Phase diagram of thin antiferromagnetic films in strong magnetic fields

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A theoretical study of the phase diagram of antiferromagnetic thin films is presented. The theory is based on a numerical self-consistent local-field calculation that allows for size and surface effects. The properties of magnetic structures as functions of the temperature and external field are calculated. Spatially nonuniform canted states are shown to intermediate the transition from the antiferromagnetic to the spin-flop phases. The model is applied to thin films of FeF_2 and MnF_2 as examples in order to clarify the role of anisotropy.

I. INTRODUCTION

In recent years the search for new materials for use in magnetic storage systems has driven a considerable amount of work on magnetic multilayer systems. For example enhanced magnetoresistance was found in multilayers of Fe/Cr,[1,2] Co/Cu (Ref. 3) and others. A series of experimental studies on layered structures consisting of ferromagnetic thin films with antiferromagnetic interlayer coupling [Fe/Cr,Co/Ru, (Ref. 4), Gd/Y (Ref. 5)] was followed by theoretical calculations and predictions of new equilibrium phases driven by weak external fields.[6–8]

While earlier studies concentrated on extended multilayer systems, recent work has focused on surface effects. Finite-size magnetic structures may be strongly influenced by surface effects since the low coordination at the surface allows the surface and subsurface spins to respond more easily to external magnetic fields. In fact surface phase transitions for magnetic multilayers have been predicted and observed for a number of systems including Fe/Gd (Refs. 9–11) and Fe/Cr (Refs. 12 and 13).

A very interesting study[13] has recently appeared which details the nature of a surface phase transition which takes place in a Fe/Cr (211) system. In this structure there are three key interactions which govern the equilibrium spin configuration: Zeeman interaction of the spins with the external field, the antiferromagnetic exchange interaction between Fe spins across the Cr layer, and an in-plane anisotropy field acting on the individual spins.

In many ways thin antiferromagnetic films are the microscopic counterpart of the antiferromagnetically coupled structures discussed above. The anisotropy of antiferromagnetic materials introduces an extra dimension in the analysis of equilibrium properties. This is a point of special interest because the features closely related to the anisotropy field can be checked experimentally, since the most recently studied uniaxial antiferromagnetic multilayered systems exhibit a wide range of values for the anisotropy field.[14,15] Considering the compounds FeF_2, CoF_2, and MnF_2, there is variation of the anisotropy field, in units of the exchange field, from 0.3 for FeF_2 to 0.015 for MnF_2.

Successful growth and characterization of antiferromagnetic films and superlattices of various layering patterns was reported a few years ago.[14] The dependence of the magnetic phase on the layering pattern of FeF_2/CoF_2 superlattices was reported on the basis of measurements of thermal-expansion coefficients. These results were analyzed theoretically and the observed dependence of the number of phases on the layering pattern was attributed to the coupling between the FeF_2 and CoF_2 layers and size effects.[16] In subsequent work the role of the interface coupling as well as the effect of the anisotropy on the weak-field properties of antiferromagnetic thin films and superlattices were studied theoretically.[17] It was shown that the antiferromagnetic-paramagnetic transition temperature of thin films decreases as the film is made thinner and that the downshift in the transition temperature is larger for compounds with low anisotropy.

The equilibrium spin configuration of antiferromagnetic thin films is expected to exhibit a significant temperature dependence because the low coordinated surface spins have a thermal averaged magnitude which decreases rapidly with temperature.[12] Therefore, for a given spin, the balance between exchange and anisotropy energies and the Zeeman energy is dependent upon its position with respect to the surface.

The previous analysis of spin structures included thermal effects only for systems where the external magnetic field was low enough that the antiferromagnetic phase was always stable. The present work concentrates on the analysis of equilibrium states of uniaxial antiferromagnetic films for arbitrary values of applied fields and temperature and particular attention is given to the spin-flop phase. FeF_2 and MnF_2 are chosen as examples so as to highlight the role of the anisotropy field. After a review of the theoretical method in the next section, we present the results for the spin profiles, phase diagrams, and magnetization curves in Secs. III, IV, and V. Concluding remarks are made in Sec. VI.

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II. THEORETICAL CONSIDERATIONS

The theoretical method is an extension of the method applied earlier to transition-metal–rare-earth multilayers and allows for the proper antiferromagnetic exchange between neighboring spins as well as external fields and anisotropy fields. Here we briefly review the main points. We use an iterative procedure seeking to find a structure (spin profile) in which each spin is in equilibrium with the local field imposed by the others. The magnetic structure of our model consists of a chain of spins, each of which represents a magnetic moment in the corresponding plane of the superlattice. The spins lie in the plane of the film, but the angular direction is allowed to vary from layer to layer. If we consider a spin S_n in layer n, it has an effective energy given by

$$E_n = -g\mu_B(\mathbf{H}_{ex}+\mathbf{H}_0)\cdot\mathbf{S}_n - KS_{nz}^2 \ . \tag{1}$$

Here H_0 is the applied field, directed along the z axis, and K is the anisotropy constant, with the easy axis also along the $\pm z$ directions. In the mean-field approximation the exchange field is given by

$$\mathbf{H}_{ex}=\frac{1}{g\mu_B}(z_{n,n-1}J_{n,n-1}\langle \mathbf{S}_{n-1}\rangle + z_{n,n+1}J_{n,n+1}\langle \mathbf{S}_{n+1}\rangle) \ , \tag{2}$$

where $J_{n,n-1}$ and $z_{n,n-1}$ are the exchange and coordination between a spin in the nth and $(n-1)$th layers and $\langle\ \rangle$ denotes thermal averages. The anisotropy field, H_a can be defined as

$$\mathbf{H}_a=\frac{2K}{g\mu_B}\langle S_{nz}\rangle\hat{z} \ . \tag{3}$$

With these definitions, the effective field acting on spin S_n is given by

$$\mathbf{H}_n=\mathbf{H}_{ex}+\mathbf{H}_a+\mathbf{H}_0 \ . \tag{4}$$

An iterative procedure is used to obtain the directions and thermal averaged values for the spins. For a given temperature (T) one initially assigns values for the angular direction and magnitude of each spin. Then a particular spin is chosen, say in layer n, and its angular position θ_n is rotated so the spin lies in the direction of the local effective field. The thermal equilibrium value of $\langle S_n\rangle$ is then found from

$$\langle S_n\rangle = SB_s(g\mu_B S_n H_n/kT) \ , \tag{5}$$

where $B_s(x)$ is the Brillouin function, and the effective field H_n is given by Eq. (4). A new spin is then chosen and the process is repeated until convergence is achieved, i.e., until a self-consistent final state emerges. By letting the spin system adapt itself to the local field at any particular site in the film, we obtain a spin profile that takes into account the finite structure.

Our model is applied to thin films of FeF_2 and MnF_2 with (001) surfaces.[14] These are bcc structures so the number of nearest neighbors $z=6$. The parameters used in calculation are as follows. For FeF_2 the spin is $S=2$, the exchange field is $H_e=434$ kG and the anisotropy field is given by $H_a=149$ kG. For CoF_2 the spin is $S=1.5$, the exchange and anisotropy fields are $H_e=324$ kG and $H_a=32$ kG and the values for MnF_2 are $S=2.5$, $H_e=465$ kG, and $H_a=6.97$ kG. These values are mean-field parameters and do not exactly match up with the measured values obtained through antiferromagnetic resonance. The values used here are those which give approximately correct transition temperatures for each compound and which have the correct ratio between exchange and anisotropy fields. The exchange parameters, J, are given by the usual expression $J=g\mu_B H_e/(2zS)$ for each material.

It is worthwhile to recall some of the general features of the spin-flop transition in *bulk* antiferromagnets. When the external field is small the magnetic structure is simple with the spins on opposite sublattices rigidly antiparallel. As the external field is increased the spin-flop state occurs where the spins on opposite sublattices are canted toward the applied field. The transition from the antiferromagnetic state to the spin-flop state can, in principle, take place at different values of the external field depending on the exact nature of the transition. The thermodynamic critical field occurs at a value

$$H_{th}=\sqrt{2H_eH_a-H_a^2} \ . \tag{6}$$

At H_{th} the energy of the spin-flop state and the antiparallel state are equal. The antiferromagnetic state, however, is still stable (i.e., in a local minimum of energy) until a larger field is applied. The stability limit of the antiferromagnetic state is given by

$$H_{af}=\sqrt{2H_eH_a+H_a^2} \ . \tag{7}$$

Finally, if the system is in the spin-flop phase and the external field is reduced, the spin-flop phase becomes unstable at a critical field given by

$$H_{sf}=\left[\frac{2H_e-H_a}{2H_e+H_a}\right]\sqrt{2H_eH_a-H_a^2} \ . \tag{8}$$

The exact field at which the phase transition from the antiferromagnetic state to the spin-flop state depends on the existence of a mechanism which could remove the system from a local minimum of energy to a global minimum of the energy. The motion of domain walls in ferromagnets provides such a mechanism, for example. Experimental results in antiferromagnets also seem to indicate that the phase transition takes place at the thermodynamic critical field. Thus in our calculations we concentrate our studies on the phase transition which occurs when the free energies for the two phases are equal.

We note that a certain amount of care must be taken in calculating the free energy numerically. It has been pointed out previously[8] that one has to avoid double counting the average exchange energy. Here we must also avoid double counting the anisotropy energy. In the mean-field approximation the partition function for the spins in layer n is given by

$$Z_n=\frac{\sinh[(2S_n+1)g\mu_B H_n/2kT]}{\sinh[g\mu_B H_n/2kT]} . \quad (9)$$

The total partition function is then

$$Z=\prod_n Z_n . \quad (10)$$

Finally, the free energy for the entire structure is given by

$$F=-kT\sum_n \ln Z_n-\frac{1}{2}\sum_n g_n\mu_B\langle \mathbf{S}_n\rangle\cdot(\mathbf{H}_{ex}+\mathbf{H}_a) , \quad (11)$$

where the second term on the right eliminates the "double counting" of the average exchange and anisotropy energies. We note that this treatment of the anisotropy energy is only an approximation, but it does give the correct results at $T=0$ and for high temperature. A detailed discussion of this point will be given elsewhere.

Surface spin-flop transitions were predicted in antiferromagnets many years ago.[18] In that work it was shown that the antiferromagnetic phase would become unstable at a field which was considerably lower than that of the bulk critical field. The surface spin-flop field for the limit where the antiferromagnetic state becomes unstable is given by

$$H_{surf}=H_{af}/\sqrt{2} . \quad (12)$$

Keffer and Chow later showed that the surface flop would evolve into a bulk spin-flop state as the magnetic field was increased.[19] Although the validity of these earlier calculations has been challenged recently,[20] the experimental results on metallic multilayers[13] has indeed shown that a surface spin flop takes place at approximately the value given by Eq. (12).

III. SPIN PROFILES: SIZE, FIELD, AND TEMPERATURE EFFECTS

The spin structure of an antiferromagnetic film depends critically on a number of parameters including applied field, temperature, and number of atomic layers. As indicated in previous studies, a number of possible phases exist. These include an antiferromagnetic (AF) phase where the spins in neighboring layers are strictly antiparallel, a bulk spin-flop (SF) phase where the spins in neighboring layers are canted with respect to the applied magnetic field, and a surface spin-flop phase where the spins in the outer atomic layers are canted but those deep inside the film are essentially antiparallel.

Before we discuss the details of the phase diagram, it is helpful to examine some typical spin profiles which shed some light on the nature of the different phases which can exist in thin films. In a thin film the interplay between the Zeeman and exchange energies is significantly influenced by the reduction of coordination near the surface and the value of the anisotropy field. In general the surface spins are more susceptible to the orientation of the applied field and the value of the temperature. The overall effect in the film, though, depends also on the way the spin system as a whole adapts itself to minimize the total free energy. High-anisotropy compounds tend to have more uniform spin profiles and restrict the surface effects to the immediate neighborhood of the surface region. On the other hand the equilibrium configuration of low-anisotropy compound films relies almost entirely on the exchange field and the applied field. This has two major consequences: first the surface effect itself is stronger for low-anisotropy materials because the lack of coordination produces a larger impact on the stability of surface spins; and second the spin system as a whole is almost isotropic and therefore can adapt itself more easily to any orientation. Therefore, small deviations from either the antiferromagnetic or the spin-flop state are possible and the surface effects may extend deep into the middle of the film.

These facts determine the way the finite spin systems respond to applied fields. Thin films of high-anisotropy compounds have nearly the bulk properties whereas thin films of low-anisotropy compounds show significant deviations from bulklike behavior.

Figures 1(a) and 1(b) display the canting angle away from the applied field as a function of position for each layer of spins in FeF_2 films containing 11 magnetic planes, respectively. The profiles for each temperature are presented for applied fields just above the spin-flop transition H_{th}. It is seen that the effect of increasing the temperature is to produce a less uniform profile in that the canting angle becomes more strongly position dependent. We also see that the nonuniformity in canting angle is confined primarily to spins near the surface. For $T=0$, only three spins near each surface are not near a normal canted state. For $T=39$ K, half the Neél temperature, the inhomogeneity penetrates deeper in the film. In these figures only the absolute value of the angles have been shown so that one may easily compare the canting angles for the two sublattices.

Figure 1(c) shows the deviations in the thermal averaged magnitude of the spins as a function of atomic layer.

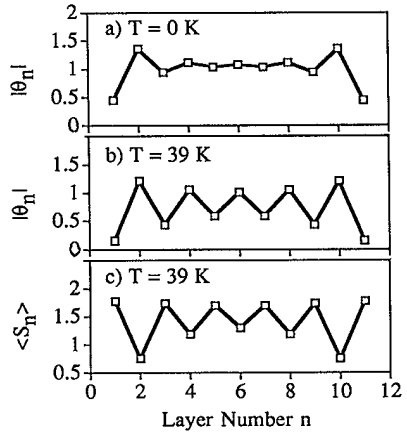

FIG. 1. Orientation and magnitude of the spins in an 11-layer FeF_2 film at the spin-flop transition at two temperatures. (a) $H_{th}=350$ kG, (b) $H_{th}=382$ kG, and (c) $H_{th}=382$ kG. The absolute value of the angle away from the magnetic field $|\theta_n|$ is shown at each temperature for visual convenience. The actual angles alternate in sign.

The variation in magnitude follows from the canting structure. Those spins which are pointing in a direction close to the applied field have a larger thermal averaged magnitude, while those spins which point away from the applied field see a smaller effective field and as a result have a smaller thermal averaged magnitude.

We note that the thermal dependence for magnetic multilayers such as Fe/Cr may be quite different. In our model, all of the exchange coupling takes place between spins in different planes. In Fe/Cr multilayers, in contrast, the Fe moments are coupled strongly within a film, both in-plane and between atomic planes. As a result, the Fe moments are not as likely to show strong variations from film to film at moderate temperatures.

In Fig. 2 we explore how the spin profile depends on the applied field for a 21-layer FeF_2 film at $T=39$ K. We see that at the lower field there is considerable oscillation in the canting angle, with large deviations occurring near the surfaces. The penetration depth for surface perturbation is about six atomic layers at this field. In Fig. 2(b) the external field has been increased. Now nearly all the interior spins have taken on the bulk canting angle. Furthermore the penetration depth for the surface perturbation has decreased significantly. This is similar to a result found previously in magnetic multilayers in the absence of anisotropy.[12]

The equivalent results, spin profiles as functions of temperature and applied field, for MnF_2 are presented in Figs. 3 and 4. In contrast to the FeF_2 results, the spin profile for MnF_2 is highly inhomogeneous near the spin-flop transition. The curves in Fig. 3 show a large nonuniform distribution of angles at the transition from the antiferromagnetic to the spin-flop phase. The canting angle on one sublattice is close to zero while the canting angle for the other sublattice is close to π. This is very different from the bulk spin-flop phase where the canting angles on the two sublattices are the same. The reason that the results for MnF_2 are so different from those of FeF_2 is that the anisotropy is much weaker in MnF_2. Thus the spin system is comparatively looser (as far as direction is concerned) in MnF_2, and the structure can change in a nearly continuous manner from an antiferromagnetic state to the uniformly canted spin-flop state as the magnetic field is increased. Again these results are similar to those for systems with no anisotropy.[12]

In Fig. 4 it is shown that even a strong field (about 50% larger than the bulk spin-flop field) is not enough to produce canted state in MnF_2. Only at much higher fields (numerical results now shown), is the uniform cant-

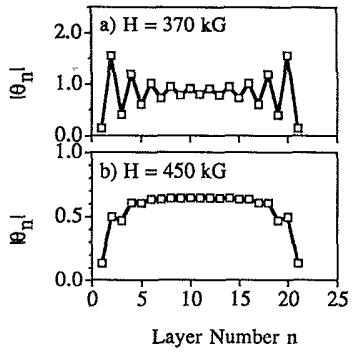

FIG. 2. Orientation of the spins with respect to the applied field for an FeF_2 film with 21 magnetic planes at $T=39$ K for different applied fields. The absolute value of the canting angle away from the applied magnetic field is shown.

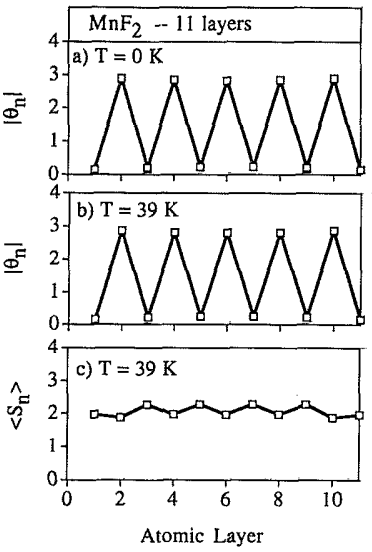

FIG. 3. Orientation and magnitude of the spins in an 11-layer MnF_2 film at the spin-flop transition at two temperatures. (a) $H_{th}=115$ kG, (b) $H_{th}=129$ kG, and (c) $H_{th}=129$ kG. The absolute value of the angle away from the magnetic field is shown at each temperature for visual convenience. The actual angles alternate in sign.

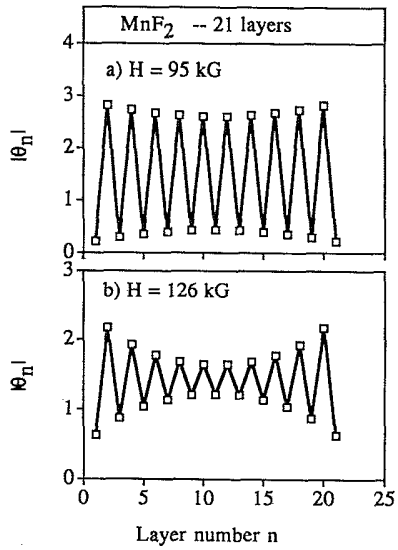

FIG. 4. Orientation of the spins with respect to the applied field for a MnF_2 film with 21 magnetic layers for two different fields at $T=0$ K. The absolute value of the angle away from the c axis is shown in units of the bulk canting angle at $T=0$ K for each field. Note the long penetration depths of the surface effects.

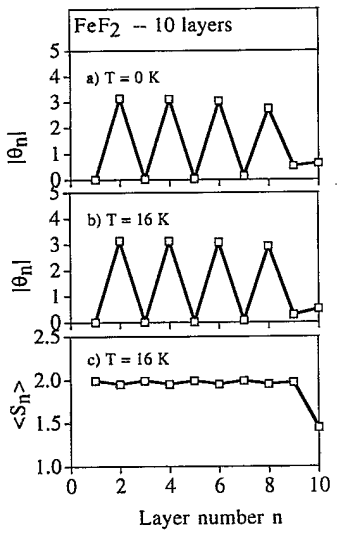

FIG. 5. Orientation and magnitude of the spins in a ten-layer FeF_2 film at the spin-flop transition at two temperatures. (a) $H_{th}=214$ kG, (b) $H_{th}=214$ kG, and (c) $H_{th}=214$ kG. The absolute value of the angle away from the magnetic field is shown at each temperature for visual convenience. The actual angles alternate in sign.

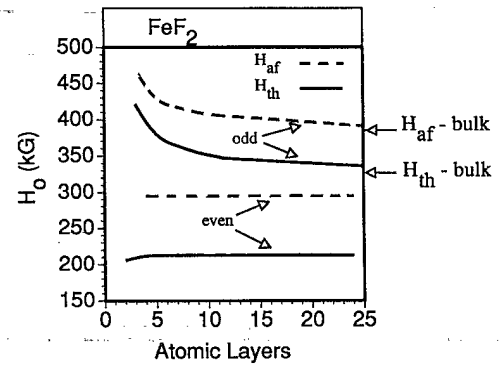

FIG. 6. Thickness dependence of the critical fields H_{sf} and H_{th} for FeF_2 films at $T=0$ K. The films with an even number of layers never reach the bulk values because there is always a surface spin-flop transition.

ed state reached. Again, this result produces an interesting contrast to the results for FeF_2 shown in Fig. 2.

In the above results we have concentrated on films with an odd number of atomic planes. The results are significantly different for systems with an even number of planes since a surface spin flop can take place. In Fig. 5 we present results showing the angular position for a ten-layer FeF_2 film. Again we have chosen the applied field to be just above that necessary to cause a spin flop. In Fig. 5(a) we see that the outer spin on the right is nearly reversed from the position it would have in the antiferromagnetic state. There are small shifts in the angular position of the nearby spins resulting in a penetration depth of 3–4 atomic layers. The angular positions at a temperature of 16 K [Fig. 5(b)] are similar, with a slightly smaller penetration depth for the surface spin flop. It is interesting to note that the thermal averaged magnitudes [Fig. 5(c)] show a significant deviation only for the outermost spin. Since the detailed study of the evolution of the surface spin-flop state to the bulk spin-flop state has been given recently,[13] we do not pursue this topic further.

IV. PHASE DIAGRAMS

As noted previously, earlier studies have detailed the nature of the surface spin-flop phase and its transition to the bulk spin flop. In this paper we deal with thin films and there is nearly always some surface character to the spin-flop phase. As a result we will often only identify the transition from the antiferromagnetic phase to a spin-flop phase without identifying whether the spin-flop phase is primarily a surface phase or a bulk phase.

We first examine the phase boundaries as a function of the number of atomic layers at $T=0$. In Fig. 6 we show the dependence of the critical fields H_{th} and H_{af} for films with both even and odd numbers of atomic layers. The films with an even number of layers have a spin-flop field significantly reduced from the bulk value. In fact, the critical field in the films with an even number of layers is close to the previously calculated value for the surface spin-flop field H_{surf}. In contrast, the spin-flop field for the films with an odd number of layers is considerably enhanced above the bulk value. The reasons for this are simple. For a system with an even number of layers, the spin at one end must initially point opposite to the external field. This is an energetically unfavorable position since it costs Zeeman energy, and it is at this end that the surface spin flop is nucleated. For a system with an odd number of layers one can have the spins at both ends of the film point in the direction of the applied field. This leads to a net magnetic moment in the direction of the field, even in the absence of the spin-flop state, and a corresponding lowering of the total energy of the system. As a result, the spin-flop state is not as energetically favorable at low fields and the critical fields are increased.

In Fig. 7 we present results for H_{th} as a function of atomic layers for MnF_2. MnF_2 has a very small anisotropy field compared to FeF_2 and so by comparing Figs. 6 and 7 we can see how anisotropy influences the phase diagram. We note that H_{th} and H_{af} are very close in MnF_2

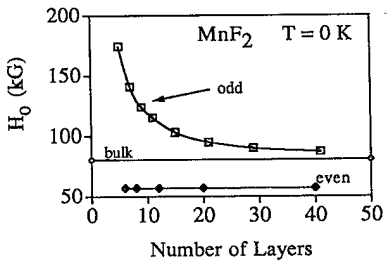

FIG. 7. Thickness dependence of the critical field H_{th} for MnF_2 films at $T=0$ K. The value of H_{af} is not shown since it is very close to H_{th} for this low-anisotropy material. Note that it takes more atomic layers for MnF_2 to reach the bulk values than for FeF_2. This is also due to the low anisotropy of MnF_2.

because of the small value for the anistropy field and we just show H_{th}. The most obvious difference between the two systems is that for MnF_2 it takes many atomic layers before the critical fields reach their bulk values. The reason for this can be understood in relation to domain walls in ferromagnets. In a domain wall the anisotropy energy tends to try to make the wall narrower since it is energetically favorable for spins to lie in particular directions. Similarly, in the antiferromagnetic film with high anisotropy the surface spin-flop phase tends to be more strongly confined to the surface region when compared to the low-anisotropy material. We have seen this explicitly in Figs. 1–4 which show that surface perturbations extend much more deeply into the bulk for the low-anisotropy material MnF_2.

We now turn to general field-temperature phase diagrams for films with a fixed number of atomic planes. The phase diagrams for FeF_2 films are presented in Figs. 8 and 9. For the 11-layer film in Fig. 8, the external field was applied parallel to the surface spins. The field dependence of the temperature of the antiferromagnetically aligned–paramagnetic (A) transition is very nearly quadratic. The value of the tricritical temperature (around $0.6T_N$) is in agreement with the available experimental data[21] for the bulk or thick films and the change for thin films is not appreciable. The AF-SF boundary, a relatively flat curve with a small upward curvature, is also in qualitative agreement with the experiments.

The phase diagrams for FeF_2 films with an even number of magnetic planes have very different features as can be seen in Fig. 9. As has been noted previously the transition fields are reduced considerably. Furthermore, the spin-flop region is now very small, and a new state, labeled A1, emerges. The A1 state corresponds to an antiferromagnetic state except that the outer spin on one side has been reversed so as to point along the external field instead of opposing it as it would in the antiferromagnetic state. At higher temperatures and fields there is an aligned state (A) where all the spins point in the direction of the external field.

For MnF_2 films we again see a rather different set of phase diagrams in Figs. 10 and 11. Since the anisotropy is weaker in MnF_2, the spin-flop state, which involves canting at arbitrary angles rather than alignment of spins in particular directions, can exist over a larger portion of the phase diagram. This pushes the temperature of the tricritical point to around $0.9T_N$, in agreement with the available experimental data[22] for thicker samples. The A1 phase seen in FeF_2 occurs only in a very narrow region of the phase diagram which does not show in the figure. In comparing Figs. 8–11, we also see that thin films containing an even number of magnetic planes have a clear reduction in the small field AF–A transition temperature. Odd numbered films do not show this effect as a result of the stabilization induced by the external field.

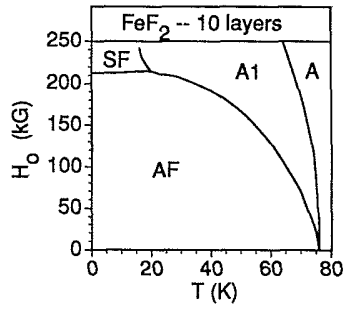

FIG. 9. Phase diagrams for a ten-layer FeF_2 film. The solid lines all indicate transition points where the free energies of the two phases are equal. The significant differences between the results for the 11-layer film and the ten-layer film are due to surface phase transitions. The phase A1 indicates that all of the spins are in the antiferromagnetic phase except for the outer layer of spins originally pointing opposite to the applied field has been reversed.

V. MAGNETIZATION FOR THIN FILMS

The existence of the phase transitions in the previous section can be seen in a number of different ways. Previous work has shown that the static susceptibility can demonstrate both bulk and surface phase transitions.[13]

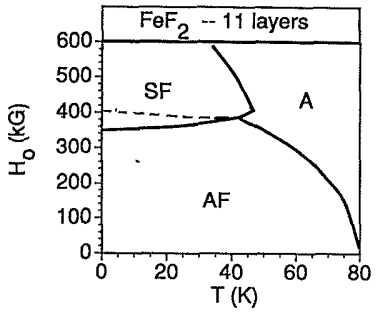

FIG. 8. Phase diagrams for an 11-layer FeF_2 film. The dashed line indicates the critical field H_{af} where the antiferromagnetic phase becomes unstable. The solid lines indicate transition points where the free energies of the two phases are equal. SF indicates the spin-flop phase; AF indicates the antiferromagnetic phase, and A indicates the aligned-paramagnetic state where all spins point along the applied field.

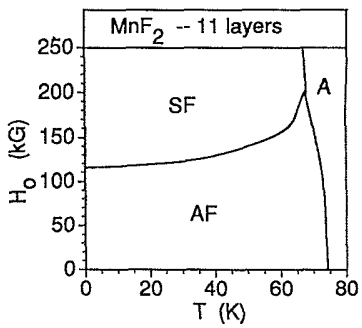

FIG. 10. Phase diagrams for an 11-layer MnF_2 film. The solid lines all indicate transition points where the free energies of the two phases are equal. Note that the spin-flop phase extends considerably farther in temperature for MnF_2 than in FeF_2.

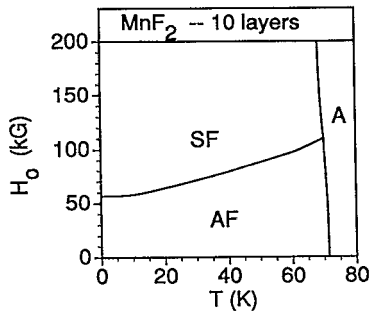

FIG. 11. Phase diagrams for a ten-layer MnF_2 film. The solid lines all indicate transition points where the free energies of the two phases are equal. The reduction in the critical field (compared to Fig. 10) is due to a surface spin flop.

In addition, heat-capacity measurements can also indicate phase transitions.[14,16] Here we look at the results of the phase transitions directly on the magnetization.

In Fig. 12 we examine the magnetization of an 11-layer FeF_2 film as a function of applied field at different temperatures. This corresponds to sampling the phase diagram of Fig. 8 along different vertical lines corresponding to the different temperatures. We see that the phase transitions from the antiferromagnetic state to the spin-flop state are accompanied by rapid changes in the magnetization. In contrast, a phase transition from the antiferromagnetic state to the aligned state shows a continuous magnetization.

It is somewhat surprising that there is a transverse magnetization as well as a longitudinal magnetization. We note that due to the anisotropy and the finite structure it is possible to have a net moment that is not aligned with the external magnetic field. As we will see, the transverse magnetization exists for films with both odd and even numbers of atomic layers. However, the origin of this transverse moment depends on whether the number of layers is odd or even. For an odd number of layers, we have seen that the spin structure is symmetric about the midplane. However, the number of atomic layers for the two sublattices is different. As a result, it is not too surprising that the canting of the two sublattices is also different. The transverse magnetization occurs because of the nonequivalent canting of the two sublattices. We note that the transverse magnetization extends over a large range of magnetic fields and slowly decays to zero as the field is increased.

In Fig. 13 we examine the magnetization of a ten-layer FeF_2 film as a function of applied field and again at different temperatures. This corresponds to a sampling of the phase diagram of Fig. 9. Again we see rapid changes in magnetization as one crosses the boundary between the spin-flop phase and the antiferromagnetic phase. The existence of the transverse magnetization is now due to the surface nature of the spin-flop phase. The magnetic structure in the surface spin-flop phase is not symmetric about the midplane and this is reflected in the existence of the transverse magnetization. As the magnetic field is increased the structure becomes slightly more symmetric and the transverse magnetization is reduced. The rapid drop in transverse magnetization and small increase in longitudinal magnetization at $H=235$ kG for the $T=0$ curve shows a transition to a state very close to the A1 state.

We have studied similar magnetization curves for MnF_2. Unfortunately, in this small anisotropy compound it appears that near the phase transitions a number of self-consistent final states occur, all at about the same energy, but with slightly different longitudinal magnetizations and significantly different transverse magnetizations. It is for this reason that we do not present figures for MnF_2 magnetizations. The general trends for the MnF_2 results seem qualitatively to match those for FeF_2, however the transverse moments are considerably smaller in magnitude than in FeF_2 because the anisotropy is much smaller in MnF_2.

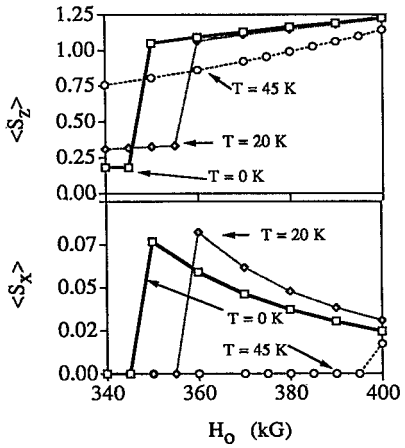

FIG. 12. Magnetization as a function of applied field for different temperatures for an 11-layer FeF_2 film. (a) longitudinal magnetization and (b) transverse magnetization. Note that the rapid changes in magnetization correspond to crossing a phase transition line in Fig. 8.

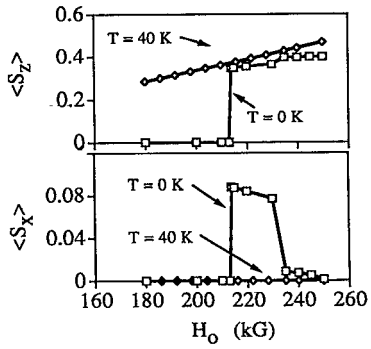

FIG. 13. Magnetization as a function of applied field for different temperatures for a ten-layer FeF_2 film. (a) longitudinal magnetization and (b) transverse magnetization. The rapid changes in magnetization correspond to crossing a phase transition line in Fig. 9.

VI. CONCLUSIONS

Our results indicate that anisotropy plays a significant role in determining the equilibrium properties of antiferromagnetic thin films. Compounds with low anisotropy are much more sensitive to surface effects, since high value anisotropy tends to localize surface perturbations to the region right near the surface.

Spatially nonuniform canting states were found to intermediate the transition from the antiferromagnetic phase to the spin-flop phase. For high-anisotropy compounds, such as FeF_2, the spin profile was shown to be nearly uniform, with surface effects restricted to the spins in the immediate neighborhood of the surface. The bulk canting angle as well as the spin-flop field are reached for films with a few tens of magnetic planes.

Thin films of low-anisotropy compounds (MnF_2 as a typical example) are strongly affected by surface effects, which are present even for thick films (101 magnetic planes, numerical results not shown here). For these compounds there is a marked influence of nonuniform canting states. No matter how thick the film is, near the antiferromagnetic–spin-flop transition, the spin angle profile is far from uniform and the bulk properties are not regained even for thick films. This reflects the fact that, since the anisotropy field is small, all the stability of the spin system relies on the exchange field. Therefore the low coordination of the surface spins strongly affects the equilibrium properties. Furthermore, contrary to intuition, the surface effects do not disappear for thick films.[12] Instead, the softer coupled surface spins influence not only the surface region but also the whole film.

The general shape of the phase diagrams for both compounds studied here is in agreement with the available experimental data. We see a significant difference in the phase diagrams between films with an even number of layers and an odd number of layers. This reflects the existence of the surface spin-flop state for the films with an even number of layers. Finally, we find that the transition from the spin flop to an aligned state occurs at a significantly higher temperature for compounds with low anisotropy.

Although the surface effects are stronger for low-anisotropy compounds, thin films in general display finite-size effects. This feature is expected to affect the spin-wave spectrum. The possible oscillatory modes of the spin system around the equilibrium configuration is expected to be affected by surface and size effects. The exact ground state, incorporating the finite-size effects, is essential to determine the elementary excitations. Characteristic frequencies of thin films may differ from the bulk values. It will be interesting to see further experimental results in thin antiferromagnetic films, including antiferromagnetic resonance, compared to the results of the present theory.

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