Magnetic phases in Ising square lattices with mixed bonds

Eugenio E. Vogel*
Centro Atómico Bariloche, 8400 San Carlos de Bariloche, Provincia Río Negro, Argentina

Patricio Vargas
Departamento de Física, Universidad de Santiago de Chile, Santiago, Chile

Graciela Pérez
Departamento de Ciencias Físicas, Universidad de La Frontera, Temuco, Chile
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CuO_2 planes are modeled by means of a square lattice with antiferromagnetic coupling provided by the oxygen between two copper ions and ferromagnetic coupling when the oxygen is absent. The magnetic interaction is described using an Ising Hamiltonian with interacting first-nearest neighbors. The ferromagnetic exchange interaction is supposed to be larger than the antiferromagnetic one. A Monte Carlo routine is then defined in order to minimize the energy and to calculate physical parameters such as correlation to nearest neighbors, time correlation to the same site, and a schematic of a phase diagram.

I. INTRODUCTION

The recent progress on the superconductivity of layered perovskite-structure compounds has led to the study of other related properties, such as the magnetic phases that can arise in these materials. It has been proposed[1] that compounds such as $La_{2-x}(Ba,Sr)_xCuO_{4-y}$ may have mixed ferromagnetic and antiferromagnetic bonds which can be responsible for a spin-glass phase which appears as the oxygen content diminishes.

As a first attempt to model this behavior, we studied the CuO_2 planes under the assumption that the oxygen between the two magnetic Cu ions provides an antiferromagnetic bond, while its absence provides a ferromagnetic bond. We then modeled the CuO_2 planes in the manner shown in Fig. 1, where only spin-up and spin-down states are pictured in accordance with the Ising Hamiltonian to be introduced later on. If we neglect the small differences between the orthorhombic and tetragonal phases,[2] we can think of this system as a square lattice with magnetic ions at the corners and mixed bonds along the sides in a proportion which is related to the oxygen content.

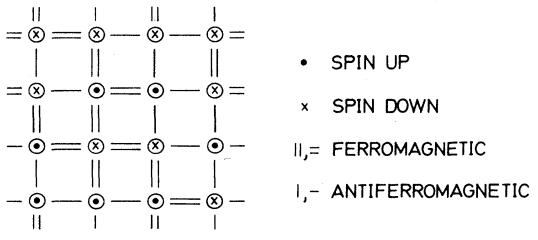

FIG. 1. Schematic of a 4×4 square lattice with mixed bonds for an arbitrary state. Periodic boundary conditions on the exchange constants are clearly shown.

Systems similar to the one just described have been considered.[3-5] Special attention has been given to the system for which the number of ferromagnetic bonds equals the number of antiferromagnetic ones, and the magnitude of the coupling is the same for both of them.[6] A result of these studies is that no stable magnetic phase (such as spin glass) is obtained at finite temperatures where only metastable states are possible.[7] However, as the temperature approaches 0.0, the relaxation times required to change to other metastable states grow beyond any practical importance (100 yr or more).[8] If we consider the critical temperature for superconductivity in these compounds (40 K) and the estimated magnitude of the antiferromagnetic exchange constant A (1300 K),[1] the relative temperature T/A of interest to us is restricted to the interval [0,0.03], where Monte Carlo simulations are reliable in the sense that finite observational times are acceptable for practical purposes.

II. MODEL AND CALCULATIONS

We restrict ourselves to the possible magnetic phases present in one single CuO_2 layer. The Ising Hamiltonian up to the nearest-neighbor interaction can be written as

$$H=\tfrac{1}{2}\sum_i S_i^z\left[\sum_j J_{ij}S_j^z\right], \tag{1}$$

where we sum over the four nearest neighbors to the ith spin, which runs over the $N=L^2$ spins of the square lattice with L spins per side. The exchange constants J_{ij} can be either A or $-F$ depending on whether the spins are coupled antiferromagnetically or ferromagnetically. The real physical system loses the antiferromagnetic phase at low concentrations of ferromagnetic bonds which suggests ferromagnetic coupling stronger than the antiferromagnetic one. As an example of this unequal

coupling the case $F=3A$ will be studied particularly. For the sake of comparison, the well-known case $F=A$ will be also included.

Periodic boundary conditions are imposed on the bonds at the edges, so that each spin is always surrounded by four nearest neighbors. One important variable is the concentration of ferromagnetic bonds (c) and the complementary concentration of antiferromagnetic bonds ($1-c$). Moreover, for each given concentration there are many distributions of bonds in the lattice. In order to avoid giving excessive weight to some very unlikely distribution of bonds (such as clusters of the same kind of bonds) we performed some basic statistical analysis on several distributions consistent with a given concentration. The number of distributions varied from 50 when the method was being tested to 5 for calculations where the dispersion of the whole data is also a good test for the stability of the solution.

The distribution of bonds is randomly assigned at the beginning of each calculation and is kept fixed for the rest of it. Namely, no migration of the bonds is allowed. However, the several initiations already described will allow us to obtain a kind of average description for a given concentration of bonds.

Next, the initial state is randomly chosen by simply assigning values to the third component of spin ($+1$ or -1) to each of the Cu sites of the lattice. The energy is then minimized by means of a Monte Carlo algorithm based on the flip of a single spin.

There are at least two possibilities for choosing the next site for flipping its spin: sequential order through the lattice or random choice. Both possibilities are appropriate when just one kind of bond is present. However, the random choice could be advantageous in the case of mixed bonds, since the sequential order would always run through the same circuit of bonds. Most of the results presented here were based on the random generation of next site. As a consequence of this, "time" will be measured in units of "spin flips" (SF's).

Temperature T enters by means of the Metropolis algorithm, which essentially compares the probability factor $\exp(-\Delta/kT)$ with a random number between 0.0 and 1.0. Δ represents here the energy difference of the new state with respect to the state prior to the spin flip. If Δ is negative, the probability factor is taken to be 1.0. It is convenient to measure T in units of the smallest between the two exchange constants (in terms of A for the purposes of this paper).

One major concern in any thermalization process is the role of the randomly picked initial states in the rest of the calculations. In order to minimize this effect, we allow the system to fluctuate in a wider band of states before beginning the actual minimization of energy. This is simply achieved by increasing the temperature of the system to $T'>(T+A)$ from which a slow cooling to the actual temperature T is performed. In all the results reported here $T'=T+1.5$ A, and the system was gradually brought back to temperature T after 10 000 spin flips (SF's). In any case this mechanism does not lead always to the same "valley" in the Hilbert space and it is only intended to avoid casual shallow "valleys" due to a trapped initial state.

The concern now is with respect to the number of SF's required in order to reach thermalization. Some basic analysis of this point is performed for the delicate case $c=0.5$, $F=A$ at low temperatures. In most of the results reported below 50 000 SF's were used to reach thermalization, otherwise available data for 60 000 SF's were also included.

The number of spins in the lattice was also investigated within ranges of interest for the computing facilities available (several personal computers). No noticeable differences were found between lattices 20×20 and 32×32 when periodic boundary conditions as already defined are used. This is the reason why we restrict ourselves from now on to lattices with $L=20$, namely, 400 spins and 800 bonds.

Once the thermal equilibrium is reached, parameters of physical interest can be calculated. One of them is the correlation to the νth nearest neighbor $C(\nu)$ which can be defined as

$$C(\nu)=\left\langle S_i^z \sum_j^{n_\nu} S_j^z/n_\nu \right\rangle , \tag{2}$$

where n_ν indicates the number of neighbors of order ν to a given site i, the sum over j runs over all those neighbors and the angular brackets represent an average with respect to all sites i in the lattice. We will be particularly interested in the correlation to next-nearest neighbors that in our case can be calculated by means of the simplified relationship

$$C(1)=\left\langle S_i \sum_j^4 S_j/4 \right\rangle . \tag{3}$$

If the spins are normalized to unity, $C(1)$ is restricted to the range $[-1,+1]$.

The other parameter of physical importance is the time correlation at a given site in accordance with the q parameter defined by Edward and Anderson[9] in the following way:

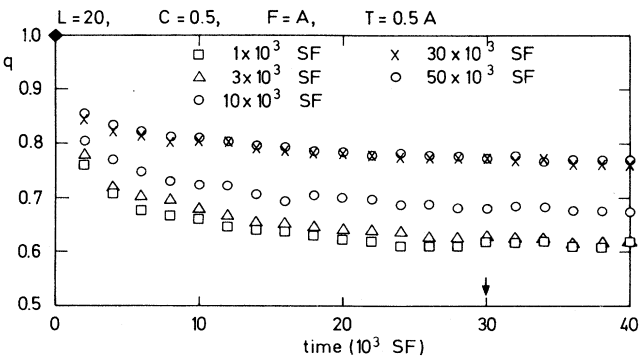

FIG. 2. Time correlation q for different thermalization times, for equal amount of both kind of bonds at a temperature half the strength of the magnitude of the exchange constants.

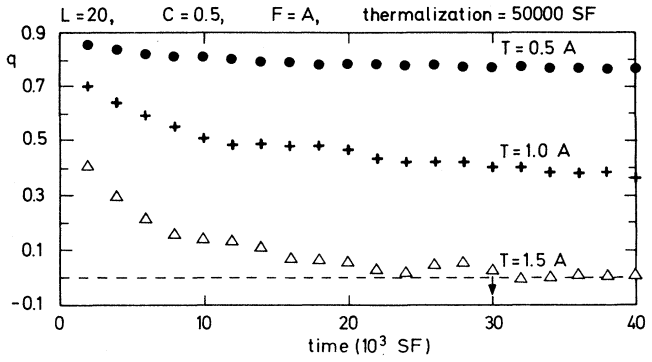

FIG. 3. Using 50 000 SF's for thermalization time the minimum time required for evaluating $q(\infty)$ is shown for three different temperatures $0.5A$, A, and $1.5A$. For the low temperatures used in most of the remaining discussions 30 000 SF's leads to acceptable stability for the parameter q.

$$q(t)=\langle S_i(t)S_j(0)\rangle \ . \tag{4}$$

The condition $t=0$ is defined at any time once the thermal equilibrium is reached; t is the number of spin flips after $t=0$ for which the calculation is performed. The angular brackets represent average over the lattice. The real interest is over $q(\infty)$, defined in the limit of SF's going to infinity (after $t=0$). For practical reasons we must restrict ourselves to finite times but large enough to give stable values for q. For the $L=20$ lattice we use an average of a few values taken around 30 000 SF's after the initial time. The stability of these results will be discussed below.

One of the importances of the parameter q is that it allows us to define the spin-glass phase under the conditions $C(\nu)=0$ and $q(\infty)\neq 0$. Since $|C(1)|$ is the largest among the correlations, it is enough to consider $C(1)$ in the way already given by Eq. (4). We are interested in investigating the possibility of a spin-glass phase, so most of the results presented here are related to the q parameter. We vary the concentration c of ferromagnetic bonds, the relative magnitudes of the exchange constants F/A and the temperature T of the system aiming to a phase diagram. We fix the parameters of thermalization, and the size L of the lattice in the way already described. The particular case $c=0.5$ was also studied as a reference for other less sensitive cases provided by unequal coupling.

III. RESULTS AND DISCUSSION

Let us first discuss the details of the numerical calculation employed. We have chosen the very sensitive case $c=0.5$ and $F=A$ in order to select the appropriate times. Actually it is believed that no true phase transition occurs except in the limit of very low temperatures (essentially in the limit of T going to 0).[7] As already pointed out, the temperatures of interest in our case are actually very low. On the other hand we have already mentioned the need to consider $F>A$ which stabilizes the magnetic lattice. This can be seen by simply realizing that less frustration is likely to occur when there are dominant links.[9] Therefore, any reasonable thermalization process acceptable at low temperatures for equal coupling in the $c=0.5$ case will be more advantageous for unequal coupling.

In Fig. 2 we report the variation of the q value at low temperature ($T=0.5$ A) for five different thermalization times: 1000, 3000, 10 000, 30 000, and 50 000 SF's. The statistics are based on 30 experiments. It is quite clear that the difference between the last two cases is quite small.

The finite observational time required in order to get

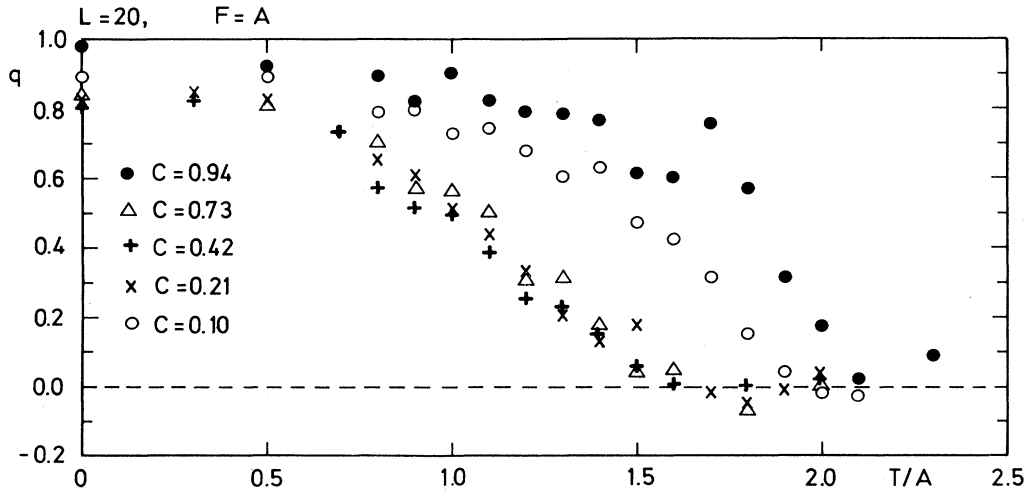

FIG. 4. Evaluation of $q(\infty)$ as a function of temperature for equal strength of ferromagnetic and antiferromagnetic bonds and five different concentrations of ferromagnetic bonds.

stable values of $q(t)$ which can be taken as $q(\infty)$ is investigated in Fig. 3. We adopt here the criterion that q can be evaluated 30 000 SF's after thermalization. As we do not attempt here a very precise calculation at intermediate temperatures or around T_c we shall restrict ourselves to evaluate q 30 000 SF's after thermalization.

In Fig. 4 we present the variation of the q value as a function of temperature for five different concentrations of ferromagnetic bonds, for the case of equal coupling $F = A$. It is clear that the value of q is more stable when any of the exchange mechanisms dominates. The critical temperature (if any) increases when c departs from the value 0.5 in any sense.

Another way of looking at these systems is presented in Fig. 5, where the variation of both correlation to nearest neighbors $C(1)$ and correlation to the same site q are calculated at very low temperature for the condition $F = A$. The spin-glass condition is confined to $c \approx 0.5$ which is a fairly large concentration of ferromagnetic bonds (absence of oxygen in the real system). Our results for this two-dimensional Ising lattice agree with several other works with respect to the q values and energy of the final state.[7–13]

This kind of calculation allows us to schematically draw a phase diagram like the one shown in Fig. 6. The main purpose of this scheme is to show that the conditions for the onset of a spin-glass phase are not present at low values of the concentration c. At least one other condition must be met in order to realize such a system. It is obvious that we must allow $F > A$. There is no physical reason for the magnitude of the exchange constant being the same after the sign has been reversed due to the lack of oxygen. On the other hand, it is natural to assume $c < 0.5$ since the system is antiferromagnetic when all the oxygen bonds are saturated. Fewer ferromagnetic bonds will require stronger coupling in order to produce conditions for spin-glass phase. Most of the rest of the present work will consider the case $F = 3A$ as an illustrative example.

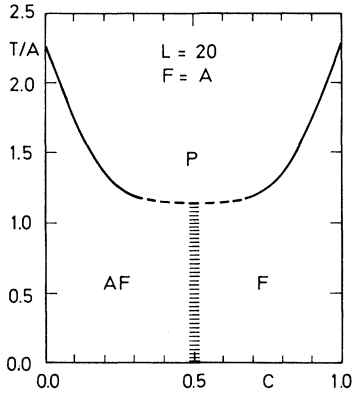

FIG. 6. Schematic magnetic phase diagram for temperature T/A and concentration of ferromagnetic bonds c as free parameters. Equal coupling $F = A$ is assumed here.

Let us first look at the correlations at low temperatures for the system $F = 3A$, as shown in Fig. 7. The conditions for a spin-glass phase are present for $c \approx 0.33$, namely, when one third of the bonds are ferromagnetic while the remaining two thirds are antiferromagnetic. A comparison with Fig. 5 indicates that the value of g is higher for unequal coupling which is an indication for more stability of a spin-glass phase.

Next we want to examine briefly the general behavior of q as a function of temperature for this particular system. We would like to emphasize that we do not attempt

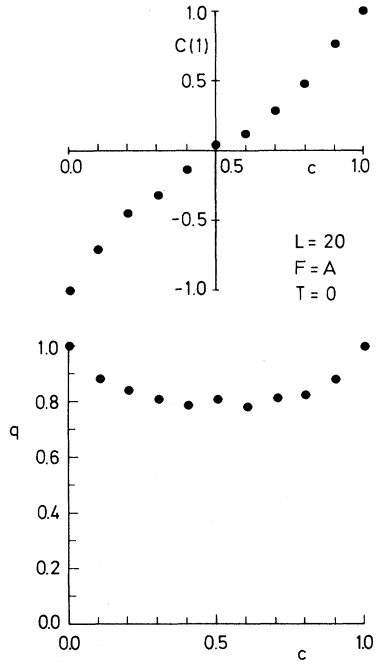

FIG. 5. Dependence of both correlation to first-nearest neighbor $C(1)$ and time correlation to the site q with respect to the concentration of ferromagnetic bonds c. Equal strength of the bonds is assumed here while temperature is zero. (Actually $T = 0.01$ A.)

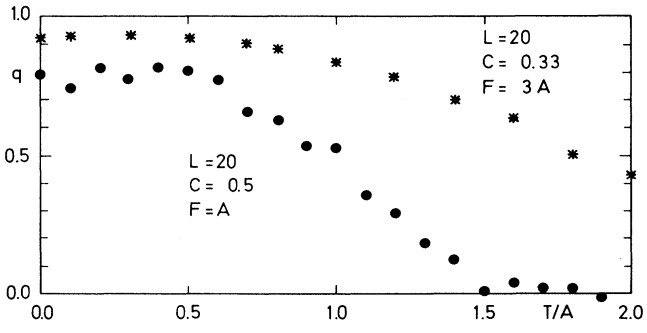

FIG. 7. Dependence of both correlation to first-nearest neighbor $C(1)$ and time correlation q with respect to the concentration of ferromagnetic bonds c. Ferromagnetic bonds are taken three times stronger than antiferromagnetic ones ($F = 3A$). Temperature is zero.

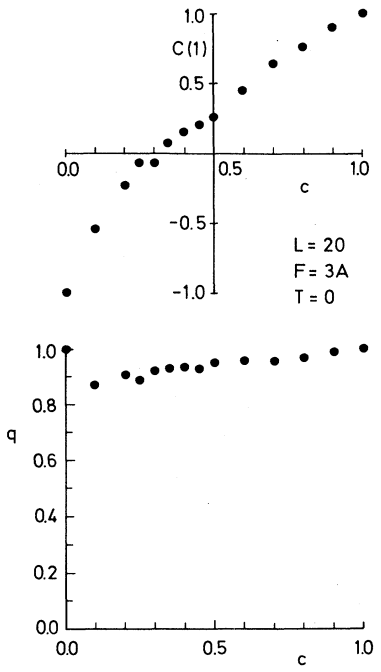

FIG. 8. Correlation to the site q as a function of temperature for two different Ising lattices with mixed exchange interactions. (Thermalization times used in the Monte Carlo processes are not enough to draw conclusions about freezing temperatures.) The parameter q is higher and more stable for the case of unequal coupling, particularly at the low temperatures discussed in the text.

here a final description of optimal Monte Carlo routines to describe a possible freezing transition, due mainly to limitations on the computer facilities available. However, Fig. 8 shows clearly that $q(T)$ remains high and stable over a wide range of temperatures. For the purposes of comparison we have also included in Fig. 8 the case of equal coupling using the same thermalization process.

Finally, let us mention a few words about the minimum energy found after the Monte Carlo process. For the case $F=3A$ under consideration we found energies as low as -1.28 A per bond for the ground or metastable state after thermalization at low temperatures. A theoretical expression based on a straightforward way of considering the frustrations of this system would lead to[10] $E_0=-1.42A$ as the average lower bound for ground energy per bond. This result compares well with the case of equal coupling where Monte Carlo simulations give $-0.70A$ for the ground energy per bond,[10-13] while the lower bound expression gives $-0.75A$.

IV. CONCLUSIONS

The admixture of ferromagnetic and antiferromagnetic bonds, with unequal strength and different complementary concentrations in an Ising square lattice defines a very interesting system which can be associated to some real systems such as the CuO_2 planes which are present in La_2CuO_4 and its related compounds.

Unequal strength of exchange constants moves the spin-glass condition to different concentrations of ferromagnetic bonds. Thus if $F>A$, then $C(1)=0.0$ for $c<0.5$ and vice versa.

The particular case of $F=3A$ leads to $c\approx 0.33$ in order to eliminate correlations to nearest neighbors while the correlation to the same site q in nonzero. There is a clear indication that the value of q is higher and more stable with respect to temperature as compared with the well-known case of equal coupling. A spin-glass phase could eventually be stabilized by the presence of interplanar interaction (three-dimensional Ising lattice) as well as by unequal coupling to ferromagnetic and antiferromagnetic states.

In an Ising square lattice the spin-glass phase is restricted to an interphase between antiferromagnetism and ferromagnetism. However, the present work is the basis for considering interplanar interactions which can lead to a three-dimensional Ising lattice where an actual spin-glass phase could be possible. This is beyond the aim of the present article and will be considered later on.

Other extensions of the present work are the possibility of pairing of ferromagnetic bonds in accordance with some existing proposals of pairing of holes at the Cu sites which would be responsible for the superconductivity in these kinds of compounds.[14] This is to say that the localization of the holes due to the oxygen vacancies will provide the ferromagnetic exchange interaction. The action of external fields would be also interesting. It would be worthwhile to study the thermalization of system of mixed bonds with unequal both exchange constants and concentrations since most of the work done so far deals with the equal coupling and equal concentrations of the two kinds of bonds.

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