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Monte Carlo investigation of the correlation between magnetic and chemical ordering in NiFe alloys

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ABSTRACT

The Ni_xFe_{1-x} alloys close to the stoichiometric Ni_3Fe composition are modeled by means of Monte Carlo simulations. To describe the atomic and magnetic configurations, the Ising and Heisenberg models with nearest-neighbor interactions have been used, respectively. The pairwise interactions have been fitted to the experimentally measured Curie and Kurnakov temperatures, the Fe–Fe magnetic exchange interaction has been considered antiferromagnetic. The mutual influence of the magnetic and chemical ordering is evidenced and a good agreement with the phase diagram is obtained. Our numerical results show that the magnetic order is able to increase the Kurnakov temperature and, reciprocally, the chemical order is responsible for a rise in the Curie temperature. Also, the influence of the applied magnetic field on the chemical order is investigated and an increase of the Kurnakov temperature with the external field is observed.

1. Introduction

The Ni–Fe alloys are soft magnetic materials that find technological applications in, for example, magnetic recording heads and spin valve devices. The equilibrium phase diagram is complicated and is still being elaborated. In the Ni-rich region, the alloy undergoes a second-order ferromagnetic–paramagnetic phase transition at T_C (Curie temperature) and a first-order order–disorder structural transition at a lower temperature T_K (Kurnakov temperature) [1]. The Curie and Kurnakov temperatures both peak near the Ni_3Fe composition. Also, a large change in the magnetocrystalline anisotropy appears due to $L1_2$ ordering [2]. As shown experimentally [3,4], the Curie temperature of the ordered Ni_3Fe alloy (940 K) exceeds the one of the disordered alloy (870 K). However, let us mention the difficulty in measuring the Curie temperature of the ordered alloy. Indeed, it is higher than the order–disorder transition temperature T_K (773 K) and, therefore, the alloy is not completely ordered due to the atomic diffusion. These features strongly suggest a mutual influence between magnetic and chemical ordering.

Ni–Fe and other alloys that contain magnetic elements (for example, Ni–Cu, Fe–Co) have been widely studied using different theoretical approaches [5–13]. The cluster-variation method (CVM) has been applied to the investigation of such alloys [5,6] as well as the mean-field (MF) approximation [7,8]. It was shown that both types of order (chemical and magnetic) are coupled and influence each other. This has been noticed from the behavior of the thermodynamic quantities and the calculated phase diagrams. It should be mentioned that due to the absence of short-range order in MF calculations (in contrast to Monte Carlo simulations), the influence of the short-range chemical order on the Curie temperature cannot be observed. *Ab initio* calculations (see Ref. [9] for example) have also confirmed an important role of the simultaneous action of the magnetic and chemical ordering in Ni–Fe alloys. Moreover, it was proposed that the chemical ordering in this system is entirely of magnetic origin. The Monte Carlo (MC) method has been also applied using both Ising and Heisenberg models [10–13]. For the Ni–Fe alloys, the Ising model has been mainly used for both atomic and spin systems [10,11]. To our knowledge, the spin system in these alloys has been modeled by the Heisenberg model only once [12]. In this paper, no thermal variation of the thermodynamic quantities is presented and the separate description of the spin and atomic systems has not been performed. Therefore, no conclusion about the interplay between the two kinds of ordering in the framework of the Heisenberg model can be drawn. Also, let us mention that the influence of an external magnetic field has not been studied in these works.

In this paper, our goal is to investigate the mutual influence of the chemical and magnetic ordering in the Ni-rich Ni–Fe alloys and, in particular, close to the Ni_3Fe composition. To that purpose we have considered the classical Heisenberg spin model, which is the most suitable for investigating the magnetic transition in such alloys. Indeed, the critical exponents of the 3D Heisenberg model ($\gamma \approx 1.39$, $\beta \approx 0.36$ and $\alpha \approx -0.11$) [14] are very close to those experimentally measured for pure Ni [15]. Moreover, from

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the Harris conjecture [16], the configurational disorder changes the numerical values of the critical exponents only if the specific heat exponent is positive, i.e. for the 3D Ising model unlike the 3D Heisenberg one. Consequently, the choice of the spin model is very important for the investigation of the effects of chemical order on magnetic transition. Also, the influence of an external field on the order–disorder transition temperature has been studied. We have chosen the MC method in which, unlike mean-field techniques, the chemical and magnetic short-range order exists above the transition. The model and the simulation method are described in Section 2. The numerical results are presented and discussed in Section 3 in the following order: firstly, simulations with only magnetic interactions, secondly simulations with chemical and magnetic interactions for the stoichiometric Ni_3Fe alloy, then the investigation of the phase diagram in the Ni_3Fe region and finally the influence of an external magnetic field on the order–disorder transition temperature. The conclusions are given in Section 4.

2. Model and simulation

Our model consists of a face centered cubic (fcc) lattice with periodic boundary conditions. Each site is occupied by an atom of type α or β (Ni or Fe, respectively) and by a classical Heisenberg spin $\mathbf{S}_i=(S_i^x,S_i^y,S_i^z)$ with $S_{Ni}=0.308$ and $S_{Fe}=1.4$.

To model the binary system we consider an Ising-type Hamiltonian:

$$H_{chem}=-\sum_{\langle ij\rangle}\sum_{\alpha\beta}V_{ij}^{\alpha\beta}\xi_i^{\alpha}\xi_j^{\beta} \tag{1}$$

where the first sum is taken over all interacting pairs, ξ_i^{α} (ξ_j^{β}) is the occupation variable which is equal to 1 if the site $i(j)$ is occupied by an $\alpha(\beta)$ atom and equal to 0 otherwise. $V_{ij}^{\alpha\beta}$ are the pairwise interaction parameters between atoms i and j of types α and β, respectively. The chemical interactions are limited to the nearest-neighbors (NN), and their values have been taken from Ref. [10]: $V^{Fe-Fe}/k_B=8400$ K, $V^{Ni-Fe}/k_B=9200$ K, $V^{Ni-Ni}/k_B=8590$ K.

To model the spin system we have used the classical Heisenberg Hamiltonian:

$$H_{magn}=-\sum_{\langle ij\rangle}J_{ij}(\mathbf{S}_i\cdot\mathbf{S}_j)+\mu_B\mathbf{B}\sum_i g_i\mathbf{S}_i \tag{2}$$

where J_{ij} are the NN "exchange" interactions. The second term represents the Zeeman energy, which is caused by the external magnetic induction $\mathbf{B}$, μ_B is the Bohr magnetron and the Landé factor g_i is equal to 2 for both Ni and Fe atoms. The average magnetic moment (in μ_B units) per atom is defined as

$$\mathbf{m}=-\frac{1}{N}\sum_i g_i\mathbf{S}_i$$

where N is the number of spins. The exchange parameters J_{ij} were defined in the following way. J^{Ni-Ni} has been chosen so that the model provides the experimental Curie temperature of pure Ni (627 K). Since the critical temperature of the Heisenberg model on the fcc lattice with NN interactions is given by $k_BT_C/(JS^2)\approx 3.16$ [14], then $J^{Ni-Ni}/k_B\approx 2093$ K. The Fe–Fe interaction was determined in order to model the antiferromagnetic γ-Fe with $T_N\approx 70$ K [17], that is $J^{Fe-Fe}/k_B\approx -80$ K. The Ni–Fe interaction has been fitted by numerical simulations in order to obtain the experimental Curie temperature (≈ 870 K) of the disordered Ni_3Fe alloy. A good agreement was obtained with $J^{Ni-Fe}/k_B\approx 1060$ K. It has to be noted that the system is frustrated due to the antiferromagnetic Fe–Fe interaction, so the stable magnetic configuration at 0 K could be no more collinear with Fe additions.

To include both magnetic and chemical interactions simultaneously we have combined the two Hamiltonians (1) and (2):

$$H_{total}=H_{chem}+H_{magn}=-\sum_{\langle ij\rangle}\sum_{\alpha\beta}V_{ij}^{\alpha\beta}\xi_i^{\alpha}\xi_j^{\beta}-\sum_{\langle ij\rangle}\sum_{\alpha\beta}J_{ij}^{\alpha\beta}(\mathbf{S}_i^{\alpha}\cdot\mathbf{S}_j^{\beta})\xi_i^{\alpha}\xi_j^{\beta}+\mu_B\mathbf{B}\sum_i\sum_{\alpha}g_i^{\alpha}\mathbf{S}_i^{\alpha}\xi_i^{\alpha} \tag{3}$$

The numerical procedure we used is the importance-sampling Monte Carlo (MC) method in the canonical ensemble [18,19] based on the standard Metropolis algorithm [20]. During the simulations with only magnetic interactions (the chemical configuration is frozen), the trial step consists in a single spin rotation. When both magnetic and chemical interactions have been considered, the following procedure has been applied: two sites i and j occupied by two atoms of different types are randomly selected, then the two atoms with their spins are exchanged ($\xi_i^A\leftrightarrow\xi_j^B$ and $\mathbf{S}_i\leftrightarrow\mathbf{S}_j$) and finally two rotation angles, $\Delta\theta_i$ and $\Delta\theta_j$, are randomly determined. So, in case of acceptance, the chemical and magnetic configurations are updated simultaneously. In case of rejection, the single spin rotation without atom exchange is proposed. Such trial step has been chosen in order to equilibrate the spin system at low temperatures because the magnetic moments still fluctuate whereas the chemical configuration is frozen.

As usual, in MC simulations the thermodynamic quantities have been calculated by averaging over the MC steps. The quantities of interest are the internal energy, the order parameters (magnetization and chemical order parameters), the specific heat and the zero-field susceptibility. The simulations with only magnetic interactions have been performed on different frozen chemical configurations: from completely disordered (random) to perfectly $L1_2$ ordered. The final thermodynamic quantities have been obtained by averaging over several configurations in order to reduce the statistical errors. For the simulations including the chemical interactions (only or with the magnetic ones), two procedures have been used: either by starting from a perfect ferromagnetic $L1_2$ structure and increasing the temperature from $T_1=1$ K or by starting from a completely chemically and magnetically disordered configuration and decreasing the temperature from $T_1>T_C$. This was done to check the hysteresis phenomenon, which is expected at a first-order transition.

Our simulations have been carried out on a $20\times20\times20$ fcc lattice, i.e. 32×10^3 atoms. Far from T_K and T_C, 10^4 MC steps have been made at each temperature. This number has increased tenfold in the vicinity of the phase transitions in order to improve the accuracy. About 2×10^3 MC steps have been discarded at each temperature for equilibration.

3. Numerical results

Three types of simulations have been performed. In this paper only the results of the simulations with magnetic interactions and with both chemical and magnetic interactions will be discussed in details since the chemical simulations on such systems have been presented elsewhere [10].

3.1. Simulations with magnetic interactions only

Firstly, the completely disordered and the perfectly $L1_2$ ordered crystals (Ni_3Fe) have been considered. The Curie temperature has been estimated from the location of the zero-field susceptibility, since the specific heat behavior of the 3D classical Heisenberg model is cusp-like rather than divergent at