

# Monte Carlo study of the change of critical temperature in a diluted Ising model due to configuration disorder

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## Abstract

Monte Carlo simulations on a diluted Ising Hamiltonian were used to obtain the susceptibility of virtual binary samples (conformed by ferromagnetic atoms and non-magnetic atoms) on a bcc lattice. Samples size of  $L = 10$  were constructed with different configuration order using random mixtures. The susceptibility curves illustrate that the critical temperature decreases when the disorder in the samples increase. From fittings of the exchange interaction versus the concentration of non-magnetic atoms it was possible to reasonably describe the magnetic phase diagram of the  $\text{Fe}_{1-q}\text{Al}_q$  alloys.

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It is well known that impurities and lattice distortions in magnetic systems play a central role in their corresponding thermodynamic properties such as magnetization and critical temperature. For instance, elastic lattices can change the character of the phase transition (from second order to first order) on magnetic materials [1]. On the other hand, one of the most important *defects* in crystalline materials is the presence of non-magnetic impurities on lattice sites which inhibits the magnetization of the specimen. In particular,  $\text{Fe}_{1-q}\text{Al}_q$  alloys are a physical realization of such process because the Al atoms have no magnetic moment and act as a dilution in terms of spin components. For  $q = 0$ , the pure iron has a ferromagnetic–paramagnetic second order phase transition at a temperature  $T_c = 1040$  K and this transition temperature  $T_c$  decreases as  $q$  increases. Another important microscopic aspect in such systems regards the atomic position of such im-

purities, which strongly depend on the heat treatment of the sample. For instance, the disorder  $\text{Fe}_{1-q}\text{Al}_q$  alloys can be obtained up to  $q = 0.5$  when the samples are given a heat treatment at around 1000 K for several days and then quenched in ice [2]. The rather complex magnetic behavior of these binary alloys have been reported in the literature [2,3] and references therein.

Efforts have also been made to explain the phase diagram of  $\text{Fe}_{1-q}\text{Al}_q$  alloys [2,4–9] but the theoretical results do not explain the whole behavior. For instance, the experimental critical temperatures reported by different authors have discrepancies of about 50 K (or more) in relation to the theoretical predictions and these uncertainties can be attributed to the different preparation processes to obtain the samples.

Motivated by this peculiar behavior we study, in the present work, a Monte Carlo simulation on bcc lattices of a diluted Ising model by taking a dynamic configuration of the non-magnetic atoms. Despite having in mind  $\text{Fe}_{1-q}\text{Al}_q$  alloys, the present approach can also be applied to any binary system as well as magnetic heterostructures.

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To begin with we have to deal with the magnetic impurities and simulate the corresponding heat treatment. To do so, virtual samples were constructed in the bcc phase with two displaced simple cubic lattices of size  $L = 10$ , each one having  $10^3$  atomic sites, with a total of  $2 \times 10^3$  atoms per sample. The lattice sites were labeled by  $\varepsilon_i = 0$  or  $\varepsilon_i = 1$ , depending if the site is occupied by aluminum or iron atoms, respectively. Beginning with all sites occupied by Al atoms, we take the following steps:

- (1) Fe atoms, in a sequential form, substitute the initial Al atoms until the desired concentration is obtained (this substitution is made in alternate form in each simple cubic lattice). This corresponds to an initial diluted configuration.
- (2) The lattice is now revisited in a typewriter form and, for each site a random number  $r$  ( $0 \leq r \leq 1$ ) is generated. If  $r$  is smaller than a pre-established “migration probability  $P_r$ ”, the atom of the site is randomly interchanged by another atom of the lattice. This procedure is done just once.

A scheme of this mixture process is shown in Fig. 1 for Fe concentration  $p = 0.9$  and Al concentration  $q = 0.1$ . In the top part of the figure for  $P_r = 0$  all the first 900 sites of each sub-lattice are occupied by Fe atoms and the remaining sites by Al atoms. For  $P_r = 0.2$ , one sweep through the lattice starts to mix the impurities, but a higher concentration of them is still noticeable in the last 100 sites of each sub-lattice (due to the great number of lattice sites the line for  $\varepsilon_i = 1$  is, within the present scale, continuous and does not show the actual vacancies; also, the vertical lines are drawn for a clearer illustration of the configuration). Nevertheless, it is easy to see that the mixture degree increases with the migration probability  $P_r$ . Of course, with a small  $P_r$  and more visits on the lattice (twice or more) one can reach a similar configuration as for higher values of the migration probability. However, our intent here is to

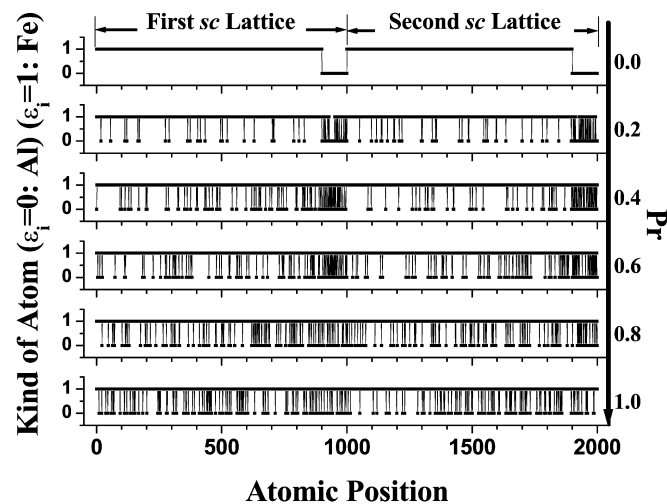


Fig. 1. Increasing of disorder degree with migration probability in the  $p = 0.9$  and  $q = 0.1$  sample. The lattice sites (atomic position) between 1 and 1000 represent the first simple cubic (sc) lattice; the sites between 1001 and 2000 represent the second simple cubic lattice. The two simple cubic lattices are interpenetrated to form the bcc lattice.

study different configurations of impurities and seek for their effects on the critical temperature. Moreover,  $P_r$  can simulate the temperature of the heat treatment. So, while  $P_r \approx 1$  means a high temperature treatment with a high mobility,  $P_r \approx 0$  is equivalent to a low temperature treatment where the mobility is quite low.

Next, with the sample built in this way, an Ising model Hamiltonian is defined on this lattice in the form

$$H = -J \sum_{\langle i, j \rangle} \varepsilon_i \varepsilon_j S_i S_j, \quad (1)$$

where  $\langle i, j \rangle$  means that the sum is made over nearest neighbors,  $J$  is the corresponding exchange interaction,  $\varepsilon_i = 0$  or 1, for Al or Fe atoms, respectively, and  $S_i = \pm 1$ . The applicability of this model to the present alloys can be justified by noticing that: (i) as already discussed in Ref. [9] only nearest-neighbor interactions should be taken into account since magnetic studies of amorphous alloys show evidence that the value of the next-nearest-neighbor interactions is one order of magnitude smaller [10] and (ii) although these systems should be better described by the Heisenberg model, the results by treating them with simple Ising spins provides no significant changes in the physical behavior of their magnetic properties [13].

Monte Carlo simulations are now performed in the Hamiltonian (1) for every diluted lattice with the single spin Metropolis dynamic. We have used  $4 \times 10^4$  Monte Carlo steps per spin (MCS) with  $2 \times 10^3$  MCS discarded for thermalization on the bcc lattice with size  $L = 10$  and periodic boundary conditions.

Thermodynamic quantities such as energy, magnetization, heat capacity and magnetic susceptibility have been obtained following the above procedure for different values of  $P_r$  in the range  $0 < \Theta \leq 8$ , where  $\Theta = k_B T / J$  is the reduced dimensionless temperature. Some graphs of the magnetic susceptibility as a function of the dimensionless temperature are shown in Fig. 2. It can be seen that the susceptibility peaks move to low temperatures when the migration probability increases and the corresponding values at the peaks have a tendency to increase when  $P_r$  increases. For small amount of impurities, for instance  $q = 0.1$ , the shift of the peak to the region of smaller temperatures as  $P_r$  increases is not so sensitive, since it does not matter whether the non-magnetic atoms are clustered or not. For higher values of  $q$ , when the non-magnetic atoms are clustered they destroy much less bonds than when they are separated in the lattice, a fact that explains the decrease of the transition temperature for increasing values of the mixing probability. This effect is much more pronounced in cases  $q = 0.3$  and  $q = 0.6$ . On the other hand, for still higher values of  $q$  (like  $q = 0.9$ ) the mixing can even destroy the peak, since we are well above the percolation threshold ( $q_c \approx 0.25$  for the bcc lattice [11]).

A similar behavior, as shown in Fig. 2, is also seen for other lattice sizes. In addition, the peaks of the susceptibility curves give the critical temperatures of the system. For the present case with  $L = 10$  one has  $\Theta_C = 6.30(5)$  for the pure Fe alloy, a value quite close to better estimates from long runs Monte Carlo simulations  $\Theta_C = 6.35408(8)$  [12]. Finite size effects must also be taken into account in this case. By considering lattice sizes

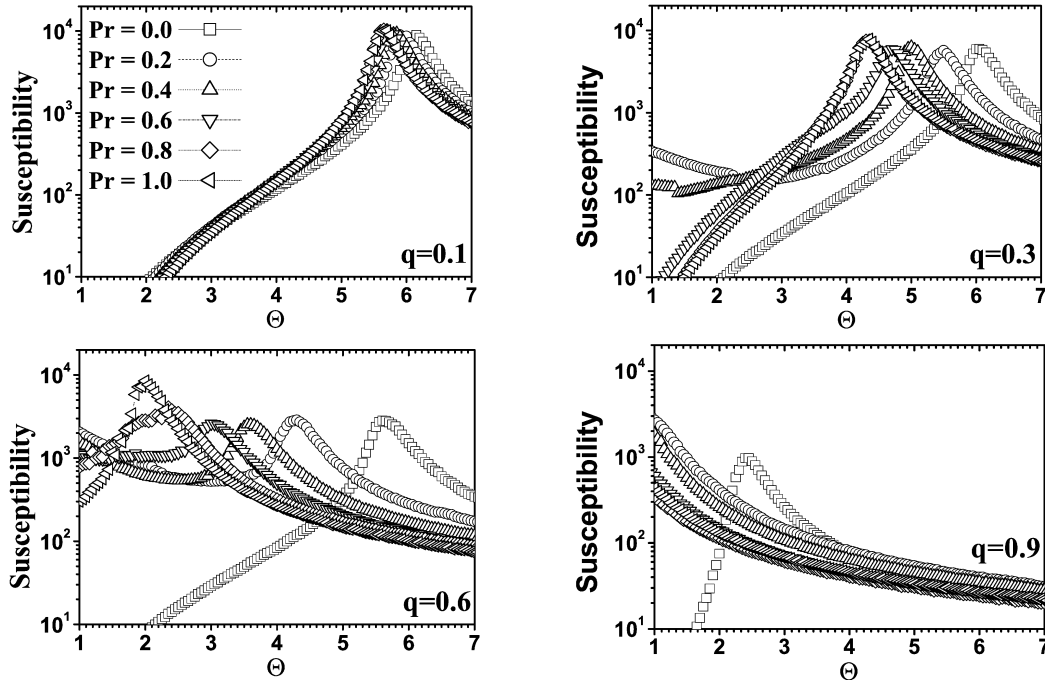


Fig. 2. Magnetic susceptibility as a function of the reduced temperature for different Al concentration obtained by Monte Carlo method for a lattice size  $L = 10$ . In each graph the curves to the right correspond to low migration probability and they move to the left as it increases. The same legend applies to all figures. The error bars are smaller than the symbol sizes.

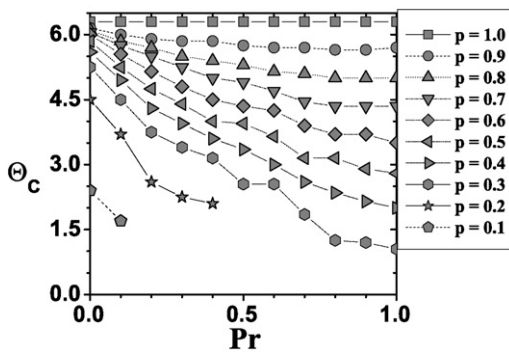


Fig. 3. Dependence of dimensionless critical temperature ( $\Theta_C$ ) with migration probability ( $Pr$ ), obtained by Monte Carlo method, for various iron concentrations ( $p$ ). The decreasing of critical temperatures with  $Pr$  is more evident for low values of Fe concentration (high values of Al concentration). The error bars are smaller than the symbol sizes.

$L = 4, 8, 10, 12, 14, 16$  and  $32$  a finite size scaling, within our simulations for the maximum of the susceptibility in the  $q = 0$  case, yields  $\Theta_C = 6.33(2)$ , which is actually closer to that value from Ref. [12]. However, this difference turns out to be unimportant for the global phase diagram of the present systems (the only difference is a change in the value of the exchange interaction from  $14.22$  to  $14.15$  meV, see also Ref. [13]). The previous comparison, in some sense, justifies the use of  $L = 10$  in the present work.

The dimensionless critical temperature  $\Theta_C$  as a function of the migration probability, as shown in Fig. 3 for different Fe concentrations, gives us a better view of the role of mixing in these binary alloys. It is interesting to notice that it is possible to obtain critical temperature for  $q = 0.8$  ( $p = 0.2$ ) and  $q =$

$0.9$  ( $p = 0.1$ ), values of  $q$  well above the percolation threshold, if the system is bad alloyed to a nanoscopic scale. However, by increasing the migration probability the critical temperature vanishes for  $Pr < 1$ , as expected.

It would now be interesting to make a closer comparison to the real  $\text{Fe}_{1-q}\text{Al}_q$  alloys in the structural disordered phase. First of all the previous results do not take into account the lattice dilatation due to the presence of Al atoms neither its effect over the exchange interaction  $J$  between iron atoms. In order to introduce such corrections we suppose that the exchange interaction  $J$  depends on aluminum concentration  $q$  through the relation

$$J(q) = J_0(1 + J_1q + J_2q^2 + J_3q^3), \quad (2)$$

where  $J_0 = 14.22$  meV,  $J_1 = 1.24$ ,  $J_2 = -5.81$  and  $J_3 = 1.94$  are theoretical parameters already adjusted from the experimental data in Ref. [13]. A comparison for different values of  $Pr$  is made in Fig. 4. This figure shows the curves for different migration probabilities of the critical temperature versus Al concentration obtained from Monte Carlo simulations (with the details explained above) and the lattice size  $L = 10$ . From this figure one can note that; (i) the majority of experimental data (filled circles) are in fact within the region delimited by the Monte Carlo data; (ii) no mobility at all ( $Pr = 1$ ) is quite away from the experimental data; and (iii) for  $q < 0.23$  the agreement is better for small values of the mixing probability while for  $q > 0.23$  the better agreement is for  $Pr \approx 1$ . As most of the previous theoretical treatments did not attempt to describe the actual positions of the Al atoms (except from a work by Schmid and Binder [4]), it seems that

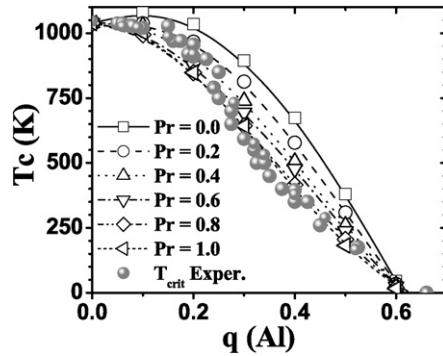


Fig. 4. Phase diagram of  $\text{Fe}_{1-q}\text{Al}_q$ . The solid circles are the experimental data from Refs. [3,14,15], the unfilled labels are results from the present work for some migration probabilities. The critical temperature was obtained supposing that exchange interaction  $J$  has a cubic dependence with Al concentration (see text). The error bars are smaller than the symbol sizes.

the mobility of Al in these samples for small concentrations  $q$  is lower than for high values of  $q$ ,  $q \approx 0.23$  being the transition region. In fact, experimental results indeed show that in this region there is a change from two different bcc structures [16]. In summary, the discrepancies between the theoretical and the experimental data in  $\text{Fe}_{1-q}\text{Al}_q$  alloys at low Al concentration seem to be related to the configuration of Al atoms on the lattice instead of some different magnetic behavior due to the Fe–Fe exchange interaction. Finally, we have in this work analyzed the properties of a diluted Ising model. However, we expect that the same qualitative behavior should also be obtained by using the more realistic model for this system, namely the Heisenberg model [13].

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