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1999 J. Phys.: Condens. Matter 11 5045

(http://iopscience.iop.org/0953-8984/11/26/307)

J. Phys.: Condens. Matter **11** (1999) 5045–5056. Printed in the UK PII: S0953-8984(99)00896-6

The competition of bulk and surface fields in a transverse Ising model thin film

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Received 13 January 1999, in final form 4 May 1999

Abstract. Monte Carlo simulations have been performed to investigate the equilibrium phase behaviour of a transverse Ising model in a thin-film geometry under the action of competing surface fields. This model is a classical isotropic vector spin model with a ferromagnetic Ising Hamiltonian and a transverse field applied perpendicular to the Ising z-axis. For finite values of the transverse field strength, Ω, the model displays an interface localization transition with a spontaneous magnetization of the film below a critical temperature T_c. However, in the limit $\Omega \to \infty$, no spontaneous magnetization of the film is observed. In the two-phase regime, below T_c, a degeneracy exists in the magnetization profiles across the film between states of positive and negative total magnetization. The interface localization transition temperature lies below the bulk critical temperature of the film for competing surface fields.

1. Introduction

The transverse Ising model was first introduced by de Gennes [1] in a pioneering study of the order–disorder phase transition in order–disorder, KDP-type ferroelectrics. It has subsequently been used to investigate the ferromagnetic–paramagnetic phase transition in anisotropic magnetic materials as well as in further studies of ferroelectric materials. Recent developments in the fabrication of thin ferromagnetic and ferroelectric films have revealed some novel physics and stimulated theoretical studies of the transverse Ising model in a thin-film geometry [2–8].

The phase behaviour of a thin ferroelectric film described by an Ising model in a transverse field was recently investigated by Wang *et al* [2] using a mean-field approximation. The dynamical properties of this model film were subsequently explored within the random-phase approximation [3]. As in other studies [4–8], attention focused on thin films in which surface effects were included through a modification of the exchange interaction for spins in the surface layers. Thus all of the observed properties were appropriate to systems in which the mean spin orientations within the film were symmetric about the mid-point of the film. Comprehensive Monte Carlo simulation studies [9–11] of thin ferromagnetic Ising films with symmetric surface fields have shown that the inclusion of symmetry-breaking surface fields gives rise to wetting phenomena in the film. Despite the deceptive simplicity of the model, the phase diagram is extremely complex, with first-order wetting, critical wetting and layering transitions all being observed for appropriate values of the model parameters. Subsequent studies [12–14] of thin Ising ferromagnetic films in the presence of competing surface fields have revealed further novel phase behaviour arising from the interplay of the surface fields and the confining

0953-8984/99/265045+12$19.50

geometry of the thin film. In this paper we explore the modifications to the phase behaviour of a thin ferromagnetic film arising from the introduction of a bulk transverse external field.

The Ising model of magnetism represents one of the landmark models of theoretical physics—not only for its application to the description of magnetic materials, but more generally as a test bed for the study of the statistical mechanics of phase transitions. Thus, the model has been widely studied by many theorists. However, the model is very idealized in that it utilizes a discrete set of anisotropic spin states. Ising spins do not rotate through all possible orientations, but instead are restricted to alignments along a particular axis, conventionally the z-direction. In general the magnetic spins in a magnetic material are not simply restricted orientations along a single easy axis, but are able to orient themselves over a large range of directions, as in easy-plane magnets for example, if not all possible directions, as in isotropic magnets. For an isotropic system, the magnitude of the z-component of the spins is continuously variable and under these circumstances the study of the collective spin behaviour is much more complicated than for an anisotropic spin model such as the Ising model.

Prelovšek and Sega [15] have performed a Monte Carlo simulation study to determine the equilibrium properties of a classical isotropic spin model with a ferromagnetic Ising Hamiltonian and a transverse field. The transverse field, Ω, was applied in the x-direction, perpendicular to the Ising z-axis, and it was shown that the temperature of the paramagnetic–ferromagnetic phase transition varied as a function of Ω. In the absence of the transverse field, for $\Omega = 0$, the system behaviour is consistent with that of an Ising spin system which possesses a non-zero transition temperature. As the magnitude of Ω is increased up to a critical value Ω_c, the spins are forced to orient in the x-direction and the transition temperature is lowered. When $\Omega = \Omega_c$, the transition temperature is zero and for $\Omega > \Omega_c$, the system displays no ferromagnetic ordering.

The influence of anisotropy on the behaviour of the Heisenberg spin system in a thin-film geometry has been studied for different types of anisotropy [16, 17]. In a recent paper [16], the phase behaviour of the Heisenberg spin system subject to a single-site perpendicular anisotropy in a thin film with competing surface fields was studied. In the absence of the single-site perpendicular anisotropy, the model behaviour was that of a classical Heisenberg spin system. But for large values of the single-site perpendicular anisotropy, the behaviour of the system became equivalent to that of an Ising model. Similarly, the inclusion of an exchange anisotropy in the Hamiltonian could also transform the phase behaviour of the Heisenberg thin film from that typically associated with a classical Heisenberg spin system to that identified with an Ising spin system as the strength of the exchange anisotropy was altered [17]. In this paper, we investigate the effect of the addition of a transverse field to the phase behaviour of thin ferromagnetic film. This is of particular interest as the phase behaviour of the thin film is an interplay of the confinement effects arising from the film geometry and their competition with applied bulk and surface fields. The detailed specification of the model and the simulation method are given in the following section. The dependence of the equilibrium phase behaviour of the model film with competing surface fields on both the transverse field and the temperature are discussed in sections 3 and 4. The observed phase behaviour for films with competing surface fields is contrasted with that for cooperative symmetric surface fields in section 5. The paper concludes with a summary of the key findings.

2. Model

We consider a simple cubic lattice of size $L \times L \times D$, in units of the lattice spacing. In the Monte Carlo simulation we 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 described by the Hamiltonian

$$\mathcal{H} = -\Omega \sum_i S_i^x - J \sum_{\langle i,j \rangle} S_i^z S_j^z - \sum_{i \in \text{surface } 1} \boldsymbol{H}_1 \cdot \boldsymbol{S}_i - \sum_{i \in \text{surface } D} \boldsymbol{H}_D \cdot \boldsymbol{S}_i \qquad (1)$$

where S_i^x and S_i^z are the x- and z-components of the unit vector $\boldsymbol{S}_i$ representing the spin at lattice site i. The notation $\langle i, j \rangle$ indicates that the sum is restricted to nearest-neighbour pairs of spins, each pair being counted only once. The coupling constant J characterizes the exchange interaction between nearest-neighbour spins and $J > 0$ for ferromagnetism. Ω is the strength of the transverse field which is applied only to the x-component of the spins. $\boldsymbol{H}_1$ and $\boldsymbol{H}_D$ are surface fields which are applied to layers $n = 1$ and $n = D$ of the film. The system is subject to surface fields which have same magnitude but are applied in opposite directions, i.e.

$$\boldsymbol{H}_1 = h\hat{\boldsymbol{z}}\delta_{i1} \qquad (2)$$

$$\boldsymbol{H}_D = -h\hat{\boldsymbol{z}}\delta_{iD} \qquad (3)$$

giving a Hamiltonian

$$\mathcal{H} = -\Omega \sum_i S_i^x - J \sum_{\langle i,j \rangle} S_i^z S_j^z - h\left(\sum_{i \in \text{surface } 1} S_i^z - \sum_{i \in \text{surface } D} S_i^z \right). \qquad (4)$$

A film thickness $D = 12$ and surface field strength $h = -0.55$ were used throughout to aid comparison with the corresponding Ising and Heisenberg films investigated elsewhere [12–14, 16, 17]. As noted for the thin Ising film [13], the results do not depend significantly on the value of h and the choice of $D = 12$ corresponds to a crossover regime, between wall- and bulk-dominated behaviour of the film. In thinner films it is difficult to distinguish between

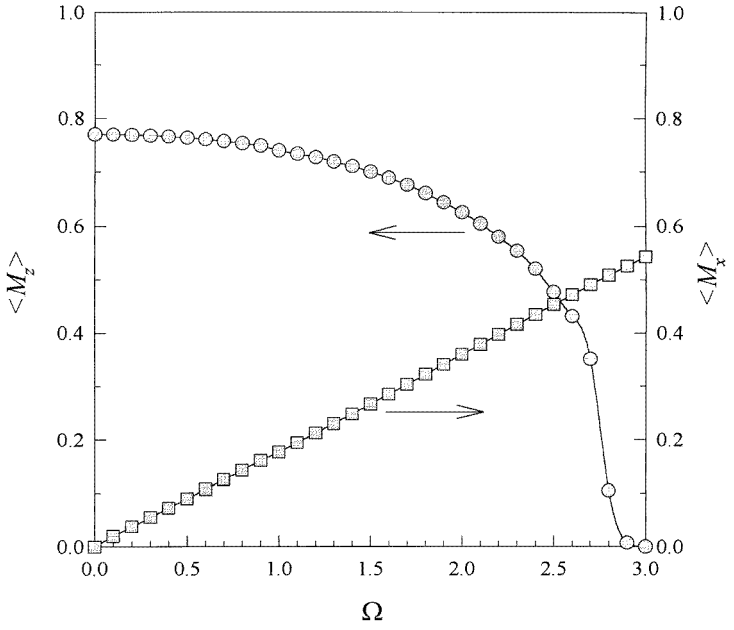


Figure 1. The x- and z-components of the mean magnetization per spin, $\langle M_x \rangle$ and $\langle M_z \rangle$, as functions of the transverse field Ω at a temperature of $T^* = 1.0$ with an initial spin configuration of $S_i^z = +1$ for all i.

'interface' and 'bulk' phases in the film, since all layers of the film 'feel' the effect of the competing surface fields rather strongly, while for much thicker films the surfaces of the film only interact close to the bulk critical point.

Results are reported for lattices of size $L = 32$. A number of additional simulations were performed for $L = 64$ and $L = 128$, but no significant differences were found from the results presented here for non-critical values of Ω and T. The Metropolis algorithm [18] was used in the Monte Carlo simulations with trial configurations generated from Barker–Watts [19] 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.

The x- and z-components of the magnetization of the film

$$M_x = \frac{1}{D}\sum_{n=1}^{D} M_n^x \tag{5}$$

$$M_z = \frac{1}{D}\sum_{n=1}^{D} M_n^z \tag{6}$$

together with the x- and z-components of the magnetization for the nth layer of the film

$$M_n^x = \frac{1}{L^2}\sum S_i^x \tag{7}$$

$$M_n^z = \frac{1}{L^2}\sum S_i^z \tag{8}$$

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

$$\chi_n = L^2(\langle M_n^{z2}\rangle - \langle M_n^z\rangle^2)/k_B T \tag{9}$$

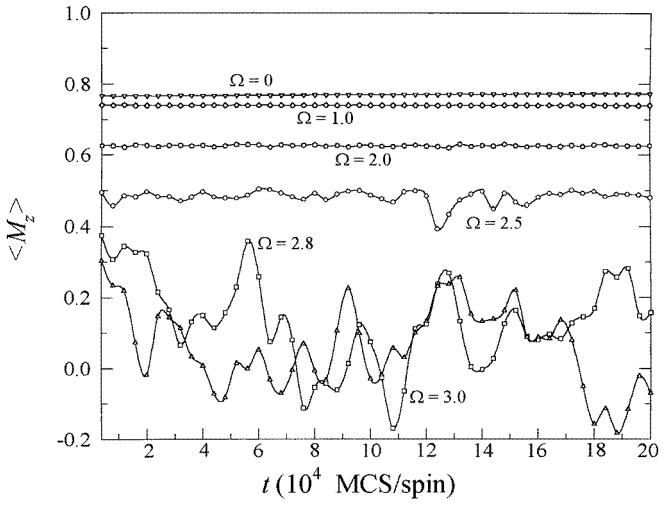


Figure 2. The time evolution of the z-component of the mean magnetization per spin, $\langle M_z \rangle$, for different values of the transverse field in the range $0 \leqslant \Omega \leqslant 3.0$ at a temperature $T^* = 1.0$. All of the simulations used an initial spin state of $S_i^z = +1$ for all i. Time is measured in units of Monte Carlo steps per spin. The curves through the points are only guides to the eye.

where k_B is Boltzmann's constant. Simulations were performed for up to 10^6 Monte Carlo steps per spin (MCS/spin). Equilibrium averages were typically taken over 2×10^5 MCS/spin with initial transients ignored. For systems in the Ising limit, $\Omega = 0$, much shorter runs could be performed, while retaining the same accuracy in the measured properties.

3. The role of the transverse field

In figure 1 the x- and z-components of the mean magnetization per spin, $\langle M_x \rangle$ and $\langle M_z \rangle$, are presented as functions of Ω for a reduced temperature of $T^* = k_B T/J = 1.0$. As might be anticipated, larger values of the transverse field Ω tend to align the spins in the x-direction and the figure shows that $\langle M_x \rangle$ increases linearly with Ω. As a consequence, $\langle M_z \rangle$ decreases with increasing Ω. However, the dependence of $\langle M_z \rangle$ on Ω is non-linear and shows the characteristic form associated with an Ising ferromagnet. $\langle M_z \rangle$ plays the role of the order parameter for the ferromagnetic–paramagnetic phase transition, being zero above Ω_c, but with a finite non-zero value below Ω_c. Figure 1 shows a critical transverse field, $\Omega_c(T^* = 1.0) \simeq 2.9$.

The temporal evolution in simulations of $\langle M_z \rangle$ from an initially ordered state with $S_i^z = +1$ is shown in figure 2 for different values of Ω at a reduced temperature of $T^* = 1.0$. For $\Omega < \Omega_c$, the system rapidly equilibrates and equilibrium states of non-zero magnetization of the film persist. This behaviour is consistent with that of Ising-like spin systems [17]. However, the result for $\Omega = 3.0$ shows that, for $\Omega > \Omega_c$, the continuous rotation of the spins results in large magnetization fluctuations and slow equilibration to a state of zero net film magnetization. No spontaneous magnetization of the film is observed for $\Omega > \Omega_c$ even though T is less

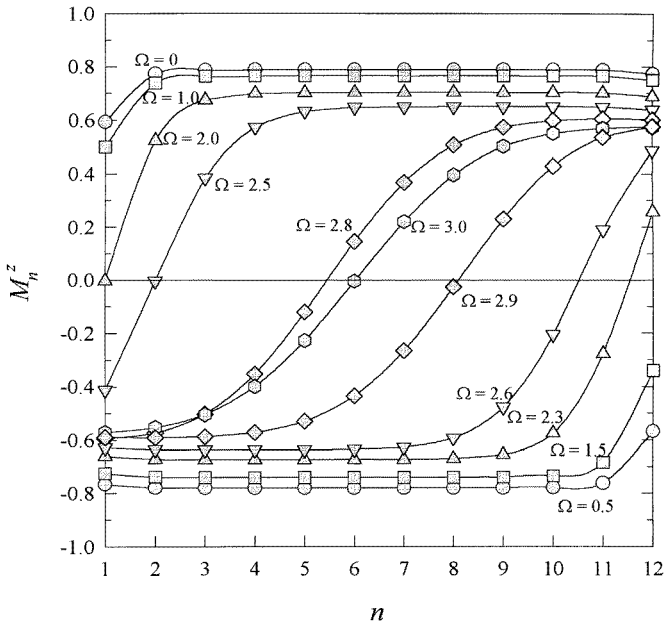


Figure 3. The z-component of the magnetization across the film, M_n^z, versus layer number n, for $D = 12$ with competing surface fields $H_1/J = -H_D/J = -0.55$ at a temperature $T^* = 1.0$ for different values of the transverse field Ω. For $\Omega = 0$, 1.0, 2.0, 2.5, 2.8, 3.0 an initial spin configuration of $S_i^z = +1$ for all i was used, while an initial spin configuration of $S_i^z = -1$ for all i was used for $\Omega = 0.5$, 1.5, 2.3, 2.6, 2.9.

than $T_c(D)$, the interface localization temperature for the corresponding Ising system where $T_c^*(D = 12) = 4.0$ [12].

Further information on the nature of the phase transition occurring in the film in the presence of competing surface fields is contained in the magnetization profiles across the film, M_n^z, as plotted for different Ω at a temperature $T^* = 1.0$ in figure 3. An initial spin state of $S_i^z = +1$ for all i was used for $\Omega = 0$, 1.0, 2.0, 2.5, 2.8, 3.0, while an initial spin state of $S_i^z = -1$ for all i was used for $\Omega = 0.5$, 1.5, 2.3, 2.6, 2.9. The competing surface fields constrain the spins in the surface layers to align in the negative magnetization direction near one surface and positive magnetization direction near the other surface. In the bulk of the film, the mean spin orientation of the layers varies smoothly from one surface to the other. For $\Omega > \Omega_c$, the interface between regions of negative and positive magnetization is located at the centre of the film. The equilibrium magnetization profile across the film is antisymmetric and is obtained from any initial spin configuration. However, for $\Omega < \Omega_c$, the symmetry of the magnetization profile is broken. The interface between regions of negative and positive magnetization sharpens and moves from the centre toward the surface of the film. The direction of the interface displacement depends on the initial spin configuration and a degeneracy exists between states of positive and negative total magnetization. Well below Ω_c, for $\Omega < 2.0$ in figure 3, the interface between regions of negative and positive magnetization disappears and all of the spins contribute in the same sense to a large value of the film magnetization.

Figure 4 shows the profile across the film of the susceptibility χ_n at a temperature $T^* = 1.0$ for $\Omega = 2.4$, 2.6, 2.8, 3.0 from an initial configuration of $S_i^z = +1$. Note that the peaks in the susceptibility profiles for each Ω are located in the same layer as the interface between regions of negative and positive magnetization in the magnetization profiles M_n^z. This indicates the

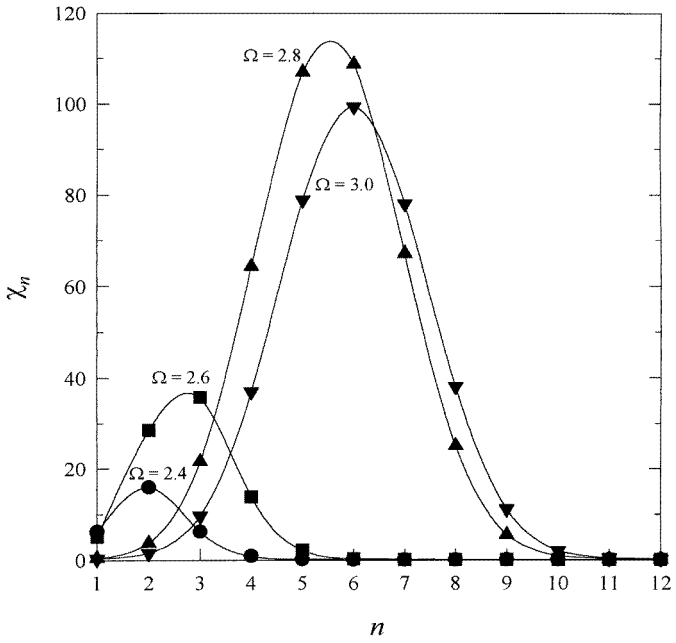


Figure 4. Layer susceptibility χ_n versus layer number n for $D = 12$ at a temperature $T^* = 1.0$ from an initial spin state of $S_i^z = +1$ for all i. The curves drawn are only guides to the eye.

existence of large fluctuations in the spin orientations within the interfacial region.

4. Temperature dependence of the phase behaviour

The temperature dependence of the magnetization profile across the film is shown in figure 5 for $\Omega = 2.0$. Here, for clarity, results are only shown for spin systems with an initial state of $S_i^z = +1$ for all i. At the highest temperature $T^* = 1.4$, the figure shows that the interface between regions of negative and positive magnetization is located at the centre of the film 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 $T^* < 1.4$, the interface is shifted toward the surface, and the film has a finite value of $\langle M_z \rangle$ at those temperatures. This behaviour can be regarded as a remnant of the Ising model behaviour seen by Binder and co-workers [12–14]. The large shift in the interface location between $T^* = 1.1$ and $T^* = 1.2$ indicates that phase transition occurs between these temperatures.

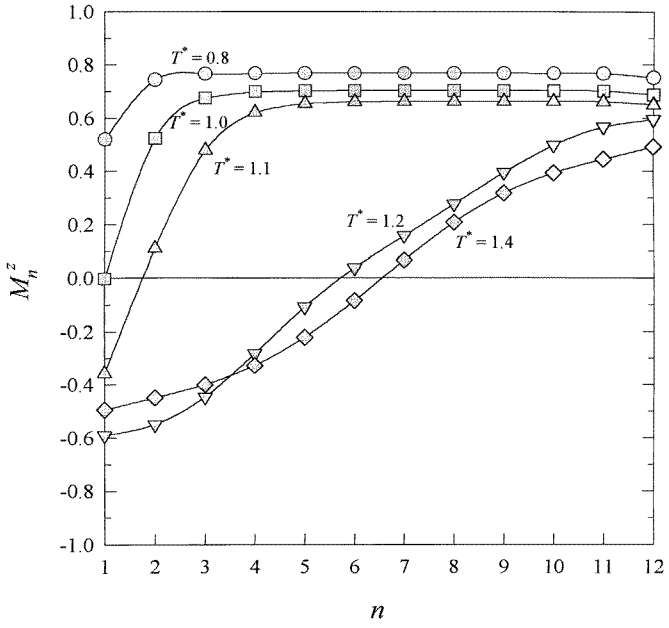


Figure 5. The z-component of the magnetization across the film, M_n^z, versus layer number n, for $D = 12$ with $\Omega = 2.0$ at different temperatures with competing surface fields $H_1/J = -H_D/J = -0.55$ from an initial spin state of $S_i^z = +1$ for all i.

To locate the critical temperature $T_c(\Omega, D)$ we have determined $\langle M_z \rangle$ as a function of temperature. For $T > T_c$ the film shows no spontaneous magnetization with $\langle M_z \rangle = 0$, while for $T < T_c$ a spontaneous magnetization of the film with $\langle |M_z| \rangle > 0$ is observed. Figure 6 shows that the critical temperature $T_c(\Omega, D)$ increases as Ω decreases. Note that for the system in the Ising limit $\Omega = 0$, we find that $T_c^*(\Omega = 0, D = 12) \simeq 1.5$. This is not equivalent to the value obtained from the corresponding Ising spin system where $T_c^*(D = 12) = 4.0$ [12]. The discrepancy is a result of the Ising spin system using a discrete set of spin states with $S_i^z = \pm 1$, while the present model makes use of a continuously orientable spin with $|S_i^z| < 1$. Thus the system with the continuously orientable spin yields a lower value of T_c than the Ising

spin system. For $\Omega = 3.0$, Monte Carlo simulations of the transverse Ising model in the bulk by Prelovšek and Sega [15] found a critical temperature of $T_c^*(\Omega = 3) \simeq 1.25$. By way of comparison, our results for the same system in a thin-film geometry with competing surface fields give a critical temperature of $T_c^*(\Omega = 3, D = 12) \simeq 1.0$. Thus the interface localization temperature is lower than the critical temperature of the bulk system. This is a result of the competing surface fields which provide a local constraint on the orientations of the spins in the two surface layers. At higher temperatures this gives rise to an antisymmetric magnetization profile across the film with a net zero magnetization of the film. When the temperature drops below the bulk critical temperature the system will tend to order and produce a net non-zero magnetization. But the symmetry-breaking transition is resisted by the competing surface fields that act to maintain an antisymmetric magnetization profile across the film and zero net magnetization. Only upon reductions to temperatures significantly below the bulk critical temperature is the symmetry broken by the interface localization transition.

5. Symmetric surface fields

In the preceding section we demonstrated that the presence of competing surface fields in a thin film could substantially modify the ferromagnetic–paramagnetic phase transition and generated an interface localization transition at temperatures significantly below the bulk critical temperature. Studies of wetting [9–11] in thin Ising films have shown that the addition of symmetric surface fields can relocate the ferromagnetic transition of the film to temperatures higher than the bulk critical temperature. In this section we shall compare the magnitudes of the temperature shift of the phase transition for competing and symmetric surface fields.

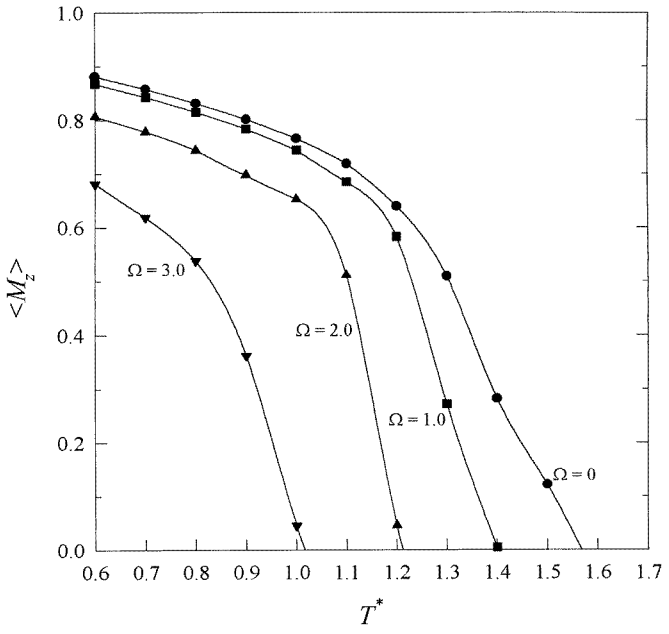


Figure 6. The temperature dependence of the z-component of the mean magnetization per spin, $\langle M_z \rangle$, for different values of the transverse field Ω from an initial spin state of $S_i^z = +1$ for all i with competing surface fields $H_1/J = -H_D/J = -0.55$.

The system introduced is simply modified to include symmetric surface fields by reversing the sign of the surface field $\boldsymbol{H}_D$. This gives a revised Hamiltonian

$$\mathcal{H} = -\Omega \sum_i S_i^x - J \sum_{\langle i,j \rangle} S_i^z S_j^z - h\left(\sum_{i \in \text{surface } 1} S_i^z + \sum_{i \in \text{surface } D} S_i^z \right). \tag{10}$$

Once more, $h = 0.55$ is used for the magnitude of the surface fields which are applied in layers $n = 1$ and $n = D$ of the film. Figure 7 shows the z-component of the film magnetization, $\langle M_z \rangle$, as a function of temperature for two different values of the transverse field strength. For $\Omega = 3.0$, the result can be compared directly with that for the corresponding competing surface field system given in figure 6. The critical temperature for the case of the film with symmetric surface fields can be estimated to be $T_c^*(\Omega = 3, D = 12) \simeq 1.3$. This is close to, but above, the bulk critical temperature $T_c^*(\Omega = 3) \simeq 1.25$ [15]. It is notable in figure 7 that $\langle M_z \rangle$ shows a long high-temperature tail corresponding to a small net film magnetization above the critical temperature. This indicates that even at high temperatures the surface spins are constrained by the surface fields to contribute in the same sense to $\langle M_z \rangle$ and so produce a small finite value of the film magnetization.

The transverse field dependence of $\langle M_z \rangle$ and $\langle M_x \rangle$ for symmetric surface fields is shown in figure 8 at a temperature $T^* = 1.0$. The corresponding result for the system with competing surface fields was given earlier in figure 1. A simple comparison of the two figures shows that the critical transverse field Ω_c is larger for symmetric surface fields.

The symmetric surface fields locally constrain the surface spins to align in the positive magnetization direction at both surfaces. The magnetization profiles across the film, M_n^z, for several temperatures are shown in figure 9 for $\Omega = 2.0$ and $\Omega = 3.0$. At higher temperatures, the spins in the middle of the film are disordered and the layer magnetization

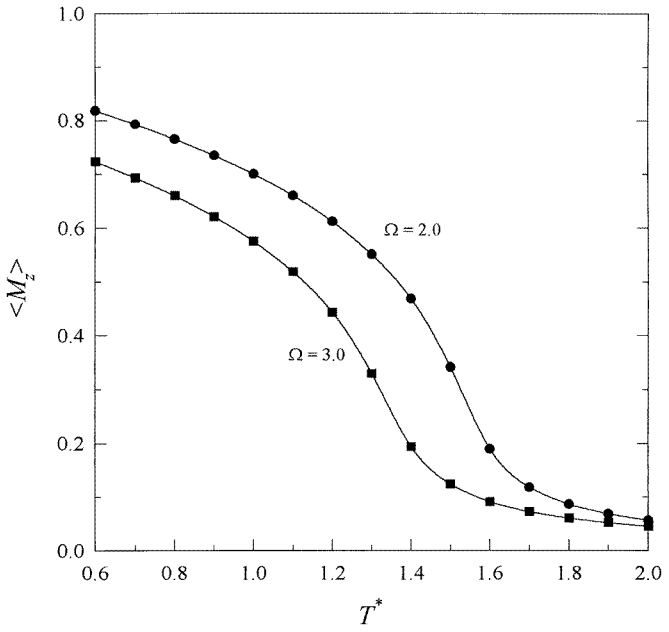


Figure 7. The temperature dependence of the z-component of the mean magnetization per spin, $\langle M_z \rangle$, for two different values of the transverse field Ω from an initial spin state of $S_i^z = +1$ for all i with symmetric surface fields $H_1/J = H_D/J = 0.55$.

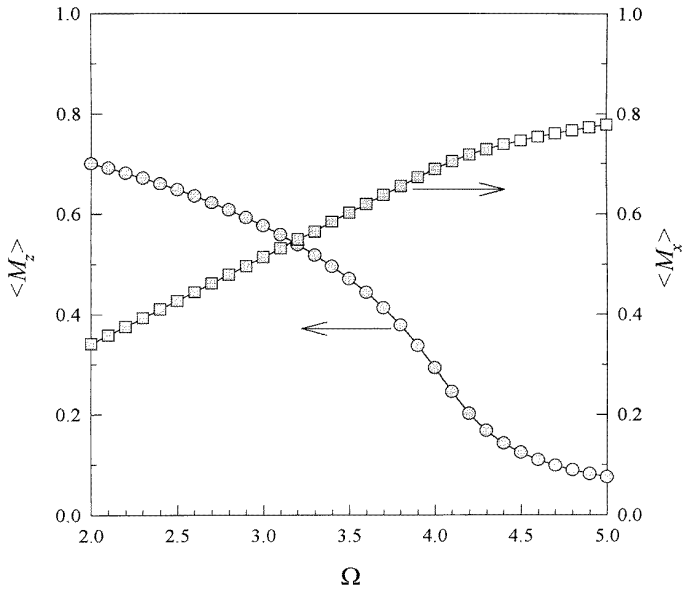
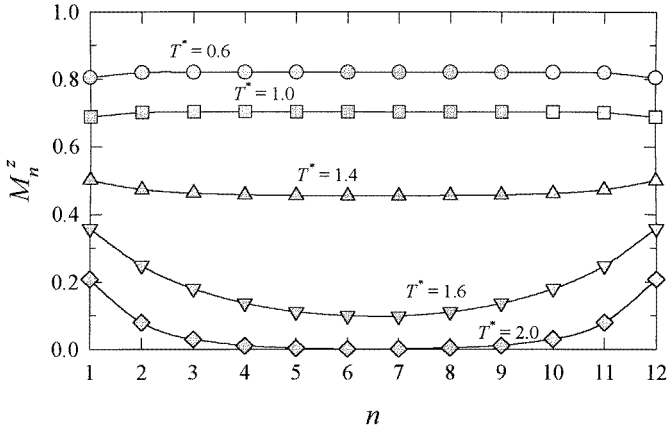


Figure 8. The x- and z-components of the mean magnetization per spin, $\langle M_x \rangle$ and $\langle M_z \rangle$, as functions of the transverse field Ω from an initial spin state of $S_i^z = +1$ for all i at a temperature of $T^* = 1.0$ with symmetric surface fields $H_1/J = H_D/J = 0.55$.

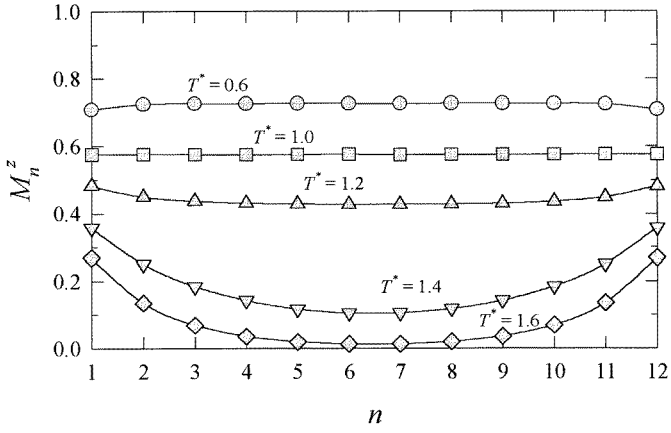
is zero. But the symmetric surface-induced ordering ensures that the film shows a small spontaneous magnetization. As the temperature falls toward the bulk critical temperature, the surface-induced order propagates into the bulk of the film and gives rise to a ferromagnetic film. However, it should be noted that the transition temperature of the film with cooperative symmetric surface fields is much closer to the bulk critical temperature than the interface localization temperature is in the corresponding competing surface field case.

6. Conclusions

In this paper, we have studied the phase behaviour of thin ferromagnetic films subject to competing surface fields within the framework of the transverse Ising model. The transverse field Ω in the Hamiltonian is shown to be an important factor in controlling the phase behaviour of the film. The model displays a spontaneous magnetization below a critical field strength Ω_c, but no spontaneous magnetization of the film is observed above Ω_c. In the limit $\Omega = 0$, Ising-like behaviour was recovered. Note however that since the model involves a continuously orientable spin and not a discrete set of spin orientations, the numerical values obtained in the zero-field limit differ from those obtained in Ising model simulations. The critical temperature characterizing the ferromagnetic–paramagnetic phase transition of the film strongly depends on the magnitude of Ω and the nature of the surface fields. Competing surface fields give rise to an interface localization transition in the film whose critical temperature is below the bulk critical temperature of the model, but cooperative symmetric surface fields lead to a phase transition in the film whose transition temperature is above the bulk critical temperature. However, the magnitude of the shift in the transition temperature away from the bulk critical temperature is greater for competing surface fields.



(a)



(b)

Figure 9. The z-component of the magnetization across the film, M_n^z, versus layer number n, for $D = 12$, at different temperatures from an initial spin state of $S_i^z = +1$ for all i with symmetric surface fields $H_1/J = H_D/J = 0.55$: (a) for $\Omega = 2.0$ and (b) for $\Omega = 3.0$.

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