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Phase Transitions in Confined Antiferromagnets

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Confinement effects on the phase transitions in antiferromagnets are studied as a function of the surface coupling v and the surface field h for b.c.c.(110) films. Unusual topologies for the phase diagram are attained for particular combinations of v and h. It is shown that some of the characteristics of the finite-temperature behavior of the system are driven by its low-temperature properties and consequently can be explained in terms of a ground-state analysis. Cluster variation free energies are used for the investigation of the finite temperature behavior.

In recent years the theoretical interest on two-sublattice, uniaxial antiferromagnets has been renewed [1 to 5]. This interest stems from the experimental work on Fe/Cr(211) multilayers by Fullerton and coworkers [6], where an antiferromagnetic coupling between the Fe layers is possible for a suitable choice of the Cr layer (11 Å). For an even number of layers, the spin-flop phase nucleates at the surface and, as the external field increases, this surface phase evolves into the bulk spin-flop phase [1]. The small anisotropy-to-exchange interaction ratio that characterizes the Fe/Cr(211) system, makes this particular sort of multilayers amenable to the theoretical modeling. Since this type of magnetic multilayers are isomorphic to the MnF_2 class antiferromagnets with (100) surfaces, a classical one-dimensional XY model is adequate to model their magnetic properties at low temperatures [1]. The early work of Mills [7] and Keffer [8] on the one-dimensional XY model, that lead to the identification of the surface spin-flop transition, have been complemented and extended recently. The surface and finite-size effects on the ground-state properties of an XY chain have been investigated in terms of discommensuration transitions [3, 5] and the analogy between the one-dimensional XY model and Frenkel-Kontorova-type chains has been elucidated [4]. These investigations have set our understanding of the rich magnetic behavior in Fe/Cr multilayers, where finite-size and surface effects are equally important, on solid physical grounds. It is appropriate to note that the passage from an inherently three-dimensional structure, such as the Fe/Cr multilayers, to a one-dimensional structure is based on the assumption that lateral fluctuations within each layer can be disregarded with respect to the interlayer fluctuations. The effect of confinement (surface plus finite size) in antiferromagnets for which the intralayer fluctuations are important has also been the subject of previous investigations [9 to 13].

In this paper we provide a brief survey of a recent study of ground-state properties of b.c.c. films with surfaces oriented in the [110] direction [14] and relate the previously

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observed topological features of the phase diagram [9, 11] to the zero temperature properties of the system. With these objectives in mind, we consider body-centered antiferromagnetic Ising films with surfaces orientated in the [110] direction. In the (110) planes of a b.c.c. structure, each site in one sublattice has nearest-neighbors in the other sublattice[2]). For nearest-neighbor pair interactions, the Hamiltonian is the following:

$$\mathcal{H} = J_{\mathrm{b}} \sum_{ij \in \text{bulk}} \sigma_i \sigma_j + J_{\mathrm{s}} \sum_{ij \in \text{surf}} \sigma_i \sigma_j - H \sum_{i \in \text{bulk}} \sigma_i - (h + H) \sum_{i \in \text{surf}} \sigma_i \,, \tag{1}$$

where the spin variable σ_i takes the value of $+1$ or -1 depending if the spin at site i is pointing up or down, respectively. We have assumed that surface sites, in layers 1 and N for an N-layer film, experience a surface magnetic field h in addition to the external field H. We can think h as the surface perturbation on a highly anisotropic antiferromagnet (Ising-like) slab, caused by the presence of ferromagnetic layers in a FM/AFM superlattice. We can also consider h as the wall-particle interaction in a fluid confined between two parallel plates, when the usual transformation $p_i = \frac{1}{2}(1 + \sigma_i)$ is used to cast Hamiltonian (1) into a lattice-gas model. The wall-particle interaction (h) is responsible for the condensation of the liquid phase at lower chemical potential than it is necessary in the bulk (capillary condensation)[3]) [15 to 18].

Phase equilibrium in confined systems is very sensitive to the boundary (interface) conditions defined by the surface field h and by the surface coupling J_{s} [19, 20]. In this paper we specialize ourselves to the case of nearest-neighbor interactions and localized symmetric surface fields; that is, the field at each surface is the same and acts only at the surface sites [see Eq. (1)]. In the remaining of the paper, the effective pair interactions, the surface field (h), and the bulk external field (H) shall be expressed in terms of the bulk AF coupling ($J_{\mathrm{b}} > 0$). The ratio of surface to bulk coupling is restricted to positive values and it is denoted by v.

Even when h is zero (neutral boundary conditions), the disruption of the translation symmetry due to the surfaces results in a "missing neighbors" field h_{m}. The surface field h_{m} is responsible of the inhomogeneities in the magnetization profile near the surfaces. When Eq. (1) is reinterpreted as a binary-alloy Hamiltonian, the missing-neighbors field, along with h, accounts for the surface segregation phenomenon, i.e., the enrichment of the surfaces with one component[2]) (see [3]) and [20, 21]). In the following we shall consider only the case of $h > 0$, since the results for $h < 0$ can be obtained straightforwardly from the symmetry properties of Hamiltonian (1), as it is discussed next. For zero surface field, Hamiltonian (1) is invariant under the transformations $\sigma_i \to -\sigma_i$, $H \to -H$. For neutral boundary conditions ($h = 0$) the ground-state and the finite-temperature properties of the Hamiltonian are symmetric about $H = 0$. A positive value of h breaks this symmetry by favoring the spin-up states at surfaces. Both zero- and finite-temperature properties of Hamiltonian in Eq. (1) become asymmetric with the applied field H. However, for nonzero h the Hamiltonian is still invariant if we extend the above transformation to include $h \to -h$.

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[2]) In Ref. [12] and [13] a (110) surface in a b.c.c. antiferromagnet is called symmetry-preserving while (100), where each layer belongs to one of the two sublattices, is denoted by symmetry-breaking orientation.

[3]) In the context of binary alloys, the surface field h can also be regarded as proportional to the difference in the enthalpies of formation of the pure elements.

The selective nature of the surface field has varied consequences on the properties of Hamiltonian (1), since the equilibrium states are defined by the competition between the Zeeman and the ordering energies. The ordering contribution to the Hamiltonian, first term in the rhs of Eq. (1), favors AFM structures whereas the Zeeman energy in third sum of the rhs of (1) promotes FM structures. An additional Zeeman contribution arises from the surface field [last term in the rhs of Hamiltonian (1)], which competes with the surface ordering tendencies [second sum in the rhs of (1)] to define the equilibrium state in the film. Thus, for positive and large values of H, applying a surface field is of little consequence since the stable state is one with spin-up at the surfaces. For low and negative values of H, where spin-down states are likely to occur, the surface field actually may give rise to an antiferromagnetic surface state.

An analysis of the ground states of Hamiltonian (1) singles out the ground-state (GS) sequence in Fig. 1(a) as the stable sequence for large h [14]. The nomenclature in Fig. 1 is as follows: the intra- and interlayer coordination numbers are represented by z_0 and z_1, respectively, with the bulk coordination number expressed as $z = z_0 + 2z_1$. The parameter $z_s = z_0 v + z_1$ can be regarded as the surface coordination number but actually accounts for the surface energy [recall that all quantities in Eq. (1) are normalized to J_b]. Label $\uparrow\downarrow / \downarrow / \uparrow\downarrow$ stands for a N-layer film with AFM surfaces and down magnetization in the remaining $(N-2)$ layers.

From Fig. 1(a) one can see that a GS structure with AFM surface coexists with a ferromagnetic bulk for $H \in (-z_s - h, -z)$, while the contrary occurs for $H \in (z_s - h, z)$. In between, i.e. for $H \in (-z, z_s - h)$, the GS is AFM in both the surfaces and the bulk. The film dis-

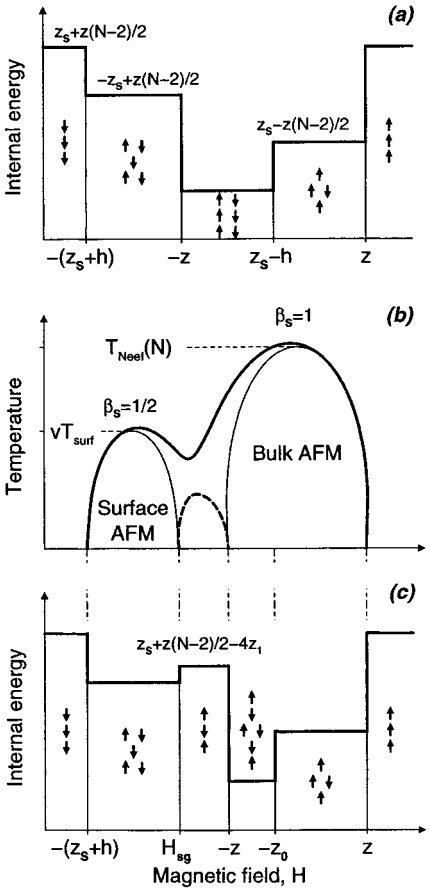

Fig. 1. Schematic representation of the ground-state and finite-temperature phase diagrams for films under intense surface fields. As a function of the external field H, a film of N layers transits between the ground states (GS) displayed in (a) if $h < h_g$ or in those showed in (c) if $h > h_g$. For the former case, the corresponding $H-T$ critical line is shown in (b) in a thick solid line. (c) For $h > h_g$ a ferromagnetic (disordered) gap intervenes between $\uparrow\downarrow / \downarrow / \uparrow\downarrow$ ground state and the zero-magnetization structure. The characteristic field between the disordered gap and the GS with surface AFM order is $H_{sg} = z_s - 2z_1 - h$. For h slightly above h_g, thermal excitations turns the ferromagnetic gap into a disordered region in the $H-T$ plane [dashed line in (b)]. For very intense surface fields the otherwise connected AFM region splits into two separate critical curves [thin solid lines in (b)]. See the text for further explanations

plays an ordered, compact domain from $H = -(z_s + h)$ to $H = z$. In Fig. 1(b), in thick solid line, we show the critical curve (schematic) in the H–T plane associated with the GS sequence of Fig. 1(a).

For negative values of the external field ($H \leqq -z$), the surface field favors the AFM ordering at the surfaces but also promotes the decoupling of the surface layers from the rest. Therefore, the inner layers closely behave as a $(N-2)$-layer film with neutral boundary conditions. In Fig. 1(b) with thin lines and appropriately shifted, we have plotted the corresponding critical curves for a 2D square lattice (left) and the corresponding $(N-2)$-layer film at $h = 0$ (right). Observe that the shoulder shows a maximum temperature $\sim vT_{\text{surf}}$, where T_{surf} is the Néel temperature of the 2D square lattice.

The ground-state sequence in Fig. 1(a) becomes unstable upon an increment in the surface field, and the GS sequence of Fig. 1(c) is then adopted by the film. Note the a disordered gap (GS $\uparrow / \downarrow / \uparrow$) and a new zero-magnetization ground structure ($\uparrow / \downarrow / \uparrow\downarrow / \downarrow / \uparrow$: an up-magnetization state at the surfaces, subsurface layers down magnetization and the rest in the AFM state) appear in lieu of the homogeneous AFM-GS structure. It can be shown [14] that the transition between GS sequences, from the sequence in Fig. 1(a) to the one in Fig. 1(c), occurs at a surface field value h_g given by

$$h_g = z_0 v + (z_0 + z_1). \tag{2}$$

For a surface field slightly above h_g, the disordered gap transforms itself, via thermal excitations, into a disordered region in the plane H–T, right in the middle of the ordered region [Fig. 1(b), dashed line]. In other words, the phase diagram is composed by two critical lines [dashed and thick solid lines in Fig. 1(b)]. Upon high-temperature cooling and for $H \in (z_s - 2z_1 - h, -z)$, the system undergoes a phase transition from the high-temperature disordered state to an AFM state. A further decrease in temperature drives the system into a low-temperature disordered state.

Intense surface fields increase the disordered gap at zero temperature and, as a consequence, the height of the associated disordered region rises. At $h = h_s$, the AFM domain splits into the surface and the bulk critical curves. In Fig. 1(b) with thin lines, the critical curves associated with the surfaces and the bulk are presented for $h > h_s$. It is shown in Ref. [14] that the splitting point corresponds to a saddle point in the Hessian of the free energy as a function of T and H[4]). We have used the pair approximation of the cluster-variation method (CVM) [22] to evaluated the finite-temperature properties of Hamiltonian (1). Previous work have shown that for nonfrustrated lattices, such as the b.c.c. and simple cubic, the PA gives reliable results for the qualitative aspects of the phase equilibrium in restricted geometries [9 to 11].

In a sense, the splitting value of the surface field, h_s, represents at finite temperatures the role of h_g. Both characteristic values of the surface field h_g and h_s, are the answer for the following question: How intense need the surface field be, in order to split the otherwise compact AFM domain, into separate surface and bulk ordered regions? At zero Kelvin, the answer is independent of the film thickness: When surface field reaches the value of $h_g = z_0 v + (z_0 + z_1)$, the surface splits from the bulk independently

[4]) The matrix of second derivatives of the free energy with respect of the long-range order parameters is called the Hessian of the free energy. The Hessian is a function of the external fields and the order parameters.

of the number of layers. At finite temperatures, the answer is more involved since now thermal excitations enhance the coupling between the bulk and the surface layers. Fig. 2 shows two regimes of behavior for h_s as a function of the film thickness: for thin films the value of h_s increases with N while the contrary occurs for thick films. This peculiar behavior of $h_s(N)$ results from the balance between the surface Zeeman energy and the ordering energy. For very thin films the surface Zeeman energy easily overcomes the contribution of a bulk made of a few layers. In this regimen, increasing the thickness in the film is equivalent to enhancing the bulk contribution to the free energy. Thus, it is necessary to apply more intense surface fields to split the surfaces from the bulk.

For very thick films the splitting value of the surface field shows virtually no change as the thickness in the film is reduced. Near the splitting point, the magnetization profile decays very fast towards the bulk state as we move from the surfaces to the inner layers (see inset in Fig. 2). The surfaces are too far away to affect each other and, therefore, h_s corresponds to the semiinfinite value of the surface field h_s^{∞}. However, if the film thickness is reduced enough ($N \sim 50$ in Fig. 2), the perturbation introduced by the surfaces reaches the middle layers. The interplay between the surface and the finite-size effects is reflected as an increment in the value of h_s as the thickness is decreased.

In summary, we have shown that the rich magnetic behavior, previously reported in AFM thin films [9, 11], is directly related to the ground state properties of the films. We focused on the thermodynamic behavior for intense surface fields, since in that case the otherwise compact antiferromagnetic regions splits into surface- and bulk-driven critical curves. In the bulk-driven critical curve, the surfaces are less ordered than the layers in the bulk. On the other hand, along the line of phase transitions driven by the surface, the bulk layers are less ordered than the surfaces. For surface fields such as $h > h_s > h_g$, in which the critical curve is well separated into the bulk- and surface-driven AFM regions, the surface order parameter vanishes with exponent β_s, which in the mean field approximation used here, equals 1 for the bulk-driven critical line, whereas $\beta_s = \frac{1}{2}$ for the surface-driven phase transitions. However, our preliminary re-

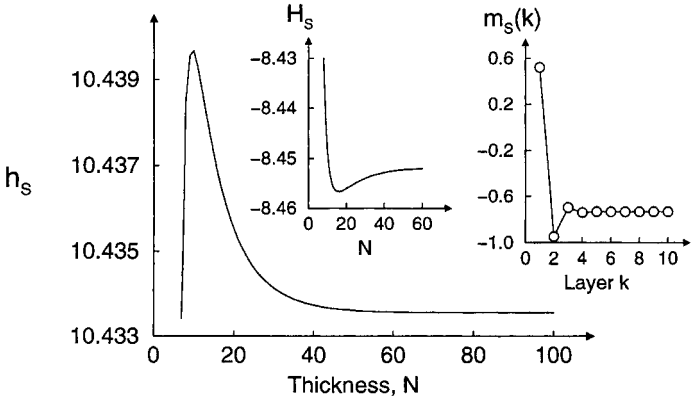

Fig. 2. Surface and bulk fields corresponding to the splitting point h_s and H_s (left inset), respectively, as a function of the thickness of the film. A magnetization profile for $N = 100$ film at $h = h_s$ is also shown in the right inset. Calculations were performed in the pair approximation of the CVM for b.c.c.(110) films with $v = 1$

sults show that *even for $h < h_g$ the surface exponent changes from $\beta_s = 1$ to $\beta_s = \frac{1}{2}$ as the external field is varied from positive to negative values.* The investigation of the critical behavior will be considered in the future.

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