Solid State Communications, Vol. 82, No. 3, pp. 161-164, 1992.
Printed in Great Britain.

0038-1098/92$5.00+.00
Pergamon Press plc

SIZE AND INTERFACE EFFECTS IN THE PHASE TRANSITIONS OF ANTIFERROMAGNETIC SUPERLATTICES

A.S.Carrico* and R.E.Camley

Department of Physics
University of Colorado, Colorado Springs CO 80933-7150

(Received 16 December 1991 by A. A. Maradudin)

A self-consistent mean field theory is used to study composition effects on the phase transitions of FeF_2/CoF_2 and FeF_2/MnF_2 superlattices under weak applied fields. The temperature dependence of the magnetization is shown to depend strongly on the superlattice layering pattern and also on the interface coupling. Thin films of low anisotropy compounds display stronger shifts of transition temperature. Comparison with bulk results shows that size effects persist even for very thick layers for weakly coupled systems.

The excellent control of experimental techniques to grow layered systems has recently motivated a great deal of both experimental and theoretical work on the properties of magnetic multilayers. The freedom to chose the layering pattern of superlattices makes it possible nowadays to tailor their magnetic properties significantly.

In the literature on magnetic layered systems considerable attention has now been paid to multilayers made of ferromagnetic materials coupled antiferromagnetically at the interfaces [1-7]. A number of magnetic phases have been reported for these systems along with careful studies of the magnetic phase diagram[3]. Of particular interest is the fact that phase transitions in these systems can be obtained relatively easily, requiring only small applied fields or small changes in temperature. The interplay between the exchange and Zeeman energies is the fundamental mechanism governing these phase transitions[5]. As a result, the antiferromagnetic interfacial coupling plays a key role in these materials.

Ionic antiferromagnetic superlattices in a sense materialize the above features in a much smaller scale. The competition between exchange and Zeeman energies involves spins in neighboring layers instead of the bigger ferromagnetic constitutive blocks of transition metal multilayers. There is therefore a more delicate balance between the tendency of alignment with external fields and the natural antiferromagnetic structure. For a review of theoretical work on phase transitions of antiferromagnetic superlattices for T=0, and on nonlinear effects on the dynamical susceptibility see reference 8.

A systematic experimental study of antiferromagnetic multilayers at finite temperatures was started in recent years[9-10]. The measured temperature dependence of thermal expansion coefficients of FeF_2/CoF_2 superlattices indicated strong size effects[10]. Most interesting was the observation that the number of phase transitions in the absence of applied fields depends on the layering pattern. For superlattices with thicker films two phase transitions were found. For superlattices with thin films only a high temperature phase transition was measured. We recently reported a possible theoretical interpretation[11] of these results indicating that the interface exchange coupling in the FeF_2/CoF_2 superlattices is strong as compared to the CoF_2 exchange. In this case the low temperature phase transition observed in samples with thick CoF_2 films involves the middle CoF_2 spins and reflects the more or less bulk environment away from the superlattice interface. For thinner films, the strong interface exchange stabilizes the CoF_2 spins and only the high temperature transition, associated with the FeF_2 films, is seen.

In the present paper we report a more detailed study of composition effects on the magnetization of ionic antiferromagnetic superlattices. The study of magnetization as a function of temperature is a valuable source of information on these structures as it can indicate the presence or absence of phase transitions and the nature of the phases as well. We consider only weak applied fields so as to emphasize the purely geometrical and interface effects and thus we do not consider any spin flop transitions in this paper. We also stress the role of the anisotropy field, and superlattices and films composed of antiferromagnets FeF_2, CoF_2 MnF_2 are considered.

Our results show that weak interface coupling favors an almost independent behavior of the two spin systems composing the superlattice and therefore size effects dominate the phase transitions. In this case each antiferromagnetic thin film undergoes the antiferromagnetic - paramagnetic transition at a temperature lower than its Neél temperature reflecting the reduction in transition temperature imposed by size effects, as reported recently[10]. As noted previously, strong interface coupling inhibits the low temperature transition for multilayers with thin films. Here we examine these features for structures different than those which were considered earlier. In particular we calculate the

* On leave from Departamento de Fisica
Universidade Federal do Rio Grande do Norte
59072 Natal/RN Brasil

magnetization as a function of temperature for single or double period superlattices FeF_2(m layers)/XF_2(n layers), with X=Co or X=Mn. For compositions with m+n=odd the magnetic unit cell is twice as large as the chemical unit cell and in the absence of external fields the magnetization is zero. For structures with m,n=even the cancellation of the sublattice magnetizations yields no net magnetization for the superlattice. In these cases we consider weak external fields to examine the phase transitions. Since double period superlattices and superlattices with m,n=even have similar dependence of the magnetization as a function of temperature, in response to applied fields, only the results for the even/even case are presented here.

The theoretical method is an extension of the theory applied earlier to transition-metal/rare-earth multilayers[5] allowing for the proper antiferromagnetic exchange between neighboring spins and including 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. Only the magnetic unit cell of the superlattice need to be considered and cyclic boundary conditions are imposed at the boundaries of the unit cell. (We note that the magnetic unit cell may sometimes be double that of the chemical unit cell.) The magnetic unit cell consists of a chain of spins, each of which represent a magnetic moment in the corresponding plane of the superlattice. The direction of each spin is fixed by the anisotropy field and they all lie in the c-axis, since only weak applied fields are considered.

The effective field acting on spin S_n is given by

$$H_n=(Z_{n-1}J_{n-1}\langle S_{n-1}\rangle+Z_{n+1}J_{n+1}\langle S_{n+1}\rangle + H_a\langle S_n\rangle)/g\mu_B + H_{ext} \qquad (1)$$

where J_{n-1} and Z_{n-1} are the exchange and coordination numbers between a spin in the n-th and (n-1)th layers. H_a and H_{ext} are the anisotropy and external fields respectively and $\langle\rangle$ denotes the thermal averages.

The thermal equilibrium values of $\langle S_n\rangle$, n within the magnetic unit cell, are found from the Brillouin function in the usual way

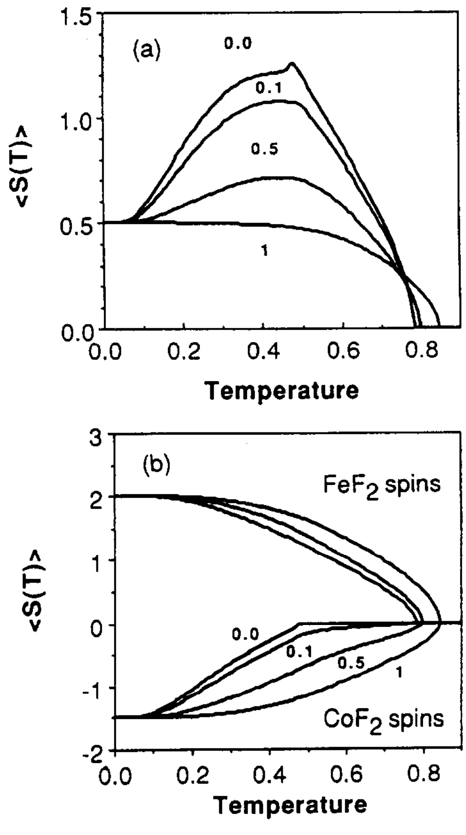
$$\langle S_n\rangle=B_s(g\mu_B S_n H_n/kT) \qquad (2)$$

where the effective field H_n is given by Eq.(1).

The numerical procedure to obtain the thermal values for the spins consists of solving Eq.1 and Eq.2 iteratively. For a given temperature (T) one assigns values to $\langle S_n\rangle$ and calculates the effective fields acting on all spins. Then a particular spin is chosen and Eqs. (1) and (2) are used to obtain the thermal averaged magnitude of that spin. A new spin is then chosen and the process is repeated until convergence is achieved, ie until a self-consistent final state emerges. By letting the spin system adapt itself to the local field at any particular site in the unit cell we obtain a spin profile that takes into account the composition of the superlattice. Not only the size of each film is considered but also the nature of the interface coupling.

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=6.97kG. 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 the 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.

The interaction of the two spin systems composing the superlattice is investigated by varying the interface exchange between weak and strong values. Here strong exchange is the case where the interface exchange constant is equal to that in FeF_2, the antiferromagnet with the higher Neél temperature. In Fig.1a we show the magnetization of the (3/7) FeF_2/CoF_2 system for four different values of the interface exchange. The temperature axis in this figure and the following ones is scaled to the bulk Neel temperature of FeF_2. It is readily seen that if the interface exchange is strong this results in a very slow decay of the superlattice magnetization. Therefore only the high temperature transition is seen. However if the interface coupling is weak, the spin



1) (a) Thermal average value of net spin per unit cell as a function of temperature for the (3/7) FeF_2/CoF_2 structure. (b) Magnetization of each compound in the (3/7) FeF_2/CoF_2 superlattice. The value of the interface exchange, J_I/J_{FeF_2} , is indicated by the numbers by the curves.

systems behave more or less independently and two transitions are clearly displayed, each occurring near the Neel temperature of one of the compounds. The low temperature transition is seen as a rapid increase in the net moment. This transition is not generally sharp because of the local exchange field produced by the high Neél temperature compound at the interface is different from the exchange field in the "bulk" of the film. All curves displays the same partial cancellation of the sublattice magnetizations at low temperature.

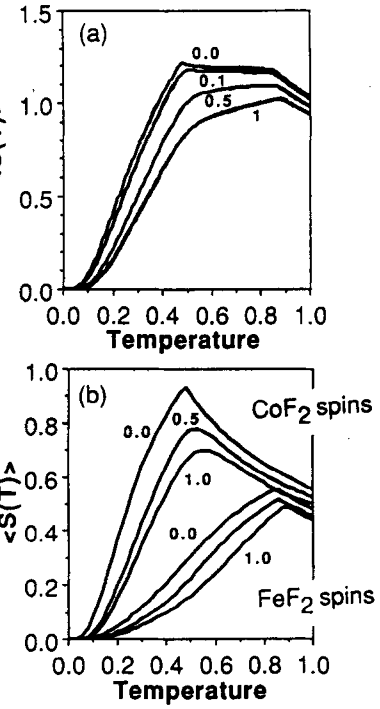
In order to display the details of the above features we show in Fig.1b the magnetization of each compound in the (3/7) FeF_2 superlattice as a function of temperature. Again we investigate this as a function of interface exchange. For no interface coupling the CoF_2 magnetization drops to zero near T=0.5. As the interface coupling is increased, the exchange field from the FeF_2 stabilizes the CoF_2 spins and the CoF_2 magnetization persists until the high temperature phase transition.

The FeF_2 magnetization seen in Fig. 1b is very close to that of a uncoupled 3 layer film, and we see in Fig. 1a and 1b that the FeF_2 magnetization displays clear size effects in that the transition temperature is reduced from 1. This reduction is less significant in the strong coupling limit than in the weak coupling limit. As we have pointed out, in the strong coupling case the CoF_2 spins are stabilized. The CoF_2 spins in turn produce an exchange field at the ends of the FeF_2 film which reduces the size effect in the FeF_2 film by helping to stabilize the FeF_2 spins at the ends of the FeF_2 film.

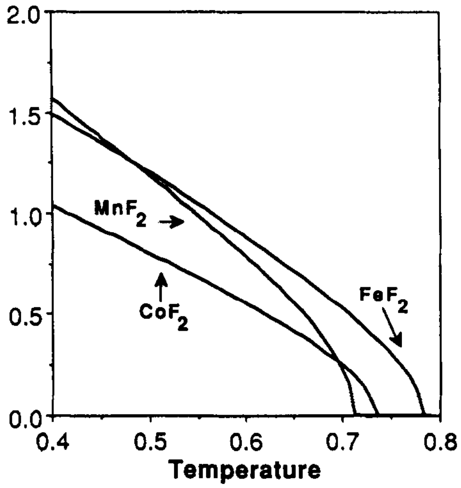
In order to examine the phase transitions of superlattices with no net magnetization (m,n=even), we applied external fields which are weak compared to the spin-flop fields. In Fig.2a we show the magnetization of the (4/8) FeF_2/CoF_2 superlattice subjected to an applied field of 0.1 in units of the FeF_2 exchange field. This amounts to approximately 43 kG. The numbers by the curves indicate the value of the interface exchange in units of the FeF_2 bulk exchange. The upper two curves, representing weak interface coupling, display clearly the CoF_2 transition at around T=0.5, as a change in slope. The third and fourth curves are for strongly coupled films. In this case the CoF_2 transition virtually disappears. In all cases the FeF_2 transition is seen at high temperatures (T=0.9).

The relatively low slope of the upper two curves above the CoF_2 transition temperature reflects the fact that in this region the CoF_2 magnetization is decreasing while the FeF_2 magnetization is still increasing in response to the applied field. This can be seen more clearly in Fig. 2b where we present the magnetization curves for the FeF_2 and CoF_2 films separately. We note that the net magnetization in each film does not decay to zero until well above the transition temperatures. This is a result of the application of the external magnetic field.

In the limit of extremely weak interface coupling the characteristics of each compound are separately observed in the superlattice magnetization. It is therefore of interest to explore the properties of films of the selected compounds. One point of interest is the possible role of the anisotropy in determining the magnetization. It is expected that compounds with smaller values for the anisotropy fields will show stronger size effects. In Fig.3 we examine the effect of



2) (a) Thermal average value of net spin per unit cell as a function of temperature for the (4/8) FeF_2/CoF_2 structure. (b) Magnetization of each compound in the (4/8) FeF_2/CoF_2 superlattice. The value of the interface exchange J_I/J_{FeF_2}, is indicated by the numbers by the curves. The curves represent the reponse to an applied field of 43 kG.



3) Magnetization of thin films of FeF_2, CoF_2 and MnF_2 as a function of temperature. All films have three layers of spins

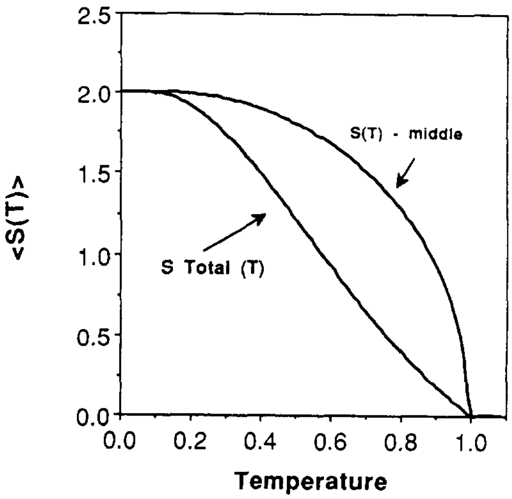
anisotropy on the phase transition of thin films. It is readily seen that CoF_2 and MnF_2 thin films have a bigger transition temperature shift as compared to that of a thin FeF_2 thin film. All films have three layers. We have also considered other thicknesses but we have not found any considerable effect for thick films.

Size effects are normally expected to disappear for thick films. We recently reported[11] that the magnetization of antiferromagnetic thin films decreases rapidly at low temperatures and vanishes at a temperature below the bulk Neel value. The shift in transition temperature was shown to vanish for thick films but the net magnetization as a function of temperature is far from the behavior of a single Brillouin function. This is somehow surprising since up to the transition temperature the majority of the spins in the film (those in the center portion) experience the bulk exchange field. In Fig.4 we therefore investigate more closely the size effect for thick films by comparing the average moment for a spin in the middle of the film with the net moment for the entire film.

Several features in Fig. 4 are worth mentioning. For the 21 layers film of FeF_2 the middle spin has a temperature dependence practically equal to the bulk except near the transition temperature. However it is evident that the net magnetization has a much more rapid decay than the the middle spin thermal average and is very small at around 0.7 of the transition temperature. The reason for this is that the net magnetization is dominated by what happens at the edges of the film. In contrast to a ferromagnet, the contribution of the spins in the middle part of the antiferromagnetic film to the net magnetization is relatively small because of the cancellation of moments due to spins on the two sublattices of the antiferromagnet. As a result when we make the film thicker, the middle part of the film (where the magnitude of the spins are independent of position) has the same contribution irrespective of thickness. Thus edge effects (where the magnitude of the spin depends on position) continue to play a significant role in the net magnetization even for thick films. Clearly this effect will also occur in superlattices with weak interface coupling.

In summary, we have examined the magnetization of FeF_2/CoF_2 superlattices for a variety of structures. In general structures with weak interface coupling show two phase transitions which can be related to the phase transitions of the two component materials. Structures with strong interface coupling and thin films display only the higher temperature phase transition. We also showed, in contrast to intuition, that edge effects can remain important in thick films and superlattices with thick films. A study of the effect of strong applied fields and the phase diagram of antiferromagnetic superlattices is now in course and will be reported soon.

ACKNOWLEDGEMENTS -- This work was supported by the U.S.Army Research Office under grant #DAAL03-91-G-0229. ASC also acknowledges financial support form the Brazilian Research Council (CNPq) and the Fulbright Commission.



4) Central spin and net spin thermal value (magnetization per unit cell) for a 21 layer FeF_2 film.

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