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Ab initio-Monte Carlo Studies on the Finite-Temperature Properties of $L1_0$ FeAu Superlattice

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Based on a Heisenberg model with exchange parameters extracted from *ab initio* calculations, Monte Carlo (MC) simulations are carried out to study the finite-temperature properties of the $L1_0$ ordered FeAu superlattice. The magnetization and the specific heat are calculated as the functions of temperature, and the Curie temperature is determined by the *ab initio*-MC method. These results are discussed in connection with experiments and previous theoretical studies.

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Keywords: *exchange interaction, Curie temperature, FeAu, Monte Carlo simulation*

1. Introduction

FeAu systems have received much attention both experimentally[1–6] and theoretically.[7–13] Although neither intermediate phase nor intermetallic compound exists naturally in the equilibrium phase diagram, the metastable $L1_0$ superlattices and the fine layered Fe/Au multilayers can be fabricated artificially by the molecular beam epitaxy technique.[1–3] It has been reported that the $L1_0$ FeAu ordered superlattice is ferromagnetic with enhanced magnetic moment and perpendicular anisotropy.[1,2] Several theoretical studies including our previous works[7–12] have been performed on the magnetic and electronical properties of $L1_0$ type 3d/(Au, Ag, Cu) monatomic superlattices.[8,10–12] These theoretical works have revealed the ground state behaviors such as the stabilities of spin configurations, the interplay between magnetization and equilibrium volume, and so on. From these theoretical studies the stable magnetic structures, especially some antiferromagnetic configurations can now be predicted by comparing total energies, and equilibrium atomic geometry and lattice constants[10,11] can also be determined. However, although these theoretical works have given good understanding to the ground state magnetism, the finite-temperature magnetism, which is more important for practical use, is still a challenging problem to theoretical research.

On the finite-temperature magnetism, several pioneering theoretical works have been successfully performed on the bulk magnetic metals. You *et al.*[14] calculated exchange parameters of bcc Fe from the selected four types spin arrangements, and estimated the Curie temperature by mean-field theory. Uhl and Kübler[15] investigated the finite-temperature properties of Fe, Co, and Ni by employing an exchange-coupled spin-fluctuation theory. Recently, Rosengaad and Johansson[16] examined the finite-temperature properties of ferromagnetic bcc Fe, fcc Co and Ni by using the Monte Carlo (MC) simulation with the exchange parameters deduced from the linear-muffin-tin orbital atomic-sphere approximation total energy of selected spiral spin-density wave magnetic structures. Zhou *et al.*[17] used a similar procedure to study the magnetic phase transitions in fcc Fe and Mn antiferromagnets with exchange parameters obtained by fitting to total energy of frozen collinear magnetic states. The method proposed by above two groups, namely the *Ab initio*-MC method, may be the only one approach of the first-principles calculations at present stage, so that the Curie temperatures determined by this method are the best estimations compared with experiments.[16,17]

In the present study, we apply the *Ab initio*-MC method on the $L1_0$ ordered FeAu superlattice. First we use the self-consistent full-potential linearized augmented-plane-wave (FLAPW) method to calculate the magnetic structure in the $L1_0$ FeAu superlattice, and extract the exchange parameters from this obtained *ab initio* total energies. Then, based on the Heisenberg model, we use the exchange parameters to perform MC simulations to study the finite-temperature magnetic properties.

2. First-principles Calculations on The Structure and Exchange Interaction

First-principles calculations in the present study are performed by using the self-consistent full-potential linearized augmented-plane-wave (FLAPW) method[18] under generalized gradient approximation (GGA)[19,20] in a scalar relativistic version without spin-orbit coupling. This method is one of the most accurate schemes for the electronic structure calculations and magnetic properties of crystals.[21] For the tetragonal $L1_0$ ordered structure, which will be considered in this work, five antiferromagnetic configurations[10,11,13] are selected. In spin-polarized calculations, the Brillouin zone sampling is performed using $90 \sim 156$ special k-points in the irreducible Brillouin zone. The muffin-tin radii of Fe and Au are set to $R_{\mathrm{Au}} = \sqrt{2}\,a/4$ and $R_{\mathrm{Fe}} = \sqrt{a^2 + c^2}/2 - R_{\mathrm{Au}}$. The energy cutoff parameter is fixed to be $R_{\mathrm{MT}}K_{\max} = 8.0$ in the present calculations. To find the ground state structures of these sys-

tems, the total energies are fully minimized with respect to lattice constants a and c. The most stable magnetic states, as reported in our previous papers for FeAu[8] and FeAg[10,11] are ferromagnetic with enhanced magnetic moments. In Fig. 1 the estimated total energies versus the lattice parameter c are plotted. It is seen that GGA gives more accurate lattice constant ($a = 0.399$ nm, $c = 0.375$ nm, $c/a = 0.94$) than LSDA ($a = 0.397$ nm, $c = 0.357$ nm, $c/a = 0.90$)[8] compared with the experimental data ($a = 0.399$ nm, $c = 0.383$ nm, $c/a = 0.96$) given by Takanashi *et al.*[1,2] It is noted that the in-plane lattice parameters a is mainly determined by the lattice parameters of Au with similar values in both GGA and LSDA and the out-plane lattice parameters c is much dependent on the magnetic moments of Fe atoms, so that different c are obtained with both GGA ($m_{\mathrm{Fe}} = 2.99\mu_{\mathrm{B}}$) and LSDA ($m_{\mathrm{Fe}} = 2.76\mu_{\mathrm{B}}$).

In Table 1, the relative total energies for five antiferromagnetic states with the lattice constants $a = 0.399$ nm, $c = 0.375$ nm are summarized. By using these relative total energies, the exchange parameters of the following Heisenberg model are determined:

$$E_{\mathrm{T}} = E_{\mathrm{PM}} + \sum_i E_{\mathrm{M}} - \frac{1}{2}\sum_{i,j} J_{ij}\sigma_i\sigma_j,$$

where E_{PM} is the total energy in the PM state, E_M the single ion magnetizing energy, σ_i the projection of the unit vector in the spin direction, J_{ij} exchange integral, and the sum runs over all pairs of lattice sites $\langle i, j\rangle$. We consider here the nearest neighbor (NN) and the next nearest neighbor (NNN) for the in-plane (J_1 and J_2) pairs and between interlayer (J_{1L} and J_{2L}) planes, and estimate these reduced parameters by least square fitting.[10,11] In Table 1, the obtained exchange parameters J_1, J_2, J_{1L} and J_{2L} are listed. These exchange parameters are used in the MC simulations to study the finite-temperature properties.

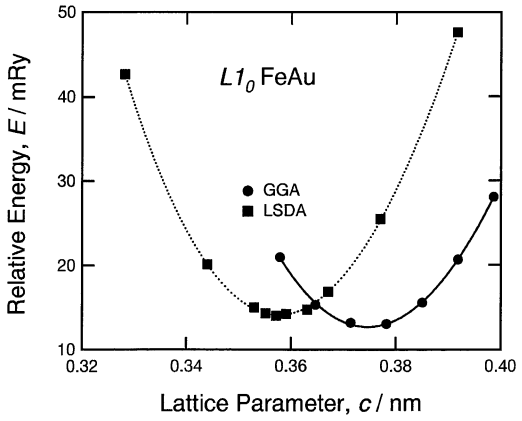

Fig. 1 Relative total energies as functions of lattice parameter c for the $L1_0$ ordered FeAu in the ferromagnetic states, where lattice parameter $a = 0.399$ nm for GGA, and $a = 0.397$ nm for LSDA.[7]

Table 1 Relative total energies ($\Delta E_i = E_{AF_i} - E_{FM}$), exchange integrals ($J_n$), Curie temperature ($T_c$) and critical exponent (β) for $L1_0$ FeAu superlattices.

Magnetic structure	$AF1$	$AF2$	$AF3$	$AF4$	$AF5$	
ΔE_i (mRy)	62.1	25.7	2.4	16.1	26.2	
	J_1 (mRy)	J_2 (mRy)	J_{1L} (mRy)	J_{2L} (mRy)	T_c (K)	β
FeAu	6.383	−0.574	2.080	−0.447	698	0.365
FePd[23]					728.6	0.377

3. Monte Carlo Simulation of Phase Transition

Using the exchange parameters given in Table 1, classical MC simulations with system size $N \times N \times N$ ($N = 16$, 24, 32) were performed to 6000 MC steps. For each system, the magnetization M, the specific heat $C = \frac{\langle E^2\rangle - \langle E\rangle^2}{k_{\mathrm{B}}T^2}$, and the susceptibility $\chi = \frac{\langle M^2\rangle - \langle |M|\rangle^2}{k_{\mathrm{B}}T}$ are calculated as functions of the temperature. We extrapolate these calculations to the thermodynamic limit using standard finite-size scaling theory. In principle, Curie temperature T_c can be estimated from the peak of the specific heat, or from the magnetic susceptibility according to the Curie-Weiss' law. While the actual values of the specific heat and magnetic susceptibility depend on the number of the system sizes, the fourth-order cumulant

$$U_L = 1 - \frac{\langle M^4\rangle_L}{\langle M^2\rangle_L^2}$$

are used in order to accurately determine the transition temperature. Theoretical results indicate that for $T < T_c$, U_L tends to 2/3, and for $T > T_c$, U_L decreases towards zero.[22] This behavior of the cumulant makes it very useful to obtain estimates of T_c itself which are not biased by any assumptions about critical exponents. In the present study, U_L versus temperature for various system sizes are plotted and T_c are estimated from the common intersection point of the U_L curves.[22]

The magnetization (M), the fourth-order cumulant (U_L), and the specific heat (C) for $L1_0$ FeAu as the functions of temperature are shown in Fig. 2. From the calculated $U_L - T$ curve, T_c for $L1_0$ FeAu is estimated to be 698 K. To our knowledge, experimental report on the T_c value of $L1_0$ FeAu superlattice is still lacking. However, this value is close to 728.6 K of $L1_0$ FePd superlattice,[23] which is believed to be similar to the $L1_0$ FeAu alloy. It is noted that the calculated T_c is larger than ~550 K of the fcc disorder $Fe_{50}Au_{50}$ alloy.[24] The difference of T_c between the disordered and ordered alloys were also found in FePt and FePd systems.[23,25] For instance, T_c for disordered fcc FePt is 530 K with respect to 750 K for ordered $L1_0$ FePt.[25] The reason of the difference in T_c is regarded as that some Fe atoms or clusters may loss their magnetization in the disordered fcc alloys[26] due to segregation fluctuation.

Finite-size scaling also provides information on the static critical exponents of the magnetization. The magnetization (M) as a function of the temperature near T_c is generally expressed as the form $M(T)/M_0 \propto (1 - T/T_c)^{\beta}$, where M_0 is the magnetization at 0 K and β is the critical exponent. By using T_c extracted from the $U_L - T$ curve, the exponent β is deduced from the slope of the $\log \frac{M}{M_0} - \log \frac{T_c - T}{T_c}$ plot with the least square fitting technique, as is shown in Fig. 3 for the $L1_0$ FeAu superlattice. From this method, the value of β is estimated to be 0.36 ± 0.02 (with $T_c = 695 \sim 702$ K), which

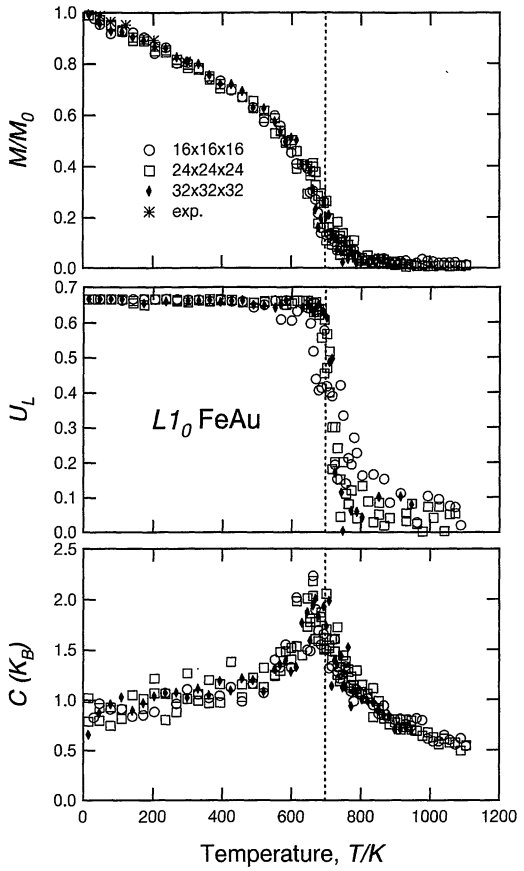

Fig. 2 Temperature dependence of the magnetization (M), fourth-order cumulant (U_L), and specific heat (C) for $L1_0$ ordered FeAu with system size changed from 16 × 16 × 16 to 32 × 32 × 32. * indicates the experimental data.[1,2]

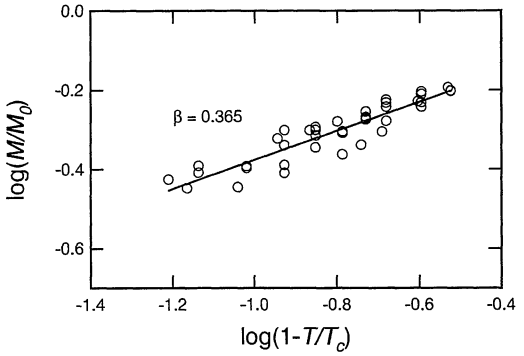

Fig. 3 Plot of $\log \frac{M}{M_0}$ versus $\log \frac{T_c - T}{T_c}$ for $L1_0$ ordered FeAu with $T_c = 698$ K.

agrees well with 0.3646 of the renormalization group scaling theoretical calculation for the three dimensional Heisenberg model[27] and also close to 0.377 of $L1_0$ FePd alloy reported by Longworth.[23] The former verifies our MC calculations with the Heisenberg model, and the latter demonstrates that the Heisenberg model can work well to describe the critical phenomenon.

4. Summary

In summary, we have performed first-principles calculations on the $L1_0$ ordered FeAu superlattice. It is shown that GGA gives more accurate lattice constants than LSDA compared to the experimental data. By using MC simulations with exchange parameters extracted from *ab initio* results, the magnetization and specific heat as functions of temperature are obtained. The Curie temperature T_c and critical exponent β are estimated to be 698 K and 0.36, respectively. These results indicate that the *ab initio*-MC method works well to describe the critical phenomena of magnets.

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