New surface structures and spin-reorientation phase transitions in anisotropic magnetic superlattices

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Spin-reorientation phase transitions in anisotropic ferrimagnetic superlattices are analyzed. Expressions for the critical lines for second-order phase transitions are derived in the limit of an infinite number of layers for the case of magnetization along a hard axis. A μ–h phase diagram is constructed. Here μ is the relative magnetic moment of a unit cell of the superlattice, and h is the reduced external field. The external surface has a strong effect on the magnetic state of the superlattice. In particular, it leads to new surface magnetic phases. Whether the surface layer is isotropic or anisotropic is also an important factor.

Magnetic superlattices consisting of alternating layers of different magnetic materials are new artificial magnetic materials and thus of much fundamental and applied interest. Some new properties were recently discovered in these superlattices: a giant magnetoresistance[1] and spin-reorientation effects, among others. Although there have been an extremely large number of experimental studies of magnetization processes and spin-reorientation phase transitions (e.g., Refs. 2 and 3), the existing theoretical models either operate with an infinite medium or use numerical methods[4] and thus do not give a complete picture of the behavior of the magnetic structure of the superlattice in an external magnetic field. In particular, there has been essentially no study of how a magnetic anisotropy (bulk or surface) affects magnetic phase transitions of superlattices. The role of the external surface in the reorientation processes also deserves a close look. These questions are the focus of the present letter.

In this letter we present the results of calculations which incorporate the actual geometry of the superlattice. As we will see below, surface layers may strongly influence the magnetization process and the magnetic state of the superlattice.

The thermodynamic potential of a superlattice in which one component is isotropic, while the other is uniaxially anisotropic (e.g., a Gd/Co superlattice), can be described at low temperatures by

$$F=-\sum_{i=1}^{N/2+1} h\cos\vartheta_{2i-1}-\sum_{i=1}^{N/2}\frac{1}{\mu}h\cos\vartheta_{2i}+\sum_{i=1}^{N}\cos(\vartheta_i-\vartheta_{i+1})+\sum_{i=1}^{N/2}\frac{K}{2}\cos^2\vartheta_{2i}, \quad (1)$$

where the dimensionless parameters μ, h, and K are given by

$$\mu=d_1M_1/d_2M_2,\quad h=d_1H/\lambda M_2,\quad K=\tilde{K}/\lambda M_1M_2.$$

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Here d_1, d_2, M_1, and M_2 are respectively the thickness of the magnetic layers and the magnitudes of the magnetizations of the atomic planes of the superlattice components; $\tilde{K}$ is the second-order anisotropy constant; $\lambda > 0$ is proportional to the integral representing the exchange interaction across the interface between layers; and the expression $\lambda M_1 M_2$ is proportional to the energy of this interaction. We assume that the superlattice is an unbounded platelet. We denote the total number of layers in it by $N+1$. We assume that the first and last layers are identical, i.e., that N is even (this is an important point; in the opposite case, the phase diagram would be different). We denote by ϑ_k the angle between the external field H and the magnetization of the kth monolayer (of the composition-homogeneous part of the superlattice), assuming that the field is directed in the plane of the superlattice. Qualitatively, however, the results remain the same if the field is directed along the normal to the plane.

In writing (1) we assumed that the magnetizations of the monolayers are "saturated" and depend on only the temperature. We also assumed that the intralayer exchange interactions are significantly stronger than the interaction across an interface. We can thus assume that the magnetization is uniform within each monolayer.

Thermodynamic potential (1) describes a superlattice in which the surface layer is isotropic. We restrict the discussion here to the case in which the magnetization is along a hard axis; i.e., we assume $K>0$.

We have derived (in this model) conditions for the stability of various collinear magnetic phases, which depend on the parameters μ, h, and K and also on the total number of layers, $N+1$. "Collinear" here means that the magnetizations of the layers are collinear with **H**. In this case it is possible to analyze exactly the conditions under which the second-derivative matrix $A=\|\partial^2 F/\partial\vartheta_i\partial\vartheta_j\|$ is positive definite; in the process one finds two stability conditions.[5] One of them corresponds to stability of the inner layers of the superlattice and leads to expressions analogous to those for a ferrimagnet. The other incorporates the particular features of the boundary layers, which are weakly exchange-coupled. In this letter we will report results for the case in which the number of layers tends toward infinity; this limit corresponds to the approximation of a semi-infinite medium. If we ignore the presence of an external surface, the expressions derived here are similar to those for ferrimagnets (Ref. 6, for example).

Figure 1 shows the phase diagram in the coordinates $\mu = d_1M_1/d_2M_2$, $h = d_1H/\lambda M_2$.

The "ferrimagnetic" phase ($\vartheta_i = 0$) is stable under the conditions

$$h>2,\ \mu<h(h-2)/[h(K+2)-2K] \tag{2}$$

(line AB) or, in dimensional variables,

$$H>2\lambda M_2/d_1,$$

$$1/d_2M_2<H(H-2\lambda M_2/d_1)/[H(\tilde{K}+2\lambda M_1M_2)-2\tilde{K}\lambda M_2/d_1]. \tag{2a}$$

For "ferrimagnetic" phase 1 ($\vartheta_1 = \vartheta_3 \ldots = 0$, $\vartheta_2\ \vartheta_4 \ldots = \Pi$), which exists only under the condition $K<2$, we have

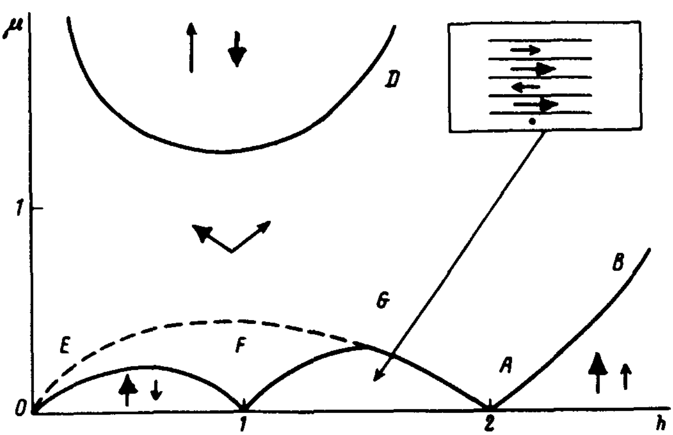

FIG. 1. Phase diagram of a ferrimagnetic superlattice in the limit of an infinite number of layers in a field h perpendicular to the easy axes. There is an isotropic layer at the surface. Solid lines—Lines of second-order phase transitions; dashed line—line of a transition from a surface-angular phase to an angular phase. The orientations of the layer magnetizations with respect to the external field are shown schematically. The smaller arrows represent the isotropic component, and the larger arrows the anisotropic component. The inset is a schematic diagram of the orientation of the magnetizations of the layers near the surface in the surface-reoriented phase.

$$h>2K/(2-K), \quad \mu>h(h+2)/[h(2-K)-2K] \tag{3}$$

(line CD) or, in dimensional variables,

$$H>2\lambda M_2\tilde{K}/[d_1(2\lambda M_1M_2-\tilde{K})],$$

$$1/d_2M_2>H(H+2\lambda M_2/d_1)/[H(2\lambda M_1M_2-\tilde{K})-2\tilde{K}\lambda M_2/d_1]. \tag{3a}$$

For "ferrimagnetic" phase 2 ($\vartheta_1=\vartheta_3=\ldots=\Pi$, $\vartheta_2=\vartheta_4=\ldots=0$) the stability conditions become

$$h<1, \quad \mu<h(1-h)/[h+K(1-h)] \tag{4}$$

(line EF) or

$$H<\lambda M_2/d_1,$$

$$1/d_2M_2<H[(\lambda M_2/d_1)-H]/[H(\lambda M_1M_2-\tilde{K})+\tilde{K}\lambda M_2/d_1]. \tag{4a}$$

Finally there exists a "surface-reoriented" phase ($\vartheta_1=0$, $\vartheta_3=\vartheta_5=\ldots\vartheta={}_{N-1}$ $=\Pi, \vartheta_{N+1}=0$, $\vartheta_2=\vartheta_4=\ldots=0$). In other words, the magnetizations of the boundary layers are oriented parallel to the external field, while the structure of ferrimagnetic phase 2 persists in the interior of the superlattice. The observation of this phase is a substantially new result; this phase has not been seen previously. The stability conditions become

$$1<h<2, \quad \mu<h(h-1)[1-2(h-1)^2]/[h+K(h-1)(2-h)] \tag{5}$$

(line FG) for $h<3/2$ and

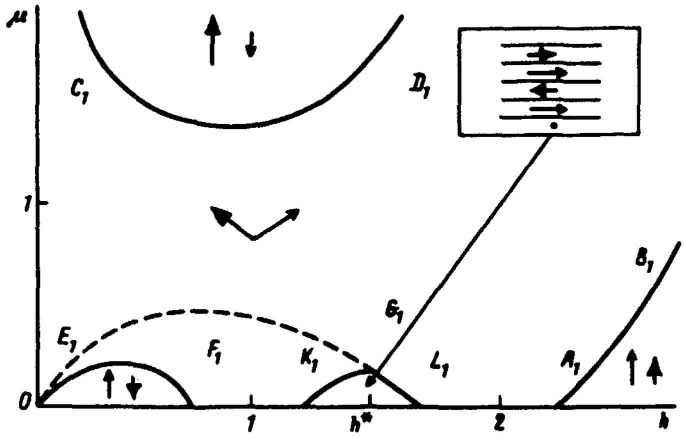

FIG. 2. Phase diagram of a ferrimagnetic superlattice in the limit of an infinite number of layers, in a field perpendicular to the easy axes. There is an anisotropic layer at the surface. The notation is the same as in Fig. 1.

$$\mu < h(2-h)/[h(K+2)-2h] \tag{6}$$

for $h > 3/2$ (line GA).

The rest of the phase diagram is occupied by an angular phase in which the moments make various angles with the field **H**. A "subphase" exists within the angular phase. In this subphase, the maximum deviation of the moments from a collinear orientation is observed near the surface; the deviation vanishes with distance into the interior of the superlattice. The region in which this subphase exists is bounded from above by curve EGA, which is given by Eq. (6), and from below by lines EF and FG [Eqs. (5) and (6), respectively]. Where EGA is intersected, the deviation of the moments is observed throughout the superlattice. This result can be interpreted as a hysteresis-free first-order phase transition in which two phases—collinear and angular—coexist. This "subphase" has been seen in some numerical simulations.[7] The other lines on the diagram are lines of second-order phase transitions.

We turn now to the case in which the surface layer is anisotropic. In the last sum in thermodynamic potential (1), we then need to replace the subscript $2i$ by $2i-1$, and we need to replace the upper limit on the summation, $N/2$, by $N/2+1$. Figure 2 shows the phase diagram of the superlattice in terms of the variables μ, h (for the case $K<1/2$). Let us examine the conditions for the stability of the same phases as in the former case.

The "ferromagnetic" phase is stable under the conditions

$$h > 2+K, \quad \mu < h[h-(2+K)]/[2(h-K)] \tag{7}$$

(line A_1B_1).

For "ferrimagnetic" phase 1 we have

$$h>K, \quad \mu>h[h-(K-2)]/[2(h-K)] \tag{8}$$

(line C_1D_1). "Ferrimagnetic" phase 2, which exists only under the condition $K<1$, is stable in the region

$$h<1-K, \quad \mu<h(1-K-h)/(h+K) \tag{9}$$

(line E_1F_1).

The "surface-reoriented" phase exists only under the condition $K<1/2$. Conditions for its existence are

$$1+K<h<2-K,$$

$$\mu<h[h-(1+K)][1-2(h-1)^2+2K(K-1)]/[h(1-4K)+K(1+4K)] \tag{10}$$

(line K_1G_1) for $h < h^*$ (Fig. 2) and

$$\mu<h(2-K-h)/[2(h+K)]$$

(line K_1L_1) for $h > h^*$. Conditions in terms of dimensional variables can be found by substituting the corresponding values for μ, h, and K into Eqs. (7)–(10). As K increases, there is a progressive "suppression" of the "surface-reoriented" phase (in the case $K>1/2$), of "ferrimagnetic" phase 2 ($K>1$), and of the "surface-angular" subphase, which is bounded from above by line $E_1G_1L_1$ and from below by lines K_1G_1, E_1F_1, and $\mu=0$ (for $K>2$).

Comparing Figs. 1 and 2, we see that if there is an anisotropic layer at the surface, the regions of stability of the "ferromagnetic" phase, of "ferrimagnetic" phase 2, and of the "surface-reoriented" phase are pushed apart, and the field region in which the angular phases are stable becomes broader. In the highly anisotropic case ($K>2$), the distinctive features are suppressed, and as a result the picture is analogous to a ferrimagnet (with a renormalized constant of the exchange interaction between sublattices).

In summary, we have studied the magnetic state of an anisotropic superlattice. We have derived stability conditions and the critical lines for second-order spin-reorientation phase transitions for various magnetic phases. We have found some new surface phases. We have shown that two second-order phase transitions can occur in the course of the magnetization. There is a certain topological similarity between these superlattice phase diagrams and the phase diagram of an anisotropic bulk ferrimagnet[6] in the case in which the field is perpendicular to the easy axis. When the spin reorientation begins at the surface, however, there are important differences. Furthermore, in superlattices we have the flexibility of being able to vary the ratio of the parameters of the exchange between layers and the magnitude of the effective anisotropy over wide ranges. We can thus realize a broad spectrum of possible spin-reorientation phase transitions in comparatively weak magnetic fields. Some interesting new possibilities arise when the actual crystallographic anisotropy between layers is taken into account. This topic will be the subject of future research.

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