

Exact results for randomly decorated magnetic frustrated models of planar CuO_2 systems

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We present *exact* results for a random *annealed* decorated square lattice in which the nodal spin interacts antiferromagnetically ($J_A < 0$) with its nearest neighbors and ferromagnetically ($J > 0$) with a decorated spin on the bond between them. For the n -vector version of the model, we present an exact calculation of the effective coupling of a decorated bond. Moreover, for the Ising version we obtain exactly the magnetic phase diagram. In particular, we find that this model exhibits a critical decorated bond concentration $p_c = (1 - \sqrt{2}/2)/2 \approx 0.1464$. . . above which, for small values of the ratio $|J_A/J|$, the antiferromagnetic phase ceases to exist, i.e., the paramagnetic phase extends to $T=0$. This intriguing exact result for the magnetic phase diagrams may possibly have relevance for the planar copper oxide superconducting materials; although the CuO_2 planes in these materials are believed to be described by quantum Heisenberg spins, our results for the Ising model may capture some of the relevant qualitative physics.

I. INTRODUCTION

The important structural feature common to all high- T_c superconducting materials discovered so far is the existence of CuO_2 planes. Currently, there are some theoretical arguments¹⁻⁸ and growing experimental evidence, including especially neutron scattering,⁹⁻¹³ muon-spin rotation,^{14,15} nuclear-quadrupole-resonance (NQR) technique,¹⁶ transport,¹⁷ and Mössbauer¹⁸ measurements, that magnetism plays a central role in the CuO_2 -based superconductors. For instance, consider the structurally simplest superconductor system, $\text{La}_{2-x}(\text{Sr},\text{Ba})_x\text{CuO}_{4-y}$. For a range of concentrations x of doping material (Sr,Ba), the three-dimensional (3D) antiferromagnetic long-range order undergoes a phase transition to a disordered phase (some authors find evidence of a spin-glass phase^{5-7,16}). However, the experimentally observed¹³ 2D antiferromagnetic correlations strongly suggest the existence of large domains of antiferromagnetic 2D order in the superconducting state.

Of course, superconductivity in the new materials appears when antiferromagnetism decreases. However, from the theoretical point of view—as has strongly been argued by Schrieffer *et al.*⁶—it is reasonable to believe that 2D spin order exists over domains characterized by large sizes compared to the size of the CuO_2 unit cell. Although this may not characterize the existence of long-range 2D order, the size of such domains could be large enough to create an antiferromagnetic underlying local order in which the pairing of the conducting holes occurs, as required by most of the pairing scenarios

presented so far. The exact connection between the magnetism of these doped CuO_2 planes and the novel superconductivity properties of these materials defies a complete and consistent explanation.

It has been shown that the effect of doping^{2(a)} or changing oxygen content^{2(b)} in pure La_2CuO_4 is to create holes (O^-) on the oxygen atoms located between the adjacent copper ions in the CuO_2 planes. With each hole is associated an unbalanced spin. This generates a local ferromagnetic exchange interaction with the two nearest-neighbor copper ions, which will compete with the otherwise antiferromagnetically coupled Cu^{2+} ions giving rise to magnetic frustration. As a result of this frustration mechanism one expects that the magnetic behavior of the CuO_2 planes should be strongly influenced by the presence of these holes on the oxygen ions which are responsible for carrying the supercurrent. This fact, by itself, is a stimulating hint for the possible connection between the magnetic and the superconducting properties of these materials. Moreover, these holes can be conceptually considered either as *quenched* or *annealed* random distributed in the *insulating* and *metallic* phase, respectively.

If we denote the spins of the two neighboring Cu^{2+} ions by \mathbf{S}_1 and \mathbf{S}_2 , and that of the O^- ion by σ , then the relevant *magnetic* Hamiltonian for each bond is $H_{12} = -J\sigma \cdot (\mathbf{S}_1 + \mathbf{S}_2)$ with $J > 0$. Since J results from overlap integrals over the Cu-O distance, one expects J to be larger than the original antiferromagnetic Cu-Cu coupling J_A . Indeed, very recently *ab initio* calculations¹⁹ have been used to estimate $J_A/J \approx -0.36$ in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. On the other hand, pure La_2CuO_4 is or-

thorhombic, but the substitution of Sr for La stabilizes a *tetragonal* phase.²⁰ The Sr-doped materials for $x \gtrsim 0.05$ were shown to be superconductors with a maximum transition temperature at about 40 K when $x \simeq 0.15$ (see Ref. 21). The superconducting properties are lost for $x = x_{\max} \geq 0.32$ —for the best analyzed samples with no oxygen vacancies (see Ref. 22)—well inside the tetragonal phase where the CuO_2 planes interact very weakly with each other.^{23,24}

In order to make contact with doped La_2CuO_4 , we present exact results for a decorated square lattice²⁵ in which the copper spin interacts (i) antiferromagnetically ($J_A < 0$) with its nearest-neighbor Cu^{2+} ions, and (ii) ferromagnetically ($J > 0$) with an oxygen hole on the bond between adjacent Cu^{2+} atoms. The doping increases the concentration of holes, which are assumed to be annealed and randomly distributed. A decorated model is a spin system in which the spins at the extremities of one bond (“nodal” spins) interact with any “physical system”,^{26,27} located on the bond that connects them. By integrating out the degrees of freedom of the intermediate physical system, one generates an effective coupling K_{eff} between the nodal spins. In general, K_{eff} depends on the temperature and on the parameters which characterize the decorating physical system. Although the decorating system may be considered as general as one desires, most of the decorated models studied so far consider only Ising ± 1 nodal spins.²⁸ Here, for the n -vector version of the model we present an exact calculation of the effective coupling of a decorated bond. Moreover, for the $n = 1$ case (Ising model), we obtain *exactly* the magnetic phase diagram for a square lattice. In particular, we find a critical decorated bond concentration $p_c = 0.1464 \dots$ above which, the phase is unstable, i.e., ceasing to exist for small values of the ratio $|J_A/J|$.

We argue that this antiferromagnetic breakdown may account for the rapid downturn of T_C observed in $\text{La}_{2-x}(\text{Sr},\text{Ba})_x\text{CuO}_{4-y}$ well inside the tetragonal phase²² for large values of x as well as favors any pairing mechanisms^{1–8} which require an antiferromagnetic underlying

order. Moreover, our calculation also provides a possible interpretation for the low current density and the strong 2D antiferromagnetic fluctuations observed¹³ in these materials.

II. MODELS AND PHASE DIAGRAM

For the n -vector model, the decorated magnetic bond Hamiltonian is

$$H_b = -J\sigma \cdot (\mathbf{S}_1 + \mathbf{S}_2) - J_A \mathbf{S}_1 \cdot \mathbf{S}_2, \quad (1)$$

and the total Hamiltonian of the system is given by $H \equiv \sum_b H_b$. Making use of the decoration transformation²⁷ we write

$$F \exp(K_{\text{eff}} \mathbf{S}_1 \cdot \mathbf{S}_2) = \text{Tr} \{ \sigma \} \exp[K\sigma \cdot (\mathbf{S}_1 + \mathbf{S}_2) + K_A \mathbf{S}_1 \cdot \mathbf{S}_2], \quad (2)$$

where $K \equiv J/k_B T$, $K_A \equiv J_A/k_B T$, k_B is the Boltzmann constant, T the temperature, and F , a factor that contributes to the free energy of the system, plays an important role in the stabilization of the antiferromagnetic phase.

The trace in (2) is a constrained n -dimensional integral and can be performed as in Ref. 29, leading to

$$F \exp(K_{\text{eff}} \mathbf{S}_1 \cdot \mathbf{S}_2) = \frac{1}{2} (2\pi)^{n/2} I_{(n/2)-1}(u) u^{1-(n/2)} \times \exp(K_A \mathbf{S}_1 \cdot \mathbf{S}_2), \quad (3)$$

where $I_m(u)$ is the modified Bessel function of the first kind of order m , and

$$u \equiv K\sigma(S_1^2 + S_2^2 + 2\mathbf{S}_1 \cdot \mathbf{S}_2)^{1/2}, \quad (4)$$

where $\sigma = |\sigma|$, $S_1 = |\mathbf{S}_1|$, and $S_2 = |\mathbf{S}_2|$. We can take $S_1 = S_2 = \sigma = 1$ and \mathbf{S}_1 as the unit vector in the z direction. Hence $\mathbf{S}_1 \cdot \mathbf{S}_2 = \cos\theta$, where θ is the angle between \mathbf{S}_1 and \mathbf{S}_2 .

From Eqs. (3) and (4) we see that $F = F(\theta)$ and $K_{\text{eff}} = K_{\text{eff}}(\theta)$. Furthermore, from symmetry considerations, $F(\theta + \pi) = F(\theta)$ and $K_{\text{eff}}(\theta + \pi) = K_{\text{eff}}(\theta)$. Using these relations we obtain

$$F(\theta) = \frac{1}{2} (2\pi)^{n/2} \left\{ I_{(n/2)-1} \left[2K \cos \left(\frac{\theta}{2} \right) \right] I_{(n/2)-1} \left[2K \sin \left(\frac{\theta}{2} \right) \right] \left[\sin \left(\frac{\theta}{2} \right) \cos \left(\frac{\theta}{2} \right) \right]^{1-(n/2)} \right\}^{1/2} \quad (5a)$$

$$K_{\text{eff}}(\theta) = \frac{1}{2 \cos\theta} \ln \left\{ \frac{I_{(n/2)-1} \left[2K \cos \left(\frac{\theta}{2} \right) \right] \left[2K \cos \left(\frac{\theta}{2} \right) \right]^{1-(n/2)}}{I_{(n/2)-1} \left[2K \sin \left(\frac{\theta}{2} \right) \right] \left[2K \sin \left(\frac{\theta}{2} \right) \right]^{1-(n/2)}} \right\} + K_A. \quad (5b)$$

The above equations are the generalization of the decoration transformation introduced in Ref. 27. Table I shows

$$\tilde{K}_{\text{eff}}(\theta) \equiv K_{\text{eff}}(\theta) - K_A$$

for the special cases $n = 1-3$, where we use the relation³⁰

$$\lim_{u \rightarrow 0} I_{(n/2)-1}(u) u^{1-(n/2)} = 2^{1-(n/2)} / \Gamma(n/2).$$

In Fig. 1 we show the variation of $\tilde{K}_{\text{eff}}(\theta)$ with respect to the reduced temperature $k_B T/J$, for several different values of the configurational angle θ , for the cases $n = 1$ (Ising), $n = 2$ (XY), and $n = 3$ (Heisenberg). Note that for $n = 1$, \tilde{K}_{eff} is θ independent (see Table I). For the $n = 2$ and 3 cases, however, there exists a weak dependence of \tilde{K}_{eff} on the angle θ which is noticeable only at low temperatures, as one can see from Fig. 1 for curves labeled

TABLE I. The effective coupling $\tilde{K}_{\text{eff}}(\theta) \equiv K_{\text{eff}}(\theta) - K_A$ for decorated Ising ($n=1$), XY ($n=2$), and Heisenberg ($n=3$) models.

n	$\tilde{K}_{\text{eff}}(\theta)$
1	$\frac{1}{2} \ln[\cosh(2K)]$
2	$\frac{1}{2 \cos \theta} \ln \left[\frac{I_0(2K \cos \theta/2)}{I_0(2K \sin \theta/2)} \right]$
3	$\frac{1}{2 \cos \theta} \ln \left[\tan \left[\frac{\theta}{2} \right] \frac{\sinh(2K \cos \theta/2)}{\sinh(2K \sin \theta/2)} \right]$

$n=2$ and 3. Hence, we should note that the nature of variation of \tilde{K}_{eff} versus $k_B T/J$ for $n=1-3$ presents a common basic behavior, which is the fact that \tilde{K}_{eff} decreases when $k_B T/J$ increases, and the θ dependence of \tilde{K}_{eff} for the XY and Heisenberg models is negligible at high temperatures and is also weak in the low-temperature region.

In the lanthanum cuprates the spins are more likely to behave as Heisenberg quantum $S = \frac{1}{2}$ spins, although some weak Ising anisotropy has been observed.^{4,9} However, the lack of knowledge of the partition function for the square lattice for the XY or Heisenberg spins prevents us from using the results of the effective coupling for $n=2$ and 3, although some approximate procedure can be used.³¹ Hence, in this paper we restrict ourselves to the Ising model, for which we will obtain the *exact* solution. Furthermore, as we are interested in the metallic phase where the holes are mobile, it is more appropriate to consider the *annealed* rather than the *quenched* case.³²

To take into account the concentration dependence of the holes, we generalize (1) to

$$H_b = -J t_b \sigma(S_1 + S_2) - J_A S_1 S_2 - \mu t_b, \quad (6)$$

where $t_b = 1$ if bond b is decorated and $t_b = 0$ otherwise and μ is the associated chemical potential. Applying the

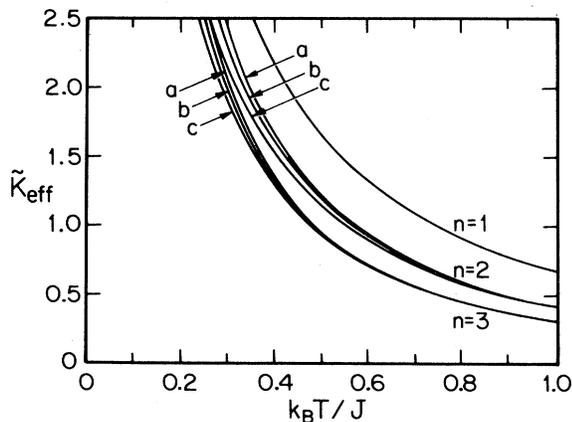


FIG. 1. The decorated effective coupling \tilde{K}_{eff} as a function of the reduced temperature $k_B T/J$ for the Ising ($n=1$), XY ($n=2$), and Heisenberg ($n=3$). The curves labeled (a), (b), and (c), are for $\theta=0$, $\theta=\pi/6$, and $\theta=2\pi/3$, respectively.

decoration transformation²⁷ we get for the Ising case

$$K_{\text{eff}}(K, K_A, \eta) = K_A + \frac{1}{2} \ln \left[\frac{1 + 2\eta \cosh(2K)}{1 + 2\eta} \right], \quad (7a)$$

$$F(K, \eta) = \{(1 + 2\eta)[1 + 2\eta \cosh(2K)]\}^{1/2}, \quad (7b)$$

where $\eta \equiv \exp(\mu/k_B T)$ is the fugacity.

For the square lattice, the partition function is $Z = F^N Z_s(K_{\text{eff}})$, where $Z_s(K_{\text{eff}})$ is the Onsager³³ partition function with effective coupling $J_{\text{eff}} \equiv k_B T K_{\text{eff}}$ and N is the total number of bonds. We can eliminate the fugacity in (7) in favor of

$$p \equiv \langle t_b \rangle = \eta \left[\frac{\partial}{\partial \eta} \ln F(K, \eta) + \epsilon \frac{\partial K_{\text{eff}}(K, K_A, \eta)}{\partial \eta} \right], \quad (8a)$$

where p is the concentration of decorated bonds and

$$\epsilon \equiv \langle S_1 S_2 \rangle = \lim_{N \rightarrow \infty} \frac{1}{N} \frac{\partial}{\partial K_{\text{eff}}} \ln Z_s(K_{\text{eff}}), \quad (8b)$$

is the nearest-neighbor pair correlation function.

Using the Onsager partition function, (8) yields

$$\eta = [A + (A^2 + 2pB)^{1/2}] / B, \quad (9)$$

where

$$A \equiv \cosh(2K)(2p - 1 - \epsilon) + 2p - 1 + \epsilon,$$

and

$$B \equiv 8(1-p) \cosh(2K).$$

Substituting (9) into (7) gives $K_{\text{eff}}(K, K_A, p)$ in terms of the concentration p .

The Onsager³³ solution, $\sinh(2K_{\text{eff}}) = -1$, can also be used to obtain the phase boundary,

$$J_A/J = \frac{1}{2K} \ln \left[\frac{(1+2\eta)(\sqrt{2}-1)}{1+2\eta \cosh(2K)} \right], \quad (10)$$

with η given by (9). Here ϵ is set equal to its critical value,²⁷ $\epsilon_c = -\sqrt{2}/2$. In Fig. 2 we show the phase diagram whose transition lines are obtained from Eq. (10) for two particular cases, $p=0.14$ and 0.15 .

III. DISCUSSION AND CONCLUSION

Here we discuss the magnetic phase diagram for the Ising model and construct a hypothetical correlation between its features and the superconducting properties of the CuO_2 planes in the lanthanum cuprates. Although for these materials the spins are more likely to be Heisenberg quantum spins, we hope that the qualitative physics still holds.

Figure 2 clearly reveals the existence of two distinct regimes for the phase diagram $k_B T_c/J$ versus $|J_A/J|$. Below a critical concentration³⁴

$$p_c = \frac{1}{2}(1 - \sqrt{2}/2) \approx 0.1464. \dots$$

the antiferromagnetic phase is stable for any value of $|J_A/J|$ in the low-temperature region, while for $p > p_c$ the antiferromagnetic ground state is stable only for

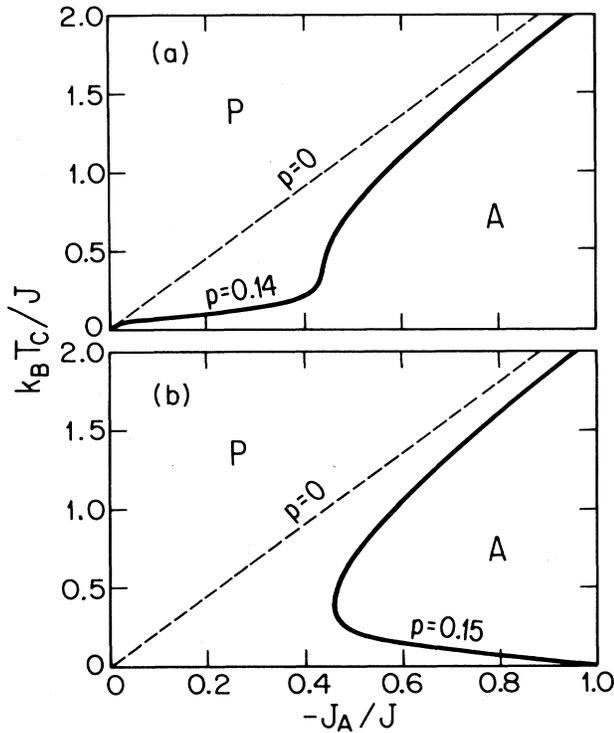


FIG. 2. The exact phase diagram of the 2D decorated Ising model for different values of the concentration p of decorated bonds. The transition line for (a) $p=0.14$ ($p < p_c$) and (b) $p=0.15$ ($p > p_c$) illustrates the drastic transition from one regime to another. Here A and P denote the antiferromagnetic and the paramagnetic regions, respectively, and the exact frontier between A and P for $p=0$ is also shown.

$|J_A/J| \geq 1$. Since the antiferromagnetic order is essential for most of the recently proposed magnetic-pairing mechanisms, the drastic transition from antiferromagnetic to paramagnetic phase, observed in Figs. 2(a) and 2(b) for small values of $|J_A/J|$, can well account for the rapid drop of $T_c(x)$ as one increases the doping concentration x beyond x_{\max} .

We note that our concentration p of decorated bonds is a number that varies between 0 and 1, while the real concentration x of doping Sr or Ba is a number between 0 and 2. As has been shown,²⁰ below $x=0.15$ there is a one-to-one correspondence between the density of holes and x , so we assume $x=2p$. If this relation holds for all values of p it will lead to $x_{\max} \sim 0.2928$, above which the superconducting state, if supported by antiferromagnetic based pairing mechanism, should disappear. However, as in the regime²⁰ $x \geq 0.15$ some of the holes may be associated with ions of Cu^{3+} or even trapped by the dopant atoms, one expects $x_{\max} \geq 0.2928$, which seems to be in good agreement with the recent experimental observations due to Torrance *et al.*²² of $x_{\max} \approx 0.32$. It is believed for the $\text{La}_{2-x}(\text{Sr,Ba})_x\text{CuO}_{4-y}$ compounds that $|J_A/J| < 0.45$; e.g., the *ab initio* calculations in Ref. 19 suggest $|J_A/J| \approx 0.36$. Then from the strong dependence of the transition lines on p we can understand *on purely*

statistical mechanical grounds why the antiferromagnetic 2D order (and, as a consequence, the superconducting state) is so sensitive to the doping concentration.

Also, the reentrant antiferromagnetic phase which we find between $|J_A/J| \approx 0.45$ and $|J_A/J|=1$, for $p > p_c$, can be understood. One must consider that the interaction Hamiltonian of the decorating spin with the nodal spins is of the form $-J\sigma(S_1+S_2)$. As p increases, at low temperatures the antiferromagnetic state will become unstable due to the “dilution” of the effective ferromagnetic coupling induced on the decorated bonds. As this concentration is still too small to support a ferromagnetic long-range order, the system will undergo a phase transition to a paramagnetic state in order to minimize its free energy. However, as the temperature increases, the system will prefer to recover its antiferromagnetic order, since this state will satisfy the direct antiferromagnetic interaction $-J_A S_1 S_2$ between the nodal spins and will free the decorating spin to make a large contribution to the entropy of the system, minimizing this way its free energy.

The existence of the reentrant antiferromagnetic phase may help in finding higher- T_c superconductors, if it is possible to find compounds in which the competing exchange ratio $|J_A/J|$ is *above* 0.45, since in that region of the phase diagram [see Fig. 2(b)] the antiferromagnetic order survives for concentrations above p_c . We also predict that in this $|J_A/J|$ regime one should expect also a reentrant superconducting state, since the antiferromagnetic order disappears as the temperature is lowered. This idea is supported by the fact that an apparent reentry behavior has experimentally been observed³⁵ in a high- T_c superconductor $\text{YBa}_2\text{Cu}_3\text{O}_7$ at low temperatures. The nature of this predicted reentrant superconducting state should not be the same as the states observed in superconductors doped with ferromagnetic impurities, for in that case the disappearance of the superconducting state is driven by the *onset of ferromagnetic order* while in our case it should occur with the *breakdown of antiferromagnetic order*.

Before concluding, we note some parallels between our model and (a) current ideas on the granular nature of the high- T_c superconductors and (b) the strong 2D antiferromagnetic fluctuations observed in these materials.¹³ For the 2D antiferromagnetic order in these grains, the *local* concentrations of decorated bonds in different domains are different. Some domains will have p closer to p_c than others. The migration of holes through the weak links from one domain to another may increase the local concentrations of some of the domains above p_c and this will destroy the local antiferromagnetic order and consequently the superconducting state. The holes will then migrate to other superconducting regions, lowering again the local concentration of the region left and thus giving way to a reconstruction of the antiferromagnetic order. The same effect will now occur in the destination domains, some of which will also undergo a superconducting to normal state transition. This picture is consistent with the strong 2D antiferromagnetic fluctuations observed¹³ in the $\text{La}_{2-x}(\text{Sr,Ba})_x\text{CuO}_{4-y}$. If one could

find compounds with $|J_A/J|$ above 0.45, we predict that the antiferromagnetic fluctuations should be reduced. Hence, the critical current should also increase since the underlying antiferromagnetic order is much less sensitive to the hole concentration.

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