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LOW-ENERGY PHYSICS OF HIGH-TEMPERATURE SUPERCONDUCTORS

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ABSTRACT- It is argued that the low-energy properties of high temperature superconductors are dominated by the interaction between the mobile holes and a particular class of collective modes, corresponding to local large-amplitude low-energy fluctuations in the hole density. The latter are a consequence of the competition between the effects of long-range Coulomb interactions and the tendency of a low concentration of holes in an antiferromagnet to phase separate. The low-energy behavior of the system is governed by the same fixed point as the two-channel Kondo problem, which accounts for the "universality" of the properties of the cuprate superconductors. Predictions of the optical properties and the spin dynamics are compared with experiment. The pairing resonance of the two-channel Kondo problem gives a mechanism of high temperature superconductivity with an uncoventional symmetry of the order parameter.

INTRODUCTION

High temperature superconductivity[1] is a robust phenomenon, occurring in a large number of materials containing CuO_2 planes. However, despite analytical arguments suggesting that there is an attractive interaction between charge carriers, numerical experiments[2] on a variety of models have so far failed to produce convincing evidence of a significantly enhanced pairing susceptibility, although the models are believed to include the essential physical interactions.[3] It is difficult to escape the feeling that something is missing.

We shall argue that the long-range Coulomb interaction is the essential piece of physics that has been ignored. In order to fully understand the basic physics of high temperature superconductors it is necessary to take account of the fact that they are doped insulators and retain in some way the memory of the undoped antiferromagnetic state. We contend that the competition between the long-range Coulomb interaction and the tendency of holes in an antiferromagnet to phase separate plays a central role in determining how such a system behaves.

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 interaction) with hopping amplitude t, and local moments with exchange interaction J. 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, it is a consequence of 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). This behavior is not restricted to the $t-J$ model. Studies of other 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) which implies 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.

In the presence of the long-range Coulomb repulsion the holes cannot macroscopically phase separate unless the counterions are mobile. 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. 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[6] 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.[7] 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,[7] which is manifested via the spin fluctuations as spin freezing.

In looking for these effects in the cuprate superconductors, it is necessary to consider two quite different situations. If the charge donors are mobile on laboratory time scales, they can be dragged along by the holes and preserve charge neutrality so that the holes can actually phase separate. This occurs in two special cases: In photo-doped materials[8] and in oxygen-doped $La_2CuO_{4-\delta}$ where the oxygen ions remain fairly mobile to low temperatures.[9] Of course, it is conceivable that the observed phase separation is driven 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.

On the other hand if the dopant atoms are absolutely frozen, then clearly phase separation can occur only as a short-distance, fluctuation effect, as we have already argued. There are strong indications of such a local fluctuating phase separation in all the superconducting cuprates. Specifically, the most striking evidence is:

a) Photoemission studies[10] of both $La_{2-x}Sr_xCuO_{4-\delta}$ and $Nd_{2-x}Ce_xCuO_4$ 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. 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.

b) In most of the high temperature superconductors (π,π) is not close to a nesting vector of the Fermi surface.[11] Nevertheless, experiments indicate that the spin susceptibility is peaked in the neighborhood of (π,π). 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.[12,13] 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.[13] 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.[13] 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[14] on $YBa_2Cu_3O_{7-\delta}$. From a microscopic point of view, this behavior is not easy to understand, since the wave vector (π,π) does not appear to be special[11], and nesting vectors are, if anything, along the $(1,0)$ and $(0,1)$ directions. This surely implies that some features of the antiferromagnetic insulating state are retained even in the heavily-doped metallic regime. Frustrated phase separation naturally accounts for the persistence of pronounced structure at (π, π), 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.[15-19] It has been found by Thomas et al[15] 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.

It is also worth noting that it is quite difficult to produce electronically homogeneous materials. We regard this as a 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.

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 the phenomenology of the normal state[20,21] and the mechanism of high temperature superconductivity . 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 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.[22,23] 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.

We now consider in greater detail the consequences of this picture, and introduce an effective model from which low-energy properties may be calculated.

IMPLICATIONS OF FRUSTRATED PHASE SEPARATION

The mobile holes create, annihilate and scatter from the low-energy 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:[7] for the present, we describe the essential ideas.

A. CHARGE DYNAMICS

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[24] and in the thermodynamic limit.[25] 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 fluctuations are either frozen (due to the disorder) or slow compared to the equilibration times of the microscopic degrees of freedom. 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[7]).

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[7] 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[26] that the behavior of the latter has much in common with the phenomenology of the normal state of high-temperature superconductors.[20,21,27] When the energy to create a collective mode may be neglected, the imaginary part of the susceptibility for $b_0^\dagger(\vec{r})$ has the form[28,29]:

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

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[15] and spin fluctuations.

The contribution $\sigma_L(\omega)$ to the optical conductivity from the local collective modes may be expressed in terms of this quantity. It is found that:[7]

$$\sigma_L(\omega) = const.\omega\chi''(\omega) \tag{2}$$

This gives a peak at Γ which should be of the order of the antiferromagnetic exchange integral J. It has been emphasized by Thomas[15] that there is a mid-infrared peak at this energy in the cuprates at all levels of doping.

There is considerable agreement on the nature of the optical absorption spectrum in superconducting materials from a few hundreths of an eV to a few eV, although the interpretation of this data is still controversial. There are two schools of thought: the one-component, or Drude school,[16] 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[15,17–19] 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.

In terms of fluctuating 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. The bound contribution is given in Eq.

(2). 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:

$$\sigma_c(\omega) = const. Re \frac{\chi(\omega)}{i\omega} \tag{3}$$

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[16]. 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.

B. SPIN DYNAMICS

For a material such as $YBa_2Cu_3O_{7-\delta}$ the dynamical structure factor $S(\omega, T)$ of the operator $b^{\dagger}_{1,\sigma}(\vec{r})b_0(\vec{r})$ determines the relaxation rate T_1 of Cu spins measured by an NMR experiment 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[11] 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[7] that, when ω is small compared to Γ, $S(\omega, T)$ is given by

$$S(\omega, T) = A(\frac{\omega_g}{kT})[exp(\frac{\omega_g}{kT})F(\omega - \omega_g, T) + F(\omega + \omega_g, T)] \tag{4}$$

where

$$F(\omega, T) = \frac{\pi}{2}\delta(\omega) + [\Gamma(1 + exp(-\frac{\omega}{kT})]^{-1} \tag{5}$$

and

$$A(x) = const.[3 + exp(x)]^{-1} \tag{6}$$

If the excitation energy of the collective modes had been included, it would have broadened the δ-function in Eq. (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. (4)-(6). It follows that:

$${}^{63}T_1 = B[4 + exp(\frac{\omega_g}{kT}) + 3exp(-\frac{\omega_g}{kT})] \tag{7}$$

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. 12), with $\omega_g = 240K$, and in $YBa_2Cu_4O_8$ (ref. 30), with $\omega_g = 285K$. The calculation of the Knight shift and T_1 for other nuclei will be presented in a future publication.[7]

Recent neutron-scattering experiments[31] 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. (4) and (5), except that the $\delta(\omega)$ term in Eq. (5) 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,[31] 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 here.

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$La_{2-x}Sr_xCuO_{4-\delta}$ 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.[32] 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.[7] Experimentally,[33] 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).

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.

C. SUPERCONDUCTIVITY

An additional feature of the fixed point of the two-channel Kondo problem is an enhancement of the 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[29,34] 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[29] that it may be odd in frequency.[35]

CONCLUSION

In the preceding sections we have introduced a new way of thinking about the physics of doped insulators and, in particular high temperature superconductors . There are several features of our approach that we feel are worth emphasizing here. First of all, it is clear, that what we have sketched here is not a fully microscopic theory; it starts from an intermediate scale model based on specific localized collective modes. 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.

Secondly, 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,[36] 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 the short-distance cutoff. 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.

Thirdly, we point out that the present theory is consistent with the unusual normal-state properties of high temperature superconductors. First of all, as was first noted by Cox,[26], the behavior of the two-channel Kondo problem has important similarities to the phenomenology emphasized in the marginal Fermi liquid theory.[21,37] 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[21] 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.[34] In particular, the electron self energy is proportional to $(\omega/\Gamma)^{1/2}$ at T=0.

Finally, it is clear that 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.[27] In particular, the essential dynamical structure is determined by a fixed point. 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.

Brief accounts of some of our ideas have previously been given in conference reports.[38] More detailed and extended accounts will be presented in papers currently under preparation.[7]

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