AIIM

Association for Information and Image Management
1100 Wayne Avenue, Suite 1100
Silver Spring, Maryland 20910
301/587-8202

Centimeter

1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 mm

1 2 3 4 5

Inches

MANUFACTURED TO AIIM STANDARDS
BY APPLIED IMAGE, INC.

NEUTRON AND X-RAY SCATTERING STUDIES OF $(FeF_2)_m(CoF_2)_n$ MULTILAYERS

D. LEDERMAN[a,b], D. P. BELANGER[c], J. WANG[c], S-J. HAN[c],
C. P. PADUANI[c,d], C. A. RAMOS[b,e], AND R. M. NICKLOW[f]
[a]Dept. of Physics, University of California, San Diego, La Jolla, CA 92093-0319
[b]Dept. of Physics, University of California, Santa Barbara, CA 93106
[c]Department of Physics, University of California, Santa Cruz, CA 95064
[d]Department of Physics, Univ. Fed. de Santa Catarina, Florianopolis, Brazil
[e]Centro Atómico Bariloche, 8400 Bariloche, Río Negro, Argentina
[f]Solid State Division, Oak Ridge National Laboratory, Oak Ridge TN 37831

ABSTRACT

We have performed elastic neutron scattering measurements of the staggered magnetization in $(FeF_2)_m(CoF_2)_n$ superlattices. Thermal expansion measurements, which are proportional to the magnetic contribution to the specific heat, were also carried out using high resolution x-ray diffraction. One of the two measured samples has thicknesses of $m = n = 4.5$ and the other $m = 26$ and $n = 28$ monolayers, as determined from high angle x-ray $\theta - 2\theta$ scans. In the $m = n = 4.5$ sample, only one transition is observed at $T_N = 62.9K$. Analysis of the neutron data, including the rounding effects, indicates an effective $\beta \approx 0.42$. This does not compare well with the 3D Ising exponent $\beta = 0.325$. The X-ray data also shows the existance of only one specific heat anomaly at $T = 62.8$ K. For the $m = 26$, $n = 28$ sample, dips in the staggered magnetization, and peaks in the thermal expansion were observed at $T \approx 40$ K and 74 K. The higher temperature anomaly, associated primarily with the FeF_2 layers, is sharper than the lower one, which is presumably rounded by the staggered ordering field imposed by the long range order in the FeF_2 regions on the CoF_2 regions.

INTRODUCTION

Multilayered epitaxial insulating antiferromagnetic films are promising systems for the study of a variety of physical models. Since they can be fabricated with overall thicknesses of a few microns, they can be used to obtain extinction-free Bragg scattering results. This has been demonstrated[1] using a single 0.8 μm thick layer of FeF_2 grown on a ZnF_2 substrate to obtain the critical behavior of the staggered magnetization for reduced temperatures of $0.002 < |t| < 0.025$. In the same manner, *multilayer* thin films can be used to obtain the staggered magnetization near phase transitions using neutron scattering techniques. Additional information may be obtained from high resolution x-ray techniques. For example, the thermal expansion coefficient of a film α, which is proportional to the magnetic contribution to the specific heat, may be measured with x-rays, thus providing the specific heat critical behavior.[2, 3, 4]

Bulk crystals of the isomorphic FeF_2 and CoF_2 antiferromagnets have been well characterized[7, 8]. Both antiferromagnets are Ising-like in their critical behavior because of the large anisotropies which force the spins to align along the c-axis. The effective exchange interaction strength in CoF_2 is much weaker that that of FeF_2. In both cases the dominant exchange is between the body-center and body-corner next-nearest-neighbor ions. The corresponding transition temperatures are $T = 78K$ and $T = 38K$ for FeF_2 and CoF_2, respectively. The FeF_2-CoF_2 *mixed* system can be well modeled by Ising spins for which the next-nearest-neighbor interactions are equal to the geometric mean of the interaction strengths of the two materials.[9]

Epitaxial *single* thin films made of these materials have been studied using a variety of techniques and their behaviors have been described in some detail.[10] An important result of these measurements is that the fundamental magnetic interactions in single thin films, ranging from

0.25 μm to 3 μm in thickness, do not measurably change from their bulk values.

Conversely, epitaxially grown *multilayers* in many instances have unique properties. For example, antiferromagnetic layers separated by nonmagnetic layers are useful for studying the crossover from three-dimensional (3D) to two-dimensional (2D) critical behavior[3, 4]. Alternatively, multilayers consisting of antiferromagnetic materials with differing nonzero exchange interactions may be fabricated to study the effects of a modulating exchange interaction along one direction.[2, 5, 6]

In the study described here, we investigated the dependence of cooperative Ising phase transitions in periodic multilayered structures, composed of two antiferromagnetic materials which have considerably different next-nearest-neighbor exchange interactions. This was done using x-ray diffraction and neutron Bragg scattering intensity measurements of antiferromagnetic $(FeF_2)_m(CoF_2)_n$ multilayered structures, whose superlattice period is composed of m monolayers of FeF_2 and n monolayers of CoF_2. The results obtained from the x-ray and neutron scattering techniques are consistent with each other. We find that for a $m = n = 4.5$ sample only one transition is observed, while for a $m = 26$, $n = 28$ sample two anomalies are detected. The anomaly at the higher temperature, which is associated with the long-range ordering of the FeF_2 regions, is sharper than the lower one, which is presumably rounded by the long range order imposed by the FeF_2 on the CoF_2 regions.

PREPARATION AND STRUCTURAL CHARACTERIZATION

The details of the multilayer growth technique has been described elsewhere.[10] Briefly, the samples were grown along the c-axis direction via MBE at a base pressure $< 5 \times 10^{-9}$, a substrate temperature of 300° C, and a rate of approximately 3 Å/sec. Polished ZnF_2 single crystal discs, 1 cm in diameter and oriented along the [001] direction, were used as substrates because ZnF_2 is both non-magnetic and has an excellent lattice match with both FeF_2 and CoF_2. Two samples were studied, with $m = n = 4.5$ and $m = 26$, $n = 28$, as determined from X-ray $\theta - 2\theta$ scans. The sample thicknesses are 0.29 μm and 0.91 μm, for the $m = n = 4.5$ and $m = 26$, $n = 28$ samples, respectively. A detailed x-ray analysis of these samples[3] indicates the presence of interf...

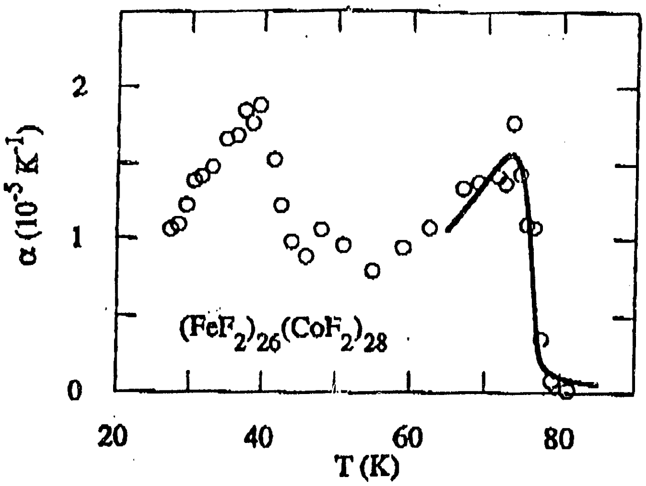

Figure 1: Thermal expansion coefficient α along the [001] direction of the $m = 26$, $n = 28$ sample, determined from x-ray scattering measurements. The solid curve represents a fit to a rounded transition. This rounding is presumed to be caused by the step disorder in the superlattice, resulting in a distribution of layer thicknesses.

EXPERIMENTAL RESULTS

Using the x-ray and neutron scattering techniques described above, we characterized the Ising-like magnetic ordering taking place in two regimes of layer thickness. For the $m = 26$, $n = 28$ sample, a phase transition takes place at $T \approx 74K$ which can be associated with the ordering in the FeF_2 layers. This can be seen in Fig. 1 for the x-ray scattering, which shows the thermal expansion coefficient versus T, and in Fig. 2 which shows the neutron scattering behavior of the staggered magnetization. The x-ray data shown in Fig. 1 clearly has two peaks, both of which are rounded. In order to obtain $T_N \approx 74K$, the higher temperature peak was fit to a rounded 3D-Ising specific heat function. This function is represented by the solid curve. The rounding was presumed to arise from the step disorder present in the samples, causing different macroscopic FeF_2 regions to have different transition temperatures due to finite-size scaling.[3, 5]. The basic form $\alpha(T) = A_{\pm}|t^{-a}|$, with $t = 1 - T/T_N$, $a = 0.11$, and the amplitude ratio $A_+/A_- = 0.54$, corresponding to the 3D Ising model, was utilized.

The neutron Bragg scattering intensity is well described by the power law

$$I \sim M_s{}^2 = M_o{}^2|t|^{2\beta} \quad (1)$$

where M_s is the staggered magnetization, $t = T/T_N - 1$, $T_N = 72.5$, and $\beta = 0.325$, as shown near the upper transition by the solid curve in Fig. 2. The slight discrepancy between the T_N values determined from x-rays and neutrons is probably due to the uncalibrated thermometer used in the neutron scattering experiment. We also show, with a dashed curve, what the second rise in intensity would look like if it were sharp and simply added to the intensity from the higher transition. The observed rise at lower temperatures, primarily from the ordering of the CoF_2 layers with the staggered field from the FeF_2 layers imposed at the layer interfaces, is rounded over a range of temperature of approximately 15 K.

The $m = n = 4.5$ sample shows only one clear anomaly, at an intermediate temperature $T = 69.2K$. The thermal expansion coefficient is shown in Fig. 3 and the neutron Bragg scattering intensity versus T is shown in Fig. 4. This sample is clearly near the limit in which the two kind of layers are strongly coupled. There is some rounding of the transition, presumably due to different macroscopic regions with slightly different values of m and n, each independently going through their phase transitions at slightly different temperatures. It is worth noting that

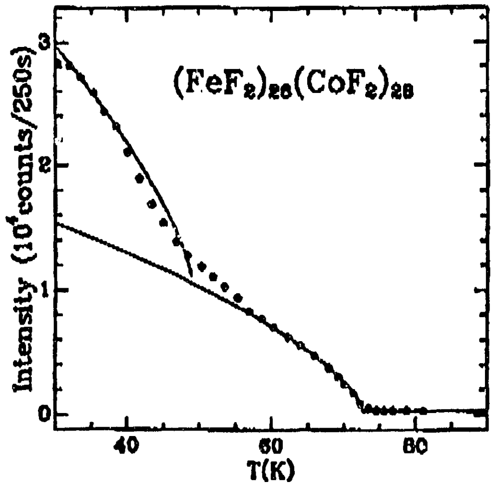

Figure 2: Neutron scattering intensity of the (100) magnetic peak for the $n = 26$, $m = 28$ sample. The solid curve represents a fit to $I \propto |t|^{2\beta}$ with $t = T/T_N - 1$ and $\beta = 0.325$. The dashed curve represents a second sharp transition added to the intensity of the solid curve.

the rounding of the neutron scattering data cannot be due to background fluctuations, since it has been experimentally shown that critical fluctuations are negligible[1]. For both the neutron and x-ray data, the solid curves are fits to appropriate power law behaviors with a Gaussian distribution in the transition temperature. From the thermal expansion data, the rounding was found to be $\delta T = 2.2$ K, while from the neutron scattering data $\delta T = 2.3$ K. Both fits yield a value of $T_N = 69.2$. However, the fitted value $\beta \sim 0.42$ is not in agreement with the well-known 3D Ising value $\beta = 0.325$. This disagreement could be explained if the order parameter in the FeF_2 and CoF_2 layers tended to grow at different rates below T_N. In this case, the intensity would not follow the expected power law behavior, except close enough to T_N so that the correlation length spans the entire superlattice period. This hypothesis is being tested via computer modeling of these multilayers, with the magnetic interactions at the interfaces equal to the geometric mean of the two intra-layer interactions.[11]

DISCUSSION

Clearly the FeF_2 layers attempt to order first as the temperature is lowered since bulk FeF_2 has a higher T_N than bulk CoF_2. If the FeF_2 layers are sufficiently thick, the CoF_2 regions will only weakly affect the critical behavior in FeF_2 layers. In this case, the value of T_N will be lowered only slightly by the presence of the more weakly interacting CoF_2 layers. On the other hand, the CoF_2 layers cannot order independently of the FeF_2 regions, since the already well ordered FeF_2 layers will act on the Co^{++} spins near the interfaces, thus producing an effective staggered ordering field on the CoF_2 layers. Just as in the case of a ferromagnet with an applied uniform field, the antiferromagnetic layers with the effective staggered field on the surface spins will *not* experience a sharp phase transition, since the ordering field induces spin order above T_N. Since the strength of the effect of one kind layer on the other will depend upon the number of interface spins relative to the spins within layers, sufficiently thick CoF_2 layers will be only slightly affected by the FeF_2. Hence, one would expect a slightly rounded transition only slightly elevated in temperature (with respect to T_N for bulk CoF_2) for thick CoF_2 layers.

As the layer thicknesses decrease, the behavior of the two kinds of layers will become more interdependent. In the extreme limit in which the the thicknesses become one atomic layer thick ($m = n = 1$), the system can be considered to be a new crystalline structure with a single magnetic exchange interaction. Thus, only a single phase transition occurs, with no remnant of

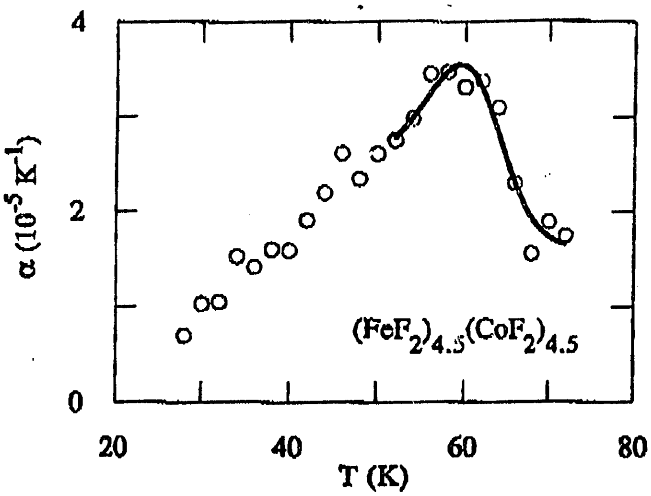

Figure 3: Thermal expansion coefficient α along the [001] direction of the $m = n = 4.5$ sample, determined from x-ray scattering measurements. The solid curve represents a fit to a rounded transition. This rounding is presumed to be caused by different macroscopic regions, with slightly different values of m and n, going through their transition at slightly different temperatures.

a second one, and the transition temperature should be intermediate between the bulk ones.

At intermediate layer thicknesses, one would expect the system to have two anomalies, as long as the strength of the FeF_2 layers' magnetic interactions is not strong enough to overcome the disorder in CoF_2 regions. If this is not the case, a single transition for the $m = n \neq 1$ system could be observed for sufficiently small values of m. The evolution of the behavior from large to small layer thickness has been described previously[3, 5, 12].

In the case of the present study, we find that for the $m = 26$, $n = 28$ sample, the neutron scattering data indicates that the higher temperature dip is much sharper than the lower temperature one. However, the position of the thermal expansion peak and the neutron scattering dip ($T \approx 74$ K), which is about 4 K lower than the transition temperature of bulk and single thin film FeF_2, indicates that the CoF_2 layers indeed affect the ordering of the FeF_2 regions. On the other hand, the $m = n = 4.5$ sample clearly shows a single rounded transition, indicating that at this value of $m = n$, the FeF_2 and CoF_2 regions order simultaneously. Hence, the $m = 26$, $n = 28$ sample belongs to the intermediate layer regime. Conversely, the $m = n = 4.5$ sample is clearly in the thin layer regime.

CONCLUSIONS

We have presented neutron and x-ray scattering data for multilayer thin films $(FeF_2)_m(CoF_2)_n$ for two layer thickness regimes. The $m = 26$, $n = 28$ sample shows a relatively sharp transition which can be associated with the ordering of the FeF_2 layers and, at lower T, a rounded transition from the CoF_2 layers. The rounding is from the staggered field imposed by the ordered FeF_2 layers. The sample with thin layers, $m = n = 4.5$, shows only one transition at an intermediate temperature, but does not yield the correct value for the exponent β, which may indicate that the order parameter grows at different rates in the two layers for $T \ll T_N$.

ACKNOWLEDGEMENTS

We would like to thank A. R. King and V. Jaccarino for discussions and for support of this work. This work was supported by the Department of Energy grant No. DE-FG03-87ER45324,

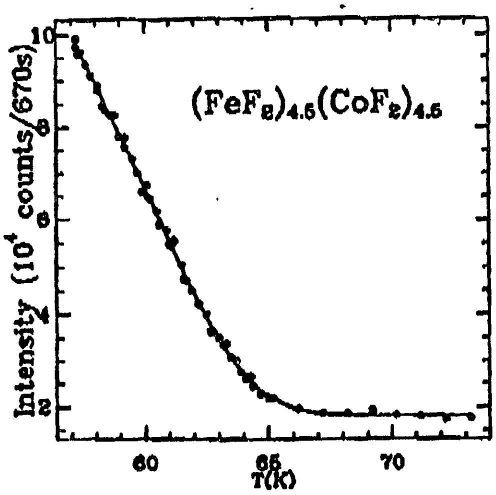

Figure 4: Neutron scattering intensity of the (100) magnetic peak for the $n = m = 4.5$ sample. The solid curve represents a fit to a rounded phase transition with $\beta = 0.42$.

DOE-Martin Marietta contract No. DE-AC05-840R21400 and NSF Grant No. DMR 88-15560.

REFERENCES

1. D. P. Belanger, M. Lui, and R. W. Erwin, these proceedings.
2. C. A. Ramos, D. Lederman, A. R. King, and V. Jaccarino, Phys. Rev. Lett. 65, 2913 (1990).
3. D. Lederman, Ph.D. Thesis, University of California, Santa Barbara, unpublished (1992).
4. D. Lederman, C. A. Ramos, V. Jaccarino, and J. L. Cardy, to be published.
5. D. Lederman, C. A. Ramos, and V. Jaccarino, presented at the Seventh Latin-American Symposium on Surface Physics, Bariloche, Argentina, 1992 (to be published).
6. For work done on antiferromagnetic oxides, see, for example, M. J. Carey, F. E. Spada, A. E. Berkowitz, W. Cao, and G. Thomas, J. Mater. Res. 6, 2680 (1991) or W. Cao, G. Thomas, M. J. Carey, and A. E. Berkowitz, Scripta Met. 25, 2633 (1991).
7. R. A. Cowley, W. J. L. Buyers, P. Martel, and R. W. H. Stevenson, J. Phys. C 6, 2997 (1973).
8. M. T. Hutchings, B. D. Rainford and H. J. Guggenheim, J. Phys. C 3, 307 (1970).
9. A. E. Nash, C. A. Ramos, and V. Jaccarino, Phys. Rev. B 47, 5805 (1993).
10. For a review, see V. Jaccarino, A. R. King, D. Lederman, M. Lui, and C. A. Ramos in Heteroepitaxy of Dissimilar Materials, edited by R. F. C. Farrow, J. P. Harbison, P. S. Peercy, and A. Zangwill (Mater. Res. Soc. Proc. 221, Pittsburgh, PA, 1991) pp. 3-14.
11. M. B. Viani, D. P. Belanger and D. Lederman, unpublished.
12. For theoretical calculations based on mean field models, see A. S. Carrico and R. E. Camley, Phys. Rev. B 45, 13117 (1992) and R. W. Wang and D. L. Mills, Phys. Rev. B 46, 11681 (1992).

DISCLAIMER

DATE FILMED

8/19/93

END