Magnetization and ordering temperature of films and multilayers

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We investigate, in this paper, a number of magnetic properties of single and multilayer thin film systems within the Ising model by application of mean field, finite cluster approximations as well as by Monte Carlo simulations. The magnetization profiles and the magnetic ordering temperature are calculated for different magnetic systems. The influence of corrugation and disorder at the surface, on the critical behavior of ferromagnetic Ising film is also studied. It is found that the critical surface exponent of the magnetization follows closely the one of a perfect surface, in the two cases: corrugated surface and random equiprobable coupling surface. However, in the case of flat surface with random interactions the surface critical exponent depends on the concentration of the strong interaction, while such critical exponent is independent on the concentration. Moreover, in the case of corrugated surface, the effective exponent for a given layer, is a function of the number of steps at the surface. The probability of a magnetic ground state is larger for low spatial dimensionality of an extended system, or lower for local symmetry of a given site in the atomic lattice. Consequently, the magnetic properties are usually more pronounced at the surface of a bulk magnet as compared to the bulk interior. The phase diagram and the characteristic behaviors of the surface magnetization, are investigated for amorphous or cristalline surfaces. Indeed, the size effects become more relevant at low temperature depending on film thickness.

I. Introduction

Recently, the ordering temperature and layering transitions of magnetic films are of high technological importance. Several models are used to study such systems and a variety of possible phase transitions has been reviewed, for example, by de Oliveira and Griffiths [1], Pandit and Wortis [2], Pandit *et al.* [3] and Ebner *et al.* [4]. Such transitions have been observed in a variety of systems including for example ^{4}He [5] and ethylene adsorbed on graphite [6]. The n^{th} layering transition from $(n\text{-}1)^{th}$ layer film to an n layer film, typically is only present at low temperatures and may terminate as the temperature increases in a layering critical point $T_{c,n}$ which belongs to the universality class of the two-dimensional Ising model. Using the perturbative theory, Harris [7] have studied the layering transitions at T=0 in the presence of a transverse field. A variation of phase diagrams with the strength of the substrate potential in a lattice gas model for multilayer adsorption is studied by Patrykiejew *et al.* [8] using Monte Carlo simulations and molecular field approximation. The effect of finite size on such transitions has been studied, in thin film confined between parallel plates or walls, by Nakanishi and Fisher [9] using mean field theory and by Bruno *et al.* [10] taking into account the capillary condensation effect. The transfer matrix method [11] and the real space renormalization group theory [12] have been applied by Benyoussef and Ez-Zahraouy to study the layering transitions of an Ising thin film spin-1/2 model. In the framework of the mean field theory, we found in a previous work [13], the wetting and layering transitions of *3D* Ising transverse model in the presence of both an external and surface fields. The critical wetting transitions have been considered in systems with corrugated periodic walls [14] by using an effective Hamiltonian study. By applying Monte Carlo simulations on thin Ising films with competing walls, Binder *et al.* [15], found that occurring phase transitions belong to the universality class of the two-dimensional Ising model and found that the transition is shifted to a temperature just below the wetting transition of a semi-infinite fluid [16]. We have established, in one of our earlier works [17], the effect of a corrugated surface on the wetting and layering transitions of an Ising model in the presence of both an external and uniform surface fields in the framework of the mean field theory (MF) and Monte Carlo (MC) simulations. The critical behaviour and magnetic properties of this model have been described in Ref. [18].
The Blume-Capel model (BC) was originally proposed to study the first-order magnetic phase transitions in spin -1 Ising systems [19]. This model was generalised to the Blume-Emery-Griffiths (BEG) to study phase separation and superfluity in ^{3}He-^{4}He mixtures [20]. Later it has been applied to describe properties of multicomponent fluids [21], semiconductor alloys [22] and electronic conduction models [23]. The (BC) model is not exactly solvable in more than one dimension, but it has been studied over infinite d-dimensional lattices by means of many different approximate techniques and its phase diagram is well known. When the theory of surface critical phenomena started developing, some attention has been devoted to the study of the (BC) model over semi-infinite lattices, with modified surface couplings. Benyoussef *et al.* [24] have determined the phase diagram in the mean field approximation, reporting four possible topologies at fixed bulk/surface coupling ratios. A similar analysis have also been done using a real space renormalization group transformation [25]. Other works referring to particular regions of the phase space are those using: the mean field approximation [26], the effective

field approximation [27] and the low temperature expansion [28]. All these works show that it is possible to have a phase with ordered surface and disordered bulk, which is separated from the completely ordered phase by the so-called *extraordinary* transition and from the completely disordered phase by the *surface* transition. When such a phase is absent, the transition between the completely ordered and the completely disordered phase is called {\it ordinary}. The meeting point of the lines of these three kinds of phase transitions is named *special* and it is generally a multi-critical point.

As discussed in Ref. [29], the strong interest in these models arises partly from the unusually rich phase transition behaviour they display as their interaction parameters are varied, and partly from their many possible applications. In most of these cases considered so far the bilinear interaction is ferromagnetic. In the anti-ferromagnetic case, the spin-1 Ising systems are used to describe both the order-disorder transition and the crystallisation of the binary allow, and it was solved in the mean field approach [30]. One of the most interesting and elusive features of the mean field phase diagram for the anti-ferromagnetic spin-1 Blume-Capel model in an external magnetic field is the decomposition of a line of tri-critical points into a line of critical end points and one of double critical points [31]. This model was also studied by transfer-matrix and Monte Carlo finite-size-scaling methods [32], but such decomposition does not occur in this two dimensional model.

On the other hand, ferroelectric films can be described by an Ising model and when the film becomes very thick, its properties are those of the semi-infinite Ising system [33-35]. From the experimental point of view, the most commonly studied magnetic multi-layers are those of ferromagnetic transition metal such as Fe/Ni, where the coupling can exist between magnetic layers [36-38]. The discovery of enormous values of magneto-resistance in magnetic multi-layers are far exceeding those found in single layer films and so exceeds the discovery of oscillatory interlayer coupling in transition metal multi-layers. These experimental studies have motivated much theoretical works to study magnetic thin films as well as critical phenomena [39-44]. This is partly motivated by the development of new growth and characterisation techniques, but perhaps more so by the discovery of many exciting new properties, some quite unanticipated. Using the mean field theory, Benyoussef *et al.* [45] and Boccara *et al.* [46] have studied the spin-1 Ising model with a random crystal field.

The effect of the surface and bulk transverse fields on the phase diagrams of a semi infinite spin-1 ferromagnetic Ising model with a crystal field was investigated in [47] within a finite cluster approximation with an expansion technique for cluster identities of spin-1 localised spin systems. On the other hand, the transverse field or crystal field effects of spin-1 Ising model has been studied by several authors [48-51]. The experimental measurements of layer-by-layer ordering phenomena have been established on free-standing liquid crystals films such as *nm*OBC (n-alkyl-4'-n-alkyloxybiphenyl-4-carboxylate) [52,53] and 54COOBC (n-pentyl-4'-n-pentanoyloxy-biphenyl-4-carboxylate) [54] for several molecular layers. More recently, Lin *el al.* [55] have used the three-level Potts model to show the existence of layer-by-layer ordering of ultra thin liquid crystal films of free-standing 54COOBC films, by adjusting the interlayer and intra-layer couplings between nearest-neighbouring molecules.

II. Disorder and corrugation of surfaces.

II.1. Critical behaviour and magnetic properties.

We are considering the surface critical behaviour of an Ising film with corrugated surfaces, see Fig. 1. If we consider only nearest neighbour interaction, the system is governed by the Hamiltonian:

$$H=\sum J_{ij}(S_{iz}S_{Jz}+S_{iz+1}S_{Jz+1}+S_{iz}S_{jz-1}) \quad (1)$$

where $S_{iz}=\pm 1$ is the spin variable of a site i of the layer z.

J_{ij} are the exchange interaction between nearest neighbour sites .

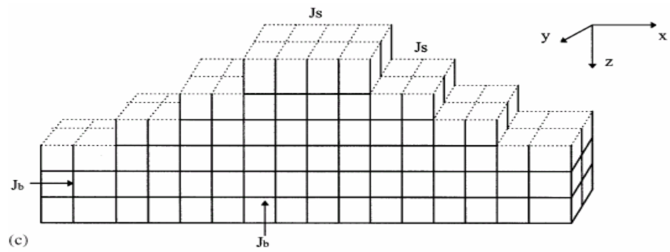


Fig. 1: Geometry of a corrugated surface system with M layers in the z-direction. The system is assumed to be infinite in the y-direction.

Typical magnetization profiles are depicted at a fixed temperature T < Tc (see Fig. 2). It is found that the magnetizations m(x,z) depend on the position x for each layer z. For a fixed layer, the magnetization decreases each time the system presents a step. But far way from the surface the magnetization keeps a constant value except at the boundaries where it decreases.

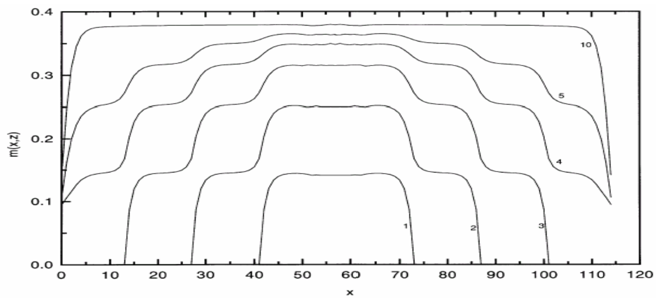


Fig.2: Magnetization profiles for the perfect surface. The number accompanying each curve denotes the value of the layer z.

Furthermore, at the vicinity of the critical temperature T_c, The spontaneous magnetizations vanish with a power low given by

$$m(x,t)\ \alpha\ t^{\beta eff(z,t)} \quad (2)$$

where the effective exponent $\beta_{eff}(z,t)$ can be expressed as

$$\beta_{eff}(z,t)=dln(m(z))/dln(t) \quad (3)$$

where t=|T-Tc|/Tc is the reduced temperature.

It is clear, from F.g.3, that near the critical temperature

the effective exponent is insensitive to the corrugation. Furthermore, for a fixed number of steps, the effective exponent of each layer decreases when the reduced temperature increases.

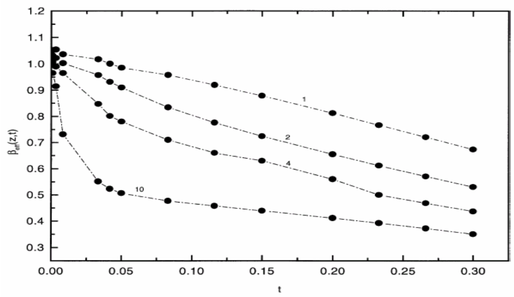


Fig.3: The dependence of the effective exponent as a function of the reduced temperature. The number accompanying each curve denotes the value of layer position z.

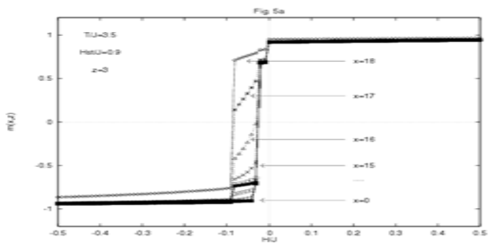
II.2. Presence of an external magnetic field.

In the presence of an external magnetic field, the Hamiltonian describing this system is written as:

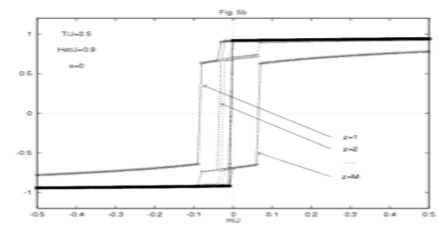
$$H=-\sum J_{ij} S_i S_j - \sum H_i S_i \qquad (4)$$

where $S_{ij}=\pm1$ are the spin variables, $J_{ij}=J$ is the exchange interaction assumed to be constant.

In order to examine the effect of surfaces in the x-direction on the local magnetizations, we plot in Fig. 4a the dependency of m(x,z) as a function of the reduced bulk field, for a fixed layer z=3 and several values of the position x. It is seen that when increasing the reduced bulk field the top surface exhibits a first order transition. Such behaviour is absent in the perfect surface case.



(a)



(b)

Fig. 4: Magnetization profiles as a function of the reduced bulk field for: (a) fixed layer z=3 and different positions x, (b) fixed position x=0 and different layers z.

To study the local magnetization behavior in the z-direction, we plot, in Fig. 4b the behaviour of m(x,z) as a function of the reduced bulk field for a fixed position x and different layers z. It is found that the deeper layers (z>1) need large values of the bulk magnetic field to transit from negative magnetizations to positive ones.

II.3. Thin film with higher values of spin and amorphous surfaces.

In the case of a magnetic film with amorphous surfaces, the Hamiltonian of this system is given by:

$$H=-\sum_{<i,j>} J_S \mu_i \mu_j - J\sum_{<mn>} S_m S_n - \sum_{<im>} J_1 \mu_i S_m - D_1 \sum_i \mu_i^2 - D_2 \sum_m S_m^2 \qquad (5)$$

Where, the summation is carried out only over nearest neighbourg pairs of spins. J_S , J_1 and J are the couplings at the surface, between the surface nearest-neighbourg and inside the film, respectively. μ_i (D_1) and S_m (D_1) are the spin operators (crystal-field interactions) at the surface and in the bulk, respectively. J_S and J_1 are assumed to be randomly distributed according to the Kaneyoshi distribution for amorphous magnets, so that:

$$P(J_k)=1/2\{\delta(J_k - J'_k - \Delta J'_k) + \delta(J_k - J'_k + \Delta J'_k)\},\ k=1,2 \qquad (6)$$

We introduce also the structure factors δ_k as

$$\delta_k = \Delta J_k / J_k\ ,\quad k=S,1 \qquad (7)$$

which is often called the ‘structural fluctuation‘ in amorphous magnets.

When the value of δ_S becomes larger than $\delta_S=1$ the structural interaction can take positive and negative values. Therefore, it is interesting to investigate whether the re-entrant phenomenon is possible or not. So, we present in Fig. 5 the behaviour of the critical temperature with the amorphization δ_S. As expected, in the absence of anisotropy, this figure shows re-entrant phenomenon when the fluctuations at the surface become strong enough. This is due to the competition between ferromagnetic and anti-ferromagnetic exchange interactions.

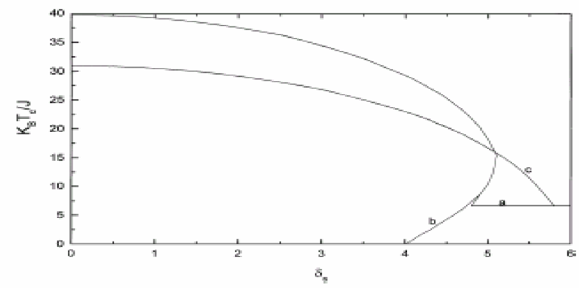


Fig. 5: The phase diagram in the plane ($K_bT_S/J,\delta_S$) for several values of anisotropy, N=20 layers, $J_S/J=4$, $J_1/J=0.1$, $D_1/J=0$, $D_2/J=0$ (curve a), $D_1/J=0$, $D_2/J=-4$ (curve b), and $D_1/J=-4$, $D_2/J=0$ (curve c),

In order to complete this study, we plot in Fig. 6, the dependence of the transition temperature as a function of the film thickness, for the crystalline case (***δ****S*=***δ****I*=0) and several values of anisotropy values $D_1$and D_2. There exists a critical value of J_S/J above which the surface magnetism appears. There exists also a critical film thickness N_C above which the critical temperature will drop down suddenly and rise slowly when the film thickness increases, see Fig. 6.

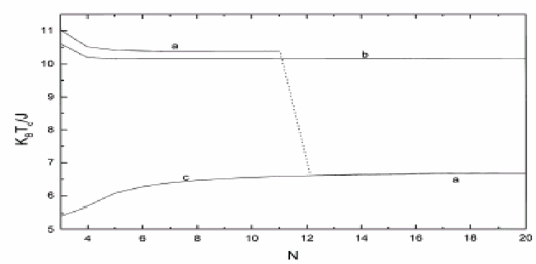


Fig. 6: The phase diagram in the plane (K_bT_c/J,***N***) for several values of anisotropy, N=20 layers, $J_S/J=J_1/J=1$, $\delta_S=\delta_I=0$, (a) $D_1/J=D_2/J=0$, (b) $D_1/J=-4$, $D_2/J=0$, (c) $D_1/J=0$, $D_2/J=-4$.

Because of the effect of layers, the transition temperature at the surface decreases when increasing the film thickness, and the surface order will be destroyed when N is large enough. This jump disappears for increasing the amorphization at surface.

III. Crystal field effect on magnetic films.

III.1. Effects of a variable crystal field.

The system we are studying here is a film formed with N coupled ferromagnetic square layers, in the presence of a crystal field. The Hamiltonian governing this is given by:

$$H=-\sum_{<i,j>} J_{ij} S_i S_j + \sum \Delta_i S_i^2 \qquad (8)$$

where, S_l (l=I,j)=-1,0,+1 are the spin variables. The interaction between different spins are assumed to be constant so that $J_{ij}=J$. The crystal field, acting on a site i of a lyer k, is so that: $\Delta_1 > \Delta_2 > \ldots > \Delta_N$ and distributed according to the law:

$$\Delta_k = \Delta_s / k^{\alpha} \qquad (9)$$

with Δ_s being the crystal field acting on the surface and α a positive constant.
The ground state phase diagram of this system is illustrated in Fig. 7. For very small values of the surface crystal field Δ_s, the system orders in the phase O^N. When increasing Δ_s the surface disorders and the phase DO^{N-1} occurs. Increasing Δ_s more and more, the second layer disorders and so on.

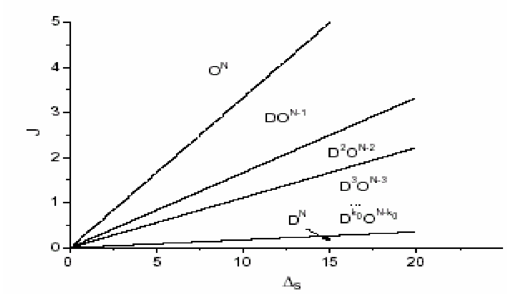


Fig. 7: The ground state phase diagram in the plane (J, Δ_s) for a film thickness N=10 layers.

It is worth to note that the re-entrant phenomenon is present for the first k_0 layers, where k_0 depends on the surface crystal field.
The profiles of the surface critical temperature in the plane (T_C/J , Δ_s/J), for several values of the constant α are given in Fig. 8. It is found that increasing α values leads to important re-entrant phenomena.

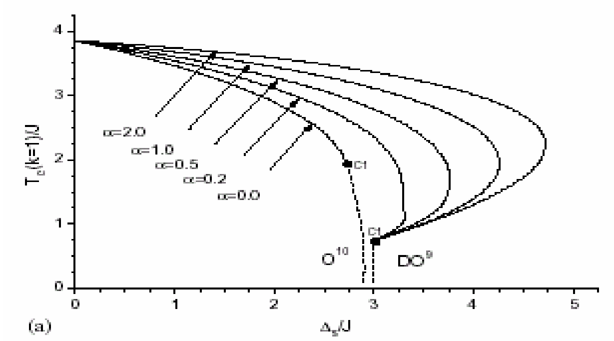


Fig. 8: The surface critical temperature behaviour as a function of the surface crystal field, for selected values of the constant α: 0, 0.2, 0.5, 1 and 2.

Indeed, for large values of α the layer with k>1 needs higher values of the surface crystal field to disorder. It is also found that, except for the special case α=0, the tri-critical point is not affected by variations in α values.

III.2. Amorphization and anisotropy of a bilayer film.

We consider a three dimensional ferromagnetic amorphous bi-layer system consisting of two magnetic monolayers A and B with different spins S_A=1/2 and S_B =3/2, respectively. The Hamiltonian of the system is given by:

$$H=-J_1\sum_{<i,j>} \mu_i\mu_j - \sum_{<mn>}' J_2 S_m S_n - \sum_{<im>}' J_3 \mu_i S_m - D\sum_m S_m^2 \qquad (10)$$

where the summation is carried out only over the nearest neighbour pairs of spins. J_1 is the nearest neighbour exchange interaction in the monolayer A. J_2' is the coupling constant if both m and n sites are in the

monolayer B, while J'_3 between spins m on the monolayer A and its nearest neighbour in the monolayer B. J'_2 and J'_3 are assumed to be randomly distributed according to the independent probability distribution functions P(J'_2) and $P(J'_3)$:

$$P(J'_l)=1/2\{\delta(J'_l-J_l-\Delta J_l)+\delta(J'_l-J_l+\Delta J_l)\},\quad l=1,2 \qquad (11)$$

where the structure factors δ_l is:

$$\delta_l=\Delta J_l/J_l,\quad l=2,3 \qquad (12)$$

In Fig. 9 we can show a paramagnetic and two ordered ferromagnetic phases: 3/2 and 1/2.

A second order transition line separates the paramagnetic phase from the two-ordered ferromagnetic phases. In fact, for a relatively small temperature, our equations admit two numerical solutions.

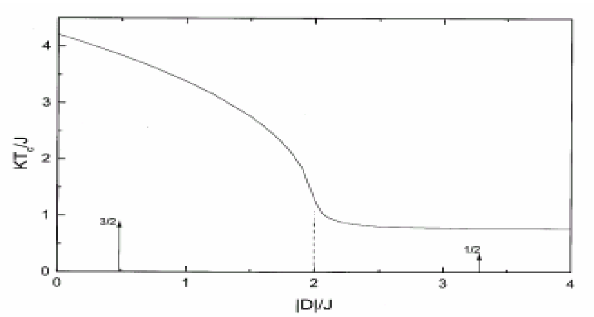


Fig. 9: Critical temperature as a function of the crystal field for spin-3/2 Blume-Capel model in a square lattice. Full curve is the second order transition, while the dotted line represents the first order line. The dashed line is an extrapolation of the first order to lower values of the temperature.

The phase diagrams in the (T/J_1,D/J_1) plane for J_1=J_2=J_3 and different values of δ_2 are represented in Fig. 10, for δ_3=0.1. As is shown, a maximum appears for the values of δ_2 greater than one.

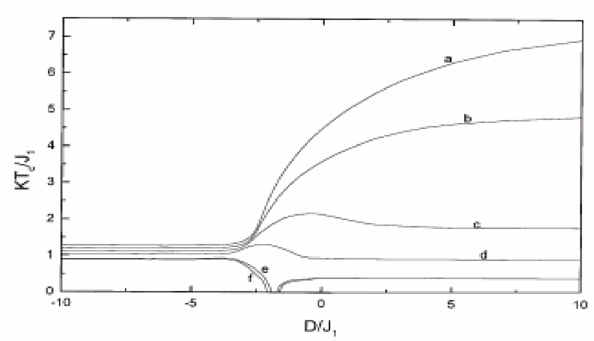


Fig. 10: Phase diagram (T/J_1,D/J_1) of the amorphous bi-layer system when the values of J_1,J_2 and J_3 are fixed at J_2/J_1=J_2/J_1=1 and the values of δ_3 and δ_2 are given by δ_3=0.1 and δ_2=0.1 (a), 1 (b), 1.5 (c), 2 (d), 2.9 (e) and 3 (f).

For δ_2>2.9 two ordered phases occur at low temperature. For large values of δ_2 and negative and large values of D, the amorphization is dominated by the crystalline field and stabilize the ½ phase in monolayer B. When D increases the effect of the amorphization δ_2 dominates and leads to the paramagnetic phase.

Conclusion

In this paper, we have studied a number of magnetic properties of single and multi-layer thin film systems. We have investigated the surface critical behaviour of Ising films with corrugated surfaces. Typical magnetization profiles are depicted at fixed temperatures. It is found that the magnetizations decrease each time the system presents a step. Furthermore, at the vicinity of the critical temperature, the spontaneous magnetizations vanish with a power low. But near the critical temperature the effective exponent is insensitive to the corrugation. It is also found that when increasing the reduced bulk field, the top surface exhibits a first order transition. While this behaviour is absent for a perfect surface case. We showed also that the deeper layers need large values of the bulk magnetic field in order to transit from negative magnetizations to positive ones. On the other hand, for films with higher values of spin and amorphous surfaces, we found a re-entrant phenomenon when the fluctuations at the surface become strong enough. This is due to the competition between ferromagnetic and anti-ferromagnetic exchange interactions. There exists also a critical film thickness value, above which the critical temperature drops down suddenly and rises slowly for increasing film thicknesses. We found also that, because of the layers effect, the transition temperature at the surface decreases when increasing the film thickness. The surface order is destroyed for sufficiently large values of the layer number. Moreover, in the case of corrugated surfaces, the effective exponent for a given layer depends on the number of steps at the surface. For low spatial dimensionality of an extended system, the probability of a magnetic ground state is larger or lower for local symmetry of a given site in the atomic lattice. Consequently, the magnetic properties are usually more important at the surface of a bulk magnet when compared to those of the bulk interior. For amorphous and crystalline surfaces, the phase diagrams and the characteristic behaviours of the surface magnetization are investigated. Nevertheless, the size effects become more relevant at low temperature depending on film thickness and crystal magnetic field values.

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