Surface Spin-Flop Transition in Fe/Cr(211) Superlattices: Experiment and Theory

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(Received 1 November 1993)

We report experimental and theoretical studies of the magnetization curves of Fe/Cr(211) superlattices with antiferromagnetic interlayer coupling and uniaxial in-plane anisotropy. There are substantial differences between structures with an even and an odd number of Fe layers, when the magnetic field is applied along the easy axis. For even layered superlattices, the inequivalence of the terminal Fe layers gives rise to a surface spin-flop transition that evolves into a bulk spin-flop arrangement with increasing magnetic field, as originally envisioned by Keffer and Chow [Phys. Rev. Lett. **31**, 1061 (1973)].

PACS numbers: 75.50.Rr, 75.30.Pd, 75.70.Fr

Magnetic superlattices exhibit rich and varied magnetic properties not found in bulk magnetic materials. An example is the giant magnetoresistance, observed first in antiferromagnetically coupled Fe/Cr superlattices [1]. Also, the exchange coupling between adjacent films is comparable to the Zeeman energies associated with modest magnetic fields, with the consequence that rich magnetic phase diagrams result [2]. In some systems, a 90° orientation of successive magnetic layers is observed resulting from biquadratic coupling [3]. Changes in temperature can induce dramatic spin reorientations as well [4]. In this Letter, we discuss experimental and theoretical studies of a novel magnetic superlattice structure with intriguing properties. Most particularly, we present the first experimental observation of a phenomenon discussed in the theoretical literature on antiferromagnets: the surface spin-flop transition [5,6]. We first summarize the early discussions.

In a classic two-sublattice antiferromagnet, such as MnF_2, a magnetic field parallel to the easy axis induces a first-order phase transition to the spin-flop phase [7]. Such materials consist of ferromagnetically aligned (100) planes of spins stacked to form an antiferromagnet. It was noted that if the surface layer of spins is antiparallel to an external magnetic field, the surface undergoes a spin-flop instability at a field well below the bulk spin flop [5]. A theory was presented by Keffer and Chow [6], who argued that the surface spin-flop state evolves continuously into the bulk spin-flop phase, as the experiments and new theory presented herein demonstrate. A precursor to the surface spin-flop transition is provided by a surface spin-wave mode discussed by Saslow and Mills [5]. This mode goes "soft" at a magnetic field just above that required to induce the (first-order) surface spin-flop transition. Experimental searches for this mode on bulk antiferromagnets were inconclusive [8]. Until now, the surface spin-flop transition has not been observed experimentally.

The new materials under study are Fe/Cr(211) superlattices grown on MgO(110) substrates. Structural, magnetic, and magnetotransport studies which characterize this system are discussed elsewhere [9]. Of present interest are Cr(100 Å)/[Fe(40 Å)/Cr(11 Å)$]_{22}$ and Cr(100 Å)/[Fe(20 Å)/Cr(11 Å)$]_N$ ($N = 20$ or 21) superlattices for which the interlayer coupling is antiferromagnetic. The epitaxial relationships to the substrate are Fe/Cr(211)∥MgO(110) and Fe/Cr[$0\bar{1}1$]∥MgO[001]. Each Fe layer in these structures is endowed with a uniaxial in-plane anisotropy which can be described by an in-plane surface anisotropy of $K_s = 0.06$ erg/cm^2 along the Fe[$0\bar{1}1$]. For Cr thicknesses of 11 Å, the interlayer coupling $J = -0.55$ erg/cm^2. Hence $|J| \gg K_s$. Therefore, these superlattices are isomorphic to the MnF_2 class antiferromagnets with a (100) surface.

Consider a sample which contains a large even number N of Fe layers. Let the easy axis (in-plane) be the $\hat{z}$ direction. If the Fe layer, say, on the top of the structure is parallel to $+\hat{z}$, then that on the bottom is necessarily parallel to $-\hat{z}$. If an external magnetic field is applied in the $\hat{z}$ direction, one of the two terminal Fe layers must be antiparallel to the applied field H. There should be a surface spin-flop transition at a field lower than that appropriate to the infinite structure by roughly a factor of $\sqrt{2}$ [5] in the limit in which the anisotropy is small compared to the exchange. (This limit is appropriate for the samples employed here.) According to the Keffer-Chow scenario, the surface spin-flop state should evolve into a bulk spin-flop arrangement as the magnetic field is increased. Our new calculations show in detail how this occurs.

On the other hand, if the sample has an odd number of layers, the magnetization of the two terminal layers will both be parallel to H, for small H. With increasing H, a bulk spin-flop transition only is realized; the spin configuration and transition field are affected by the presence of the two surfaces in the finite superlattice, similar to states discussed by Nörtemann *et al.* [10], in their studies which neglect anisotropy.

0031-9007/94/72(6)/920(4)$06.00

We illustrate these points with calculations carried out as follows. We consider a finite superlattice with N layers antiferromagnetically coupled. The lth layer has its magnetization canted with respect to H by an angle α_l. We then must minimize the energy functional

$$E = \frac{H_E}{2}\sum_{l=1}^{N-1}\cos(\alpha_l+\alpha_{l+1}) - \frac{H_A}{2}\sum_{l=1}^{N}\cos^2(\alpha_l) - H\sum_{l=1}^{N}\cos(\alpha_l)\,.$$

Here H_E and H_A are measures of the interlayer exchange coupling and intralayer uniaxial anisotropy, respectively.

In Fig. 1(a), we show calculations of the magnetization as a function of field, for a 15- and a 16-layer structure, with $H_A = 0.5$ kG and $H_E = 2.0$ kG. These parameters characterize the samples described below, within the framework of our model Hamiltonian.

The 15-layer sample displays a spin-flop transition near 1.4 kG, with saturation at 3.5 kG. This is a bulk spin flop at a field H_B; the canting extends throughout the structure. The layer magnetizations near the surfaces are aligned more closely to the field direction than the interior layers. This is a consequence of the fact that the surface layers are exchange coupled to only one neighbor, while the interior layers are coupled to two neighbors [10].

The 16-layer sample "flops" at the field $H_S = 0.93$ kG, lower than the H_B value of its 15-layer counterpart by a factor close to $\sqrt{2}$, as discussed in the original papers on the surface spin-flop transition [5]. The surface spin-flop state evolves into the bulk spin-flop state in a quasicontinuous manner, following a scenario close to that described by Keffer and Chow [6]. The manner in which this transition develops is intriguing. This is illustrated in Fig. 1(b). The field $H = 1.063$ kG is just above the surface spin-flop field; in the low-field state the right-hand layer is antiparallel to the applied field. At the surface spin flop, as argued by Keffer and Chow in their brief remarks on the antiferromagnet of finite thickness, the magnetization in the right layer rotates by an angle of close to 180°. A domain wall separates two nearly antiferromagnetic regions. With increasing field, this domain wall migrates towards the center of the structure, in a sequence of sudden jumps. The structure after the first jump is illustrated by the curve labeled $H = 1.094$ kG. Each jump produces a signature in the magnetization curve too small to perceive clearly in Fig. 1(a). As the field increases, the domain wall settles into the center of the structure, as illustrated in the curve labeled $H = 1.258$ kG. Note that the spin configuration is now symmetric. Its width then increases with increasing magnetic field. As a consequence, this state evolves continuously into a configuration one may identify with the bulk spin-flop state, modified by surface effects, as illustrated in the portion of the figure labeled $H = 1.492$ kG. While Keffer and Chow [6] argued that the surface spin-flop state evolves continuously into the bulk spin-flop configuration, and in very brief comments near the end of their paper envisioned an initial configuration for the finite structure similar to that illustrated in the curve labeled $H = 1.063$ kG, these authors did not present a detailed theory. We remark that the description of these features requires calculations of high numerical precision.

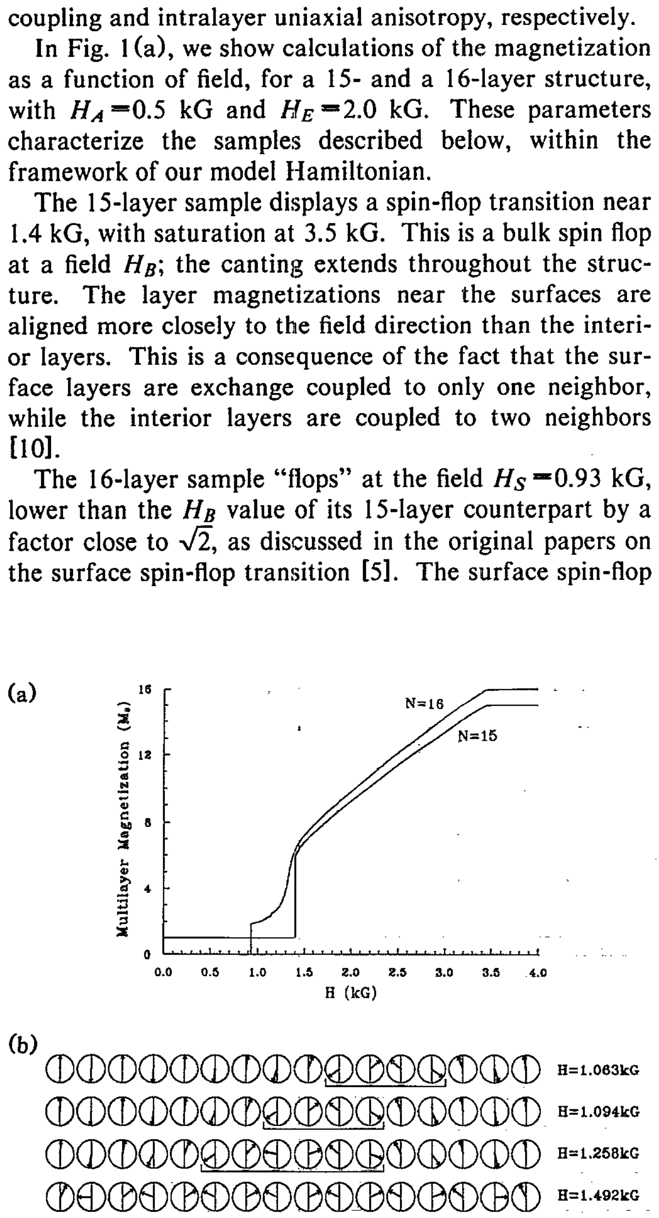

FIG. 1. (a) The field dependence of the magnetization for the models of 15- and 16-layer structures discussed in the text. (b) Spin configurations of the 16-layer structure explored in (a), for various applied fields. The underline regions are the domain walls.

The features in the scenario just outlined are illustrated nicely in calculations of dM/dH, with M the total magnetic moment of the structure. In Fig. 2(a), we give the result for the case $N = 15$; and in Fig. 2(b), that for $N = 16$. The spikes in Fig. 2(b) within the surface spin-flop regime are introduced by the domain wall jumps. Since the change in both magnetization and total energy associated with these jumps is very small, these features may be hard to detect experimentally. As the field is swept through the bulk spin-flop field, there is a sharp cusp in dM/dH, though M itself is continuous.

With these results in hand, we now turn to the experimental data. The magnetization curves were measured by means of SQUID magnetometry and the longitudinal magneto-optic Kerr effect using p-polarized, 633-nm light. Because the Kerr-effect measurements provide surface sensitivity on the scale of the optical penetration depth (of order 10^2 Å), these two techniques prove complementary in understanding the switching behavior of multilayer films. Shown in Fig. 3(a) are the room-temperature magnetization and Kerr intensity of the [Fe(40 Å)/Cr(11 Å)]$_{22}$ superlattice measured from $-M_s$ to M_s (saturation) with H directed along the easy axis. In the magnetization curve there is a plateau region

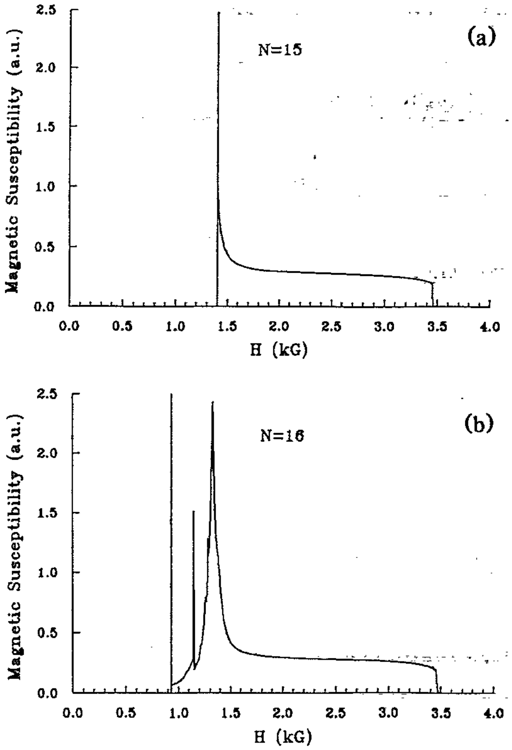

FIG. 2. Calculations of dM/dH for the (a) 15-layer and (b) 16-layer model structures described in Fig. 1.

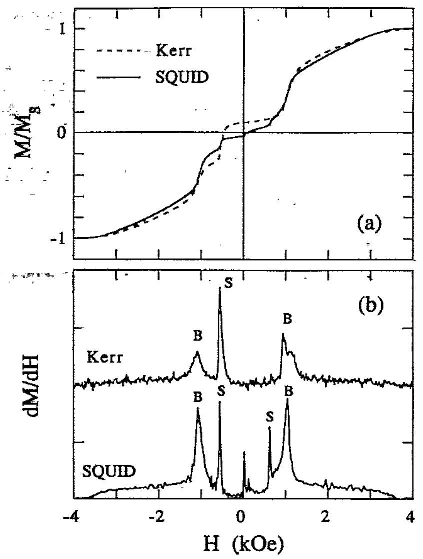

FIG. 3. (a) Magnetization curve of a [Fe(40 Å)/Cr(11 Å)]$_{22}$ superlattice from $-M_s$ to M_s with the applied field parallel to the magnetic easy axis. The solid line is measured by a SQUID magnetometer and the dashed line is measured by longitudinal magneto-optic Kerr effect. (b) The numerical derivative of the measured curves in (a). S and B refer to the surface and bulk spin-flop transitions, respectively.

near zero field in which the Fe layers are arranged in an antiferromagnetic configuration along the magnetic easy axis. At higher fields, the magnetization undergoes two transitions into canted configurations as was observed in the numerical calculations for an even number of layers and identified as surface and bulk spin flops. The two transitions also are identified in the derivative plots of Fig. 3(b). The peak at the lower field H_S corresponds to the surface spin-flop transition and the peak at higher field H_B results from the bulk spin-flop transition.

The Kerr-effect measurements confirm that the peak in the dM/dH curve at H_S is indeed a surface spin-flop transition. At the surface spin-flop transition, the right-hand layer undergoes an $\approx 180°$ change in direction. The switching of one layer should cause the magnetization to increase by $\approx 9\%$ of M_s for a 22-layer superlattice. This is consistent with the change in the measured magnetization curve at H_S observed in Fig. 3(a). In the Kerr intensity measurements which are heavily weighted towards the top few layers, for $H < 0$, the bulk spin-flop transition is reduced and the surface transition is significantly enhanced relative to the magnetization measurements, as expected. At H_S, the Kerr intensity in Fig. 3(a) switches from negative to positive which indicates that the magnetization of the top Fe layer is oriented in opposition to H for small negative fields. The expected change in the Kerr signal for top-layer switching can be calculated [11]. For our film the expected change should be 38% relative to M_s, which compares well to the measure change of 33% at H_S. As H crosses to the positive field side of the magnetization curve, the top Fe layer already is aligned with H; so the surface spin-flop transition is then initiated from the Fe layer closest to the substrate and is not observed in the Kerr measurements. Only the bulk spin-flop transition is observed in the Kerr curve for positive fields. An additional feature of the Kerr results is that in the canted region for $H > H_B$, the Kerr signal is higher than the SQUID magnetization measurement. This indicates that the surface layers are canted closer to the applied field than the bulk layers, in agreement with prediction.

The experimental results for samples with an odd number of layers also agree with theoretical expectations. Figure 4(a) compares the magnetization curves for [Fe(20 Å)/Cr(11 Å)]$_N$ superlattices with $N = 20$ and 21. As was observed in the previous example, a double transition is observed in the $N = 20$ sample. For the $N = 21$ sample, the remanent magnetization is higher (resulting from the uncompensated magnetic layer) and only a bulk spin-flop transition is observed with no evidence for a surface spin-flop transition in either SQUID or Kerr measurements.

We have carried out detailed studies of the collective spin-wave modes of the structure [12], and describe some highlights here. Interlayer dipolar interactions, of modest influence in the bulk antiferromagnets considered earlier

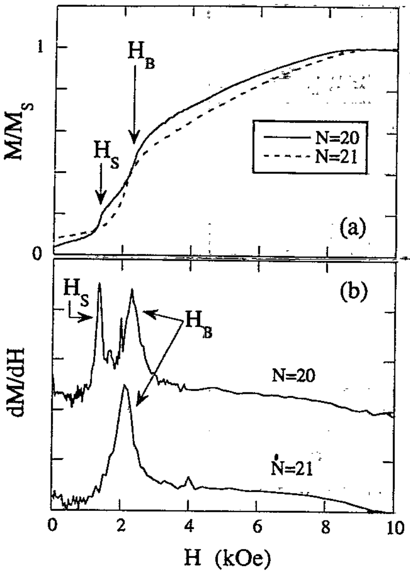

FIG. 4. (a) Magnetization curves of [Fe(20 Å)/Cr(11 Å)]$_N$ superlattices from 0 to M_s with the applied field parallel to the magnetic easy axis. (b) The numerical derivative of the measured curves in (a). H_S and H_B refer to the surface and bulk spin-flop transitions, respectively.

by Saslow and Mills [5], here modify the spin-wave spectrum importantly. For values of the wave vector $k_\parallel$ in the range probed by Brillouin scattering, in the low-field state the surface spin-wave mode lies above the standing wave modes of the structure, rather than below as found earlier [5]. In this wave vector regime, there is no "soft mode" associated with the surface spin flop. Brillouin scattering examines the region $k_\parallel D \gtrsim 1$ for these samples. For $k_\parallel D \ll 1$, with D the superlattice thickness, we do find a "soft mode," whose frequency vanishes for fields somewhat above H_S, as expected for a first-order transition. Microwave resonance studies which explore this regime would thus be of great interest.

In summary, the present study has provided the first experimental observation of the surface spin-flop transition, and of the subsequent evolution of this state with increasing magnetic field. Our experiments demonstrate that the combination of magnetization and Kerr-effect studies provide complementary information on spin-flop structures. As we have seen, the former probes the response of the entire structure, while the latter is sensitive to the vicinity of the illuminated surface, by virtue of the skin depth. Also, the measurement of dM/dH renders the spin-flop transitions visible in a vivid manner, while these are only more subtly displayed in the magnetization curves themselves. The presence of the twofold anisotropy introduced by growth of the Fe/Cr(211) structure on MgO(110) is the crucial ingredient for generating the rich magnetic phases presented herein.

Work at Irvine was supported by the Army Research Office, under Contract No. CS001028. Work at Argonne was supported by the U.S. Department of Energy, BES-Materials Sciences, under Contract No. W-31-109-ENG-38.

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