Magnetization direction of a semi-infinite Heisenberg ferromagnet with perpendicular surface anisotropy

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The phase transition of a semi-infinite Heisenberg ferromagnet having easy-axis (perpendicular) anisotropy of the form $-D_s(S^z)^2$ ($D_s>0$) in the surface and easy-plane anisotropy of the form $-D_b(S^z)^2$ ($D_b<0$) in the bulk is studied by the molecular-field approximation. For a fixed D_b, three ordered phases, i.e., canted spin, in-plane ordering, and perpendicular ordering phases, are obtained depending on the temperature and the strength of the surface anisotropy D_s.

The direction of the magnetization of films and semi-infinite magnets has attracted much attention recently. In general, the preferred direction of magnetization in such materials is parallel to the surface because of a shape anisotropy. Surfaces, however, can have perpendicular anisotropy in consequence of the reduced symmetry,[1] and if the magnitude is large enough to overcome the shape anisotropy, the spins may exhibit a perpendicular ordering or have vertical components. In recent experimental and theoretical analyses, large perpendicular surface anisotropy and perpendicular magnetization were in fact found in films and semi-infinite magnets,[2-6] and several theories considering the interplay between the perpendicular anisotropy and the shape anisotropy have been proposed.[7-10]

There have been many investigations on the thermodynamic properties of Ising and Heisenberg semi-infinite magnets.[11,12] It is well established that the surface spins with weak exchange coupling are driven by the bulk and hence have the same transition temperature as the bulk. On the other hand, if the surface exchange coupling is greater than a critical value, the surface region can order even when the bulk is paramagnetic and has a transition temperature higher than the bulk one. As the temperature is lowered, the bulk also becomes ordered at the bulk transition temperature. Recently, the study of Ising semi-infinite ferromagnets has been extended to include a single-ion anisotropy, in particular, that of the easy-plane type.[13,14] Such a system undergoes first-order transitions as well as second-order ones.

Most of the theoretical studies on semi-infinite magnets have been concerned with the case where the surface and the bulk have the same easy direction of magnetization. If the surface anisotropy is noncollinear with the bulk, as encountered in the system with dominant perpendicular surface anisotropy, spin canting will appear near the surface. Mills,[7] and O'Handley and Woods[8] have shown in a continuum approach that the ground-state spin orientation near the surface changes from an in-plane to a canted one when the perpendicular surface anisotropy exceeds a certain critical value.

In this paper we treat a semi-infinite Heisenberg ferromagnet with a single-ion surface anisotropy whose easy direction is noncollinear with the bulk single-ion anisotropy and compute the phase diagram and magnetization profiles at finite temperatures. Since the thermodynamics of semi-infinite magnets have been described qualitatively within the framework of the molecular-field theory, we also apply it to our model Hamiltonian. This would be inadequate for very thin films with noncollinear anisotropy axes because a soft mode, which will appear at the phase boundary, yields an isotropic quasi-two-dimensional feature to the thermodynamic properties.[15]

The semi-infinite ferromagnet considered here has a simple-cubic structure with a free surface. The z-axis is normal and the xy plane is parallel to the surface. The bulk spins are assumed to lie in xy plane, while the surface spins orient preferentially to the z direction. To describe such a system, we set up a model Hamiltonian,

$$H=-J\sum_{<lj,l'j'>}\mathbf{S}_{lj}\cdot\mathbf{S}_{l'j'}-\sum_{lj}D_l(S_{lj}^z)^2\ , \tag{1}$$

where J denotes the nearest-neighbor ferromagnetic exchange constant (hereafter we shall use units of $J=1$), and l the layer index; $l=1$ is the surface layer, $l=2,3,\ldots$ the inner layers, and j denotes lattice points in xy plane. The perpendicular surface anisotropy is represented by taking D_1 ($\equiv D_s$) to be positive, while we take D_l ($\equiv D_b$) to be negative for the bulk.

Our model will allow for the possibility of three types of ordered states, i.e., canted spin, in-plane ordering, and perpendicular ordering states. Since the latter two are a special case of the former, we here develop the formulation for the canted spin state, where both the parallel component $<S_{lj}^x>=S_l^{\parallel}$ and the perpendicular one $<S_{lj}^z>=S_l^{\perp}$ have finite values. Then the Hamiltonian (1) is written in the molecular-field approximation,

$$\begin{aligned}&H_M=\sum_{lj}H_{Mlj}\ ,\\ &H_{Mlj}=-(K_lS_{lj}^x+E_lS_{lj}^z)-D_l(S_{lj}^z)^2+C_l\ ,\end{aligned} \tag{2}$$

where

$$\begin{aligned}&E_l=4S_l^{\perp}+S_{l-1}^{\perp}+S_{l+1}^{\perp}\ ,\\ &K_l=4S_l^{\parallel}+S_{l-1}^{\parallel}+S_{l+1}^{\parallel}\ ,\end{aligned} \tag{3}$$

$$C_l=\tfrac{1}{2}(K_lS_l^{\parallel}+E_lS_l^{\perp})\ , \tag{4}$$

where $S_0^{\perp}=S_0^{\parallel}=0$ by the boundary condition at the surface. For the spin-1 system considered here, the energy eigenvalues $\epsilon_{li}=\lambda_{li}-D_l+C_l$ $(i=1,2,3)$ of H_{Mlj} is obtained from the roots of the secular equation

$$\lambda_{li}^3-D_l\lambda_{li}^2-(K_l^2+E_l^2)\lambda_{li}+D_lE_l^2=0 \ . \tag{5}$$

Using the corresponding eigenstates, we have layer magnetizations in the canted spin state,

$$S_l^{\parallel}=-2K_l\sum_{i=1}^{3}\frac{\lambda_{li}(\lambda_{li}^2-E_l^2)}{(\lambda_{li}^2-E_l^2)^2+K_l^2(\lambda_{li}^2+E_l^2)}P_{li} \ , \tag{6}$$

$$S_l^{\perp}=-2E_lK_l^2\sum_{i=1}^{3}\frac{\lambda_{li}}{(\lambda_{li}^2-E_l^2)^2+K_l^2(\lambda_{li}^2+E_l^2)} \ , \tag{7}$$

with

$$P_{li}=\frac{\exp(-\beta\lambda_{li})}{\sum_{i=1}^{3}\exp(-\beta\lambda_{li})} \ , \tag{8}$$

and the free energy

$$F_c=N_{\parallel}\sum_{l=1}^{\infty}\left[C_l-D_l-\beta^{-1}\ln\sum_{i=1}^{3}\exp(-\beta\lambda_{li})\right] \ , \tag{9}$$

where $N_{\parallel}$ means the number of atoms in a plane, and $\beta=1/T$, with T as the absolute temperature scaled by J/k_B, with k_B as the Boltzmann constant.

The layer magnetization and the free energy of the in-plane ordering state can be obtained by setting $E_l\equiv 0$ in Eqs. (5), (6), and (9):

$$S_l^{\parallel}=\frac{4K_l\sinh(\beta X_l/2)}{X_l\{2\cosh(\beta X_l/2)+\exp(\beta D_l/2)\}} \ , \tag{10}$$

$$F_{\parallel}=N_{\parallel}\sum_{l=1}^{\infty}[(K_lS_l^{\parallel}-D_l)/2 -\beta^{-1}\ln\{2\cosh(\beta X_l/2)+\exp(\beta D_l/2)\}] \ , \tag{11}$$

with

$$X_l=\sqrt{D_l^2+4K_l^2} \ . \tag{12}$$

On the other hand, in the perpendicular ordering state they can be obtained by setting $K_l\equiv 0$ in Eqs. (5), (7), and (9):

$$S_l^{\perp}=\frac{2\sinh(\beta E_l)}{2\cosh(\beta E_l)+\exp(-\beta D_l)} \ , \tag{13}$$

$$F_{\perp}=N_{\parallel}\sum_{l=1}^{\infty}[E_lS_l^{\perp}/2 -\beta^{-1}\ln\{2\exp(\beta D_l)\cosh(\beta E_l)+1\}] \ . \tag{14}$$

The formulation developed here is a generalization of that for the infinite uniaxial ferromagnets, studied by Khajehpour, Wang, and Kromhout,[16] in the case of a semi-infinite ferromagnet, and then all the quantities become layer dependent. The bulk magnetization $S_b^{\parallel}$ is computed from Eq. (10) by omitting the layer dependence.

We now turn our attention to the phase diagram. For the numerical calculations, we use N-layer approximation, i.e., the layers with $l\geq N+1$ are assumed to be the bulk and the bulk magnetization $S_b^{\parallel}$ forces a molecular-field boundary condition to the Nth layer. This would be a good approximation if we choose N large enough to satisfy $S_N^{\parallel}\approx S_b^{\parallel}$, and in this paper we take $N=16$. In Fig. 1 a representative phase diagram in the (T,D_s) plane is shown for $D_b=-1$. The phase designated by C, $\parallel$, $\perp$, and P refer to canted spin, in-plane ordering, perpendicular ordering, and paramagnetic phases, respectively. In the canted spin phase in Fig. 1, Eqs. (6), (7), (10), and (13) have respective solutions, but among them the canted spin state has the lowest free energy. Since the exchange interaction in the Hamiltonian (1) is assumed to be isotropic, the transition lines shown in Fig. 1 are of second order. This is not the case if we introduce anisotropic exchange interaction[16] or consider the Ising model with easy-plane single-ion anisotropy.[13,14]

For D_s smaller than D_{s2}^c, the canted arrangement turns into the in-plane ordering as the temperature increases, and then a transition to the in-plane ordering state takes place. This feature is shown in Fig. 2 for the case with $D_s=2$, where the canting angle θ_l is measured with respect to the xy plane, and $S_l=\sqrt{(S_l^{\parallel})^2+(S_l^{\perp})^2}$. The curves S_l for $l\geq 4$ lie between the curves S_3 and the bulk and are omitted in the figure. It follows that a significant canting of spins appears within four layers from the surface, and the canting angle θ_l remains almost unaltered until the temperature approaches near the transition temperature to the in-plane ordering state. For D_s smaller than D_{s1}^c, all the spins are parallel to the surface for any temperature below T_b^c. The analytic form of the phase boundary between the phases $\parallel$ and C is given using the following procedure. By expanding the roots of the secular equation (5) into power series in $S_l^{\perp}$, and substituting them into Eqs. (7) and (9), we get a set of homogeneous equations,

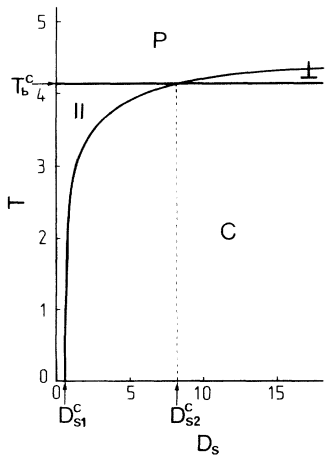

FIG. 1. (T,D_s) phase diagram for $D_b=-1$. C, $\parallel$, $\perp$, and P designate canted spin, in-plane ordering, perpendicular ordering, and paramagnetic phases, respectively. T_b^c is the bulk transition temperature.

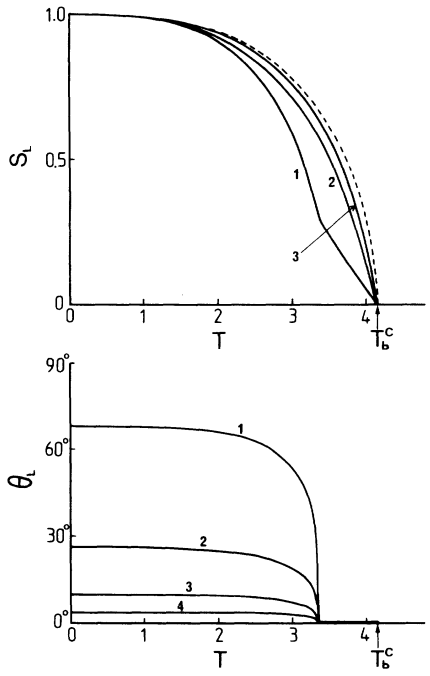

FIG. 2. Temperature dependence of $S_l=\sqrt{(S_l^{\parallel})^2+(S_l^{\perp})^2}$ and the canting angle θ_l measured with respect to xy plane for the case $D_b=-1$ and $D_s=2$. The number attached to the curves represent the layer ordering number from the surface and dashed line represents the bulk magnetization curve. T_b^c is the bulk transition temperature.

$$S_l^{\perp}+2\left[\frac{K_l^2}{X_l}\left[\frac{P_{l3}^0}{(\lambda_{l3}^0)^2}-\frac{P_{l1}^0}{(\lambda_{l1}^0)^2}\right]+\frac{D_l}{K_l^2}P_{l2}^0\right]E_l=0\ , \qquad (15)$$

where $P_{li}^0(i=1,2,3)$ is the probability given by Eq. (8) of

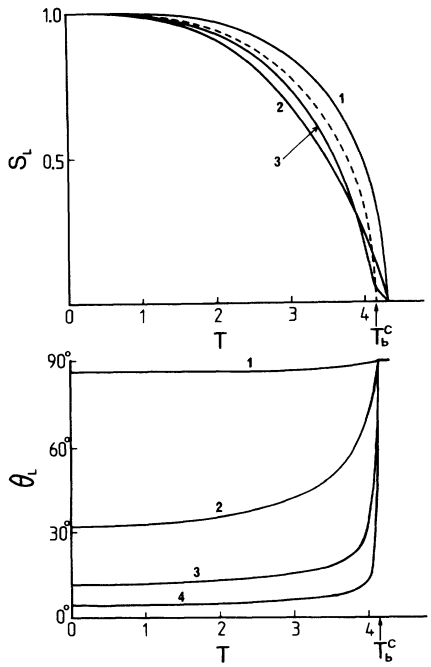

FIG. 3. Same as in Fig. 2 but for $D_s=14$.

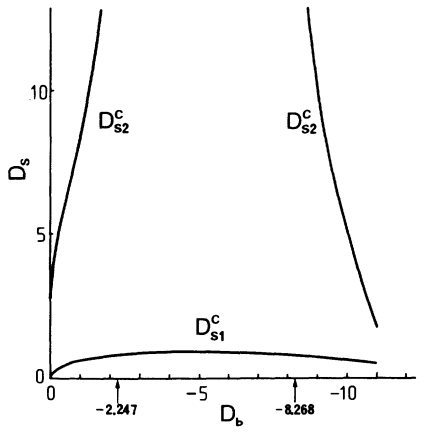

FIG. 4. Critical values D_{s1}^c and D_{s2}^c as a function of D_b.

the three energy states of the in-plane ordering state; $\lambda_{l1}^0=(D_l-X_l)/2$, $\lambda_{l2}^0=0$, and $\lambda_{l3}^0=(D_l+X_l)/2$. In the N-layer approximation, the coefficient matrix of Eq. (15) can be truncated, and the condition that the determinant of the coefficient matrix be zero gives the phase boundary. The boundaries between other phases are also found by a similar expansion for a small value of $S_l^{\perp}$ or $S_l^{\parallel}$.

For D_s larger than D_{s2}^c, as shown in Fig. 3 in the case with $D_s=14$, the canted arrangement turns into the perpendicular ordering as the temperature increases. The curves S_l for $l\geq 4$ lie between the curves S_3 and the bulk except around T_b^c and are omitted. As in the case $D_s=2$, the canted arrangement appears only near the surface, and θ_l remains almost unaltered until the temperature approaches near T_b^c. Above T_b^c, the perpendicular ordering takes place.

Finally, we discuss the phase diagrams for other selection of D_b. In Fig. 4 critical values D_{s1}^c and D_{s2}^c are plotted as a function of D_b. In the corresponding phase diagrams, the features of the in-plane ordering and the canted spin phases are qualitatively the same as shown in Fig. 1, but whether the perpendicular ordering phase exists or not depends sensitively on D_b. For $-2.247>D_b>-8.268$ no perpendicular ordering occurs, and in this range of D_b the canted spins turn into the in-plane orientation as the temperature approaches T_b^c. The appearance of the perpendicular ordering state for $D_b<-8.268$ is due to a decrease in T_b^c, which is characteristic of the system with easy-plane single-ion anisotropy.[16] Since the saturation magnetization of the bulk almost vanishes near $D_b=-12$, and consequently T_b^c becomes very low, the molecular-field approximation fails to determine the phase diagram in such a region. In this case, we should employ a method accurately taking into account a quasi-two-dimensional aspect of the surface.

Though in this paper we have not treated the dipole-dipole interactions directly and have confined ourselves within the framework of the molecular-field theory, we believe that the phase diagram shown in Fig. 1 exhibits the essential features of the semi-finite Heisenberg ferromagnet with perpendicular surface anisotropy.

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