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Invited paper

Phase transitions in magnetic multilayers; statics and dynamics

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Abstract

Magnetic multilayers which incorporate ultrathin ferromagnetic films are physical realizations of classical, one dimensional spin systems, with spins coupled via exchange mediated by spacer layers or interactions at interfaces, and subject to anisotropy. Here by 'spin', we refer to the total spin angular momentum of an ultrathin film in the structure. Such systems can undergo a rich range of phase transitions, in reponse to an external magnetic field, or change in temperature. Since interfilm exchange is weak, modest magnetic fields can induce spin reorientation phase transitions. We thus have a new and diverse class of magnetic materials, with phase diagrams subject to design, since both thickness, composition, or growth conditions. The paper reviews selected examples, including recent studies of the dynamic response (AC susceptibility) of an antiferromagnetically coupled Fe/Cr system which undergoes the surface spin-flop transition. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Magnetic multilayers have attracted very considerable attention in recent years, as evidenced by the attendance at the present conference. It is fair to say that transport phenomena and the structural or compositional aspects which influence these properties have been the primary focus of both theorists and experimentalists, for good reason, thanks to the exciting applications of the giant magnetoresistance found in these materials.

In this paper, we place emphasis on other aspects we believe have yet to be studied or exploited sufficiently. These are phase transitions which involve reorientation of the magnetic moments in a magnetic superlattice structure, in response to either an applied magnetic field, or changes in temperature. Rich magnetic phase diagrams can be realized, and these are subject to design and manipulation, since the interfilm exchange couplings and intrafilm anisotropies which control them can be varied over a wide range, both in sign and magnitude by variations in film thickness, composition and growth conditions. When these materials are viewed from the macroscopic perspective, we have a new class of 'designer magnetic materials' remarkably diverse in nature. In contrast, in magnetic crystals, the interion exchange couplings are fixed in each case, as are the anisotropy energies. The opportunities for manipulating phase diagrams are thus limited. Finally, in the multilayers, interfilm exchange couplings are weak, so spin reorientation phase transitions occur at very modest externally applied magnetic fields. This will be useful for device applications since, as we shall see, the dynamic response of the structures can be tuned by driving the system through such a transition.

2. Examples

In this section, we discuss and review selected examples from the literature. We begin with the Fe/Gd superlattice structure [1]. Both Fe and Gd are ferromagnetic,

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and there is antiferromagnetic coupling between the two at the interfaces between films. Thus, in zero magnetic field, the ground state is 'ferrimagnetic', with Fe and Gd film moments antiparallel; the material has a net ferromagnetic moment, except for the special case where moments in the Fe and the Gd films exactly compensate. This is a superlattice with a magnetic structure that reminds one of YIG.

Suppose the Gd film moments are larger that those in the Fe films. Application of a magnetic field then aligns the Gd film moments along the field, with, in weak fields, the Fe moments antiparallel to the field. As the temperature is increased, since the Curie temperature of Gd is much lower than that of Fe, the Gd magnetization decreases, so one has a compensation point where the net magnetization vanishes. One may adjust the compensation temperature over a wide range, by simply varying the film thicknesses. Above the compensation point, the Fe magnetization is parallel to the external field. At higher applied fields, a new magnetic phase intervenes between the Gd-aligned, and the Fe-aligned phase. This is the 'spin twist' state, where the Fe moment, antiparallel to the applied field in low fields, cants away from the anti-parallel direction, and the interfilm exchange coupling leads to a canting of the Gd magnetization. One may think of this phase as similar to the spin flop-phase of an antiferromagnet, rendered asymmetric by the fact that the two sublattice magnetizations are unequal in magnitude. Thus, this rather simple structure displays a fascinating magnetic phase diagram. In Ref. [1], one finds a detailed account of the phase diagram is given by rather straightforward application of mean field theory. We know of no studies of the dynamical response of these structures in the various regions of the phase diagram. Ferromagnetic resonance or Brillouin light scattering data should prove most intersting.

Magnetic superlattices with films of the magnetically ordered rare earths exhibit exotic and intersting spin structures, and illustrated by studies of superlattices fabricated from the spiral spin material Dy, and ferromagnetic Gd [2]. At low temperatures, the structures have a ferromagnetic moments. With increasing temperature, the magnetization is a highly non-monotonic function of temperature, with one or sometimes a greater number of minima reminiscent of a compensation point. Theoretical modeling reproduces this behavior nicely, and shows that as temperature is increased, the system undergoes a sequence of transitions though rather exotic spin states. Changes in film thickness alter the phase diagram dramatically, including the nature of the states traversed as temperature increases. It is possible to synthesize magnetic superlattices from diverse combinations of rare earth metals. The Urbana group and their collaborators have carried out extensive studies of various combinations, to realize a remarkable range of intriguing states (for review see Ref. [3]).

In a number of much studied superlattices, the 'magnetically active' constituents are ultrathin Fe films. The Curie temperature of these films is typically far above room temperature. The individual magnetic moments within a given film are then tightly locked together via effective exchange couplings. In response to external magnetic fields, the total magnetic moment rotates as a rigid entity, with all atomic moments locked parallel by the very strong intra film exchange. Thus, each film may be viewed as a single spin, with a very large total spin equal to the sum of that associated with each ion in the film. Since the total spin S is very large, the total spin may be viewed as a fully classical object. An individual spin, as it rotates perhaps in response to an external magnetic field, is subject to intra film anisotropy. We may describe a multilayer or superlattice by an energy functional or Hamiltonian which consists of a line of such spins, each subject to single site anisotropy (the intra film anisotropy), and each coupled to nearest neighbors via nearest-neighbour exchange (the inter film exchange, possibly mediated by spacer layers, of bilinear or biquadratic character). The magnetic multilayer or superlattice is thus a physical manifestation of a purely classical, one-dimensional spin system. Viewed in the context of statistical mechanics, the spin configurations realized are those appropriate to the magnetic ground state, in this picture. Temperature enters indirectly through the temperature variation of the interfilm exchange, and the anisotropies.

An example of such a structure is the Fe/Cr(2 1 1) superlattices, synthesized at Argonne Laboratory by Fullerton and his colleagues [4]. The Fe magnetizations lie in plane, within which there is two-fold anisotropy. One may realize antiferromagnetic exchange coupling between adjacent Fe films, via the intervening Cr layers. The energy functional of this system is thus identical to that of the classical antiferromagnets MnF_2 and FeF_2. Thus, if a magnetic field is applied parallel to the easy axis, these 'artificial antiferromagnets' will undergo a spin-flop transition, very similar to that in the bulk crystals.

Within these superlattices, it has proved possible to confirm [5] a theoretical prediction made three decades ago [6,7]. This is that a magnetic field applied antiparallel to the surface moment of a terminated antiferromagnet will initiate a surface spin-flop transition, at a field lower than that of the bulk spin-flop field by a factor of $\sqrt{2}$, if the anisotropy is small in strength, compare to the exchange. In the next section, we discuss a theoretical approach we have developed recently [8], which allows us to examine both the static spin structures realized in this state, along its dynamic response.

3. The surface spin-flop state; statics and dynamics

One may elucidate the nature of the surface spin-flop state, by finding the configuration which minimizes the total energy of the spin array, as a function of external magnetic field. Such calculations [5] provide the following picture of the evolution from the low-field, antiferromagnetic ground state, to the high-field ferromagnetic spin arrangement.

When the external field exceeds the surface spin-flop field, the surface moment, initially antiparallel to the field, rotates nearly 180°, in efffect, a twist is applied to one end of the structure. A domain wall is then set up, in an off center position in the finite structure. With further increase in field, the wall undergoes a series of discontinuous jumps, as it migrates to the center of the structure. In this field regime, $(\mathrm{d}M/\mathrm{d}H)$ acquires a sharp spike, with each such jump. Interestingly, Trallori and co-workers [9] have shown that as the number of films $N \rightarrow \infty$, this field regime acquires a chaotic character. The domain wall becomes centered in the structure, and then with further increase in field broadens, to open up as a flower to evolve into a bulk spin-flop state. The angle between the spins and the external field is less at and near the surface than in the center of the structure. This is caused by the fact that the endmost moments are exchange coupled to only one neighbor. There is no longer a bulk spin-flop transition. As just described, the surface spin-flop state evolves continuously into the surface modified bulk spin-flop state. There is one intersting aspect of the symmetry of the spin states. The surface spin-flop transition occurs only in structures with an even number of ferromagnetic moments [5]. In zero field, the spin structure is odd under reflection through a point at the line of spins. At the point where the domain wall is just centered in the finite structure, one has a spin structure even under reflection through the midpoint. The migration of the domain wall provides a mechanism for evolution from an odd parity to an even parity spin arrangement (of course, the final ferromagnetic state is even under this reflection).

We find calculations based on minimization of the energy of the spin array are tedious, and must be carried out to high accuracy. Also, only a small number of spins may be probed (15–20), in circumstances where complex spin states are realized. We describe now a method we have used which can be applied to hundreds of spins, and which yields information on the static and dynamic response of the system, in one computation. We begin by writing down the Landau–Lifschitz equation of motion for the spin system, placed in a static magnetic field H_0 parallel to the easy axis in our case, and a time-dependent transverse field $h\cos(\Omega t)$:

$$\frac{\partial \boldsymbol{M}(i)}{\partial t} = \boldsymbol{M}(i) \times H_{\mathrm{eff}}(i) - g\boldsymbol{M}(i) \times \frac{\partial \boldsymbol{M}(i)}{\partial t}. \tag{1}$$

Here $\boldsymbol{H}_{\mathrm{eff}}(i)$ is the effective field which acts on the ith spin. This includes the external DC field, the single site (intrafilm) anisotropy, nearest-neighbor exchange, and the externally applied transverse AC field. The second term is a damping term which, it will be noted preserves the length of each spin.

We start with a small external DC field, where we are surely in the low-field antiferromagnetic state. We integrate the equations of motion of N spins forward in time. After an initial transient stage, a steady state is reached. Then we very slowly increase the DC field in time. When we reach the surface spin-flop field, by virtue of the dissipation term, the spin system seeks and spirals down into the lower energy surface spin flop-state. We keep increasing the field slowly, and we can follow the system's evolution to the final ferromagnetic state. The DC susceptibility can be obtained from the total longitudinal magnetization, which may be generated for each value of the DC field, as this is increased very slowly. We find the real and imaginary part of the AC susceptibility from a numerical Fourier transform of the transverse moment, which oscillates at the frequency Ω. The surface spin-flop transition is a first-order phase transition. We may generate hysteresis curves by sweeping the DC field first upwards, then downwards, through the critical field. We can also generates a snapshot of the configuration for any H_0. We have now applied this technique to the study of hundreds of spins, though in applications to the finite superlattices studied in the laboratory, only 22 are required [8].

Here we confine our attention to the dependence on H_0 of the low-frequency AC susceptibility. In Fig. 1, the upper panel shows data on the field dependence of X_2 taken by Fullerton, on the Fe/Cr (2 1 1) structure which displays the surface spin-flop transition. The lower panel is generated theoretically, as described above. We hasten to add that in the experiment, the AC field is applied parallel to H_0, while we have a transverse field in the theory. In the flopped states, where the 'active' spins make a large angle with the easy axis, the principal features are rather insensitive to the direction of the AC field. The agreement between theory and experiment is excellent, except the feature in the data near $H_0 = +0.5$ kG is, in the theory, the modest bump near -0.5 kG. This discrepancy has its origin in the large difference in hysteresis loop in the theoretical model, and in the actual sample, as discussed in Ref. [8]. Here we focus on the three other structures.

The sharp, dramatic peak $H_0 = -0.6$ kG is the signature of the surface spin-flop transition, while the two features at $H_0 = \pm 0.9$ kG occur at the bulk spin-flop field. While there is no true field-induced phase transition at the bulk spin-flop field in such a sample, as explained above, nonetheless there is a dramatic enhancement of X_2 here. The system remembers the bulk spin-flop transition, so to speak.

We see from the data and the theory, that near a spin reorientation phase transition, there is dramatic enhancement in the dynamic response of the superlattice

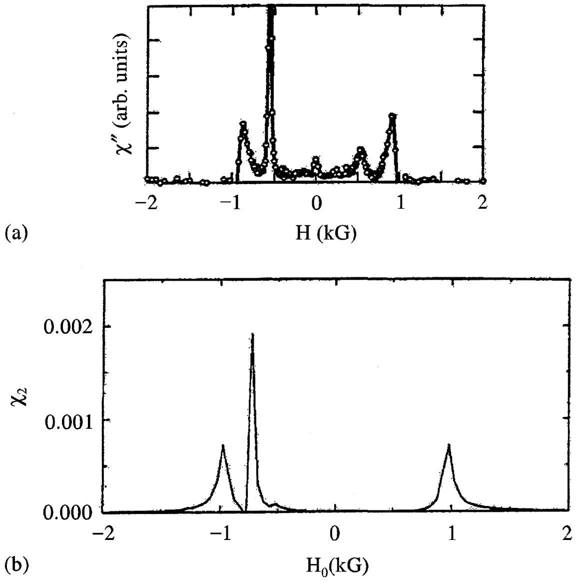

Fig. 1. We show (a) experimental data and (b) theoretical simulations of the field dependence of X_2 at low frequencies, for the Fe/Cr (2 1 1) superlattice.

structure. Since, as discussed above, the surface spin-flop field may be 'tuned' over a considerable range through variations in the microstructure of the superlattice, one can locate this feature where desired.

4. Concluding remarks

We have seen that diverse magnetic superlattices may be synthesized, with a remarkable variety of exotic spin structures. Phase transitions may be induced by changing the temperature, or through application of very modest external magnetic fields. Near spin reorientation transitions, as we see from the X_2 data on the Fe/Cr (2 1 1) structures, the dynamic response of the material is affected dramatically. It is the view of this author that in the future, far more experimental effort should be devoted to the study of the dynamics of these fascinating materials, through AC suscptiblity, ferromagnetic resonance studies, and Brillouin light scattering.

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