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Effects of atomic ordering on the Curie temperature of FePd $L1_0$ type alloys

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In this work, we systematically studied the effects of atomic ordering on the Curie temperature of FePd alloys in the composition region of $L1_0$ phase. The Curie temperatures of the bulk Fe–50 at. % Pd, Fe–55 at. % Pd, Fe–60 at. % Pd alloys for both ordered and disordered structures have been measured. Unlike the Curie temperatures reported in the literature, atomic ordering decreases the Curie temperature by about 30 K for each of the compositions. Using a molecular field model, we calculated the Curie temperatures of Fe–Pd alloys. The simulation results show that the difference of the Curie temperature of the disordered and ordered states arises from the change of the nearest-neighbor occupation probabilities and the change of atomic interactions. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1669272]

I. INTRODUCTION

$L1_0$ type magnetic materials, such as Fe–Pt and Fe–Pd, are potential recording materials for ultrahigh density magnetic recording. The magnetic properties of these materials are closely related to the atomic ordering of the crystal structure. Among these properties, the Curie temperature is an important parameter. Accurate measurements of the Curie temperatures for $L1_0$ materials are necessary, because they can help us to understand the magnetic properties at elevated temperatures for thermally assisted magnetic recording.[1] However, in the literature (Table I), the Curie temperatures of both $L1_0$ ordered and fcc disordered Fe–Pd alloys show striking discrepancies.[2–6] This encouraged us to carry out a comprehensive study of the Curie temperature for FePd alloys.

For ferromagnetic materials, the temperature dependence of the magnetization can be derived by finding the equilibrium free energy of the magnetic system.[7] Both the Ising model and the Heisenberg model have been developed to describe this free energy and the microscopic spin configurations.[7] The solutions of these two models are commonly simplified using the molecular field approximation (mean field), where the interatomic interaction is averaged and replaced by an effective molecular field acting on each individual spin. Although the temperature dependence of the spontaneous magnetization can be well described by the molecular field model, there are significant deviations close to the Curie temperature, due to the critical behavior.[7] Nevertheless, the molecular field model provides a simple solution of the Curie temperature and therefore is used herein to study the effect of atomic ordering on the Curie temperature.

This article is organized as follows. We first present the measurement of the Curie temperature for FePd alloys in Sec. II. The molecular field model used in the simulation is described in Sec. III. Finally, the experimental results and their comparison with the simulation are presented in Sec. IV.

II. EXPERIMENTS

The bulk Fe–Pd alloys with the composition of Fe–50 at. % Pd, Fe–55 at. % Pd, and Fe–60 at. % Pd were prepared using the arc-melting process. The disordered fcc phase was obtained by homogenizing at 1200 °C for 24 h and then water quenching to room temperature. The ordered $L1_0$ phase was obtained by annealing the fcc phase at 500 °C for 48 h. The alloy structures were then studied using a Rigaku x-ray $\theta/2\theta$ diffractometer with Cu K_α radiation.

The Curie temperatures were obtained by measuring the magnetization change with temperature using a Lakeshore vibrating sample magnetometer (VSM). This VSM is equipped with a vacuum electric furnace to reduce the oxidation at elevated temperatures. During the Curie temperature measurements, the VSM was programmed to ramp down at a rate of 5 °C/min from 550 to 150 °C under a constant magnetic field. It is necessary to apply a magnetic field in order to obtain large enought signals for measurements. However, the magnetic field shifts the moment-temperature curve. In such case, the Curie temperature can be determined by finding the point of inflection from the moment-temperature curve.[8] We found this point of inflection by taking the derivative of the moment-temperature curve. An error of 5 °C is estimated for this Curie temperature determination due to the temperature fluctuation.

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TABLE I. The Cure temperatures of FePd alloys with ordered L1$_0$ structure and disordered fcc structure.

	fcc Fe–0.5 Pd (K)	L1$_0$ Fe–0.5 Pd (K)	fcc Fe–0.55 Pd (K)	L1$_0$ Fe–0.55 Pd (K)	fcc Fe–0.6 Pd (K)	L1$_0$ Fe–0.6 Pd (K)	Reference
Fallot[a]	713	743	643	643	633	593	Ref. 2.
Kuprina[b]		693		623		573	Ref. 3.
Massalski[c]		763		718		643	Ref. 4.
Longworth[d]		729					Ref. 5.
This work	756±5	723±5	703±5	673±5	623±5	593±5	

III. MOLECULAR FIELD MODEL

In order to understand the effect of atomic ordering on the Curie temperature, we adopted a phenomenological atomistic model.[6] In this model, the magnetic moment of each atom depends on its local environment. For simplicity, we assumed that the local environment of an atom in an A–B alloy system can be described by the number of A and B atoms occupying its nearest-neighbor sites. With such an assumption, we ignored the influence of more distant neighbors. Using the molecular field model, the magnetic moments for alloy $A_{1-x}B_x$ near the Curie temperature can be expressed as[6]

$$\bar{\mu}_A(x)=[T_{AA}\bar{\mu}_A(x)+T_{AB}\bar{\mu}_B(x)]/T_C, \tag{1}$$

$$\bar{\mu}_B(x)=[T_{BB}\bar{\mu}_B(x)+T_{BA}\bar{\mu}_A(x)]/T_C, \tag{2}$$

where $\bar{\mu}_A(x)$ and $\bar{\mu}_B(x)$ are the average magnetic moment per A or B atom, and

$$T_{AA}=\frac{j+1}{6jk_B}J_{AA}\sum_{n_B=0}^{N}(N-n_B)P(n_B,x)[\mu_A^{(0)}(n_B)]^2, \tag{3}$$

$$T_{AB}=\frac{j+1}{6jk_B}J_{AB}\sum_{n_B=0}^{N}n_BP(n_B,x)[\mu_A^{(0)}(n_B)]^2, \tag{4}$$

$$T_{BA}=\frac{j+1}{6jk_B}J_{BA}\sum_{n_A=0}^{N}n_AP(n_A,1-x)[\mu_B^{(0)}(n_A)]^2, \tag{5}$$

$$T_{BB}=\frac{j+1}{6jk_B}J_{BB}\sum_{n_A=0}^{N}(N-n_A)P(n_A,1-x)[\mu_B^{(0)}(n_A)]^2. \tag{6}$$

Here k_B is Boltzmann constant and j is the spin quantum number. J_{AA}, J_{AB}, J_{BA}, and J_{BB} are the exchange-coupling constants. $\mu_A^{(0)}(n_B)$ is the magnetic moment of A atoms with n_B B atom nearest neighbors at 0 K; $\mu_B^{(0)}(n_A)$ is the moment of B atoms with n_A A atom nearest neighbors at 0 K. The probability that any atom has n_A A-type neighbors and n_B B-type neighbors is written alternatively as $P(n_A,1-x)$ and $P(n_B,x)$, where

$$P(n,x)=N!(1-x)^{N-n}x^n/(N-n)!n!. \tag{7}$$

The Curie temperature is then calculated by solving Eqs. (1) and (2) together[6]

$$T_C=\tfrac{1}{2}(T_{AA}+T_{BB})+[\tfrac{1}{4}(T_{AA}-T_{BB})^2+T_{AB}T_{BA}]^{\frac{1}{2}}. \tag{8}$$

IV. RESULTS AND DISCUSSION

Figure 1 shows the magnetization-temperature curves of six FePd alloys at 100 Oe applied magnetic field. This applied field strength is chosen because the measured magnetization-temperature curves are close to the ones at zero applied field. In addition, the coercivities of all the specimens were measured to be less than 500 Oe at room temperature, and they decrease rapidly with increasing temperature. Therefore, the applied field of 100 Oe is sufficient to saturate the specimens at temperatures close to the Curie temperature. The Curie temperatures are estimated from the point of inflection in the moment-temperature curves. The results are listed in Table I and also plotted in Fig. 2. Unlike the data in the literature,[2–6] our experimental results show a consistent effect of atomic ordering on the Curie temperature. It can be seen that the atomic ordering decreases the Curie temperature for FePd alloys in the L1$_0$ phase region.

Before we can do the simulation, we need to know how the magnetic moment changes with its local environment, more specifically, the functions of $\mu_{\mathrm{Fe}}^{(0)}(n_{\mathrm{Pd}})$ and $\mu_{\mathrm{Pd}}^{(0)}\times(n_{\mathrm{Fe}})$. The average atomic moment can be calculated as, $\bar{\mu}_{\mathrm{Fe}}(x)=\Sigma_{n_{\mathrm{Pd}}=0}^{12}P(n_{\mathrm{Pd}},x)\mu_{\mathrm{Fe}}(n_{\mathrm{Pd}})$, $\bar{\mu}_{\mathrm{Pd}}(x)=\Sigma_{n_{\mathrm{Fe}}=0}^{12}P(n_{\mathrm{Fe}},1-x)\mu_{\mathrm{Pd}}(n_{\mathrm{Fe}})$, and $\bar{\mu}(x)=(1-x)\bar{\mu}_{\mathrm{Fe}}(x)+x\bar{\mu}_{\mathrm{Pd}}(x)$. If we assume $\mu_{\mathrm{Fe}}^{(0)}(n_{\mathrm{Pd}})=\mu_{\mathrm{Fe}}(n_{\mathrm{Pd}})$ and $\mu_{\mathrm{Pd}}^{(0)}(n_{\mathrm{Fe}})=\mu_{\mathrm{Pd}}(n_{\mathrm{Fe}})$, the magnetic moment changing with its local environment can be obtained by fitting $\bar{\mu}_{\mathrm{Fe}}(x)$ and $\bar{\mu}_{\mathrm{Pd}}(x)$, where $\bar{\mu}_{\mathrm{Fe}}(x)$ and $\bar{\mu}_{\mathrm{Pd}}(x)$ can be found in the literature.[9]

The following exchange coupling constants are used in the simulation to fit our experimental data: $J_{\mathrm{Fe-Fe}}=0.0022\,\mathrm{eV}$, $J_{\mathrm{Pd-Pd}}=0.00086\,\mathrm{eV}$, and $J_{\mathrm{Fe-Pd}}=0.0108\,\mathrm{eV}$.

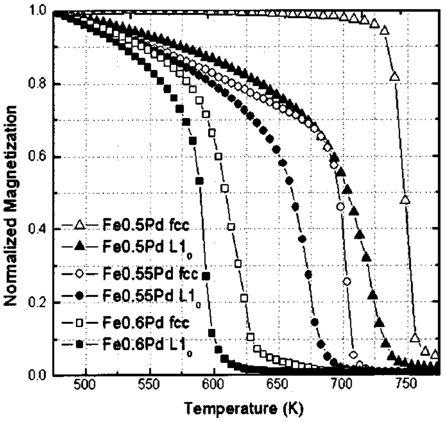

FIG. 1. (a) Magnetic moment vs temperature curve for the fcc and L1$_0$ FePd alloys. All the magnetic moments were normalized.

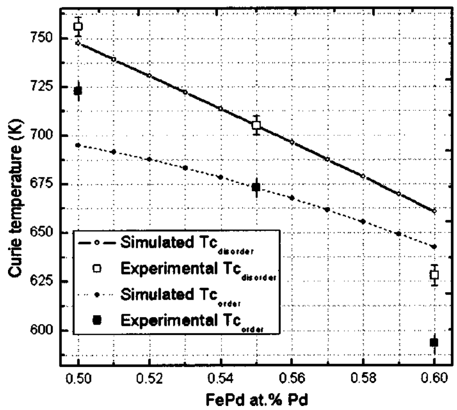

FIG. 2. The Curie temperature (T_C) changes with composition for the fcc and L1_0 Fe–Pd alloys. The open squares are the experimental T_C for the disordered phase. The solid squares are the experimental T_C for the ordered phase. The solid line is the simulated T_C for the disordered phase. The dashed line is the simulated T_C for the ordered phase.

The positive signs of all three exchange coupling constants indicate that the Fe–Fe, Pd–Pd, and Fe–Pd are ferromagnetically exchange coupled.[6] The simulated Curie temperatures are plotted in Fig. 2. From Fig. 2, we can see that the simulated results predict the same effect of atomic ordering on the Curie temperature as the experimental data. The difference between the experimental data and the theoretical fitting results is probably due to the critical fluctuations,[7] and also the model simplification. In this model, we only include the interactions with the nearest-neighbor atoms and ignore the interactions with other neighbors. In addition, the energy differences among different atomic configurations are not distinguished. In the future work, we will include these two factors into our model.

During the simulation, we assumed the exchange coupling constant of Fe–Fe ($J_{\text{Fe-Fe}}$) as 0.0022 eV. This value is smaller than the exchange coupling constant of Fe–Pd ($J_{\text{Fe-Pd}}=0.0108\,\text{eV}$), and seems to be contrary to fact the Fe is a strong ferromagnetic material. However, the strong ferromagnetic coupling of Fe atoms is in the bcc Fe phase. In the Fe–Pd alloys, Fe atoms are in the fcc arrangement. The bulk fcc Fe becomes antiferromagnetically exchange coupled at low temperature (67 K) and is paramagnetic at room temperature.[4] It has weak ferromagnetism in the form of ultrathin films.[10] Therefore, we think these exchange coupling constants are reasonable.

In FePd, CoPt, and NiPt alloys, atoms are ferromagnetically coupled for both the ordered phase and the disordered phase. Studies have shown that in NiPt and CoPt alloys the atomic ordering also decreases the Curie temperature.[11] A cluster variation method, with tetrahedron approximation, has been used to simulate the phase diagram of CoPt and NiPt alloys.[12] The simulation parameters were chosen as following: $J_{\text{Ni-Ni}}=0.005\,25\,\text{eV}$, $J_{\text{Ni-Pt}}=0.003\,82\,\text{eV}$, and $J_{\text{Co-Co}}=0.012\,\text{eV}$. These parameters are within the same order of magnitude as our data. Compared with the cluster variation method, the molecular field model provides simplified equations to calculate the Curie temperatures.

V. CONCLUSION

We measured the Curie temperatures of fcc disordered and L1_0 ordered Fe–Pd alloys in the L1_0 composition region. The results show a consistent atomic ordering effect on the Curie temperature: atomic ordering reduces the Curie temperature. This ordering effect can be explained using a molecular field model. The simulation results match well with the experimental data.

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