Ising model for disordered ferromagnetic Fe-Al alloys

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A simple site-diluted Ising model is proposed to study the magnetic properties of Fe_pAl_q alloys (with $p+q=1$) in the structural disordered phase. It is assumed that Al atoms, which are nonmagnetic, can induce an extra ferromagnetic interaction between second-neighbor Fe atoms. It is further assumed that this second-neighbor interaction, as well as the nearest-neighbor one, decreases as q increases. The critical properties are obtained by the variational approach based on Bogoliubov inequality for the free energy in the pair approximation. Quite good fittings to the experimental results of the ordering temperature are obtained as a function of q. A negative value of the extra exchange interaction for some range of the Al concentration is obtained. It is argued that this negative exchange can drive a spin-glass-like phase in these compounds, a fact that should be sought experimentally.

The study of the effects of disorder on the critical behavior of magnetic systems has been the subject of a great amount of investigations during the last few decades, both theoretically and experimentally.[1,2] From the theoretical point of view it is known that Ising-like models are well suitable to describe the critical behavior of insulating anisotropic magnetic systems.[1] However, for band magnets[3] the situation is not so clear due to the lack of theoretical results on models that could be applicable to real experimental systems.

In particular, the structural and magnetic properties of Fe_pAl_q alloys (with $p+q=1$) have been widely studied in the literature.[4–20] They are arranged on a bcc structure, which can be viewed as two cubic interpenetrating sublattices. When prepared by slow cooling or quenching, from temperatures lower than 700 °C, they are of the ordered type for $0.18<q<0.5$, with the Al atoms entering only on one specific cubic sublattice of this bcc lattice (the other cubic sublattice is always occupied by Fe atoms in this ordered structural phase[8]). From $q=0$ to $q\sim0.18$ they are all disordered, independent of the heat or any other treatment[14] (in this disordered phase any site of any sublattice of the whole bcc structure can independently be occupied by Fe or Al atoms). The structural ordered alloys present an anomalous behavior in the critical temperature and mean hyperfine field as a function of the Al concentration q, in the composition range $0.2<q<0.3$. Some theoretical works based on the assumption of a superexchange antiferromagnetic interaction between Fe atoms separated by an Al atom have been developed for this ordered system.[7] However, no experimental evidence of an antiferromagnetic phase has been found and the existence of a spin-glass phase near this composition range is currently more accepted instead[12,13] (though no spin-glass phase have already been seen in these systems as well).

On the other hand, the extension of the existence region of the disordered ferromagnetic solid solution beyond concentrations $q\sim0.18$ has been achieved through the use of many treatments such as cold-working by rolling,[11] rf sputtering,[15] quenching,[16] coevaporation[17] and mechanical grinding.[18] It has been shown that these alloys exhibit the same anomalous behavior in the critical temperature as the ordered ones (but not in the hyperfine field). Regarding the magnetic ordering, the ferromagnetic transition temperature $T_c(q)$ decreases as the Al concentration q increases. The decreasing in T_c for $q<0.2$ is very slow and one has a small value for

$$\alpha=-1/T_c(dT_c/dq), \tag{1}$$

as $q\to0$. However, for $q>0.2$ the critical temperature falls down rather abruptly and α is definitely different from zero. It should be stressed that such anomaly in these alloys is far from being fully explained as yet.

Earlier theoretical results,[20] based on simple Ising models, were not able to explain this rather unexpected behavior of T_c for small concentrations q. Since the Al atom has no magnetic moment it plays the role of a site-dilution in the system. Indeed, exact theoretical results for the diluted Ising model on two-dimensional lattices, as well as reliable approximations in three dimensions, give values of $\alpha\sim1$.[1] This is in complete disagreement with the experimental data and shows that dilution is certainly not the only mechanism played by the Al atoms in such disordered systems.

In this work we propose an extended version of the simple site-diluted Ising model, in the same lines as the previous works on Fe-Al (Ref. 20) as well as on Fe-Al-Mn systems,[21] in order to study the phase diagram of these disordered alloys. We further assume herein that the Al atoms, though being nonmagnetic, can induce an extra superexchange like ferromagnetic interaction between second-neighbors Fe atoms, which are separated by an Al atom. It is also assumed that this superexchange interaction, as well as the ferromagnetic nearest-neighbor one, decreases as q increases, since the lattice expands when Fe is substituted by Al.[16] To obtain

the phase diagram we employ a variational approach based on Bogoliubov inequality for the free energy associated to a two-cluster-like approximation. Such a procedure allows one to obtain closed form expressions for the critical temperature and the phase diagram is easily obtained as a function of the theoretical parameters. The present approach, although still rather simple, is better than the usual mean-field approximation.

The proposed Hamiltonian for this system can be written as

$$H = -J_1 \sum_{\langle \mathrm{NN} \rangle} \epsilon_i \epsilon_j \sigma_i \sigma_j - J_2 \sum_{\langle \mathrm{NNN} \rangle} \epsilon_i \epsilon_j \sigma_i \sigma_j , \qquad (2)$$

where J_1 and J_2 are the first- and second-neighbor interactions, respectively, and $\sigma_i = \pm 1$. $\epsilon_i = 1$ or 0 whether the site i is occupied by an Fe or Al atom, respectively. The first sum in Eq. (2) runs over all nearest-neighbor (NN) pairs and the second sum over the next-nearest-neighbor (NNN) pairs having an Al atom as NN, i.e., for an NN site k common to the sites i and j one has $\epsilon_k = 0$ (this Al atom is inducing the extra NNN J_2 interaction). It is also assumed that any site can independently be occupied by an Fe or Al atom so that the probability distribution for ϵ_i is

$$P(\epsilon_i) = p\,\delta(\epsilon_i - 1) + q\,\delta(\epsilon_i), \qquad (3)$$

where p is the Fe concentration and q is the Al concentration.

In order to obtain the approximate thermodynamic properties of the present system we employ the pair approximation based on Bogoliubov inequality for the free energy.[22] It closely follows the approach of Ferreira *et al.*[23] and has been previously applied to the Fe-Al-Mn alloys[20,21] as well as to other quenched disordered classical[24,25] and quantum[26] models. Following the same procedure of Ref. 20 we obtain for the critical temperature

$$\frac{z}{z-1}(K_1 + 3qK_2) = p(K_1 + 3qK_2)(1 + \tanh K_1) + q\left(K_1 + 3qK_2 + \frac{3K_2}{z-1}\right), \qquad (4)$$

where $K_1 = \beta J_1, K_2 = \beta J_2$ with $\beta = 1/k_B T$, k_B the Boltzmann constant and T the temperature. z is the coordination number of the lattice. For the one-dimensional model ($z = 2$) the above procedure gives the exact free-energy of the system and for $z > 2$ it is equivalent to the Bethe approximation. In the limit $J_2 \to 0$ one recovers the same result of Ref. 20.

In the previous treatment of this system it was considered a nearest-neighbor exchange interaction depending on the Al concentration of the form[20]

$$J_1(q) = J(1 - Lq), \qquad (5)$$

where $J = 0.013$ eV and $L = 0.95$ were the theoretical values that best represented the phase diagram (the dashed line in Fig. 1 shows the early theoretical curve together with the experimental data). It can be seen that good agreement is obtained only for $q > 0.3$, which is clearly out of the anomalous region. Moreover, one obtains a very high value $\alpha \sim 1.6$ for $q \to 0$ from this dashed line, in complete disagreement with the experimental results.

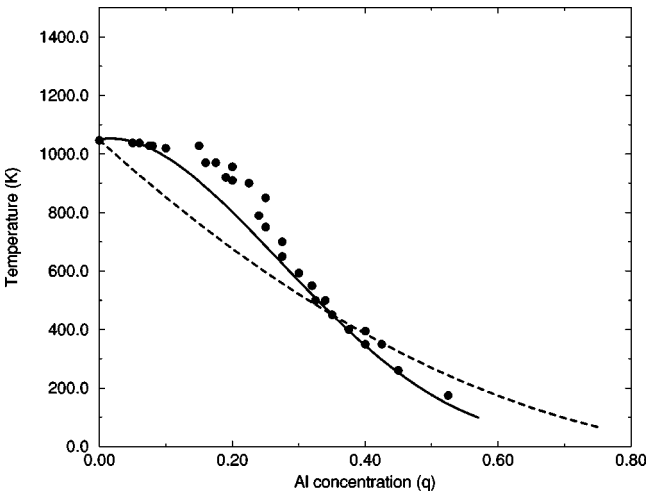

FIG. 1. Critical temperature as a function of the Al concentration according to Eqs. (4)–(6) and the parameters given in the text (solid line). The experimental results were taken from Refs. 4,5,16,18 and 19. The dashed line is the previous result corresponding to $J_2 = 0$ (Ref. 20).

In the present model, besides the nearest-neighbor interaction given by Eq. (5) we also assume

$$J_2(q) = J(A - Bq)(C - q), \qquad (6)$$

where $A, B,$ and C are additional theoretical parameters. The above form for J_2 has been chosen in an attempt to preserve the quality of the previous fitting for $q > 0.3$ by choosing C around the value 0.35 (in the case where J_2 is small).

Figure 1 exhibits the ordering critical temperature as a function of the Al concentration according to Eq. (4) for $A = 2.2$, $B = 2.9$, and $C = 0.35$ and taking the *same* values as before for J and L. One can see now that the agreement is far better than by considering just the NN interaction J_1. Although the value of the slope $\alpha \sim -0.16$ is positive (due to the fact that the superexchange interaction is suddenly switched on by the Al atoms) it is quite small and the curve is almost constant for $q \sim 0$. In Fig. 2 we show the behavior of the interactions J_1 and J_2 as a function of the Al concentration in the whole range $0 < q < 1$. One can see that in the interval $0.35 < q < 0.76$ the extra exchange interaction J_2 becomes negative, i.e., an antiferromagnetic interaction (which has already been proposed for the ordered system[7] as well as for the disordered one in a different manner.[27,28])

One can see that quite good results are obtained from the present approach. However, some few remarks concerning the fittings and the model are now in order.

(i) In comparing the present model to the experimental results for the disordered Fe-Al alloys we have first tried a linear decay for the superexchange interaction J_2 in a similar way as Eq. (5) for J_1. The corresponding fittings, however, were not so good as those obtained by assuming the second-order function of q in Eq. (6). On the other hand, we have not tried higher degree functions for J_2 in order not to proliferate theoretical parameters (that is why we have also chosen the same values for J and L from the previous model).

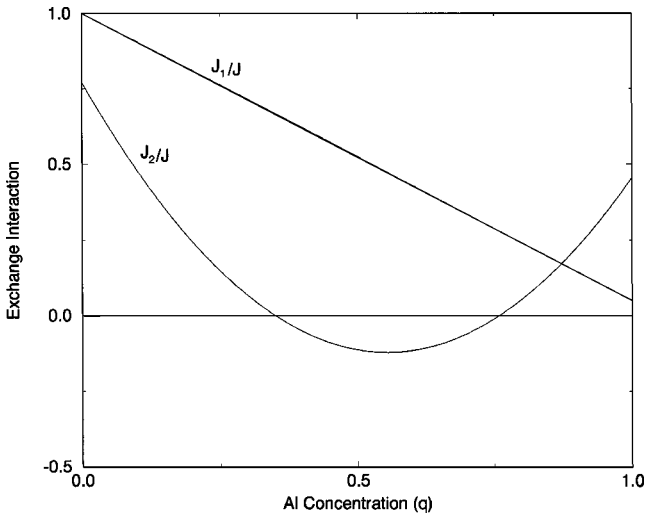

FIG. 2. Ratio J_1/J and J_2/J as a function of the Al concentration obtained from Eqs. (5) and (6) and the parameters in the text.

(ii) We have neglected the direct ferromagnetic exchange interaction between next-nearest-neighbor Fe atoms (that is, that one which is not induced by Al atoms) once this interaction is expected to be one order of magnitude smaller than the nearest-neighbor J_1.[20] From Fig. 2 one can in fact see that J_2 is indeed comparable to J_1 for q in the anomalous region range and also greater than the NNN direct ferromagnetic interaction.

(iii) The fittings of the phase diagram naturally give rise to an antiferromagnetic interaction J_2 for some range of q. This has been previously proposed in the literature in a different context for the ordered alloys[7] and also for the present disordered system[27,28] (with no ferromagnetic coupling between NNN). Moreover, in the present case we have also dilution (and competition if we remember that J_1 is always positive) in such a way that we have theoretically the basic ingredients for the presence of a spin-glass like phase. Besides, this approach is capable of giving a better agreement with the experimental results for small values of q than the previous ones.[27,28]

(iv) The behavior of J_2, shown in Fig. 2 in the entire range of the concentration, turns out to be positive again for $q>0.76$. It should be stressed here, however, that the present results do not depend on this ''reentrant'' behavior of the induced J_2 coupling as function of q. In fact, for Al concentrations greater than $q\sim 0.6$ these alloys do not present the structural disordered phase any more and can, moreover, change its bcc lattice[4–11]. It means that the Hamiltonian model given by Eq. (2) is valid while the disordered bcc structural phase is maintained by the system. As a result, the extra exchange interaction J_2 has indeed, in the interested $0<q<0.6$ region, its most physical and expected behavior, being positive at small q and becoming negative at some intermediate concentration with no ''reentrance''

(v) Better theoretical approaches will certainly improve the critical values. However, the qualitative behavior, and we also believe as well that the quantitative values of the theoretical parameters for the fittings, will not be so drastically changed in this case. That is what happens by performing Monte Carlo simulations on the model with $J_2=0$.[29]

(vi) Finally, something has to be said regarding the choice of the Hamiltonian (2). Although these compounds are all metalic the Mössbauer spectroscopy obtained from these disordered alloys on the bcc structural phase makes clear their ferromagnetic character and also indicates the most probable sites that appear in the hyperfine field distribution.[16] This fact suggests one to think, at least at first sight, of localized spins on the lattice. For this reason the Ising model has been extensively used in describing the magnetic properties of such systems.[20,21,27–29] A different situation occurs in the same alloys on the fcc phase, which present a magnetic behavior where, according to Ishikawa,[30] would be more reasonable to be described by an itinerant electron model.

As a final remark it should be said that the present model is still rather simple to account for the total interesting properties of these disordered alloys. However, it seems indeed that the superexchange character induced by the Al atoms are in fact relevant in order to describe the main magnetic critical behavior of such systems. We believe that the present results will provide an additional motivation for further more rigorous theoretical effort on these disordered (as well as on the ordered counterpart) alloys. On the other hand, seeking for a spin-glass-like phase on these systems seems to be quite interesting from the experimental point of view.

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