BNL-62496
CONF-9510281--1

Charge Ordering in High Temperature Superconductors

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

S. A. Kivelson*
Dept. of Physics, University of California Berkeley, CA 94720, USA

Abstract

The reasons to expect the formation of ordered or fluctuating charge stripes and antiphase spin domains in high temperature superconductors are surveyed. Evidence for such behavior is described, and some of the consequences for the physical properties of high temperature superconductors and the mechanism of superconductivity are presented.

I. INTRODUCTION

High temperature superconductors and other synthetic metals, such as organic conductors and alkali-doped C_{60} are strongly-correlated electron systems with a poor electrical conductivity and (often) a rather low effective carrier density. In general, screening is poor in such materials and the long-range part of the Coulomb interaction cannot be neglected, although it usually is omitted from the models which are studied in the theory of strongly-correlated electron systems.

There are a number of important consequences of this observation. First of all, an examination of the systematics of charge transport in synthetic metals, strongly suggests that any theory based on conventional quasiparticles with more or less well defined crystal momenta suffering occasional scattering events does not apply [1]. Secondly, both classical and quantum fluctuations of the phase of the superconducting order parameter depress the transition temperature, and together they may even prevent the establishment of long-range phase order [2]. In this picture the pseudogap, observed above T_c in underdoped high temperature superconductors, occurs below the mean-field transition temperature, where the amplitude of the order parameter becomes well-defined but there is no phase coherence [2].

In this paper, we shall focus on another aspect of the problem– the fact that strong electron correlations and poor screening lead to an intrinsic inhomogenity of the conduction electrons, in the form of droplets or of various charge and/or spin ordered states. The latter may may exhibit long-range order (as in doped La_2NiO_4 and La_2MnO_4 or $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$) or dynamical fluctuations (as in superconducting samples of doped La_2CuO_4), and they may occur in conjunction with droplet formation. It is important to realise that this behavior, which has profound consequences for the physical properties of synthetic metals, is intrinsic to the conduction electrons and is *not* a consequence of chemical or other kinds of inhomogeneity. Sections II and III will consider high temperature superconductors in particular and show that charge stripes together with antiphase spin domains are expected to occur in the CuO_2 planes as a consequence of the competition between the long-range part of the Coulomb interaction and the tendency of holes in an antiferromagnet to phase separate. Section IV will describe the recent discovery of striped phases in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ by Tranquada *et al.* [3], which provides strong and specific support for this point of view, and clearly indicates a "fluctuating-stripe" interpretation of the so-called incommensurate peaks observed in $La_{2-x}Sr_xCuO_{4-\delta}$ by neutron scattering. Section V will consider the consequences for other experiments and for the mechanism of high temperature superconductivity.

II. CHARGE INHOMOGENEITY

A central feature of high temperature superconductors is that they are doped insulators, obtained by chemically adding charge carriers to a highly-correlated antiferromagnetic insulating state. By now there is a good deal of theoretical evidence that, in the absence of long-range Coulomb interactions (*i.e.* for neutral holes), a low concentration of holes is unstable to phase separation into a hole-rich "metallic" phase and a hole-deficient antiferromagnetic phase [4].

It has always been clear that macroscopic phase separation is suppressed by the long-range Coulomb interaction [4]. However this does *not* mean that the Coulomb interaction merely stablises a state of uniform density in the neutral system. Indeed, for jellium, the Coulomb interaction favors *local* charge inhomogeneity (a Wigner crystal) whereas it is the *kinetic energy* that forces 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.

A simple qualitative argument shows that charge inhomogeneity is the likely consequence of the competition between phase separation and the long-range part of the Coulomb interaction. In linear response theory, the Debye screening length λ_D is given by

$$\lambda_D^2 = \frac{1}{4\pi e^2}\frac{\partial\mu}{\partial n}, \tag{1}$$

where μ and n are the chemical potential and number density of the neutral system, and e is the charge. Now $\frac{\partial n}{\partial \mu} = n^2\kappa$, where κ is the compressibility. Thus, between the pseudospinodals of the neutral system, $\kappa < 0$ and λ_D is imaginary. This implies that the uniform state is unstable to the formation of periodic structures, with a period determined by the value of $|\lambda_D|$. 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. The dynamical character of this state of frustrated phase separation typically stems from the *quantum* nature of the problem and is not easily displayed by solving the microscopic many-body problem. Consequently we consider two versions of a coarse-grained representation of the problem; a classical Ising pseudospin model [5] and a quantum version of the corresponding spherical same model [7]. In particular, it will be 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.

III. COARSE-GRAINED MODELS

The Hamiltonian for the Ising pseudospin model is given by:

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

Here $S_j = \pm 1, 0$ is a coarse-grained variable, representing the local density of mobile holes [6]. 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 [5]. 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.

In the spherical version of the model [7], the S_j are real numbers in the range $[-\infty, \infty]$, and quantum conjugate "momenta" P_j are introduced. Momentum order corresponds to superconductivity. The Hamiltonian includes a term proportional to $\sum(P_i - P_j)^2$ and a constraint in which the mean value of $\sum[S_i^2 + P_i^2]$ is equal to a constant. Thus the model is Gaussian, so it may be solved exactly and correlation functions and other properties may be evaluated at finite temperature. The constraint guarantees a non-trivial phase diagram, in which superconductivity competes with charge density wave order. The disordered region displays crossovers to fluctuating hole-free droplets and to orientationally-ordered stripes, as the temperature is lowered [7].

The solution of these simple models 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 unless they are destroyed by quantum effects and/or frustration. A specific example is $La_{2-x}Sr_xNiO_{4+\delta}$, which is identical in structure to $La_{2-x}Sr_xCuO_{4+\delta}$ with Ni replacing the Cu. When undoped, this system is a spin-one antiferromagnet, so it is expected that quantum fluctuations will be considerably less important than in the cuprates. When doped, La_2NiO_4 is known to form a variety of modulated phases [8]. It is evident that high temperature superconductivity can occur as a consequence of frustrated phase separation only if the charge ordering itself is suppressed, for example by the environment or 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 phonons, the atomic states of the doped holes, and the stripe orientations may also play a role.

In real systems, we expect to find static or dynamical stripes with wave vectors that are not simple nesting vectors of the Fermi surface and, in general are *metallic* because the hole concentration is governed by the energetics of phase separation. The hole-free regions should display antiferromagnetic correlations, which are coupled across the charge stripes. The consequence is an antiphase spin domain with a wave vector π parallel to the stripes, and a period equal to twice the stripe period in the perpendicular direction. In this sense, the charge order is driving the spin order.

IV. EVIDENCE FOR STRIPES.

Recent neutron scattering experiments by Tranquada *et al.* [3] have shown that the suppression of superconductivity in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ in the neighborhood of $x = \frac{1}{8}$ is associated with the formation of ordered charge and spin-density waves in the CuO_2 planes. The ordered state consists of an array of charged stripes which form antiphase domain walls between antiferromagnetically ordered spin domains. This observation explains the peculiar behavior of the $La_2CuO_{4-\delta}$ family of compounds near to $\frac{1}{8}$ doping [9], and strongly supports the idea that disordered, or fluctuating stripe phases are of central importance for the physics of high temperature superconductors [6].

In the neutron scattering experiments the principal signature of the antiphase spin domains in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ is a set of resolution-limited peaks in the spin structure factor at wave vectors $(\frac{1}{2} \pm \epsilon, \frac{1}{2})$ and $(\frac{1}{2}, \frac{1}{2} \pm \epsilon)$ in units of $2\pi/a$. The associated charge stripes are indicated by peaks in the nuclear structure factor at wave vectors $(0, \pm 2\epsilon)$ and $(\pm 2\epsilon, 0)$. Thus it is natural to interpret the *inelastic* peaks in the magnetic structure factor previously observed in similar locations in reciprocal space for superconducting samples of $La_{2-x}Sr_xCuO_{4-\delta}$ [10] as evidence of stripe *fluctuations* in which the stripes are oriented along vertical or horizontal Cu-O bond directions respectively.

Two mechanisms for producing stripe phases have been suggested by theories of doped Mott-Hubbard insulators: a Fermi-surface instability [11–17] and frustrated phase separation, as described above. The former mechanism typically relies on Fermi surface nesting and produces an insulating state, with a reduced density of states at the Fermi energy (an energy gap). On the other hand, in the frustrated phase separation picture, the period of the ordered density wave is generally unrelated to any nesting vector of the Fermi surface. The charge forms a periodic array of *metallic* stripes, with a hole density determined by the energetics of phase separation. The spin order has twice period of the charge order, and consists of undoped antiferromagnetic regions, which are weakly antiferromagnetically coupled across the charge stripes. The experiments clearly favor the latter point of view. The relevant wave vectors do not nest the Fermi surface, the stripes are partially filled, and the ordered system is not an insulator. The peaks associated with magnetic ordering develop below the charge-ordering temperature [3], which shows that the transition is driven by the charge, rather than the spin.

V. SOME CONSEQUENCES OF CHARGE ORDERING

In this section we describe some of the consequences of charge ordering an the low effective carrier concentration of high temperature superconductors.

A. Angle-Resolved Photemission Spectroscopy

The single-particle properties of a disordered striped phase also account for the peculiar features of the electronic structure of high temperature superconductors observed by angle-resolved photoemission spectroscopy (ARPES) in $Bi_2SrCaCu_2O_{8+x}$, the best studied of the hole-doped high temperature superconductors [18]. In particular, the spectral function of holes moving in a disordered striped background reproduces the experimentally-observed shape of the Fermi surface, the existence of nearly dispersionless states at the Fermi energy ("flat bands") [19], and the weak additional states ("shadow bands") [20], features which have no natural explanation within conventional band theory. In our picture, the "flat bands" arise as follows: The ordered system has energy gaps at specific points on the Fermi surface that are spanned by the wave vectors of the charge and spin structures. An energy gap serves to flatten the energy bands in its vicinity. In the case of disordered but slowly-fluctuating stripes, the energy gaps are smeared, leaving a region of dispersionless states, which give the appearance of flat bands in the ARPES experiments, although they do not correspond to quasiparticle states.

B. Magnetic Resonance

Since the stripes are charged, they are easily pinned by disorder. Thus, if the temperature is not too high, we can think of the system as a quenched disordered array of stripes, which divides the Cu-O plane into long thin regions, with weak antiphase coupling between the intervening hole-deficient regions. This picture rationalises the observation [21] that NQR sees two distinct species of Cu nuclei, which we would associate with those in a pinned stripe and those between the stripes. Since the antiphase coupling between regions is frustrating, this picture gives a microscopic justification for the observation of a "cluster-spin-glass" phase in samples with $x < 15$ % [22]. Moreover, there is evidence that the creation of dilute meandering stripes can account for the rapid supression of the Nèel temperature for $x < 2$ % [23].

C. Mechanism of High Temperature Superconductivity

The phase diagram which follows from the role of classical phase fluctuations in underdoped high temperature superconductors [2] strongly suggests that there is a very high energy scale for pairing in lightly-doped but metallic high temperature superconductors. In other words, a single stripe in an undoped antiferromagnetic environment should manifest the mechanism of pairing, although full phase coherence and long-range order could not be established. As a model for this problem, we have analysed the behavior of a one-dimensional electron gas (the stripe) in an active environment (the undoped antiferromagnet). This is a generalization of the theory of the one-dimensional electron gas. We have found several processes that involve the coupling between the mobile holes and the environment and lead to pairing, *even though the basic Hamiltonian contains only repusive interactions* [24]. Here we mention one which involves a pair of holes hopping from the stripe into a bound state of the environment. This process has a number of advantages for high temperature superconductors. A pair of holes in the medium may have a large binding energy, but such a tightly bound pair is typically immobile, since it cannot easily move without breaking up. Thus it does not, by itself, lead to high temperature superconductivity. However, the holes in the stripe are able to utilise this large binding energy to form pairs, without losing their own mobility, and in this way they achieve a high superconducting transition temperature [24]. Secondly, a stripe phase has already incorporated the long range part of the Coulomb interaction, and a pair may hop into the close neighborhood of a stripe without too much cost in energy. Thus the poorly-screened Coulomb force, which is especially damaging to pairing in systems with a small coherence length (such as the high temperature superconductors), is not a severe problem.

It is important to note that this model provides an important counterexample to the argument that magnetic effects cannot be relevant for true high temperature superconductivity, because they are so difficult to see by neutron scattering experiments on optimally-doped $YBa_2Cu_3O_{7-\delta}$. If the "environment" is simply two coupled spin chains (a spin ladder), the magnetic excitations have a spin gap of about $0.5J$ (where J is the exchange integral and is about 100meV in the high temperature superconductors) [25]. Thus there would be no spin excitations in the range of energies that have been used in most neutron scattering experiments; nevertheless two holes on a ladder form a bound state [26] of just the kind required to account for high temperature superconductivity.

Acknowledgements: SK was supported in partby 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.

*Permanent address Dept. of Physics, UCLA, Los Angeles, CA 90095, USA

REFERENCES

[1] V. J. Emery and S. A. Kivelson, Phys. Rev. Lett. 74 (1995) 3253.

[2] V. J. Emery and S. A. Kivelson, Nature 374 (1995) 434.

[3] J. M. Tranquada *et al.*, Nature 375 (1995) 561.

[4] V. J. Emery, S. A. Kivelson, and H-Q. Lin, Phys. Rev. Lett. 64 (1990) 475.

[5] U. Löw, V. J. Emery, K. Fabricius, and S. A. Kivelson, Phys. Rev. Lett. 72 (1994) 1918.

[6] V. J. Emery and S. A. Kivelson, Physica C 209 (1993) 597.

[7] L. Chayes, V. J. Emery, S. A. Kivelson, Z. Nussinov, and G. Tarjus, Physica A, in press.

[8] See, for example, J. M. Tranquada, D. J. Buttrey, and D. E. Rice, Phys. Rev. Lett. 70 (1993) 445; V. Sachan, D. J. Buttrey, J.'M. Tranquada, J. E. Lorenzo, and G. Shirane, Phys. Rev. B**51**, (1995) 12742; J. M. Tranquada, J. E. Lorenzo, D. J. Buttrey, and V. Sachan, Phys. Rev. B**52** (1995) 3581.

[9] A. R. Moodenbaugh *et al.*, Phys. Rev. B**38** (1988) 4596 .

[10] S.-W. Cheong *et al.*, Phys. Rev. Lett. **67** (1991) 1791; T. E. Mason, G. Aeppli, and H. A. Mook, Phys. Rev. Lett. **68** (1992) 1414; T. R. Thurston *et al.*, Phys. Rev. B **46** (1992) 9128.

[11] H. Schultz, Phys. Rev. Lett. **64** (1990) 1445. J. Phys. (Paris) **50** (1989) 2833.

[12] J. Zaanen and O. Gunnarson, Phys. Rev. B **40** (1989) 7391.

[13] D. Poilblanc and T. M. Rice, Phys. Rev. B **39** (1989) 9749.

[14] J. A. Vergés *et al.*, Phys. Rev. B **43** (1991) 6099

[15] M. Inui and P. Littlewood, Phys. Rev. B **44** (1991) 4415.

[16] T. Giamarchi and C. Lhuillier, Phys. Rev. B **42** (1990)10641.

[17] G. An and G. M. J. van Leeuwen, Phys. Rev. B**44**, 9410 (1991).

[18] M. Salkola, V. J. Emery, and S. A. Kivelson, to be published.

[19] D. S. Dessau *et al.*, Phys. Rev. Lett. 71 (1993) 2781.

[20] P. Aebi *et al.*, Phys. Rev. Lett. 72 (1994) 2757.

[21] B. Statt *et al.* (unpublished).

[22] J. H. Chou *et al.*, Phys. Rev. B**46** (1992) 3179.

[23] F. Borsa *et al.* Phys. Rev. B**52** (1995) 7334; A. Castro-Nieto and D. Hone (unpublished).

[24] V. J. Emery and S. A. Kivelson (unpublished).

[25] S. R. White, R. M. Noack, and D. J. Scalapino, Phys. Rev. Lett. **73** (1994) 886.

[26] H. Tsunetsugu, M. Troyer, and T. M. Rice, Phys. Rev. B**51** (1995) 16456.