Magnetic frustration model and superconductivity in doped planar CuO_2 systems

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We present a model for the magnetic phases and superconductivity in doped planar CuO_2 systems. Electronic holes on the oxygen ions introduce local ferromagnetic exchange couplings between the Cu spins. The resulting frustration destroys the antiferromagnetic state characterizing the undoped planes, and generates a new spin-glass phase. This frustration also yields an attractive interaction between the holes, whose range decreases with increasing doping. We use the BCS approximation to obtain an excellent estimate of the superconducting transition temperature $T_c(x)$ for $La_{2-x}Sr_xCuO_4$.

1. Introduction

Neutron scattering [1–3], muon rotation [4], and transport phenomena [5] show that magnetism plays an essential role in the CuO_2-based superconductors. In what follows we discuss specifically the most studied, $La_{2-x}Sr_xCuO_4$, but we believe the same phenomena to occur in $YBa_2Cu_3O_{7-\delta}$ [6] and in $Bi_2Sr_2CaCu_2O_{8+\delta}$ [7].

The T–x phase diagram of $La_{2-x}Sr_xCuO_4$ is shown schematically in **Figure 1** [8, 9]. The magnetic frustration model which predicted this phase diagram is briefly reviewed in Section 2. Spin-glass phenomena in the predicted region of the phase diagram were in fact observed independently in parallel to our work [10, 11], although they were not clearly identified as such. More details were subsequently observed by a number of experimental groups [12, 13].

The magnetic frustration model also predicts an attractive pair interaction between the hole spins which is reviewed in Section 3. In particular, recent neutron-scattering experiments [3] show that the antiferromagnetic spin correlations which mediate this interaction decay as x increases. The attractive pair potential is therefore effectively short-ranged [14]. In Section 4 we argue that the holes may reside in states that arise from nonbonding in-plane oxygen p orbitals and copper d_{xy} orbitals, thus making the Cu^{++}–O^- exchange ferromagnetic. Using the simplest approximation, that the holes may be described with a free-particle approximation in which the potential and kinetic energies are decoupled, we proposed the BCS approximation [15] described in Section 5, which yields an excellent fit for the superconducting transition temperature $T_c(p)$, where p is the hole concentration. In ideal samples, $p = x$. In real systems, p and x may differ. In our model, the appropriate parameter is p.

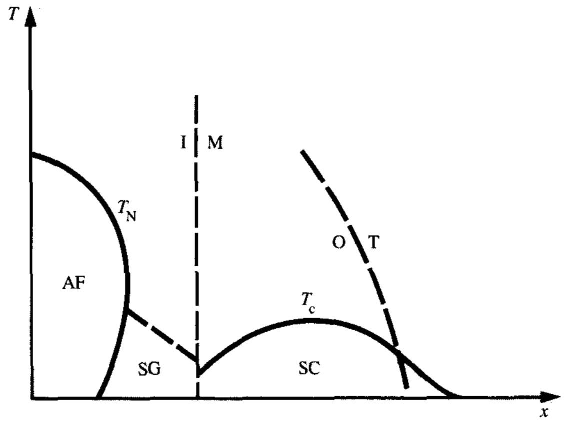

Figure 1

Schematic temperature–concentration phase diagram. AF = antiferromagnetic; SG = spin glass; I = insulator; M = metal; SC = superconductor; O = orthorhombic; T = tetragonal.

2. Frustration and spin glass

The charge carriers are the electronic holes which reside on the oxygen ions [16, 17]. For concentrations $p \leq 0.05$, the holes are localized [18]. Consider first an instantaneous configuration with a single hole on one O^- ion. The spin of the hole, $\vec{\sigma}$, will have strong exchange interactions with the two neighboring Cu spins $\vec{S}_1$ and $\vec{S}_2$. Writing

$$H = -J_\sigma \vec{\sigma} \cdot (\vec{S}_1 + \vec{S}_2), \tag{1}$$

it is intuitively clear that, regardless of the sign of J_σ, the ground state of H_σ prefers $\vec{S}_1 \parallel \vec{S}_2$. Quantum-mechanically, the exact ground state of H_σ indeed has $S_{12} = 1$ (where $\vec{S}_{12} = \vec{S}_1 + \vec{S}_2$; i.e., $\langle \vec{S}_1 \cdot \vec{S}_2 \rangle = 1/4$ [8]. Similar results were recently obtained by diagonalizing larger clusters around the hole [19]. It is thus reasonable to replace H_σ with a ferromagnetic (F) interaction, $\tilde{H}_\sigma = -K(\vec{S}_1 \cdot \vec{S}_2)$, where $K = O(|J_\sigma|) \gg |J|$. Here $J \sim 1300$ K ~ 0.11 eV [2, 5, 20] is the antiferromagnetic (AF) exchange interaction between neighboring Cu spins in the CuO_2 plane, and $K \gg |J|$ because the Cu–Cu distance is twice that of Cu–O. The replacement of H_σ by $\tilde{H}_\sigma$ is exact for classical spins at low temperatures.

Since a strong F bond in the CuO_2 plane destroys the local AF order, it also influences the coupling to the neighboring planes. The Cu spins thus feel competing AF and F interactions. In the extremely localized case, the concentration of the F bonds would be x. As x increases, the localization length l_0 of each hole increases, and this increases the effective concentration of F bonds.

Competing AF and F interactions are known to yield a sharp decrease in the Néel temperature T_N, a spin-glass (SG) phase [21], and a re-entrance from the AF to the SG phase upon cooling, because of frozen random local moments [22]. This yields the magnetic parts of Figure 1. In the isostructural $K_2Cu_xMn_{1-x}F_4$, the Cu ferromagnetism is lost at $x \simeq 0.8$ [23], corresponding to a concentration 0.36 of the very weak Cu–Mn and Mn–Mn AF bonds. As recently shown by Vannimenus et al. [24], a large ratio $K/|J|$ brings the threshold concentration down. The fact that $(l_0/a) \gtrsim 3$ [18] also renormalizes the threshold. Furthermore, quantum fluctuations also seem to lower the threshold, as indicated by preliminary Monte Carlo simulations [25]. All of these explain why in $La_{2-x}Sr_xCuO_4$ the SG phase appears at the low concentration $x \simeq 0.02$.

3. Pairing potential

A strong F bond between two Cu spins turns them parallel, against the AF coupling to the other Cu spins. The details of the resulting spin configuration depend on the symmetry of the spins. For classical Heisenberg or XY models, the Cu spins around each hole will cant, perpendicular to the F bond, with a canting angle that decays as the inverse distance from that bond; this is similar to the potential around a dipole [8, 26]. This canting angle has oscillating signs, associated with the underlying AF ordering of the Cu spins. For a pair of holes, the canting of the spins costs less energy when they approach each other. This yields an oscillating attractive interaction, which decays as $1/r^2$ for an intrapair distance r [8].

For Ising spin anisotropy, there is no canting, and a K-bond simply flips one of its spins [**Figure 2(a)**], with an energy gain of $(K - 7|J|)S^2$ (compared to the AF state without the hole) [9]. When two K-bonds are placed next to each other [**Figure 2(b)**], flipping the central spin yields a gain of $(2K - 6|J|)S^2$, which is larger by $8|J|S^2 = 2|J|$ than that of two isolated holes. This implies an attractive potential energy between the holes. Similarly, a gain of $4|J|S^2 = |J|$ results for next-nearest-neighbor bonds [**Figure 2(c)**]. Comparison of Figures 2(b) and 2(c) shows, however, that the two hole spins are parallel (triplet) in the former and antiparallel (singlet) in the latter. In this Ising case, similar arguments can be applied to each of the 22 neighboring bonds denoted by 1, 2, 2′, 3, 4, and 5 in **Figure 3**. The singlet state is unfavorable for the six bonds denoted 1, 2, and favorable (with energy gain $|J|$) for the remaining 16 bonds.

Thus far, we have derived the effective potential between the two hole spins, assuming that the Cu spins are completely correlated antiferromagnetically. Although true at low T and $x = 0$, these correlations have a finite range outside the AF phase. Recent neutron-scattering experiments [3] have demonstrated that the AF correlation length is of the order of the average separation between the holes, $K_{SS}^{-1} =$

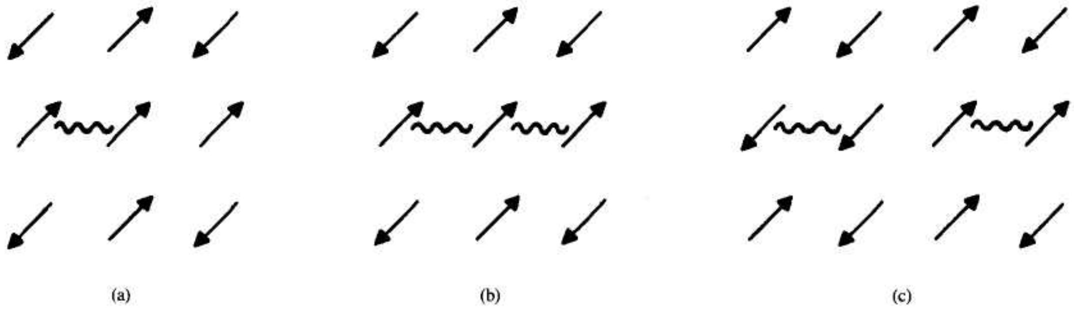

Figure 2

Ising ground state with (a) one K bond (wiggly line), (b) two nn K bonds, (c) two nnn K bonds.

$(3.8/\sqrt{p})$ Å. At distances large compared to the AF correlation length, the attractive interaction decays by an exponential factor, $\exp(-K_{ss}r)$. Extending this down to $r \gtrsim a$ eliminates the need to worry about interactions of more distant pairs not shown in Figure 3, e.g., of the kind derived for the XY case [8]. For classical spins, the energy at short distances is intermediate between the Ising and XY values. Preliminary quantum calculations, in which the coupling of the Cu–O–Cu complexes to the neighboring Cu spins is treated perturbatively, yield pairing potentials of the same order of magnitude, i.e., $|J|$. We thus expect the Ising values to give reasonable approximations of the short-range pair interaction.

Thus far we have ignored the Coulomb energy. As usual for transition-metal oxides, we write

$$V_c(r) = e^2 \exp(-K_{TF}r)/(\varepsilon r), \tag{2}$$

with $\varepsilon \sim 10$. The inverse Thomas–Fermi screening length K_{TF} is given by $K_{TF}^2 = 4(3/\pi)^{1/3} n_0^{1/3}/(a_0\varepsilon)$, with n_0 the electron concentration per unit volume and a_0 the Bohr radius. This yields $K_{TF}^{-1} = 3.6$ Å, close to the Cu–Cu spacing of 3.80 Å. The Coulomb repulsion of holes at sites 0 and 4, separated by 6 Å, is thus about 0.04 eV, which is small compared to $J \sim 0.11$ eV. On the other hand, the attractive triplet attraction of the pair 0–1 is completely overcome by the Coulomb repulsion. It should be emphasized that the screening of the Coulomb interaction is actually quite complicated. The Thomas–Fermi model overestimates the screening when K_{TF}^{-1} is close to the distance between charge carriers. On the other hand, the dielectric constant may be very large close to the metal–insulator transition. There is no doubt, however, that without screening of some sort, the Coulomb interaction will dominate unless the attractive interaction is much larger than $\sim J$.

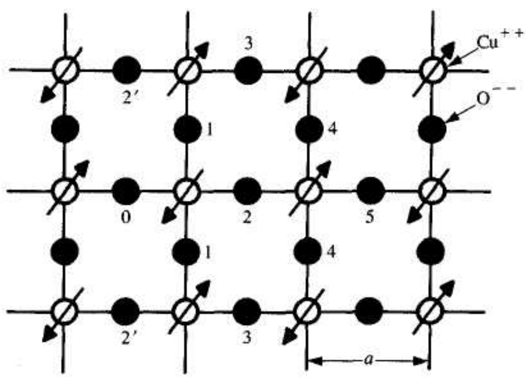

Figure 3

CuO_2 square lattice with various oxygen sites labeled.

4. Kinetic energy

Spectroscopic studies convincingly demonstrate that the effect of doping La_2CuO_4 with Sr^{++}, Ba^{++}, or excess oxygen is to place holes, which carry the current, on the oxygen atoms [16]. It is explicitly assumed in the vast majority of theoretical models for the CuO_2 superconductors that the holes are in the CuO antibonding orbitals or on the Cu atoms themselves.

Beginning from the covalent (uncorrelated) limit, it is natural to assume that the holes move in the antibonding orbital. However, the magnetism requires very strong correlations, so in [14] we began, instead, from the *ionic* limit, and argued that the holes reside in bands originating from the nonbonding in-plane oxygen orbitals. We review that argument here, but we discuss below why the situation is actually more complicated. Consider the CuO_2 layer in the ionic limit (Figure 3). Calling the two axes x and y, there are two oxygen ions per cell which we label O_x and O_y. Each site has orthorhombic symmetry which lifts the degeneracy of the three p orbitals. Because the $p_x(p_y)$ orbital of $O_x(O_y)$ is directed toward the Cu^{++}, its energy is the lowest of the three. On the other hand, the $p_y(p_x)$ orbital is directed toward the center of the square cell and the centroid of negative charge, making it the highest energy of the three. The p_z orbitals have intermediate energy. If electrons are removed from the oxygen, they must therefore come from the $p_y(p_x)$ orbitals. Using the point-ion limit, we estimate that the p orbitals are crystal-field split by 1–2 eV. This is consistent with an interpretation of Mattheiss' band calculations [31] and the quantum-chemical calculations of Guo et al. [32].

Using a tight-binding model that includes hopping-matrix elements only between the nonbonding orbitals on nearest-neighbor oxygens, the kinetic energy of the holes would be

$$E(\vec{k}) = E_0 \pm 4t \sin(k_x a/2) \sin(k_y a/2). \tag{3}$$

When we set the bandwidth $8t = 2$ eV, to agree with band structure calculations, the hole states near the maximum $\vec{k} = (1, 1)\pi/a$ are found to have an effective mass $m^*/m \simeq 2$. In fact, one should consider the hopping of the quasi-particles in which the spin complex Cu–O–Cu moves. Preliminary calculations including these effects do not modify the results in a significant way.

The idea that the holes reside in nonbonding orbitals has remained a minority view primarily because it is generally believed that the large hopping-matrix element for the bonding p orbitals would make the antibonding states highest in energy despite correlations. The latter opinion has been bolstered by some effective multiband Hubbard model calculations [33], although the difficulty in making a definitive statement about this has been emphasized by McMahon et al. [34].

In our early work, as well as that by most others, the copper d_{xy} orbital was not explicitly considered. We had assumed, as is customary, that the admixture of the d_{xy} orbital was very small because of the large energy denominator resulting from large crystal-field splittings. The overlap of the d_{xy} with the nonbonding band [Equation (3)] at $k_x = k_y = \pi/a$ is large, within a factor ~2 of the overlap of the bonding p orbital with the $d_{x^2-y^2}$ [35]. In the absence of the large energy denominator, therefore, the two bands would have comparable widths, and the Coulomb splittings of the p orbitals would probably make the nonbonding the highest energy for electrons, as discussed above. Moreover, we now know that when correlations are included, the energy denominator is actually *smaller* for the d_{xy} than for the $d_{x^2-y^2}$. The evidence is that in La_2CoO_4 the Co ions are in the high spin state [36], showing that the intra-ionic (Hund's rule) exchange is larger than the crystal-field splittings.

We emphasize that because the nonbonding/d_{xy} orbitals are orthogonal to the Cu $d_{x^2-y^2}$ orbitals, the motion of the holes does not alter the occupancy of the $d_{x^2-y^2}$, which give rise to the antiferromagnetism. This orthogonality also leads to the conclusion that the coupling of the hole spin to that of the Cu^{++} ion is ferromagnetic [32]; that is, $J_\sigma > 0$. The inclusion of the d_{xy} orbitals is expected to alter the estimate for m from the dispersion relation [Equation (3)], and the effective mass will be even smaller than the value $m^*/m = 2$ given above. However, there will be mass enhancement from the coupling of the hole spin to the Cu spins.

The assumption that the holes move in nonbonding/d_{xy} orbitals makes our analysis of the superconductivity (Section 5) simpler. However, we do not believe that the conclusions will be changed in a fundamental way if the holes are in antibonding states.

Castellani et al. [37] have recently examined the properties of a model Hamiltonian including nonbonding states as well as the antibonding bands.

5. Superconductivity

One possible solution of our model is d-state pairing of the form $\phi \sim (\cos k_x a - \cos k_y a)$, as first deduced by Emery [17] from a different model. $L = 2$ pairing seems favorable for the following two reasons: First, such a d-state wavefunction eliminates the on-site Coulomb repulsion. Second, it eliminates the repulsive magnetic interactions of site 1, leaving predominant the attractive interaction of sites 4 and 5. However, it should be emphasized that an s state with a zero at the origin in its radial dependence might have a lower energy [38], since more of the next-nearest-neighbor pairs contribute to the attractive potential. Only a detailed quantitative analysis will resolve this question.

In order to obtain a rough estimate of the dependence of T_c on x, we now use a BCS weak-coupling approximation [15]. First, we assume that the Fermi energy, which in this case provides the cutoff in the BCS integral, is simply $E_F = 15\,000$ K $(p - p_c)$ for $m^*/m = 2$, which assumes a 2D free-particle dispersion relation. Here $p_c = 0.05$ is the hole concentration at the metal–insulator transition. Second, we assume that the optimal distance for the pairing interaction is ~6 Å, corresponding to the pair 0–4 in Figure 3. Thus, $V(0) = V_0 e^{-6K_{SS}} = V_0 e^{-1.58p^{1/2}}$. Including the Coulomb repulsion, this yields the BCS mean-field expression

$$T_c(p) = 15000\,(p - 0.05)\exp[-C/(e^{-1.58p^{1/2}} - d)]\,\mathrm{K}, \tag{4}$$

where as usual $C \sim [N(0)V_0]^{-1}$ and d is the ratio of the average Coulomb to the magnetic potential for $r = 6$ Å at $p = 0$. Clearly, because of the short superconductivity coherence length [39] and the two-dimensional fluctuations, mean-field theory overestimates T_c. However, the variation of T_c with p should come out correctly. In **Figure 4** we show Equation (4), plotted versus p. To compare with the data, we chose C to give $T_c(0.15) = 36$ K and $d = 0.15$. This gives $[C/(e^{-1.58p^{1/2}} - d)]^{-1} = 0.26$, consistent with the weak-coupling approximation inherent in (4). A change in m^* in (4) leads to a small change in C, with no important change in the shape of the curve. Given the extreme simplicity of the model, the agreement with the data is excellent, providing substantial evidence that our basic approach is correct. In particular, among various theoretical models ours may be unique in predicting the remarkable re-entrant decrease of $T_c(p)$ at large p. Note that the agreement with the data would probably improve if one took into account the fact that the cutoff $E_F(p)$ must be replaced by the p-independent spin-wave-zone boundary Debye energy, ~2.4 J, for larger p when 2.4 J $\lesssim E_F$.

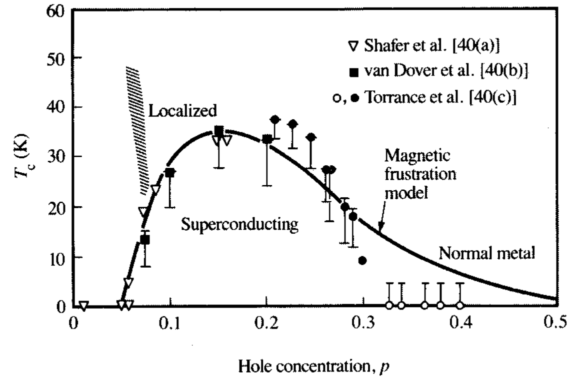

Figure 4

Superconducting transition temperature versus hole concentration in $La_{2-x}Sr_xCuO_4$. The solid line is Equation (4), with $C = 1.5$ and $d = 0.15$. Data are from [39]; the figure is adapted from Torrance et al. [40].

6. Going beyond mean-field theory

As many authors emphasize, most of the fluctuations occur in the CuO_2 planes. In [5] we have used the exact form for the order-parameter susceptibility of the planar spin-1/2 Heisenberg model [41], and have treated the weak interplane coupling using mean-field theory. The results were in excellent agreement with the measured susceptibility. This proved, first, that the planar system is indeed described well by a renormalized classical Heisenberg model [41] (justifying in retrospect our intuitive classical spin-glass picture, and making non-Néel ground states very unlikely), and, second, that only planar fluctuations seem to be important near the antiferromagnetic transition.

Following that success, we treated the transition from the smectic C to the hexatic I liquid-crystal phases using similar weakly coupled Kosterlitz Thouless [42] planar XY model systems [43]. The results gave a fast increase in the three-dimensional order parameter, and a large peak in the specific heat (which might be confused with a small order-parameter exponent and a large specific heat exponent, α).

Since the superconducting ordering is described by an XY model, and since the planes are weakly coupled, we expect such an approach to take account of most of the fluctuations near T_c. Indeed, large values of α seem to have been measured [44].

7. Conclusions

In summary, we have presented a heuristic model for the superconductivity in CuO_2 lamellar systems, with specific application to $La_{2-x}Sr_xCuO_4$. The attractive potential between the holes is based on the frustration of the Cu antiferromagnetism, which also results in our predicting a spin-glass phase which has since been observed experimentally. Our derived $T_c(p)$ curve contains a maximum at $p \sim 0.1$–0.2, due to the competition between the growth of the Fermi energy with $(p - p_c)$ and the decrease in the frustration energy with the decay of the AF correlations. Our $T_c(p)$ curve is also in excellent agreement with the data. Because of the large value of J we agree with Emery and Reiter [45] that retardation effects are probably not important.

We have provided a simple model in which the pairing of holes arises from minimization of magnetic frustration energy. If the pairing is s-wave, there is no reason why other mechanisms cannot increase the BCS gap. Electron–phonon coupling may contribute in $La_{2-x}Sr_xCuO_4$, and this may be the reason for the finite isotope effect in that material. In other materials, other mechanisms [46] such as excitonic or charge transfer may add to the magnetic one.

Our model may be applied to $YBa_2Cu_3O_{7-\delta}$ and to $Bi_2Sr_2CaCu_2O_{8+\delta}$ as soon as appropriate structural and magnetic information becomes available. Theoretically, better estimates of the frustration potential, based on quantum and classical evaluations of the spin configurations at various values of p and $K/|J|$, and more detailed BCS calculations, based on these more accurate potentials, may improve the predictive power of the model.

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