Surface Phase Transitions and Spin-Wave Modes in Semi-Infinite Magnetic Superlattices with Antiferromagnetic Interfacial Coupling

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We analyze the bulk and surface spin-wave modes of a semi-infinite Fe/Gd superlattice. If an externally applied magnetic field is increased from zero, one surface mode is driven soft, indicating a surface phase transition which occurs at a field value about 5 times lower than that necessary to cause a bulk phase transition. The phase transition is shown to nucleate at the surface and has a large penetration depth (on the order of a few hundred angstroms) even for fields just slightly above the value needed to induce the transition.

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In recent years, a great deal of attention has been given to the topic of surface reconstruction. In addition to crystallographic reconstruction where the physical structure on the surface may differ from that found in the interior, in magnetic structures the spin configuration at the surface may be different from that of the bulk.[1,2] Phase transitions in magnetic systems are particularly interesting in that one can drive the transition by application of an adjustable, external, magnetic field.

An interesting example of a magnetic surface phase transition, known now for many years, concerns the (100) surface of a simple uniaxial antiferromagnet such as MnF_2.[3,4] For that system a bulk phase transition to a spin-flop state can be induced by an external field. It turned out that a surface phase transition also occurred, but at a lower field value.

Most surface reconstructions involve only the first few outer layers. Recent examples include the surface-enhanced magnetic ordering of Gd (Ref. 5) and Tb.[6] The experimental study of such transitions can require sophisticated techniques such as spin-polarized electron spectroscopies.[7] A different way to study such phase transitions is to look for an excitation whose frequency is driven to zero. For a surface phase transition, it is appropriate to look at surface excitations. This was tried in the case of the antiferromagnet, but proved to be unsuccessful in part because the penetration length of the surface wave is also very small.

In contrast, the Fe/Gd superlattice[8,9] structure examined in this paper exhibits a long-range surface reconstruction. In fact, this region can extend several hundred angstroms into the material even for fields not much larger than that necessary to cause the phase transition. The details of the phase transition are particularly interesting. The choice of the outermost film radically alters the nature of the phase transition. If iron is chosen, the phase transition nucleates at the surface and the transition takes place at a field about 5 times lower than that required for the bulk transition. On the other hand, if gadolinium is chosen, the phase transition begins in the interior of the crystal and, in fact, can be slightly suppressed by the surface for thin enough films.

In order to study the surface phase transitions in the Fe/Ge superlattice, we first develop a method to determine the frequencies of the surface modes for the semi-infinite superlattice structure.[10] A surface phase transition occurs at a field value for which the frequency of the surface mode is driven to zero. We can study properties of the reconstructed state such as penetration length and configuration by finding the minimum-energy configuration at a given field.[11] Of course, we could also have found the phase-transition points using this method; in fact, this technique was used to corroborate the results of the spin-wave method. However, the energy-minimization algorithm suffers from two irritating defects. First, it is very slow near phase transitions. Second, it is possible that this technique will find a local minimum rather than the desired absolute minimum. Therefore, we have chosen to use the spin-wave method for the detection of the phase-transition points.

Consider a semi-infinite magnetic superlattice composed of alternating films of iron and gadolinium. While both materials are ferromagnetic, the coupling at the interface is antiferromagnetic. The z axis is in the direction of the external applied field and parallel to the interfaces while the y axis is perpendicular to the interfaces.

Depending on temperature and the magnitude of an externally applied magnetic field, such a system can exist in several distinct phases.[11,12] The first phase is the Gd-aligned phase where the Gd spins are parallel to the external field while the iron spins are antiparallel to this field. There is also an iron-aligned phase in which the iron spins are parallel to the field while the Gd spins are antiparallel. In the twisted phase, the spins are rotated away from the z axis in the x-z plane. Finally, there is a paramagnetic phase in which some interior spins in the gadolinium spins act essentially paramagnetically. The dispersion curves[13] for the bulk modes of a similar structure have been discussed earlier.

We briefly outline the method used for obtaining the surface-mode frequencies in the aligned state. Surface modes at finite temperatures and for the canted states may be calculated by a straightforward extension. The analysis begins by writing the equations of motion for spins in the outermost unit cell as well as for spins in a unit cell deep in the interior of the crystal. The equation of motion for a spin $\mathbf{S}_n$ is

$$\frac{d\mathbf{S}_n}{dt} = \gamma \mathbf{S}_n \times \mathbf{H}_{\text{eff},n}\,, \tag{1}$$

where $\mathbf{H}_{\text{eff},n}$ is the effective field that acts on this spin in layer n. For a bcc structure, the effective field is given by

$$\mathbf{H}_{\text{eff},n} = \mathbf{H}_0 + \sum_\delta J_{n,n+\delta} \mathbf{S}_{n+\delta}\,, \tag{2}$$

where δ is summed over the nearest neighbors and $\mathbf{H}_0$ is an external applied field. $J_{n,n+\delta}$ is the exchange constant that acts between spin $\mathbf{S}_n$ and one of its nearest neighbors. For a bcc structure, each (interior) spin has eight nearest neighbors.

We assume a time and spatial dependence of the form $\exp(-i\omega t + \mathbf{k}_\parallel \cdot \mathbf{x}_\parallel)$, where $\mathbf{k}_\parallel$ is the component of the wave vector parallel to the interfaces. The equations of motion are written in component form and linearized by assuming that the z components of the spins are constant in time. For the aligned phases, we can rewrite these equations in terms of S^+ and reduce the size of the matrix. Spins in adjoining unit cells are related via the Bloch theorem. If N is the number of (x-z plane) layers in a unit cell, then spins in adjoining unit cells are related according to

$$\mathbf{S}_{n+N} = \mathbf{S}_n e^{+\beta L}\,, \tag{3}$$

$$\mathbf{S}_{n-N} = \mathbf{S}_n e^{-\beta L}\,, \tag{4}$$

where L is the length of the superlattice unit cell in the y direction and β is, in general, complex. If β is imaginary, the usual oscillatory Bloch-wave solutions result. When β is real and negative, we have the exponential decrease associated with a surface wave. We thus arrive at the following system of simultaneous equations for the "bulk" system.

$$\begin{aligned}
(\omega/\gamma)S_1^+ &= -[\Delta(\mathbf{k}_\parallel)J_{0,1}S_1^z]S_N^+ e^{-\beta L} - [\Delta(\mathbf{k}_\parallel)J_{1,2}S_1^z]S_2^+ + (4J_{0,1}S_N^z + 4J_{1,2}S_2^z + H_0)S_1^+\,,\\
(\omega/\gamma)S_2^+ &= -[\Delta(\mathbf{k}_\parallel)J_{1,2}S_1^z]S_1^+ - [\Delta(\mathbf{k}_\parallel)J_{2,3}S_2^z]S_3^+ + (4J_{1,2}S_1^z + 4J_{2,3}S_3^z + H_0)S_2^+\,,\\
&\cdots\\
(\omega/\gamma)S_N^+ &= -[\Delta(\mathbf{k}_\parallel)J_{N-1,N}S_N^z]S_{N-1}^+ - [\Delta(\mathbf{k}_\parallel)J_{N,N+1}S_N^z]S_1^+ e^{+\beta L} + (4J_{N-1,N}S_{N-1}^z + 4J_{N,N+1}S_1^z + H_0)S_N^+\,,
\end{aligned} \tag{5}$$

where

$$\Delta(\mathbf{k}_\parallel) = \cos\left(\frac{k_x}{a/2}\right)\cos\left(\frac{k_z}{a/2}\right), \tag{6}$$

and a is the lattice constant in the x and z directions. For ease of notation, we use J in this paper to represent the true exchange constant divided by $g\mu_B$. Thus JS has units of magnetic field.

We obtain similar equations for the "surface" system. In fact, only the first equation is different:

$$(\omega/\gamma)S_1^+ = -[\Delta(\mathbf{k}_\parallel)J_{1,2}S_1^z]S_2^+ + (4J_{1,2}S_2^z + H_0)S_1^+\,. \tag{7}$$

The absence of a term containing $e^{-\beta L}$ in Eq. (7) reflects the fact that there are no spins above the surface layer.

We now form an eigenvalue equation $A_B\mathbf{S} = \omega\mathbf{S}$, where ω is an eigenvalue of the system, $\mathbf{S}$ is an eigenvector with components $(S_1^+, S_2^+, \ldots, S_N^+)$, and A_B is the (bulk) matrix formed from the right-hand elements of Eqs. (5). We can form similar eigenvalue equations for the surface unit cell: $A_S\mathbf{S} = \omega\mathbf{S}$. A solution for the semi-infinite structure is one which simultaneously satisfies the two eigenvalue equations above with the same eigenvector and eigenvalue. The solutions are found numerically by a procedure which will be discussed elsewhere.

In the numerical calculations we use $J_{\text{Gd}}/J_{\text{Fe}} = 0.03555$, $J_I/J_{\text{Fe}} = -0.706$, where J_I is the interface exchange constant.[14] The external field h is given in units of $J_{\text{Fe}}/g\mu_B$, where J_{Fe} is the true exchange constant and $h = 0.01$ corresponds to 7.3 kG. The temperature is given in terms of the reduced temperature $t = T_{\text{abs}}/T_{\text{Curie-Fe}}$

As an example, we consider a system with a unit cell which has thirteen Fe layers and five Gd layers. We explore the transition from the aligned state (stable at low external fields) to the canted state (stable at high fields) by presenting the bulk and surface spin-wave dispersion relations at $h = 0$ (Fig. 1) and $h = 0.01$ (Fig. 2). In each figure we see two bulk bands (shaded) and two surface modes. Which surface mode exists is dependent on which material, Fe or Gd, is on the outer surface of the superlattice. With Gd on the outside the surface mode is degenerate with the bottom of one of the bulk bands at $\mathbf{k}_\parallel = 0$. As the field is increased, both the bulk band and the Gd surface mode move upward in frequency and thus do not produce a phase transition. In contrast, with Fe outside there is a surface mode which is well separated from and *below* the bottom of the remaining bulk band. As the external field is increased (cf. Figs. 1 and 2) both the Fe surface mode and the bulk band move to lower frequency. As seen in Fig. 2, the Fe surface mode is

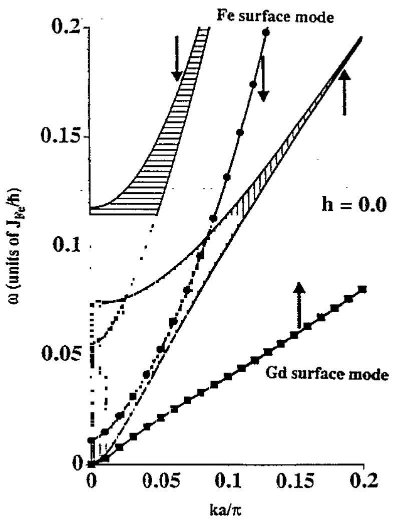

FIG. 1. Dispersion relation for bulk and surface spin waves in a semi-infinite superlattice with unit cell (13 Fe)/(5 Gd). The applied field is zero. The arrows indicate the motion of the spin-wave dispersion curves with increasing field. The Fe surface mode occurs only for the system with Fe on the outside, while the Gd surface mode occurs only for the system with Gd on the outside. The bulk modes shown are those for an infinite sample and are thus independent of the surface.

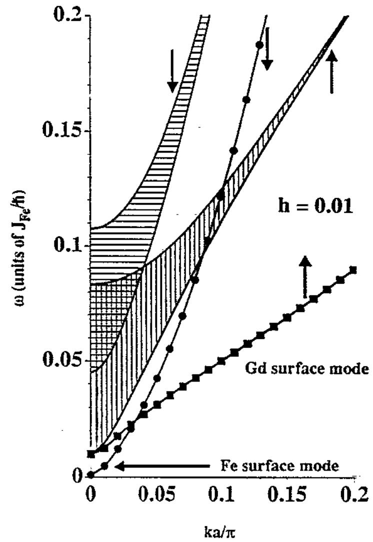

FIG. 2. Dispersion relation for bulk and surface spin waves in a semi-infinite superlattice with unit cell (13 Fe)/(5 Gd). The applied field is $h=0.01$, near the value needed to drive the lowest surface mode to zero frequency. The arrows indicate the motion of the spin-wave curves with increasing field. The Fe surface mode occurs only for the system with Fe on the outside, while the Gd surface mode occurs only for the system with Gd on the outside. The bulk modes shown are those for an infinite sample and are thus independent of the surface.

driven to zero frequency at a field given by $h=0.011$. The bulk band, in comparison, is still well above zero frequency and in fact requires a field of $h=0.055$ before it is driven to zero. We thus have a surface phase transition which takes place at a field about 5 times lower than that for the bulk phase transition.

It is easy to understand why the structure with the Fe on the outside should have a surface phase transition. In the Gd-aligned phase the Gd spins point along the external field and the Fe spins are antiparallel to the external field. Fe spins in the interior are strongly held antiparallel to the external field by the antiferromagnetic coupling to the Gd spins on both sides of the Fe film. In contrast, Fe spins at the surface are not as strongly fixed since there are Gd spins on only one side of the Fe film. As a result, as the external field increases from zero, those Fe spins in the outermost layer are the first to turn toward the direction of the applied field. Thus, the phase transition nucleates at the surface and occurs at fairly low values of field. On the other hand, if the outermost film is composed of gadolinium, the situation is very different. If gadolinium is on the outside, then (in the Gd-aligned phase) the spins in the outermost film are parallel with the external applied field. Any increase in the external field tends to stabilize these outer spins in the direction of the field. Therefore, the phase transition in this case is essentially a bulk phenomenon, and, in fact, for thin films the phase transition can actually be somewhat suppressed.

An analogous phenomenon occurs in the transition from the Fe-aligned phase to the canted phase. In the Fe-aligned phase, the gadolinium spins are antiparallel to the external field. As a result, if the outer layer is composed of gadolinium, then the phase transition nucleates at the surface. In contrast if iron is on the outside, then the phase transition nucleates in the interior of the crystal. Again, the phase transition is essentially a bulk phenomenon and so occurs near the bulk value.

It is worthwhile to examine the nature of the reconstructed state. We can obtain the static configuration of a finite system of spins by an iterative energy-minimization scheme.[11,12] The surface nature of the phase transition for the system with iron on the outside is illustrated by Fig. 3. This diagram shows the angle of deviation of a spin from the z axis at each spin site at various field values. The temperature is zero and we have a finite system with ten unit cells, each one with thirteen layers of Fe and five layers of Gd. (On this diagram, the Gd-aligned state is indicated by a value of 0° for all gadolinium sites and a value of −180° for all the iron spins.) As stated in the introduction, surface phase transitions typically do not extend beyond a few layers. However, the reconstructed region (canted region) in this system extends deeply into the material even at low field values. For instance, even at $h=0.012$, just above the phase transition, the canted region extends approximately four unit cells (on the order of 300 Å) into the interior. At the center of this (finite) crystal, the spins

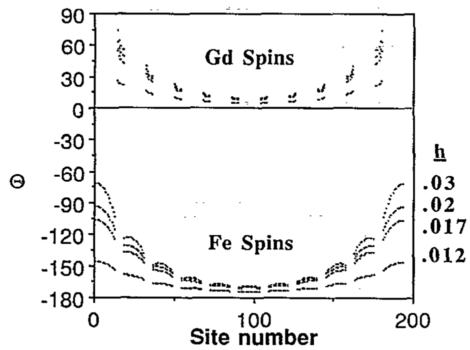

FIG. 3. The angle the spins make with respect to the applied field as a function of position for a finite (10 unit cell) superlattice. As the field is increased the reconstructed state deviates significantly from the aligned state.

are *almost* in the Gd-aligned state. This figure shows that the outer spins deviate strongly from the z axis, even at fairly low values of field. As the field increases, the deviation from the aligned state becomes more pronounced and the canted region penetrates deeper into the material. This diagram clearly shows that this phase transition nucleates at the surface.

In Fig. 4 we present a magnetic-field–temperature phase diagram for the (13 Fe)/(5 Gd) system. This is again calculated using the iterative method mentioned previously. We see that the surface phase transition (with Fe on the outside) remains well below the bulk phase transition for all temperatures where the Gd-aligned state exists. For temperatures for which the Fe-aligned state exists, the surface (Gd on outside) and bulk phase transitions are quite close to each other.

In conclusion the Fe/Gd superlattice system proves to be a fascinating system with a rich spectrum of surface phase transitions. Since the surface reconstruction region penetrates deeply into the material and occurs at external fields well below that necessary to cause a bulk phase transition, it should be possible to explore these transitions experimentally. Similarly, the fact that the choice of outer material results in a qualitative change in the spin-wave spectrum and phase-transition points also indicates this is a good choice for experimental study.

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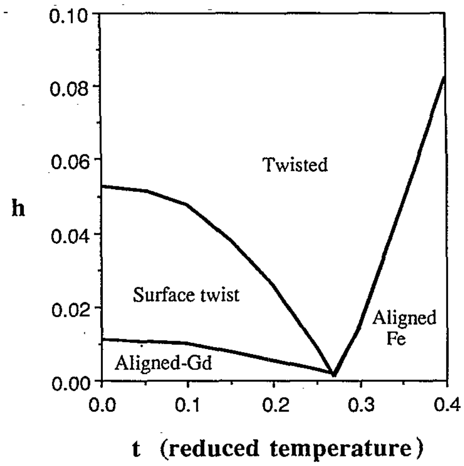

FIG. 4. Phase diagram for bulk and surface phase transitions in a (13 Fe)/(5 Gd) superlattice with the Fe film on the outside.

[1]C. L. Fu and A. J. Freeman, J. Phys. (Paris), Colloq. **49**, C8-1625 (1988).

[2]H. Hasegawa and F. Herman, J. Phys. (Paris), Colloq. **49**, C8-1677 (1988).

[3]D. L. Mills, Phys. Rev. Lett. **20**, 18 (1968).

[4]F. Keefer and H. Chow, Phys. Rev. Lett. **31**, 1061 (1973).

[5]D. Weller, S. F. Alvarado, W. Gudat, K. Schroder, and M. Campagna, Phys. Rev. Lett. **54**, 1555 (1985).

[6]C. Rau, C. Jin, and M. Robert, J. Appl. Phys. **63**, 3667 (1988).

[7]H. C. Siegman, D. Mauri, D. Scholl, and E. Kay, J. Phys. (Paris), Colloq. **49**, C8-9 (1988).

[8]T. Morishita, Y. Togami, and K. Tsushima, J. Phys. Soc. Jpn. **54**, 37 (1985).

[9]S. Tsunashima, T. Ichikawa, M. Nawate, and S. Uchiyama, J. Phys. (Paris), Colloq. **49**, C8-1803 (1988).

[10]See also, J. Barnas, J. Phys. C **21**, 4097 (1988). For bulk modes see E. L. Albuquerque, P. Fulco, E. Sarmento, and D. R. Tilley, Solid State Commun. **58**, 41 (1986). For semi-infinite Heisenberg ferromagnets, see R. F. Wallis *et al.*, Solid State Commun. **5**, 89 (1967). For semi-infinite Ising ferromagnets, see M. G. Cottam, Solid State Commun. **45**, 771 (1983).

[11]R. E. Camley and D. R. Tilley, Phys. Rev. B **37**, 3413 (1988).

[12]R. E. Camley, Phys. Rev. B **39**, 12316 (1989).

[13]J. G. LePage and R. E. Camley, Phys. Rev. B **40**, 9113 (1989).

[14]See K. Moorjani and J. M. D. Coey, *Magnetic Glasses* (Elsevier, New York, 1984), Chap. 6.