compelling evidence comes from the optical response, in which spectral features characteristic of the lightly-doped insulator are found to persist in superconducting materials. d) Neutron scattering studies of apical oxygen distances provide support for the notion that the system is inhomogeneous on some intermediate time scale.

In our opinion, these relatively gross features of high temperature superconductors confirm the relevance of phase separation in these materials. But they also *suggest* an approach to a more-detailed understanding of low-energy phenomena; specifically the temperature- and frequency dependences of the optical conductivity and the dynamical spin susceptibility, the occurence of high temperature superconductivity, and the properties of the superconducting state. We have quoted some preliminary results which are sufficiently promising that we feel they are also worth summarizing here.

1) The dynamics of the localized collective density fluctuations are governed by the same fixed-point Hamiltonian as the two-channel Kondo problem. Since this model is critical, it implies that there are singnificant fluctuations on all energy scales below some high energy cutoff Γ, the Kondo energy scale. Despite the formal similarity, the present theory should be distinguished from the one-dimensional electron gas where the low-energy behavior is determined by the fixed *line* of the Tomonaga-Luttinger model,[51] rather than a fixed *point*. The difference is important because the behavior along a fixed line is non-universal, and typically depends on coupling constants, the carrier density, and the details of short-distance cutoffs. It is an essential feature of our theory that a fixed point determines the behavior of the system in the temperature range between Γ and T_c (where new coherent phenomena come into play). This behavior accounts for the robustness and universality of many of the normal-state features of the high temperature superconductors.

2) The existence of two-channel Kondo behavior is apparent in the physical properties of the normal state and provides a plausible explanation for the optical conductivity and the spin dynamics.

3) There is an enhanced local superconducting susceptibility as a result of a pairing resonance involving the local collective mode and the metallic electrons. Superconductivity appears when coherence develops between nearby regions of the solid. Elsewhere, we and others[22,25] have examined the nature and symmetry of this resonance. While it is clear that the resulting superconducting order parameter is spin singlet, we have not yet fully explored the remaining symmetry, although it appears[22] that it may be odd in frequency.[52]

Clearly, what we have sketched in the last three points is not a fully microscopic theory; it starts from an intermediate scale model based on localized collective modes of a specific variety. In that sense it is logically independent of the central discussion of the present paper. The connection with frustrated phase separation is that this concept provides an appealing rationale for such a model.

Finally, we wish to stress that there are many significant areas of overlap between the present theory and several other existing approaches to the theory of high temperature superconductivity.

1) The spin-bag theory[53] is based on the observations that it is energetically costly to place a hole in an antiferromagnet, and that the consequent tendency of holes to share regions of supressed antiferromagnetism amounts to an effective attraction between holes. We diverge from the spin-bag picture in our belief that the consequence is phase separation, not superconductivity. Long-range Coulomb interactions are essential, in our view, to frustrate the phase separation.

2) As was first noted by Cox,[19] the behavior of the two-channel Kondo problem has important similarities to the phenomenology emphasized in the marginal Fermi liquid theory.[7,54] As a minimum, our theory of the specific low-energy localized collective modes outlined above can be viewed as a higher level model, one step closer to the

microscopic, which rationalizes the more successful aspects of that phenomenology. Important features of these collective modes are that they are dipolar and that they generate their own (two-channel Kondo) dynamics through their interaction with the mobile holes. Also they have an internal structure, which is especially important in understanding the magnetic properties of the system. Their energy scale is found to be of order J, as required to explain the experiments. On the other hand, the collective modes previously invoked in the marginal Fermi liquid theory[7] were thought to be featureless charge-density excitations, and their dynamical properties were *assumed* at the outset. We also note that, although the behavior of the two-channel Kondo problem is marginal in some respects, it is truly *non*-Fermi liquid in character.[25] In particular, the electron self energy is proportional to $(\omega/\Gamma)^{1/2}$ at T=0.

3) The present theory satisfies, for the most part, the experimental constraints on any theory of the normal state of high temperature superconductors, outlined recently by Anderson.[20] In particular, the fact that the essential dynamical structure is determined by the two-channel Kondo *fixed point* accounts for the universality and robustness of the anomalous normal-state properties (i.e. those features inconsistent with Fermi liquid theory). However, we find enhanced pairing within a single CuO_2 plane and do not need to invoke a fundamental role for the tunnelling of electrons between planes.

4) At this workshop, quite distinct approaches to the implications of phase separation, that did not invoke frustration by long-range Coulomb interactions, are considered. C. Di Castro describes models in which superconductivity either occurs in a region of the phase diagram close to phase separation, or suppresses phase separation altogether. E. Sigmund attributes the metal-to-insulator transition, which occurs with increased doping, to the percolation of ferromagnetic polarons in the otherwise antiferromagnetic material, although it has yet to be established that ferromagnetic polarons occur for the strong antiferromagnetic exchange found in the cuprate superconductors.

6. Acknowledgments

We benefitted greatly from discussions with more of our colleagues then we can possibly list. However, we must particularly acknowledge useful discussions with G. Aeppli, A. Auerbach, R. Birgeneau, J. Budnick, D.L. Cox, A. Heeger, H.Q. Lin, G. Shirane, J. Tranquada, and A.Tsvelik. Innumerable discussions with Sudip Chakravarty have been central to the development of these ideas, and to our understanding of this problem. SK acknowledges the hospitality of Brookhaven National Laboratory and IBM Almaden Research Center. Part of this work was carried out while VJE was Kramers Professor at The University of Utrecht, The Netherlands, and he thanks Professor J.E. van Himbergen for his hospitality. SK was supported in part by NSF grant #DMR -90-11803. This work also was supported by the Division of Materials Sciences, U.S. Department of Energy, under contract DE-AC02-76CH00016.

7. References

1. J. G. Bednorz and K. A. Mueller, *Z. Phys.* **B64** (1986) 89.
2. For a review, see D. J. Scalapino in *High Temperature Superconductivity*, eds. K. S. Bedell, D. Coffey, D.E. Meltzer, D. Pines, and J. R. Schrieffer (Addison-Wesley, Redwood City, 1990) p. 314.
3. P. W. Anderson, *Science*, **235** (1987) 1196; V. J. Emery, *Phys. Rev. Lett.* **58** (1987) 2794; C. M. Varma, S. Schmitt-Rink, and E. Abrahams in *Novel Superconductivity*, eds. S. A. Wolf and V. Z. Kresin (Plenum, New York 1987).
4. V. J. Emery, S. A. Kivelson, and H-Q. Lin, *Phys. Rev. Lett.* **64** (1990) 475.
5. S. A. Kivelson, V. J. Emery, and H-Q. Lin, *Phys. Rev.* **B42** (1990) 6523.
6. P. W. Anderson, in *Frontiers and Borderlines in Many Particle Physics*, eds. J. R. Schrieffer and R. Broglia (North Holland, Amsterdam, 1988).
7. C. M. Varma, P. B. Littlewood, S. Schmitt-Rink, E. Abrahams, and A. E. Ruckenstein, *Phys. Rev. Lett.* **63** (1989) 1996.
8. A. Weidinger et al, *Hyp. Int.* **63** (1990) 147; B. J. Sternlieb et al., *Phys.Rev.* **B41** (1990) 8866; E. Torikai et al, *Hyp. Int.*, **63** (1990) 271.
9. P. Imbert, lecture at this workshop.
10. E. Dagatto and J. Riera, preprint.
11. A. Bohr, B. Mottelson, and D. Pines, *Phys. Rev.* **110** (1958) 936.
12. V. J. Emery and A. M. Sessler, *Phys. Rev.* **119** (1960) 248.
13. V. J. Emery, *Hyp. Int.* **63** (1990) 13; *Physica* **B169** (1991) 17.
14. V. J. Emery and S. A. Kivelson, to be published.
15. C. Di Castro, lecture at this workshop.
16. P. Bak and R. Bruinsma, *Phys. Rev. Lett.* **49** (1982) 249.
17. See, for example F. Figueierido et al, Phys. Rev. **B41** (1989) 4619.
18. S.Chakaravarty, B.I.Halperin, and D.Nelson, *Phys. Rev.* **B39** (1988) 2344.
19. D. L. Cox, unpublished.
20. P. W. Anderson, *Science*, **256** (1992) 1526.
21. A. M. Tsvelick, *J. Phys. Condens. Matt.* **2** (1990) 2833.
22. V. J. Emery and S. Kivelson, preprint.
23. H. Monien, lecture at this workshop.
24. W. Pickett, *Rev. Mod. Phys.* **61** (1989) 433.
25. A. W. W. Ludwig and I. Affleck, *Phys. Rev. Lett.* **67** (1991) 3160.
26. P. C. Hammel; A. Heeger; and M. Mehring; lectures at this workshop.
27. B. Keimer et al, *Phys. Rev. Lett.* **67** (1991) 1930 and M.I.T. preprint (1992).
28. S. Chakravarty, private communication, has shown that an equally good fit can be obtained from an analysis of the correlation length of the non-linear sigma model[18] under the assumption that the holes simply produce a renormalized spin-stiffness parameter.
29. G. Thomas in *High Temperature Superconductivity*, eds. D. P. Tunstall and W.

Barford (Adam Hilger, Bristol, England, 1991).
30. J. W. Allen et al, *Phys. Rev. Lett.* **64** (1990) 595.
31. E. H. Lieb and F. Y. Wu, *Phys. Rev. Lett.* **20** (1968) 1445.
32. See also A. Bianconi and H. Luetgemeier, lectures at this workshop.
33. Y. Tokura, H. Takagi, and S. Uchida, *Nature* **337** (1989) 345.
34. M. Azuma et al, *Nature* **356** (1992) 775.
35. R. J. Cava et al, *Physica* **C165** (1990) 419); J. D. Jorgensen et al, *Phys. Rev.* **B41** (1990) 1863.
36. T. Egami et al, *Physica* **C185-189** (1991) 867.
37.Y. Bertier; P. C. Hammel; H. Luetgemeier; I. Mangelschots; M. Mehring; lectures at this workshop.
38. G. Shirane et al, *Phys. Rev.* **B41** (1990) 6547; J. M. Tranquada et al, *Phys. Rev. Lett.* **64** (1990) 800; J. Rossat-Mignot et al, *Physica* **B163** (1990) 4.
39. P. Burlet, lecture at this workshop.
40. M. Takigawa et al, *Phys. Rev.* **B43** (1991) 247.
41. I. Mangelschots, lectures at this workshop.
42. J. M. Tranquada et al, *Phys. Rev.* **B46** (1992): B. J. Sternlieb, M. Sato, S. Shamoto, G. Shirane, and J. M. Tranquada, unpublished.
43. P. Littlewood, J. Zaanen, G. Aeppli and H. Monien, unpublished; Y. Zha, Q. Si, and K. Levin, unpublished.
44. T. E. Mason et al, *Phys. Rev. Lett.* **68** (1992) 1414; T. R. Thurston et al, preprint.
45. S. M. Hayden, lecture at this workshop.
46. Z. Schlesinger et al, *Phys. Rev. Lett.* **65** (1990) 801; L. D. Rotter et al, *Phys. Rev. Lett.* **67** (1991) 2741.
47. D. B. Tanner and T. Timusk, University of Florida preprint (1992).
48. Y. Watanabe, Z. Z. Wang, S. A. Lyon, N. P. Ong, D. C. Tsui, J. M. Tarascon, and E. Wang, Princeton University preprint, (1992).
49. C. M. Foster, K. F. Voss, T. W. Hagler, D. Mihailovic, A. J. Heeger, M. M. Eddy, W. L. Olson, and E. J. Smith, UCSB preprint, (1992).
50. See also lectures by B. Bucher at this workshop.
51. V. J. Emery in *Highly Conducting One-Dimensional Solids*, eds J. T. De Vreese, R. P. Evrard, and V. E. Van Doren (Plenum, New York, 1979): J. Solyom, *Adv. Phys.* **28** (1979) 201.
52. V. L. Berezinskii, *Pisma Zh. Eksp. Teor. Fiz.* **20** (1974) 628 [*JETP Lett.* **20** (1974) 287]; A. Balatsky and E. Abrahams, *Phys. Rev.* **B45** (1992) 13125; E. Abrahams, A. Balatsky, and J. R. Schrieffer, preprint.
53. J.R.Schrieffer, X.G.Wen, and S.C.Zhang, *Phys. Rev. Lett.* **60** (1988) 944; *Phys. Rev.* **B39** (1989) 11663; A.Kampf and J.R.Schrieffer, *Phys. Rev.* **B42** (1990) 7967.
54. A. E. Ruckenstein and C. M. Varma, *Physica* **C185-189** (1991) 134.

DISCLAIMER

END

DATE
FILMED

9/22/92