Magnetically induced hole-hole correlations in CuO_2 sheets

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A randomly decorated Ising model is considered to study first-neighbor hole-hole correlations in CuO_2 sheets. Within a Bethe-lattice approach, two pairing mechanisms are observed: one weak, induced by magnetic fluctuations; the other, as a consequence of frustration, so strong that we argue its relevance for high-T_c superconductors. Experimental fitting for the $La_{2-y}Sr_yCuO_4$ suggests that superconductivity arises at an equicorrelation line in the temperature-versus-hole concentration plane.

INTRODUCTION

Experimental evidence supports the fact that magnetism is an important ingredient to the understanding of the behavior of copper-based metallic oxide superconductors.[1] Several mechanisms based on magnetic grounds have been proposed to explain the formation of bound states by mutually attracting particles,[2] but none of them is widely accepted or confirmed experimentally. So far, the only obvious structural feature common to all high-T_c oxides is the existence of CuO_2 planes,[3] where the supercurrent is supposed to flow.[4] According to most authors, charge transport in these compounds occurs via electron holes in the oxygen band originated by doping.[5] The holes generate an effective local ferromagnetic exchange between Cu ions which were previously antiferromagnetically coupled. Also, the holes lessen the strength of the Cu antiferromagnetic coupling.[6] Aharony *et al.*[7] have suggested that the resulting frustration yields an effective attractive interaction between the holes and that this could lead to superconductivity. Numerical calculations on finite CuO clusters,[8] t-J models,[9] and extended Hubbard models[10] have shown a pairing tendency between carriers. The incipient localization effect introduced by disordered doping[11] actually competes with the pairing attraction. However, it can be neglected in the small-doping regime as a result of the large energies involved by frustration. This pairing tendency can be enhanced by next-nearest-neighbor hopping in a t-t'-J model,[12] but the essential feature is that competition must be present. In strongly correlated models, the effective attractive potential may lead to cluster formation rather than pair formation. Consequently, this attraction could drive the system to a phase-separation instability,[13] which is an unwelcomed feature for the issue of superconductivity. In this way it is important to investigate the role played by the magnetic-induced attractive potential between holes in high-T_c superconductors.

The Ising approach has been successfully used to describe high-T_c materials,[4] although spins are believed to be more Heisenberg type near the Néel temperature T_N.[15] Neutron scattering has shown some small anisotropy in spin space in both La_2CuO_4 and $YBa_2Cu_3O_{6+\delta}$ with ordered spins confined to a particular direction or plane.[16] Moreover, experimental susceptibility data[17] suggest that, at low temperature, spins might well develop Ising or XY symmetries. Monte Carlo simulations[18] have explicitly shown a pairing tendency by using classical Ising spins on a square lattice to describe the magnetic moments of the Cu ions interacting via exchange parameters plus and minus J randomly distributed to simulate the frustration effects introduced by doping. Rigorous results[19] have shown through oxygen-oxygen correlations that the ground state of a lattice-gas model with short-range effective pair interaction between oxygens[20] can develop stable paired states. The correlation function between the charge carriers is itself a very important tool to the understanding of the pairing mechanism.

MODEL AND FORMALISM

Motivated by the interesting results of the hole-induced frustration model and having in mind all the facts described above, we have studied the behavior of a first-neighbor hole-hole correlation function for a simple model: one that yields exact results and yet still keeps the most relevant features of the pairing mechanism such as magnetic frustration and itinerancy of holes. The model consists of representing the Cu magnetic moments by $\frac{1}{2}$ Ising spins S_i localized at the vertices of a lattice. Next, we distribute at random a few holes with spin σ_{ij} between Cu ions (on bond i-j). The spin S_i interacts with its neighbor S_j via an antiferromagnetic exchange parameter $J<0$, in the absence of a hole between them. In the presence of a hole, the exchange parameter is modified and the spin S_i interacts with spin S_j through the parameter γJ and also each of them interact with the hole spin σ_{ij} through the parameter αJ. As the hopping of holes from bond to bond leads to a large energy band, we do not allow double occupation. This means that we are working in the limit of strong intrasite Coulomb repulsion. The itinerancy of holes is considered here by using a grand-canonical distribution to treat the hole configurations and therefore taking annealed averages. Although one does not consider their kinetic energy in this annealed regime, the holes are permitted to move throughout the lattice among the configurations that minimize the free energy. The kinetic energy is relevant in a BCS approach[21] in determining the density of states of a single particle near the Fermi surface, but we believe that the pairing mechanism is well described by the present approach. The stability of the superconducting state will actually be determined by the competition between the kinetic energy and effective pairing tendency. The annealing procedure is particularly suitable, for it takes into account fluctuations on the total hole number due to migration of holes from one sheet to another.

The Hamiltonian of such a model can be written as

$$H=-\sum_{i,j} JS_iS_j(1-n_{ij})+\gamma Jn_{ij}S_iS_j + \alpha Jn_{ij}\sigma_{ij}(S_i+S_j)+\mu n_{ij} \;, \tag{1}$$

where the summation runs over all nearest-neighbor sites of a lattice and μ is the chemical potential per hole. Here n_{ij} is the hole-occupation number of the i-j bond assuming only the values 0 or 1 in the absence or in the presence of a hole, respectively. It should be noted that only classical variables appear in Eq. (1), so that superconductivity rises only when the effective attraction between holes, presented by this model, is considered together with the Pauli exclusion principle. Summing up the spin configurations of holes σ, we obtain the following expression for the effective Hamiltonian H_{eff}:

$$H_{\text{eff}}=-\sum_{i,j} JS_iS_j(1-n_{ij})+\alpha_{\text{eff}}JS_iS_jn_{ij}+\mu n_{ij} \;, \tag{2}$$

in which α_{eff} is a temperature-dependent parameter given by

$$\alpha_{\text{eff}}=\gamma+(1/2K)\ln\cosh(2\alpha K) \;, \tag{3}$$

with $K=J/k_BT$. The hole concentration may be thermodynamically obtained through the evaluation of the mean number of holes per bond:

$$x=\langle n_{ij}\rangle=\frac{\sum n_{ij}\exp(-\beta H)}{\sum \exp(-\beta H)} \;, \tag{4}$$

where $\beta=1/k_BT$ and the summation runs over all Cu spin and hole configurations. Equation (4) will be used in order to eliminate the hidden variable μ. Some exact results related to the magnetic phase diagram of a quite similar model were presented by dos Santos *et al.*,[22] and connections with superconductivity were suggested. The principal aim of the present work is to look closely at the pairing mechanism through the analysis of the hole-hole correlation functions $C_{\mathbf{R}}$ defined here as

$$C_{\mathbf{R}}=\langle n_{ij}n_{kl}\rangle-\langle n_{ij}\rangle^2 \;. \tag{5}$$

Here $\mathbf{R}$ is the distance in bond units between i-j and k-l bonds. This correlation function may represent three different regimes as follows: $C_{\mathbf{R}}=0$ for uncorrelated holes, $C_{\mathbf{R}}<0$ when there is a repulsion between holes, and $C_{\mathbf{R}}>0$ when there is an attraction between them, thus leading to a pairing tendency.

It is well known that mean-field theory on the reciprocal $\mathbf{k}$ space works quite well for superconductors with large-coherence-length Cooper pairs. As copper oxide superconductors involve short-coherence-length Cooper pairs,[23] a mean-field theory on the real space should be more adequate to describe the main properties of such materials. Let us then place the Cu ions at the vertices of a Bethe lattice (with coordination number $q=4$), which belongs to the same mean-field universality class. In this case the above correlation function C_1 for two neighboring holes, whose spin correlation $\langle\sigma_{ij}\sigma_{kl}\rangle$ favors triplet pairing, is exactly evaluated.

Based on the hierarchical structure of the Bethe lattice, one is able to write the grand-partition function in terms of an effective field for the sth ring, denoted here by p_s, which is defined through a recursion relation of the form

$$\exp(2p_{s-1})=\frac{C_s^+}{C_s^-}=\frac{a_s^+ + vb_s^+}{a_s^- + vb_s^-} \;, \tag{6}$$

with

$$a_s^{\pm}=2\cosh[(q-1)p_s\pm K_{\text{eff}}] \;,$$
$$b_s^{\pm}=2\cosh[(q-1)p_s\pm K_{\text{eff}})] \;, \tag{7}$$
$$v=\exp(\mu/k_BT),\quad K_{\text{eff}}=\alpha_{\text{eff}}K \;.$$

In the thermodynamic limit, $s\to\infty$ and $p_s\to p^*$, so that one has to solve the fixed-point version of Eq. (6) to find its solutions $p^*(K,K_{\text{eff}},\mu)$ for the various phases. In this way the grand-partition function is readily obtained, and therefore the thermodynamic functions of interest may be found exactly. In this way the first-neighbor correlation function takes the form

$$C_1=v^2\frac{(b^+)^2(C^+)^{q-2}+(b^-)^2(C^-)^{q-2}}{(C^+)^q+(C^-)^q}-x^2 \;, \tag{8}$$

where the quantities without a subscript are to be calculated at the fixed point.

FLUCTUATION AND FRUSTRATION EFFECTS

The first thing we notice is that the results obtained here agree remarkably well with Monte Carlo simulations of a similar model embedded on the square lattice in the particular case where plus and minus J bonds are randomly distributed.[18] Figure 1 compares the predicted behavior of the normalized correlation function P, as a function of the hole concentration, with the Monte Carlo results. Here the normalization is done according to $P=C_1/\lambda$, where

$$\lambda=\tfrac{1}{2}x(1+x)-x^2 \;, \tag{9}$$

so that $0\le P\le 1$, as in Ref. 18. The pairing parameter we have chosen to fit represents the correlations between two collinear bonds of the square lattice because it is closer to the correlation between two neighboring bonds of a Bethe lattice. Note that the fitting is better for lower concentrations. Experimental results for $La_{2-y}Sr_yCuO_4$ (Ref. 24) have shown that the hole concentration x is a quasilinear function of dopant concentration only up to $y=0.15$. For higher dopant concentrations, oxygen vacancies are generated in the CuO_2 sheets and the hole concentration decreases. The fitting is also better for T away from T_N where superconductivity arises. Therefore, we may conclude that the Bethe lattice reveals itself quite good to describe the magnetic pairing mechanism in the CuO_2

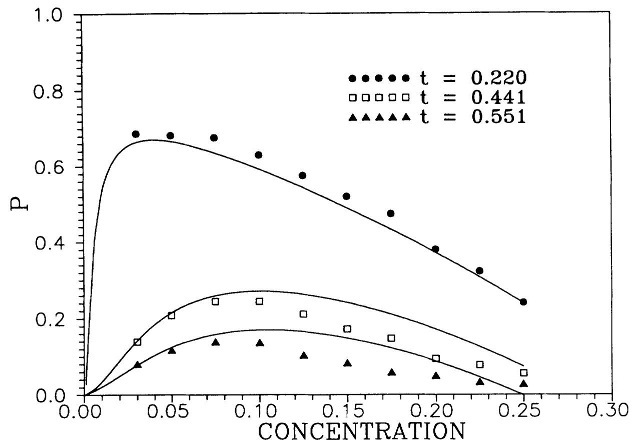

FIG. 1. Normalized nearest-neighbor correlation P-vs-hole concentration for the annealed random-bond ($\pm J$) Ising model on a Bethe lattice of coordination number $q=4$ (solid lines). Characters represent data for the correlation between a collinear pair from a Monte Carlo simulation of the above model on a square lattice (see Ref. 18). Here $t=T/T_N$, where T_N is the Néel temperature at $x=0$. The agreement is excellent, especially in the temperature and hole concentration ranges of interest to superconductivity.

sheets.

Our results show that for small levels of concentration, the maximum value obtained by the first-neighbor correlation is given by $C_1(T=0)=\frac{1}{2}(x-x^2)$. As the concentration increases, the factor on the right-hand side of the above expression is even smaller than one-half. Here it should be noted that the value above is just half of the one required for a system composed of a phase where all bonds are occupied by a hole and another one composed by hole-free bonds. Therefore, one might conjecture that the effective attraction induced by the magnetic degrees of freedom is not enough to promote a phase separation instability. To confirm this conjecture, a detailed analysis of other correlations and of the thermodynamic stability of the system is required. Nevertheless, as we shall see in the following, just C_1 gives very interesting and surprising results.

Figure 2(a) illustrates the C_1 behavior as a function of temperature for $\gamma=1.0$, $\alpha=0.5$ (dashed line), and $\alpha=1.0$ (solid line), with a hole concentration of $x=0.15$. It is observed that, if the effective hole-mediated interaction $\alpha_{\text{eff}}J$ between Cu ions is not ferromagnetic ($\alpha<1.0$), there is a positive correlation between holes at finite temperatures in the ordered phase. This means that an effective attraction induced by magnetic fluctuations exists even when frustration is absent, except for the ground state where the holes are uncorrelated. On the other hand, when the resulting hole-mediated coupling becomes ferromagnetic [$\alpha>1.0$, Fig. 2(b)], the disorder and competition between these and the pure antiferromagnetic ones introduce frustration effects which considerably enhance the hole-hole correlation. In this case the correlation has a feature not observed in the nonfrustrated case: It persists even at $T=0$. This is clear evidence that this correlation is essentially induced by a free-energy-

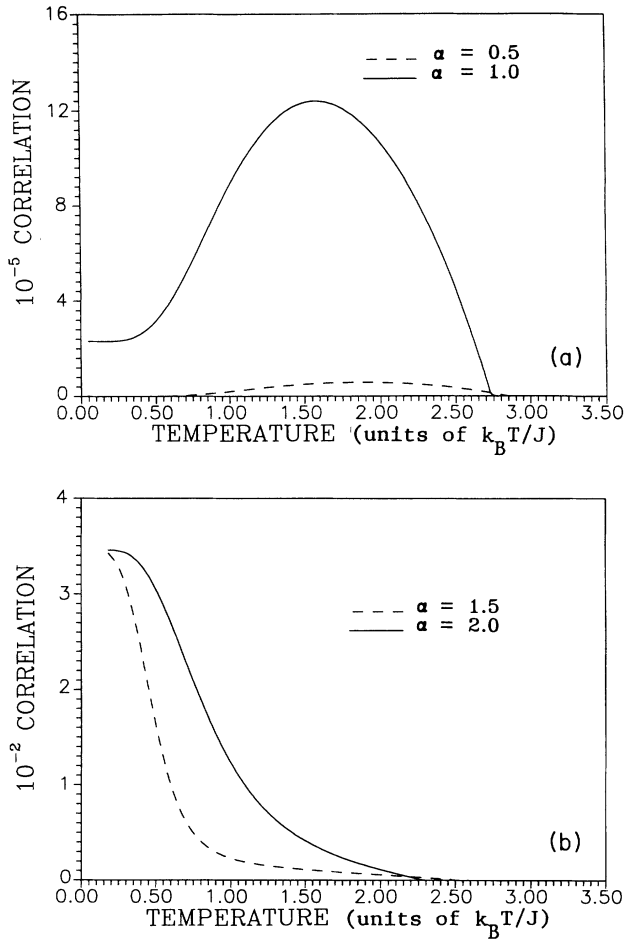

FIG. 2. Hole-hole correlations C_1 vs temperature for a concentration of holes $x=0.15$ and $\gamma=1.0$: (a) Nonfrustrated regime, where the effective coupling between Cu ions is not ferromagnetic. There is only a small correlation induced by magnetic fluctuations for $\alpha<1.0$ (dashed line). Note that for $\alpha=1.0$ (continuous line), in which case the system is diluted at $T=0$, a weak correlation persists even in the ground state. (b) Frustrated regime where the correlation is highly enhanced by a free-energy-minimization process.

minimization process. The enormous difference between the mean amplitude of the correlation function in these two cases (note the difference of a 10^3 factor in the correlation scales) reveals the fundamental role played by frustration in the pairing mechanism. We see that in the particular case of $\alpha=1.0$, when the system is diluted at $T=0$, a small correlation persists even in the ground state, indicating that both mechanisms are equally present in this case.

EQUICORRELATION LINE

There is a thermodynamic effective attractive potential between holes whether there is or not competition. This raises the question about the experimental evidence that a superconductor state is stable only when frustration effects are present. We are impelled to suggest that the reasons for that lie in the following considerations: The maximum value achieved by the correlation function C_1

in the nonfrustrated regime happens when the system is diluted at $T=0$. Note that, in this case, the correlation is of order 10^{-4}, a value which, as we shall see below, does not seem to be large enough to stabilize the bound state. The equilibrium state is one that minimizes the free energy. Here the small energy decrease in forming a bound state does not compensate the consequent entropy reduction. Furthermore, the kinetic energy of holes also contributes to break up the bound state when the latter is weakly coupled. In this way the bound state will stabilize only when the hole-hole correlation achieves a critical value, above which the system becomes superconductor. Based on the behavior of other annealed decorated systems, we suggest that this critical correlation is temperature and hole concentration independent.[25] By using our simple model based on classical variables, we cannot obtain such critical correlation. However, we may conjecture an approximate value for it by means of experimental data. In $La_{2-y}Sr_yCuO_4$, for example, superconductivity arises at a hole concentration of $x_0 \cong 0.055$.[24] For this concentration and using the values of $\gamma=0.68$ and $\alpha=1.9$ obtained by Guo, Langlois, and Goddard[6] through an *ab initio* calculation for finite CuO_2 clusters, our model yields $C_1=0.0201$ at $T=0$. Note that this value is about 200 times greater than the maximum value of C_1 in the nonfrustrated case where there is only a fluctuation-induced pairing mechanism. The equicorrelation line in the T-vs-x plane at this value of correlation is plotted in Fig. 3 together with the experimental data of Shafer, Penney, and Olson[24] and the theoretical prediction of Birgeneau, Kastner, and Aharony[26] for the superconducting transition. This line is normalized to give $T_c(0.15)=35$ K. The excellent agreement presented by the equicorrelation line with experimental data reinforces

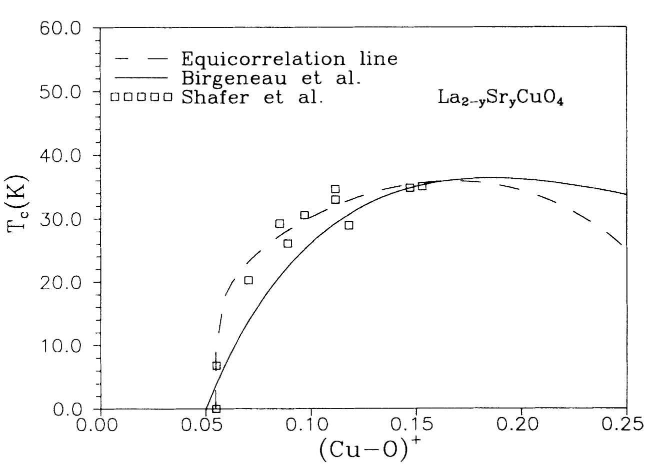

FIG. 3. Equicorrelation line is plotted in the T-vs-x plane (dashed line) using the values $\gamma=0.68$, $\alpha=1.9$ (see Ref. 6), and $C_1^*=0.0201$. Experimental data of T_c for $La_{2-y}Sr_yCuO_4$ (see Ref. 24), together with BCS-like fitting (see Ref. 26) are also plotted on the same graph. The plot strongly supports the suggestion that the superconducting phase arises at a constant value of hole-hole correlation. Note the rapid increase of T_c above the $(Cu\text{-}O)^+$ threshold predicted by this line as it has been experimentally observed.

our suggestion that superconductivity occurs at a constant hole-hole correlation. Furthermore, when x is near x_0 this line provides the following law for T_c:

$$T_c \propto |\ln(x-x_0)|^{-1} , \tag{10}$$

in opposition with the linear behavior predicted by BCS-like approaches.[26,27] This abrupt increasing of T_c is also in agreement with experiment.[24]

CONCLUSIONS

We have used an Ising approach to study first-neighbor hole-hole correlations in the CuO_2 sheets and its connections with superconductivity. In spite of the fact that we use a Bethe lattice to describe the sheet, comparison with Monte Carlo results shows that this procedure gives quite good quantitative results in the range of temperature and hole concentration where superconductivity is supposed to occur. We have found two pairing mechanisms: one weak and always present as a result of magnetic fluctuations and the other strong as a result of free-energy-minimization processes and present whenever the system is frustrated. We believe that the latter is responsible for the high-T_c superconductivity observed in the CuO_2-based ceramics. Also, we have conjectured that the effective attraction is not enough to promote a phase separation of holes. The fact that we do not consider the kinetic energy in our annealed approach means that we are working in the large-hopping-amplitude limit of extended Hubbard models. Therefore, previous results of the literature[28] confirm our conjecture on the absence of phase separation in this limit. Supported by experimental data, we have suggested that the transition to superconductivity occurs at an equicorrelation line and obtained in this way the critical value $C_1^*=0.0201$ for the $La_{2-y}Sr_yCuO_4$ compound. Other superconductors may present a slightly different critical correlation because of particular features of the density of states near the Fermi surface. For example, in $Bi_2Sr_2CuO_{6+\delta}$ the critical correlation required to give $x_0=0.10$ (Ref. 29) is $C_1^*=0.03$. Experimental data related to hole-hole correlations at the superconducting transition, if physically reliable, would be of great interest in order to confirm our results. From a theoretical point of view, one must obtain the effective pair potential[7,26,30] through a mapping onto a model containing only interacting holes. This potential can be then used in an interacting Fermi gas embedded in a square lattice to obtain explicitly the superconducting transition. The results presented here may be improved by considering spins with continuous symmetry to study the influence of canting of spins[7,31] on the hole-hole correlations. We are currently working along these lines.

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