Thin ferromagnetic films with competing surfaces: A Monte Carlo study of the classical Heisenberg model

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Monte Carlo simulations have been performed for different values of perpendicular anisotropy λ in a thin ferromagnetic Heisenberg film. In the model competing surface fields with the same magnitude but opposite direction have been used. In the Heisenberg limit, $\lambda \rightarrow 0$, no spontaneous magnetization of the film is observed. Whereas, in the Ising limit, $\lambda \rightarrow \infty$, nonzero magnetization of the film is observed below a critical temperature T_c and a degeneracy in the magnetization profiles exists between states of positive and negative total magnetization at low temperatures. The results of magnetic relaxation studies indicate that the magnetization decays exponentially with a relaxation time that increases with λ and decreases with temperature. [S0163-1829(97)13217-9]

I. INTRODUCTION

Phase transitions in thin ferromagnetic films have been investigated experimentally[1,2] and theoretically[3–9] due to their importance for applications in magnetic-recording media. The order parameter for the ferromagnetic-paramagnetic phase transition is the spontaneous magnetization vector **M**, which is zero for temperatures above a critical temperature T_c in zero external field.[10] However the inclusion of appropriate anisotropies and interactions in the energy can significantly modify the phase behavior. For a perpendicular anisotropy with long-range dipole-dipole interactions, Moschel and Usadel[3,4] have shown that the direction of **M** relative to the surface varies with increasing temperature from perpendicular to in-plane in a reorientational transition of a ferromagnetic Heisenberg ultrathin film. They note that the perpendicular anisotropy favors the spins being directed perpendicular to the surface whereas the long-range dipole-dipole interactions tend to align the spins in the in-plane direction. Moreover they showed that the spin cantings are not only affected by temperature but also by the model anisotropy parameters. This indicates that the direction of **M** in the ferromagnetic Heisenberg thin film is very sensitive to the anisotropy properties.

For ferromagnetic Ising thin films, both finite-size and surface effects can produce phase transitions as a function of temperature. Recent simulations[7–9] on the thin Ising film have shown that phase transitions may occur in the bulk region of the film due to the presence of competing surface forces which are external fields acting on the surfaces alone. In the case of a thin film with two surface fields of the same magnitude but opposite direction and zero bulk field, the surface fields favor a negative magnetization at one surface and a positive magnetization at the other surface. For sufficiently high temperatures the interface between the regions of negative and positive magnetization is located in the middle of film. However, Binder and co-workers[7–9] found that for temperatures below a critical temperature $T_c(D)$ which depends on the film thickness D, the interface is shifted from the center toward the one of the surfaces. The particular surface depends on magnetization fluctuations in the bulk region and, most especially, the initial spin configuration. The low-temperature magnetization profiles of the film $\mathbf{M_n}$ show a degeneracy between states of negative and positive total magnetization. This phase transition in the thin ferromagnetic Ising film is only observed for applying surface fields which have the same magnitude but opposite direction.

Thus, while Binder and co-workers[7–9] have shown the significance of surface effects on the phase behavior of thin Ising spin system, it is not clear how general this result is for all ferromagnetic systems, since the Ising model of magnetism uses a highly anisotropic spin-spin interaction. Ising spins do not rotate through all possible orientations, but instead are restricted to a particular axis, conventionally the z direction. Below the critical temperature, the spins tend to preferably align in the z direction and give rise to a finite value of **M** even in the absence of an external field.[11] However, in contrast, the classical Heisenberg model of magnetization, the magnetic spins are very sensitive to the temperature and only order at zero temperature in the absence of an external field. According to the investigation of Taylor and Gyorffy,[12] without any perpendicular anisotropy, there is no magnetic order at any finite temperature. The ferromagnetic order is destroyed by long-wavelength spin waves.[13,14] However different anisotropy constraints acting on spins at the surface and in the bulk can change the behavior of Heisenberg spins toward that of Ising-like spins.

This paper investigates the phase behavior and magnetization profiles of ferromagnetic thin Heisenberg films at different values of the perpendicular anisotropy and temperature. In the following section the model and simulation method are detailed. The equilibrium magnetic phase behavior of the model system is discussed in Sec. III and the temperature dependence of the magnetic relaxation is investigated in Sec. IV. The paper concludes with a summary of the key findings.

II. THE MODEL

The system under consideration is a three-dimensional ferromagnetic thin film of finite thickness D that is described by the Hamiltonian

$$\mathcal{H}=-J\sum_{\langle i,j\rangle}\mathbf{S}_i\cdot\mathbf{S_j}-\lambda\sum_i (S_i^z)^2-\sum_{i\in\text{surface }1}\mathbf{H_1}\cdot\mathbf{S_i}$$

$$-\sum_{i\in\text{surface }D}\mathbf{H_D}\cdot\mathbf{S_i}, \tag{1}$$

where $\mathbf{S_i}=(S_i^x,S_i^y,S_i^z)$ is a unit vector representing the ith spin and the notation $\langle i,j\rangle$ means that the sum is restricted to nearest-neighbor pairs of Heisenberg spins, each pair being counted only once. J is a coupling constant characterizing the exchange interaction which has a positive sign for ferromagnetism and λ determines the strength of the perpendicular anisotropy which is applied to spins throughout the whole film. As stated by Taylor and Gyorffy,[12] in the case of $\lambda=0$, the model is a classical Heisenberg spin system, while for $\lambda=+\infty$ it becomes an Ising model. It should be noted that the quadratic anisotropy term in Eq. (1) forces the spins to align along a z direction and minimizes the canting of the spins for increasing temperature. In this paper the perpendicular anisotropy was investigated over the range $0\leqslant\lambda\leqslant 0.5$ with larger values of λ only being used to facilitate comparison with the Ising model. $\mathbf{H_1}$ and $\mathbf{H_D}$ are the surface fields.

We consider a simple cubic lattice of size $L\times L\times D$, in units of the lattice spacing, and in the Monte Carlo simulation apply periodic boundary conditions in the x and y directions. Free boundary conditions are applied in the z direction which is of finite thickness D and the system is subject to surface fields applied a layer $n=l$ and $n=D$ of the film

$$\mathbf{H_1}=h\hat{\mathbf{z}}\delta_{i1}, \tag{2}$$

$$\mathbf{H_D}=-h\hat{\mathbf{z}}\delta_{iD}, \tag{3}$$

giving a Hamiltonian

$$\mathcal{H}=-J\sum_{\langle i,j\rangle}\mathbf{S_i}\cdot\mathbf{S_j}-\lambda\sum_i (S_i^z)^2$$

$$-h\left(\sum_{i\in\text{surface }1}S_i^z-\sum_{i\in\text{surface }D}S_i^z\right). \tag{4}$$

The film thickness $D=12$ and surface field strength $h=-0.55$ were used throughout and the simulations performed for lattices of size $L=16$, 32. The Metropolis algorithm[15] was used in the Monte Carlo simulations with trial configurations generated from Barker-Watts[16] spin rotations. The magnitude of the maximum spin rotation was adjusted to ensure approximately 50% of trial configurations were rejected in the bulk equilibrium state. For large values of λ, to ensure a rejection rate of approximately 50%, the Barker-Watts spin rotation was supplemented by a randomly selected spin flip. The z component of the magnetization for the film

$$M_z=\frac{1}{D}\sum_{n=1}^{D}M_n^z, \tag{5}$$

and the z component of the magnetization for the nth layer of the film

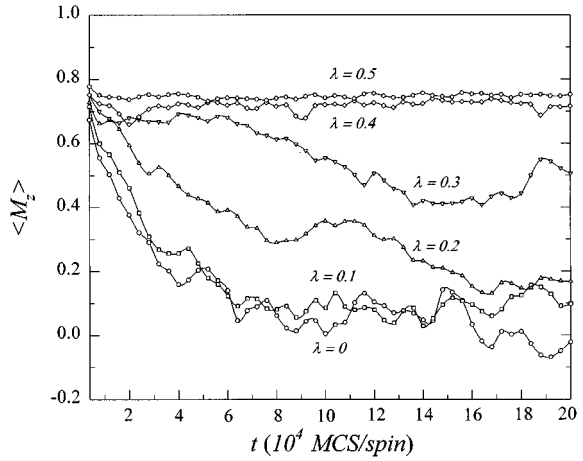

FIG. 1. Mean magnetization per spin vs time in units of Monte Carlo steps per spin for thin ferromagnetic Heisenberg films of size $16\times16\times12$ for different values of the perpendicular anisotropy in the range $0\leqslant\lambda\leqslant0.5$ from an initial spin state of $S_i^z=+1$ for all i at a temperature $T^*=1.0$. The curves through the points are only guides to the eye.

$$M_n^z=\frac{1}{L^2}\sum S_i^z \tag{6}$$

were determined for different values of λ and temperature T. The fluctuations in the magnetization were used to calculate the layer susceptibility χ_n which is given by

$$\chi_n=L^2(\langle M_n^{z2}\rangle-\langle M_n^z\rangle^2)/k_BT, \tag{7}$$

where k_B is Boltzmann's constant. Simulations were performed for up to 10^6 Monte Carlo steps per spin (MCS/spin) to ensure equilibration of systems in the Heisenberg limit ($\lambda\to0$).[17] Equilibrium averages were typically taken over 2×10^5 MCS/spin with initial transients ignored. For systems in the Ising limit ($\lambda\to\infty$), much shorter runs could be performed.

III. MAGNETIC PHASE BEHAVIOR

The simulations show that the z component of mean magnetization per spin, $\langle M_z\rangle$, depends on both λ and temperature T. Figure 1 shows the evolution of $\langle M_z\rangle$ with time from an initially ordered state at a reduced temperature of $T^*=k_BT/J=1.0$ for different values of λ from $\lambda=0$ to $\lambda=0.5$. In Fig. 1 MCS/spin is used as a unit of time and an initial spin state $S_i^z=+1$ was selected. For $\lambda=0.4$ and 0.5, the systems quickly approach equilibrium and equilibrium states of nonzero magnetization of the film persist. The temperature is well below $T_c(D)$ for the Ising system $T^*=4.0$.[8] However for $\lambda=0$, the spins continuously rotate to reach equilibrium at zero film magnetization. No spontaneous magnetization is observed even though $T<T_c(D)$ for the Ising system. For $\lambda=0$, i.e., an isotropic spin-spin interaction, the model is a classical Heisenberg spin system and the ordered spin states are quickly destroyed at finite temperature. At intermediate values of λ, $0<\lambda<0.4$, spontaneous magnetization of the film persists but the magnitude of the equilibrium magnetization of the film decreases with λ. Likewise the time to achieve equilibrium increases with λ. An-

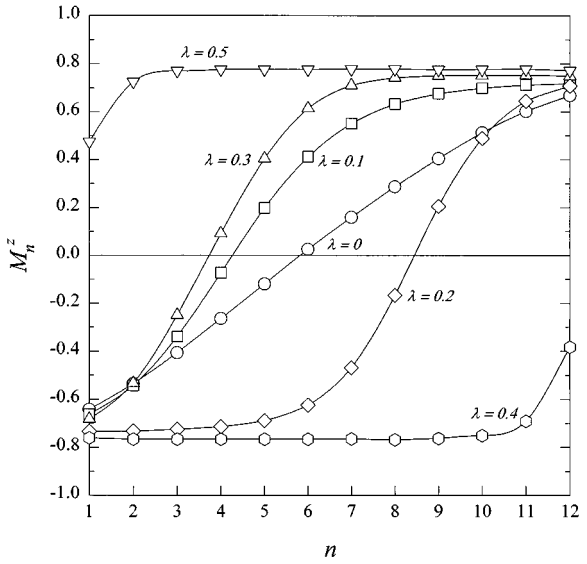

FIG. 2. Magnetization profiles across the film, M_n^z, vs layer number n for $D=12$ at a temperature $T^*=1.0$ with surface fields $H_1/J=-H_D/J=-0.55$. For $\lambda=0,0.1,0.3,0.5$ an initial spin state of $S_i^z=+1$ for all i was used, while for $\lambda=0.2,0.4$ an initial spin state of $S_i^z=-1$ for all i was used.

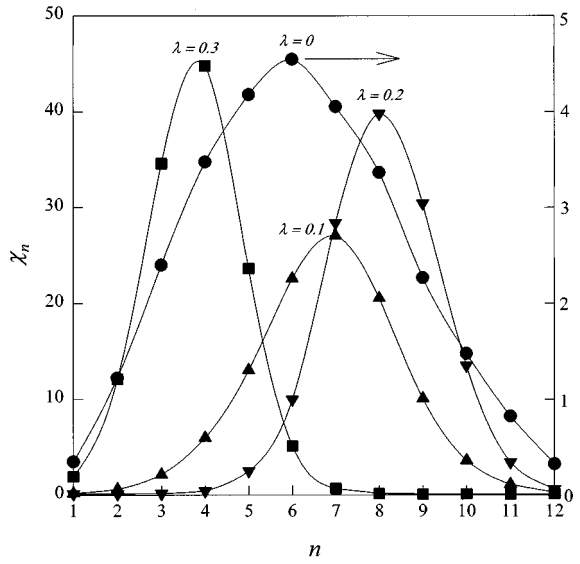

FIG. 3. Layer susceptibility χ_n vs layer number n for $D=12$ at a temperature $T^*=1.0$. For $\lambda=0,0.3$ an initial spin state of $S_i^z=+1$ for all i was used, while for $\lambda=0.1,0.2$ an initial spin state of $S_i^z=-1$ for all i was used. Note the change of scale for χ_n with $\lambda=0$. The curves drawn are only guides to the eye.

other aspect of the data in Fig. 1 for smaller λ is the pronounced fluctuations in $\langle M_z\rangle$. These arise for Heisenberg spin systems since the probability of spin flips becomes very small and metastable states occur due to strong magnetization in the x and y directions which averages to zero much quicker than for $\langle M_z\rangle$.[17]

The magnetization profiles across the film, M_n^z, for different λ at a temperature $T^*=1.0$ are shown in Fig. 2. For clarity the figure shows results for $\lambda=0$, 0.1, 0.3, 0.5 from an initial state of $S_i^z=+1$ and from an initial state of $S_i^z=-1$ for $\lambda=0.2$ and 0.4. It can be seen that the surface fields locally constrain the spins to align in the negative direction near one surface and positive direction near the other surface. In the bulk, the mean spin orientation of the layers varies smoothly from one surface to the other. For $\lambda=0$, the interface between regions of negative and positive magnetization is located in the center of the film. The interface moves from the center toward the surface for $\lambda>0$. The direction of the interface displacement depends on the initial spin configuration and a degeneracy exists between states of positive and negative total magnetization. However, for larger λ ($\lambda=0.4$ and 0.5), the interface disappears and spins are confined to one of the $\pm z$ directions according to their initial states to produce a large value of the film magnetization. The temperature is well below $T_c(D)$ for the Ising system ($\lambda\to\infty$).

Figure 3 shows the film profiles of susceptibility χ_n at a temperature $T^*=1.0$ for $\lambda=0,0.3$ from an initial configuration of $S_i^z=+1$ for all i and for $\lambda=0.1,0.2$ from an initial configuration of $S_i^z=-1$ for all i. Here for $\lambda=0$, we find a broad peak centered around the middle of the film. While, for $\lambda>0$ the peaks in the profiles are shifted toward the surface appropriate to the initial spin configuration. Moreover the peaks in χ_n for each λ are located in the same layer as the interfaces in the profiles of M_n^z, indicating larger fluctuations of spins in the interface.

The temperature dependence of the magnetization profiles is shown in Fig. 4 for $\lambda=0.2$. For clarity an initial state of $S_i^z=+1$ for all i is used for temperatures $T^*=0.8,1.0,1.4$, while an initial state of $S_i^z=-1$ for all i is used for temperatures $T^*=0.7,0.9,1.1$. At the highest temperature $T^*=1.4$, we find that the interface is located in the center of the film, between $n=6$ and $n=7$, and the mean film magnetization $\langle M_z\rangle$ is zero due to the symmetry of M_n^z about the middle of the film. However, for lower temperatures from $T^*=0.7$ to 1.1, the interface is shifted toward

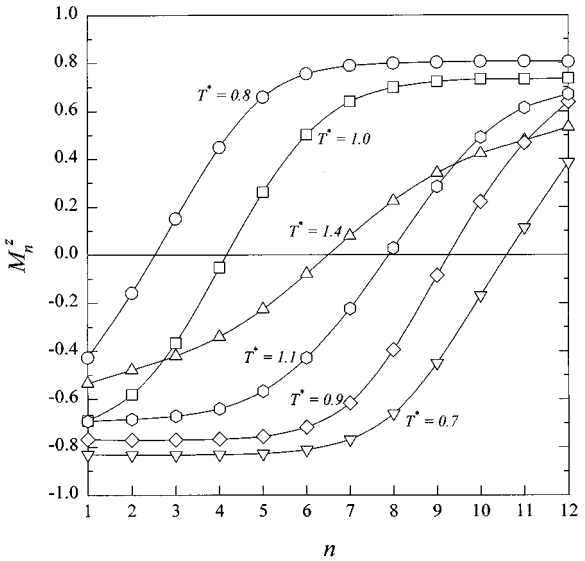

FIG. 4. Magnetization profiles across the film, M_n^z vs layer number n, for $D=12$ with $\lambda=0.2$ at different temperatures with surface fields $H_1/J=-H_D/J=-0.55$. An initial spin state of $S_i^z=+1$ for all i was used for $T^*=0.8,1.0,1.4$, while an initial spin state of $S_i^z=-1$ for all i was used for $T^*=0.7,0.9,1.1$.

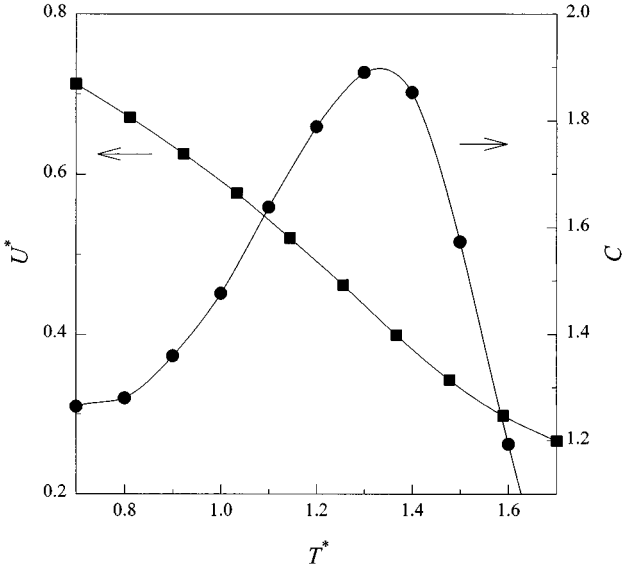

FIG. 5. Temperature dependence of the reduced energy $U^* = U/U_G$ and the specific heat $C = \partial U/\partial T^*$ for $\lambda = 0.2$.

the surface and a degeneracy exists between two states of the film magnetization. The selected state depends on the initial spin state. The film has a finite value of $\langle M_z \rangle$ at these temperatures. This behavior can be regarded as a remnant of Ising model behavior seen by Binder and co-workers.[7–9] When $\lambda = 0$, the model becomes a classical Heisenberg spin system which has no spontaneous magnetization at a nonzero temperature and the interface between negative and positive magnetization is always located in the center of the film.

It is of value to consider a critical temperature $T_c(\lambda, D)$ which is equivalent to that of Binder and co-workers,[7–9] in the Ising limit $\lambda \to \infty$. For $T > T_c$ the film shows no spontaneous magnetization with $\langle M_z \rangle = 0$, while for $T < T_c$ spontaneous magnetization with $\langle |M_z| \rangle > 0$ is observed. For $\lambda > 0.4$ we find $T_c^*(\lambda, D) = T_c^*(\infty, D) = 4.0$, while for $\lambda = 0$ we have $T_c^*(0, D) = 0$. Figure 5 plots the reduced energy $U^* = U/U_G$, where U_G is the ground-state energy, and specific heat $C = \partial U/\partial T^*$ as a function of temperature for $\lambda = 0.2$ and shows $T_c^*(\lambda = 0.2,\ D = 12) \cong 1.3$. The critical temperature $T_c(\lambda, D)$ reduces smoothly from the Ising limit value to the Heisenberg value as λ decreases from 0.4 down to zero.

IV. DEPENDENCE OF THE MAGNETIC RELAXATION ON λ AND ON THE TEMPERATURE

The time dependence of the magnetic relaxation is investigated here for different values of λ and temperature. In these studies, as elsewhere,[18] we focus on the role of λ and temperature in determining the relative magnetic relaxation behavior of the Heisenberg spin systems and do not attempt to obtain absolute relaxation times. In Fig. 6(a), for different λ from $\lambda = 0$ to $\lambda = 0.8$, the ratio of time-dependent magnetization to the initial magnetization, $M_z(t)/M_z(0)$, is shown as a function of time at temperature $T^* = 1.5$ for $h = 0$ from an initially ordered state with $S_i^z = +1$ for all i. Comparison with the results in Fig. 1 for a similar system with $h = -0.55$ shows that equilibrium was obtained in a shorter time for $h = 0$. In general the relaxation time increases with the

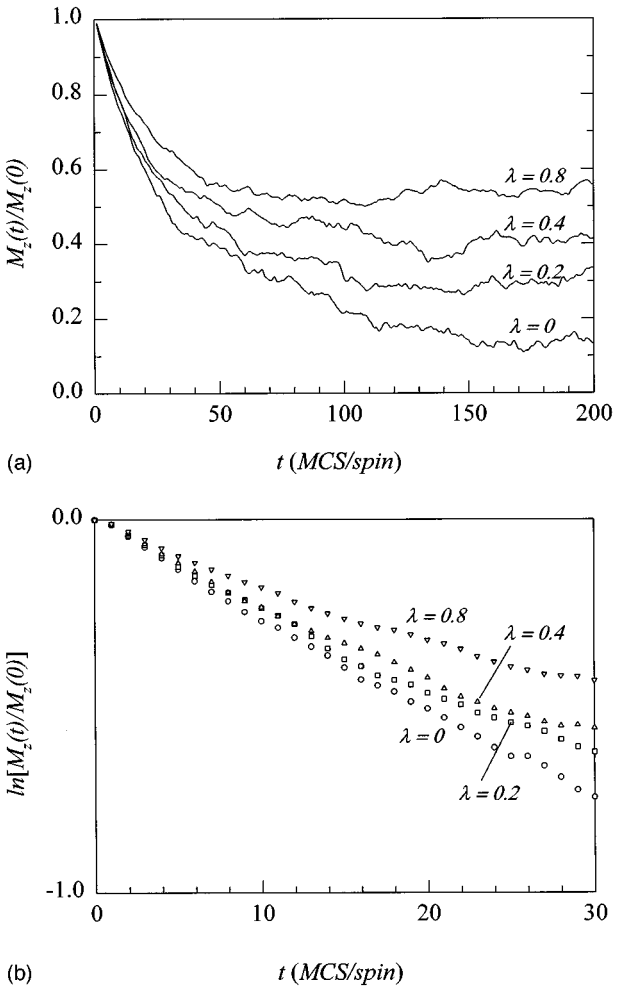

FIG. 6. Relaxation of the film magnetization with time for different values of λ with zero surface field at a temperature $T^* = 1.5$: (a) reduced magnetization $M_z(t)/M_z(0)$ vs time and (b) $\ln[M_z(t)/M_z(0)]$ vs time.

surface field. The results of Fig. 6(a) also show a faster decay of the initial state is observed for smaller λ but that the time required to achieve the equilibrium is increased. For $\lambda = 0.8$, in the Ising limit $\lambda \to \infty$, a much shorter time is required to an equilibrium and produce finite value of magnetization. Figure 6(b) shows the magnetic relaxation on a natural logarithm scale, $\ln[M_z(t)/M_z(0)]$, as function of time for different λ. The linear character of the curves for short times indicates that the initial magnetic relaxation can be characterized by an exponential decay and the magnetic relaxation can be written as

TABLE I. Relaxation time τ for thin Heisenberg film with $D = 12$, temperature $T^* = 1.5$ and zero surface field $h = 0$ for perpendicular anisotropy λ.

λ	τ (MCS/spin)
0	46.0 ± 0.6
0.2	53.1 ± 0.6
0.4	59.2 ± 0.7
0.8	75.5 ± 0.6

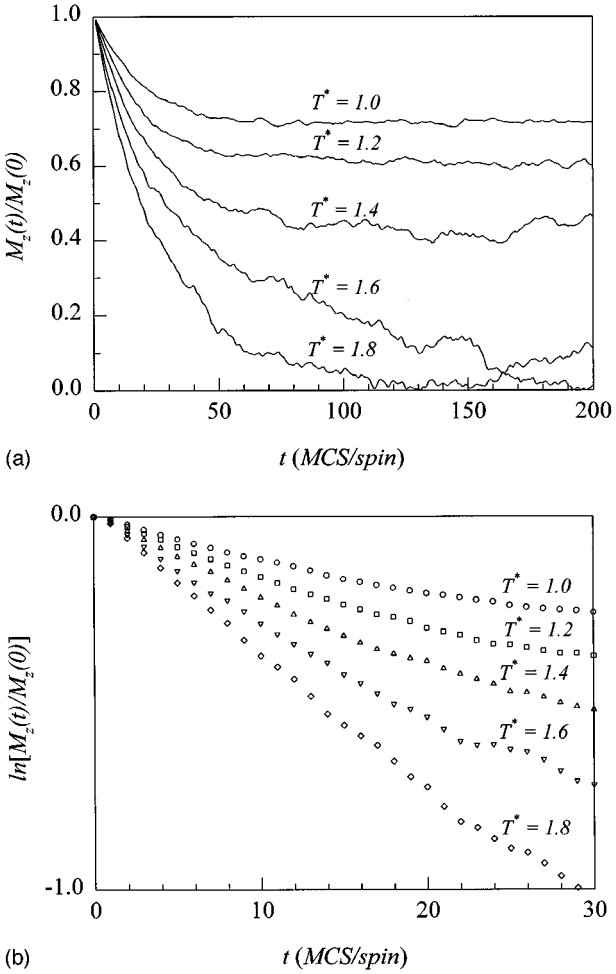

FIG. 7. Relaxation of the film magnetization with time for $\lambda = 0.2$ with different temperatures and zero surface field: (a) reduced magnetization $M_z(t)/M_z(0)$ vs time and (b) $\ln[M_z(t)/M_z(0)]$ vs time.

$$\frac{M_z(t)}{M_z(0)} = \exp(-t/\tau(\lambda, T)), \tag{8}$$

where τ is a relaxation time. Table I gives the relaxation times for different values of λ at $T^* = 1.5$ corresponding to the systems in Fig. 6 with error estimates obtained from ten repetitions with different random number sequences.

TABLE II. Relaxation time τ for thin Heisenberg film with $D = 12$, perpendicular anisotropy $\lambda = 0.2$, and zero surface field $h = 0$ at temperature T^*.

T^*	τ (MCS/spin)
1.0	133.1±1.0
1.2	92.2±1.0
1.4	63.1±1.3
1.6	41.8±0.9
1.8	28.8±0.6

The temperature dependence of the magnetic relaxation is shown in Fig. 7(a) for $\lambda = 0.2$. Again the decay of the magnetization is monitored for $h = 0$ with an initial state of $S_i^z = +1$ for all i. The rate of decay of the initial state is greater for higher temperatures, but the time to achieve equilibrium also increases with temperature. Once more the initial magnetic relaxation is governed by an exponential decay as shown in Fig. 7(b). Table II gives the relaxation times for $1.0 \leqslant T^* \leqslant 1.8$ at $\lambda = 0.2$ corresponding to the systems in Fig. 7 with error estimates obtained from ten repetitions with different random number sequences.

V. CONCLUSION

We have studied the phase behavior of thin ferromagnetic films with Heisenberg systems and competing surface fields. The perpendicular anisotropy λ in the Hamiltonian is shown to be an important factor in controlling the phase behavior of the film. For $\lambda = 0$, the model is a classical Heisenberg spin system which shows no spontaneous magnetization for $T > 0$. While for value of $\lambda > 0$, the model yields a spontaneous magnetization of the film at low temperatures. The critical temperature T_c characterizing the phase behavior of the magnetization of the film strongly depends on the magnitude of λ as does the magnetic relaxation time τ. These observations can be expected to be of relevance in studies of the phase behavior and dynamics of thin films of more complex materials such as ferronematic liquid crystals which also have a continuous spin system and show spontaneous ordering at low temperatures, but have more complicated Hamiltonians.

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