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Monte Carlo simulation of magnetic nanostructured thin films*

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Using Monte Carlo simulation, we have compared the magnetic properties between nanostructured thin films and two-dimensional crystalline solids. The dependence of nanostructured properties on the interaction between particles that constitute the nanostructured thin films is also studied. The result shows that the parameters in the interaction potential have an important effect on the properties of nanostructured thin films at the transition temperatures.

Keywords: nanostructured materials, Monte Carlo simulation, Metropolis sampling
PACC: 7540, 6860, 0500, 0550

1. Introduction

In recent years, nanostructured materials have received much attention, and investigations on nanostructured systems in the past several decades have undergone a dramatic development, covering a wide range of fields such as physics, chemistry, biology, electronics, and computer science.[1] In the technology core of information development, the physical and economic limits to the geometrical scaling of semiconductor devices also call for a substitute to win the challenge in the upcoming nanotechnology revolution. Nanostructured materials are found to be good candidates to serve this purpose in a wide range of areas. Among them, nanostructured films are the most commonly used. One can see papers on the physical properties of nanostructured materials, such as nanostructured Si thin films [2,3] and nanostructured gold [4] in the literature. Most of these works focused on experimental studies, though there are some theoretical investigations on magnetic nanostructured materials.[5,6] How to build a model for nanostructured ultrathin films still remains a challenge.

To know the physical properties of nanostructured materials, a useful approach is to apply statistical physics. It has been proved to be a successful tool in the materials science, symbolized by the renormalization group theory [7–9] developed by Wilson *et al* on the basis of the technique originating from particle physics. However, most of these great advances were applied to bulk materials like crystalline solids. Although we are discussing nanostructured materials that are characterized by their structural hetereogenity, we are, in most cases, frustrated by the difficulties involved in how to analytically apply statistical physics such as renormalization group to them. Fortunately, the development of computer science has provided us with the power to deal with what would be otherwise impossible in the study of these materials. Computational methods such as Monte Carlo simulation or molecular dynamics are examples of this improvement. There have been a number of papers on Monte Carlo simulation of two-dimensional (2D) magnetic crystals. However, most of them focused on crystalline solids,[10] though there were some reports on defects.[11] The study of nanostructured materials with the help of Monte Carlo simulation is a new topic. In this paper we apply classical Monte Carlo simulation to the study of the physical properties of nanostructured materials in thermodynamic equilibrium.

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2. Interaction energy

Nanomaterials differ from bulk materials in that they are composed of two parts: coarse grains and interfacial components. This inhomogeneity makes nanostructured materials exhibit special properties that can be possibly applied in many technology frontiers, such as nanoelectronics and biotechnology. To study such materials, it is important to start from a typical magnetic system placed in a weak external magnetic field, which is of many degrees of freedom and is composed of particles interacting with each other. Such a system can be described by a Hamiltonian [12,13]

$$H = \sum_i K_i + \sum_{i>j} A_{ij} - \sum_i h_i \cdot S_i, \qquad (1)$$

assuming that the system is incompressible. The components of Hamiltonian (1) are the kinetic energy, interaction between particles, and interaction between particles and external fields. It is useful to note that, if we are interested in the stationary states of the system, we may assume that the system varies with time in a very slow way, and the system can be treated as one that is always in equilibrium. The kinetic contribution in the above Hamiltonian will thus be cancelled out during the thermodynamic average calculation. In Eq.(1), K_i is the kinetic energy of the ith particle, h_i is the external magnetic field applied to the ith particle (in this paper, we apply a homogeneous field $\boldsymbol{h}$), and S_i is the spin vector of the ith particle. A_{ij} describes the interaction energy between the ith particle and the jth particle, which can be generally given by [13]

$$A_{ij} = J_{ij}\left[-\kappa\left(S_i \cdot S_j\right) + \alpha\right]. \qquad (2)$$

Here J_{ij} is the particle potential, and κ and α are the coefficients representing the contributions of spin and positional configuration respectively. From Eq.(2) it is apparent that $\kappa = 0$ and $\alpha = 1$ represents a nonmagnetic system, while $\kappa = 1$ and $\alpha = 0$ corresponds to an on-lattice Ising system of two-component spinoids. And $\kappa > 0$ is the ferromagnetic system and $\kappa < 0$ stands for the antiferromagnetic one. In this paper we will confine our attention to the discussion of off-lattice systems.

To study the properties of nanostructured materials, it is easy to know from Eq.(1) that the knowledge of the nonmagnetic particle potential J_{ij} in Eq.(2) becomes critically important. A convenient and most common treatment is to apply the Lennard-Jones potential

$$J_{ij} = 4\varepsilon\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right] \qquad (3)$$

to the interparticle interactions. Here r_{ij} is the distance between the ith and the jth particles normalized by a typical length σ, and ε is a constant representing the interaction strength. Figure 1 shows the dependence of the interaction on the normalized distance between particles.

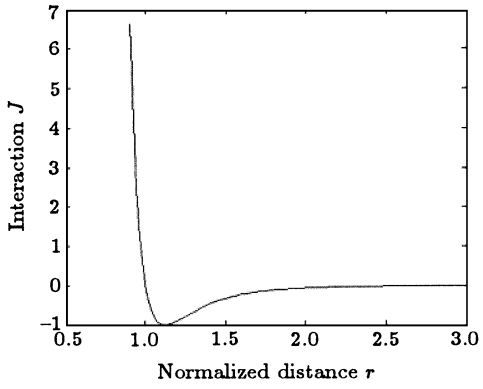


Fig.1. Interaction energy as a function of normalized distance between particles. Here we suppose $0 < \kappa < 1$.

3. Monte Carlo simulation

A simple and widely used method for the important sampling in Monte Carlo simulation is the Metropolis sampling,[14] based on the idea not of choosing the successive states $\{X_l\}$ independently of each other but to construct a Markov chain where each state $\{X_{l+1}\}$ is generated from a previous state via a transition probability that tends toward the equilibrium distribution when the sampling number is adequately large.[15] The Markovian process used in simulations ensures the system of interest to develop in the direction of low energy. For simplicity, we choose a two-dimensional (2D) magnetic nanostructured system for our simulation. It may be a solid material system such as that of metals or it can be soft material such as organic monolayers.[16−18] A 19×19 system plus a periodical boundary condition is used to simulate the 2D nanostructured films and thus the end effect can be neglected. Figure 2 shows the nanostructure from which we start the Monte Carlo simulation. It looks very much like the real nanostructured films

composed of coarse grains (CGs) with different orientations and boundary components. Trident boundaries and defects can also be found. 30,000 Monte Carlo steps (MCS) are taken into account.

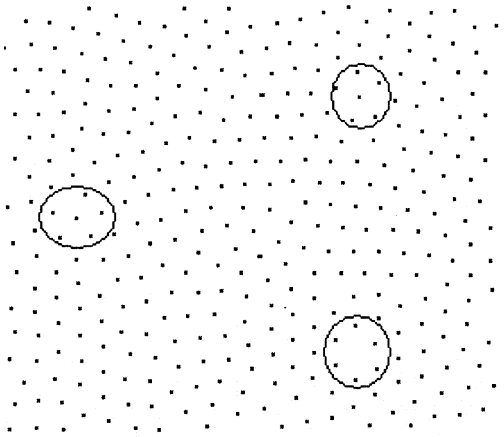


Fig.2. Nanostructure used for comparison between nanomaterials and crystalline solids. Areas enclosed are defects and interfaces.

For a system composed of N particles, $N(N-1)$ pairs of interaction energy have to be calculated per Monte Carlo step, which consumes a large amount of time in the simulation process. A time-saving and conventional treatment is to consider the interaction forces exerting on each single particle up to the 3rd nearest neighbours.[13] In addition, in order to avoid being trapped within metastable potential energy wells during the simulation process, we set the maximum displacement of each particle to be proportional to $\sqrt{T}$.[19]

To generalize the Monte Carlo simulation used in this paper, we make the following normalization to obtain dimensionless parameters:

$$r^* = r/\sigma, \quad T^* = Tk/\varepsilon,$$

$$E^* = E/\varepsilon, \quad h^* = h/(\varepsilon/m) \tag{4}$$

for distance r, temperature T, energy E and external field h respectively. Here k is the Boltzmann constant and m is a typical magnetic moment.

4. Correlation function

4.1. Density correlation function

In order to gain more insight into the structural correlation between particles, we introduce a density correlation function (DCF) [20]

$$\mathrm{DCF} = \rho_0(\rho_r - \rho_0) \tag{5}$$

to describe the structural relation among particles in the nanostructured material. Here ρ_0 is the average density of the area within radius r, and ρ_r is the density function at the distance r. For homogeneous systems, it is obvious that DCF=0.

4.2. Spin correlation function

Besides the configurational structure that is related to the DCF, the spin is also an important degree of freedom that has to be considered. Compared with the DCF, the spin correlation function (SCF) is defined as the average of the inner product of the spins S_n within the region of radius r and S_o at its centre:

$$\mathrm{SCF} = \frac{1}{N}\sum_{n}^{N_r}(S_o \cdot S_n), \tag{6}$$

where N_r is the total number of spins within the area of interest.

5. Simulation results and discussion

5.1. Comparison between nanostructured and crystalline films

Figures 3(a) and 3(b) are the magnetizations of crystalline films and nanostructured films respectively. Figure 3(c) shows their corresponding differentiations: $\mathrm{d}|m|/\mathrm{d}T$. A comparison of $|m|$ -T and their differentiations between nanostructured and crystalline films reveals that the Curie temperature of the nanostructure of interest is about 0.91, while the crystalline solid of the same average density possesses a higher Curie temperature, around 1.03. The reason for the lower Curie temperature for nanostructured films is that nanomaterials have a relatively looser structure and higher disorder, especially those located at the boundaries between crystalline grains.

From Fig.3(c), we also found two peaks at T^*=0.49 and T^*=0.91 respectively. (it can be seen from Fig.3(b) that the narrow peak at T^*=0.22 is generated by random errors.) In contrast, there is only one peak at T^*=1.03 for 2D crystalline films. The multiple peaks of the differentiations curves Fig.3(c) of nanostructured films are induced by the complexity and diversification of structures of nanomaterials, which leads to a metastable platform between 0.49 and 0.91 in $|m|$ -T. This characterizes not only the structure of interest (Fig.2) but also many nanostructured films. The number of peaks in Fig.3(c) depends on the number of boundaries of different sizes and

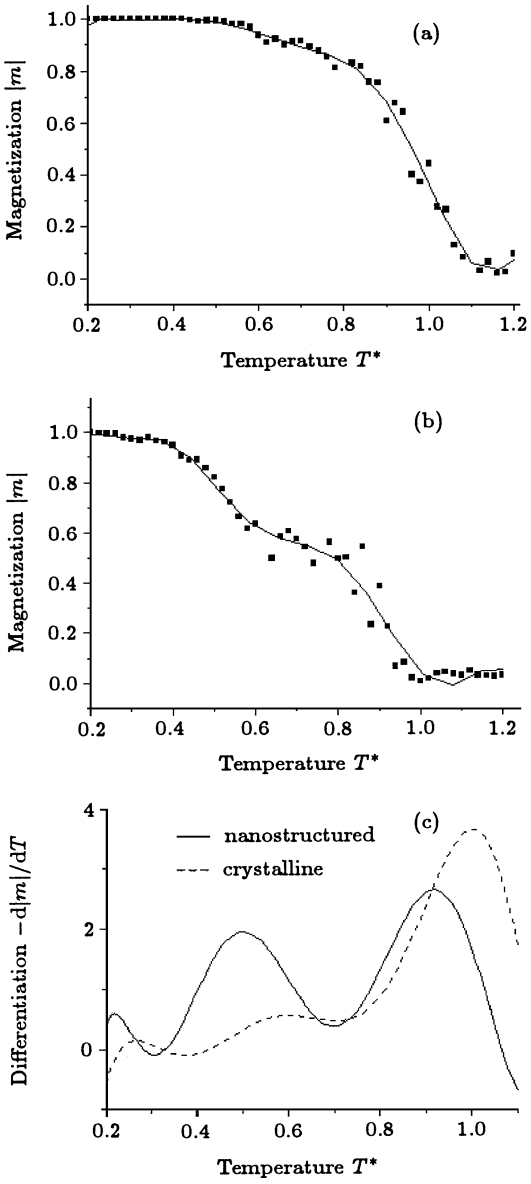


Fig.3. (a) Magnetization of crystalline films. (b) Magnetization of nanostructured films. (c) Magnetic susceptibilities of crystalline and nanostructured films.

structures as well as the distribution of the coarse grains. For further analysis, we have tracked the whole dynamic process and found that the particles at and around the boundaries and defects between crystalline grains play an important role. Figure 4 gives an intermediate state of the whole simulation process. As the temperature rises, most of the particles remain unchanged in the crystalline films, while in the nanostructured films, part of the particles at and around boundaries and defects (areas enclosed by circles) will change their orientations because of their high energy density distribution. This leads to the increase of their susceptibilities, and a peak at 0.49 appears. Raising temperature further will not generate new peaks until some other particles at and around boundaries and defects change their orientations. This process will continue until the system reorganizes itself into a similar one to that of crystalline films. The temperature 0.91 triggers the reorientation of most particles, corresponding to the Curie temperature where the magnetic phase transition occurs.

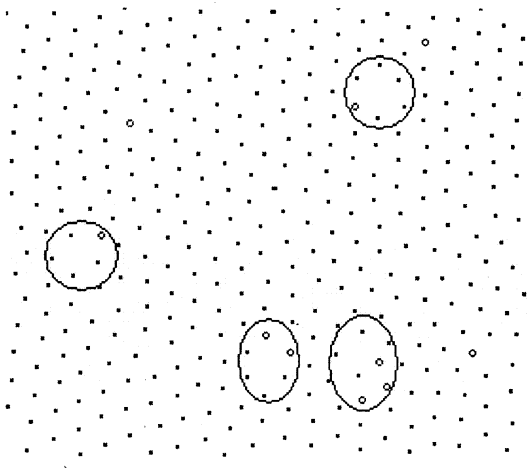


Fig.4. An intemediate state of the simulation process at T^*=0.5. Areas closed with circles stand for the particles at the interfaces.

The density of coarse grains and the dimensional size of grains, compared with the boundaries, will also influence the process of transition. Higher density requires higher temperature to destroy the structure of the grains. If the coarse grains cover absolutely larger areas than boundaries, the process of transition will resemble that of crystalline films. If the size of boundaries is comparable to that of grains, then the diversification of the system will generate some metastable states (short $|m|$ -T platforms), corresponding to the small peaks before the Curie temperature, i.e. before the transition occurs. Each peak represents a local structural change (local destruction of order) triggered by the increase of temperature.

5.2. Dependence of nanostructured properties on interparticle interaction

In order to investigate how the magnetic properties of nanostructured films depend on interparticle interactions, we start from a ferromagnetic superlattice consisting of one crystalline grain, as shown in Fig.5. A 2D periodical boundary condition is imposed on the structure to ensure that the simulation result can be extended to systems of infinite size. Three different κ are selected for simulation.

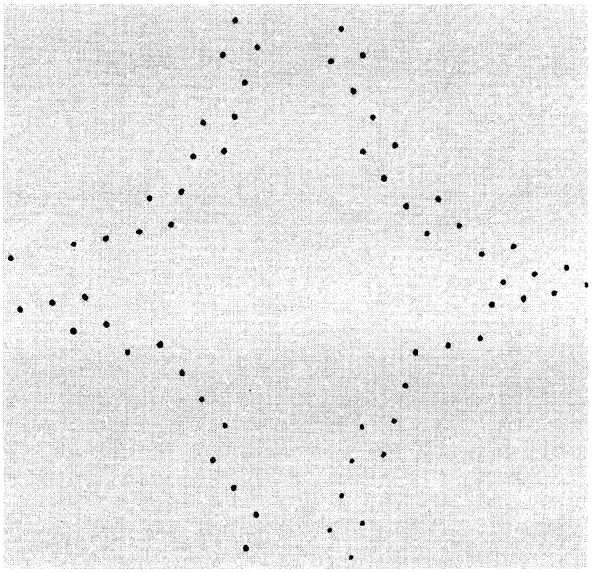


Fig.5. Initial configurations. White dots represent particles in the coarse grains and black dots are the particles in the interfacial components. Size $=18 \times 18$; lattice constant = 1.113201303556; disc number = 1; radius of discs = 8*lattice constant; MCS = 30000; h^*z=0.0001.

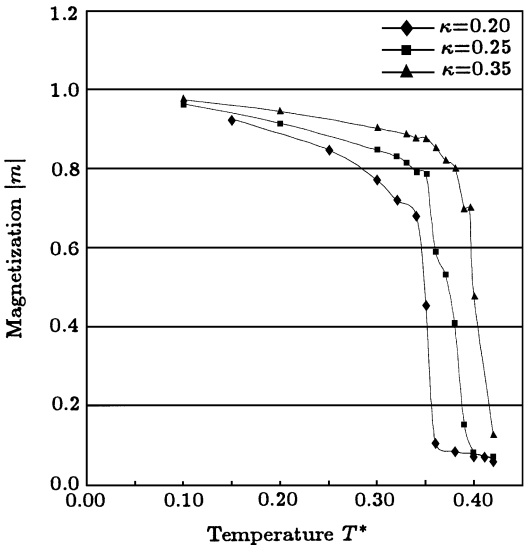


Fig.6. Magnetizations of the material with three different interaction coefficients.

Figure 6 shows the magnetizations of the nanostructured films of interest at different κ. Each point is obtained under the conditions of constant temperature during the relaxation process of simulation. The dependence of magnetization on κ in Fig.6 shows that the bigger the κ, the higher the Curie temperature. This is reasonable as κ is a coefficient exhibiting the magnetic contribution to the interaction formation. The simulation results of magnetization (Fig.6) also indicate that as the environmental temperature rises, the system becomes disordered. When the temperature crosses the transition point, the spin order vanishes and a further increase of temperature sees a collapse of the structural configuration of the system. This is nothing but the melting of the material. Further investigation by DCF and SCF (Fig.7) shows that DCF is almost invariant while SCF increases with κ. Such a feature becomes clearer as the environmental temperature increases.

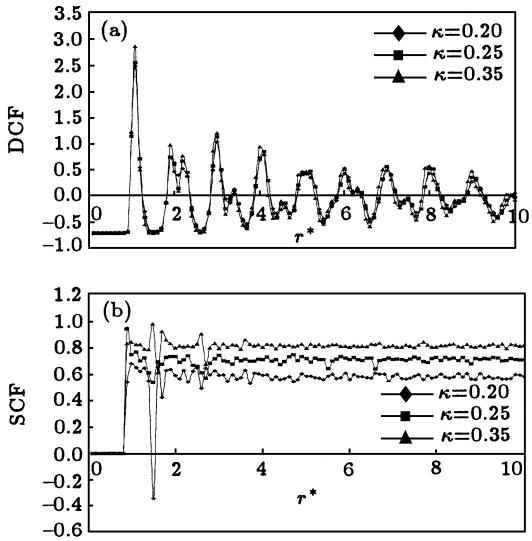


Fig.7. DCFs (a) and SCFs (b) of the nanostructured films at T^*=0.3 (α=0.75) with three different κ: κ=0.20; κ=0.25; κ=0.35.

6. Conclusion

Using Monte Carlo simulation, we found that the transition process of nanostructured magnetic films is more complicated as compared with that of crystalline ones. Several metastable states can be found. The two interaction coefficients κ and α govern the Curie temperature and the melting point of the material. It is also shown that the bigger the value κ, the higher the Curie temperature. The simulation results of the correlation functions also show that as temperature rises above Curie point, the system becomes disordered, which can be seen from DCF and SCF using different interaction coefficients.

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