Diluted and random-bond Ising model for the Fe-Al disordered alloys

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A quenched disordered Ising model with a distribution function that includes dilution and random bonds is studied through the mean-field renormalization-group approach. Random bonds are considered through a ferromagnetic exchange interaction between nearest-neighbor spins (NN) and an antiferromagnetic exchange interaction between next-nearest-neighbor spins (NNN). The phase diagrams are obtained and the results are applied to Fe-Al disordered alloys.

I. INTRODUCTION

The magnetic and structural properties of Fe-Al alloys have been experimentally studied by several authors.[1-8] These works have shown that the bcc structure of these alloys is ferromagnetic and disordered up to 20 at. % Al in iron, independent of the heat treatment. When the alloys are fast quenched from 700° C the nonstoichiometric FeAl structure type is obtained from 20 to 50 at. % Al; however, if they are very slowly cooled the nonstoichiometric Fe_3Al structure type is obtained from 20 to 33 at. % Al and the nonstoichiometric FeAl structure type is obtained from 33 to 50 at. % Al. The Fe_3Al and FeAl structure types can be considered as two interlocking simple cubic sublattices, in a way that for the FeAl structure one sublattice is occupied by iron atoms (Fe site) and the other by aluminum atoms (Al site), and for the Fe_3Al structure the aluminum site is occupied by four Fe atoms and four Al atoms.

The experimental values of the average magnetic moment $\bar{\mu}$, extrapolated to 0 K, as a function of Al concentration q follows a simple dilution model until 20 at. % Al. For larger concentrations $\bar{\mu}$ shows a rapid decrease in the range $0.25 < q < 0.30$, followed by a slower decrease to zero at $q=0.40$. The same anomalous behavior is shown for $\bar{\mu}$ as a function of the number of Al NN as well as for the effective hyperfine field with the Al concentration and the NN number.

To explain this anomalous behavior Arrot and Sato[2] proposed a ferromagnetic-antiferromagnetic transition near the critical concentration. This hypothesis has not been confirmed by the neutron scattering experiments of Pickart and Nathans.[9] Vincze[5] and Srinivasan *et al.*[10] interpreted this anomaly by considering that the Fe magnetic moment decreases with the increase of Al NN. Huffman[6] proposed a ferromagnetic-mictomagnetic transition when T decreases, hypothesis experimentally confirmed by Shull, Okamoto and Beck.[11] Shiga and Nakamura[7] proposed a spin glass as the low-temperature phase for these ordered alloys near the critical concentration.

Some theoretical studies have been done in order to interpret the magnetic behavior of these ordered alloys.[5,10-14] Shukla and Wortis[13] using the model proposed by Sato and Arrot[12] and postulating a spin-glass behavior that arises by virtue of competition between a nearest-neighbor (NN) Fe-Fe ferromagnetic exchange J and next-nearest-neighbor (NNN) antiferromagnetic superexchange given by $-\alpha J$, obtained an acceptable fit to the experimental phase diagram. They used the decimation technique proposed by Kadanoff and Houghton.[15] Grest[14] assuming the previous model and calculating by Monte Carlo method, has studied the transition from ferromagnetic to spin glass in these alloys as a function of q. Some differences between this calculation and the experimental results were obtained.

More recently Pérez Alcazar and Galvão da Silva[16] have reported an experimental study of Fe-Al alloys by Mössbauer spectroscopy at room temperature (RT). By an adequate heat treatment they obtained Fe-Al disordered alloys in the range $0<q<0.5$. At RT these alloys are all ferromagnetic and do not show the anomalous behavior of the ordered ones. Quite good fits of these experimental datas have also been obtained by using a simple site diluted Ising model.[17]

The mean-field renormalization-group method (MFRG) was proposed by Indekeu, Maritan, and Stella[18] for calculating critical properties of lattice spin systems. Droz, Maritan, and Stella[19] applied the MFRG method to the study of dilute, random fields and the symmetric random-bond Ising model. The asymmetric random-bond Ising model has been treated by Lyra and Coutinho[20] using a fenomenological competition parameter α. They obtained symmetric phase diagrams for $\alpha=1$ and asymmetric ones with reentrancies in the ferromagnetic or antiferromagnetic boundaries depending on the α value. Recently, Rosales Rivera, Pérez Alcazar, and Plascak[21] have studied a diluted and random-bond Ising model with a probability distribution for the coupling constant between the pair of nearest-neighbor spins which includes the concentrations p, x and q for the ferromagnetic, antiferromagnetic and diluted bonds, respectively. For $q=0$, the results are identical to those obtained by Lyra and Coutinho[20] in the Ising limit and for $x=0$ the equations for the diluted model studied by Droz, Maritan, and Stella[19] are recovered.

In this work we report the theoretical phase diagrams, calculated by the MFRG method, for a type of diluted and random-bond Ising model in which the ferromagnetic and antiferromagnetic bonds are taking in the NN and

NNN interactions, respectively. Then we discuss the application of this theoretical model to the binary Fe-Al disordered system.

II. MODEL SYSTEM

In order to obtain the phase diagrams for this quenched disordered model, we assume a diluted and random-bond Ising spin model with NN and NNN interactions. The Hamiltonian for this model can be given by

$$H=-\sum_{\langle ij\rangle}K_{ij}\sigma_i\sigma_j-\sum_{(i,j)}K_{ij}^{*}\sigma_i\sigma_j\ , \tag{1}$$

where $K_{ij}=J_{ij}/k_BT$ and $K_{ij}^{*}=J_{ij}^{*}/k_BT$ are the reduced coupling constants between NN spins $\langle ij\rangle$ and NNN spins (i,j), respectively, and $\sigma_i=\pm1$. For the diluted and random bond we use, the probability distributions

$$P(K_{ij})=p\delta(K_{ij}-K)+q\delta(K_{ij})\ , \tag{2}$$

and

$$P(K_{ij}^{*})=p\delta(K_{ij}^{*}+\alpha K)+q\delta(K_{ij}^{*}) \tag{3}$$

for the reduced NN coupling and the reduced NNN coupling, respectively. Here p is the ferromagnetic or antiferromagnetic bond concentration and $q=1-p$ is the diluted bond concentration. α is the fenomenological competition parameter.

III. PHASE BOUNDARY EQUATIONS

In order to obtain the phase boundary equations and the phase diagrams, we used the MFRG,[18] by considering the one- and two-particle clusters. In the MFRG the interaction within the clusters are treated exactly and that of the neighboring spins are treated by a mean field acting on the spins of the cluster boundary. In this way the Hamiltonian for the one- and two-spins clusters are given, respectively, by

$$H_1=-\sum_{i=1}^{z}b_i'K_{1i}'\sigma_1-\sum_{j=1}^{s}b_j'K_{1j}^{*\prime}\sigma_1\ , \tag{4}$$

$$H_2=-K_{12}\sigma_1\sigma_2-\sum_{i=1}^{z-1}K_{1i}b_{1i}\sigma_1-\sum_{j=1}^{z-1}K_{2j}b_{2j}\sigma_2 -\sum_{i=1}^{s}K_{1i}^{*}b_{1i}\sigma_1-\sum_{j=1}^{s}K_{2j}^{*}b_{2j}\sigma_2\ , \tag{5}$$

where z and s are the NN and NNN numbers, respectively, and b_j and b_{mj} $(m=1,2)$ are the effective fields acting on the respective boundary cluster. The magnetization per spin of the one- and two-spin clusters for small effective fields are given, respectively, by

$$\langle\sigma_1\rangle=\sum_{i=1}^{z}K_{1i}'b_i'+\sum_{j=1}^{s}K_{1j}^{*\prime}b_j'\ , \tag{6}$$

$$\langle\sigma_2\rangle=\sum_{i=1}^{z-1}K_{2i}b_{2i}+\sum_{i=1}^{s}K_{2i}^{*}b_{2i}+\left[\sum_{j=1}^{z-1}K_{1j}b_{1j}+\sum_{j=1}^{s}K_{1j}^{*}b_{1j}\right]\tanh K_{12}\ . \tag{7}$$

To perform the configurational average $\overline{\langle\cdots\cdots\rangle}$ we assume that the K_i and b_i are independent random variables with symmetry conditions for the effective magnetization and the staggered magnetization given by: $\bar{b}_i'=b';\bar{b}_{1j}=b$ and $\bar{b}_{2j}=\pm b$, where the sign $+(-)$ holds for the ferromagnetic (antiferromagnetic) order parameter. We then get

$$\overline{\langle\sigma_1\rangle}=p'K'b'(z-\alpha's)\ , \tag{8}$$

$$\overline{\langle\sigma_2\rangle}=pKb(z-1-\alpha s)(1\pm p\tanh K)\ . \tag{9}$$

To obtain the spin-glass order parameter we use the relations proposed by Lyra and Coutinho:[20] $\overline{b_i'}=0,\overline{b_i'b_j'}=h'\delta_{ij},\overline{b_{mj}}=0,\overline{b_{mi}b_{lj}}=h\delta_{ml}\delta_{ij}$. With these conditions the Edwards-Anderson spin-glass order parameter[22] for each cluster can be obtained by squaring Eqs. (6) and (7), respectively, and performing the configurational average; that is

$$\overline{\langle\sigma_1\rangle^2}=h'K'^2p'(z+\alpha'^2s)\ , \tag{10}$$

$$\overline{\langle\sigma_2\rangle^2}=hpK^2[(z-1)+\alpha^2s][p\tanh^2K+1]\ . \tag{11}$$

In order to obtain the equations of the critical lines we used the recursion relations between the order parameter according with the standard MFRG. But the complete renormalization flow in the (K,p,q,α) space cannot be fully determined by the equations for $\overline{\langle\sigma\rangle}$ and $\overline{\langle\sigma\rangle^2}$. Then we studied the fixed point solutions $K'=K=Kc$ in the subspace $\alpha=\alpha'$ and $p=p'$. We then get

$$(z-\alpha s)/(z-1-\alpha s)=1\pm p\tanh Kc\ , \tag{12}$$

which gives the ferromagnetic (upper sign) and the antiferromagnetic (lower sign) phase boundaries and

$$1/(z-1+\alpha^2s)=p\tanh^2Kc\ , \tag{13}$$

for the spin-glass boundary. For $\alpha=0$, Eq. (12) is identical to that obtained by Droz, Maritan, and Stella[19] for the diluted model.

IV. THEORETICAL PHASE DIAGRAMS AND DISCUSSION

Figure 1 shows the phase diagrams in the $Tc(p)/\tau c(1)$ against concentration p, for $z=8$, $s=6$ and two typical competition parameters $\alpha=0.2$ and $\alpha=0.5$. Where we have normalized the temperatures with the critical temperature $\tau c(1)$ obtained for $\alpha=0$ (diluted model). These NN and NNN values correspond to a bcc lattice. From this figure it can be noted that with these parameters the ferromagnetic, paramagnetic, and spin-glass phases are obtained. The antiferromagnetic phase is not stable for these conditions and this can be proved with Eq. (12) which shows that this phase is obtained only for α values larger than 1.17 which gives an impossible physical situa-

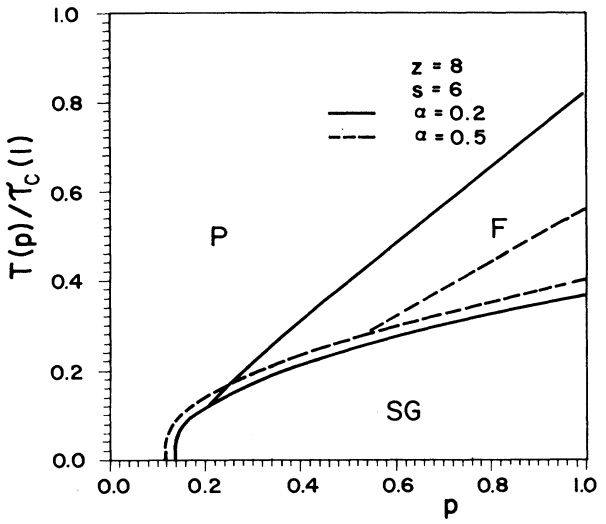

FIG. 1. Diluted and random-bond phase diagrams for different values of the competition parameter α.

tion with NNN coupling larger than that for NN coupling. We can see that the ferromagnetic region decreases and, the spin-glass region increases when the competition parameter α increases. Also we can note that the width of the spin-glass phase at $T=0$ enlarges as α increases. All these facts can be understood with the knowledge that the α increase produces a larger antiferromagnetic bond which reduces the critical paramagnetic-ferromagnetic temperature and increases the competition between the two types of bonds stabilizing in this way the spin-glass phase until $p=1$. This is a consequence of the constant α value, that permits the competition between the ferromagnetic- and antiferromagnetic interactions also until $p=1$.

The antiferromagnetic phase does not appear, because a pure antiferromagnetic region is impossible to find when ferromagnetic bonds between NN spins are always present. We found analogue results for the sc structure ($z=6, s=12$). From Eqs. (12) and (13) we can prove that for $\alpha=0.652$ only paramagnetic and spin-glass phases appear in the phase diagram. For $\alpha=0$ we obtained a typical diluted ferromagnetic phase diagram with only paramagnetic and ferromagnetic phases and a critical concentration $p_c=1/7$ for $T=0$ K.[19]

V. APPLICATION TO THE Fe-Al DISORDERED ALLOYS

For the Fe-Al disordered alloys system we obtained the phase diagram using Eqs. (12) and (13), and postulating for the competition parameter an expression given by

$$\alpha = Apq \ . \tag{14}$$

Some experimental evidences reinforce the choice of this model and this competition parameter. (1) It was shown experimentally[16] that these alloys, following an adequate heat treatment, can be obtained in a disordered bcc phase, thus we can use a random model. (2) In the Fe-Al ordered alloys the Al atom does not have magnetic moment but it contributes to the production of an antiferromagnetic superexchange bond between Fe atoms separated by it. Then we can suppose a model with dilution and ferromagnetic exchange between NN atoms and dilution and antiferromagnetic exchange between NNN Fe atoms. (3) The Fe-Al system is completely ferromagnetic for $p=1$ and not magnetic for $q=1$, respectively. Also, for amorphous alloys the NNN interaction has a maximum value which is one order of magnitude smaller than that between NN. Experimentally, for Fe-Al disordered alloys it was shown that the relation between the Al interactions is lest than 0.2. Then we postulate for α the relation (14), where A is a parameter to be adjusted in accord with the experimental datas. Amorphous and disordered magnetism are very similar. For the Fe-Al ordered alloys Shukla and Wortis[13] used a constant α value given by $\alpha=0.14$ and Grest[14] used a value $\alpha=0.4$. These results favor strongly the choice of the Hamiltonian model (1) and the probability distribution (2) and (3) in order to describe in a more complete way the magnetic properties of disordered Fe-Al alloys.

Figure 2 shows the phase diagrams obtained from Eqs. (12) and (13) using a competition parameter given by $\alpha=Apq$, with $A=1$. The phase diagram with large ferromagnetic and spin-glass phases (dashed lines) were obtained using a constant exchange parameter. In order to take into account the increases in the lattice parameter produced by the Al atoms we use an exchange parameter that changes with the Al concentration in the form $J=J(q)=J_1-J_0q$ (Ref. 17) and so, we obtained the smaller ferromagnetic and spin-glass phases showed in Fig. 2 (solid lines). The adjusted values of these parameters were $J_1=12.846$ meV and $J_0/J_1=0.95$. We can note that this change in the exchange parameter reduces the critical ferromagnetic-paramagnetic and spin-glass paramagnetic temperatures. Below the paramagnetic-ferromagnetic transition curve we do not find a spin-glass

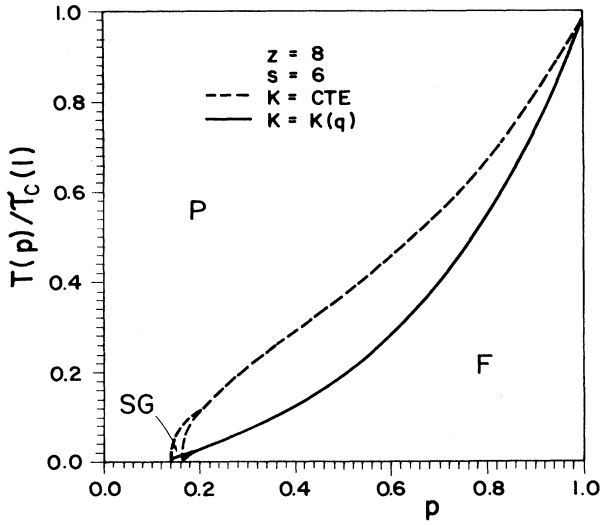

FIG. 2. Diluted and random-bond phase diagrams for $\alpha=pq$ and two different types of exchange interaction given in the text.

phase because the antiferromagnetic exchange parameter goes to zero when p increases, and then there is no competition.

Finally, in order to obtain a quantitative comparison of the present theoretical results with experimental datas, we show by dots in Fig. 3 the experimental critical temperature for the transition ferromagnetic-paramagnetic as a function of p obtained by Mössbauer spectroscopy.[16] In this figure we also show the theoretical phase diagram using Eqs. (12) and (13), with an exchange parameter $J(q)$ given previously and the competition parameter given by $\alpha = Apq$. The best agreement with the experimental data was obtained for $A = 0.2$. In this case the spin-glass phase is stable only for low temperatures, less than 20 K, and low Fe concentration, between 14 and 15.5 at. % Fe. This can be seen in the inset of Fig. 3 and is a consequence of the small α value.

In conclusion, we can see that the present model, although simple, can give a theoretical point of view of a diluted and random-bond Ising model, with ferromagnetic and antiferromagnetic interactions in the NN and NNN, respectively. Previously theoretical reports for this model, consider ferromagnetic- and antiferromagnetic interactions only in the NN. The present model postulate the paramagnetic, ferromagnetic, and spin-glass phases for α values varying from 0.0 to 0.652. For α values between 0.652 and 1.67 we have only spin-glass and paramagnetic phases and for $\alpha > 1.67$ the antiferromagnetic phase appears. Otherwise, values close or larger than 1.0 are not physically possible. When we applied this model to the Fe-Al disordered system including some experimental reports, we obtained good agreement with the experimental ferromagnetic-paramagnetic transition temperature. This good agreement also was obtained using a simple site diluted Ising model[17] considering only ferromagnetic NN interactions, so the antiferromagnetic superexchange between NNN reported for Fe-Al ordered alloys was not taken into account. Our model considering the superexchange supposes that all NNN interactions are antiferromagnetic and we show that in the Fe-Al disordered alloys it is possible to have spin-glass phase for low temperatures and Fe concentrations. Experimental work at low temperature is now in progress in order to complete the phase diagram for this system and to prove the existence of the spin-glass phase.

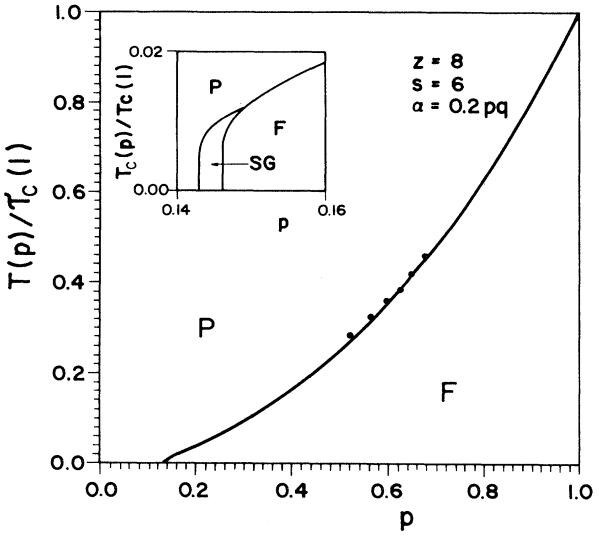

FIG. 3. Phase diagram for the binary Fe-Al disordered phase. The dots represent the experimental result of the ferromagnetic (F) to paramagnetic (P) transition. The solid lines represent the theoretical results according to the present model and with $\alpha = 0.2pq$ and the parameters previously reported for this system.

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