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Magnons in antiferromagnetically coupled superlattices

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We present an experimental Brillouin-scattering study of the collective spin-wave modes in antiferromagnetically coupled Fe/Cr(211) superlattices. We show spectra as a function of external magnetic field, where the field is swept from very small values up to those required to saturate the sample in the ferromagnetic state. We thus explore excitations in the low-field antiferromagnetic state, the surface spin-flop regime, the symmetric "bulk" spin-flop regime, and the high-field state. The data are compared with theoretical studies based on a description of the collective modes developed previously.

I. INTRODUCTION

Advances in deposition techniques have now made it possible to synthesize magnetic superlattices from diverse constituents, and thereby to realize new materials with macroscopic properties and magnetic response characteristics subject to design. Structures composed of thin films of a ferromagnetic metal, such as Fe, separated by nonmagnetic spacer layers have been found to possess many unexpected and intriguing properties. In such systems the spins within each ferromagnetic layer are coupled tightly by very strong effective exchange interactions, so for many purposes one can view the film as endowed with a fixed total magnetization M_s, whose direction may vary in response to an externally applied magnetic field.

Both the weaker interfilm exchange couplings, mediated through spacer layers, along with the intrafilm crystalline anisotropies can be controlled by the superlattice geometry, the symmetry of the substrate, and the growth conditions. This means one may produce materials with rich magnetic phase diagrams. It has been found that many of these materials exhibit interfilm exchange coupling whose origin is still somewhat controversial but is clearly mediated by the spacer layers. The strength and sign of the exchange coupling is affected by the superlattice geometry, substrate, spacer layer, growth conditions, etc., and the above factors also affect the crystalline anisotropy fields within the layers. This great variety and variability of magnetic parameters leads to a very rich and fascinating behavior of the magnetic properties, with only modest fields required to alter the nature of the magnetic ground state.

An interesting example studied in detail recently is Fe/Cr (211) superlattices.[1] Here the Fe magnetizations lie in plane, and have a dominant in-plane uniaxial anisotropy by virtue of the growth of these structures on MgO(110) surfaces. When the interfilm coupling is antiferromagnetic, the energy functional is very similar to that of the classical antiferromagnets MnF_2 and FeF_2, wherein one has ferromagnetically aligned (100) sheets of spins with antiferromagnetic coupling between such sheets, along with twofold in-plane anisotropy. Application of an external magnetic field parallel to the easy axis thus induces a spin-flop transition in the Fe/Cr (211) superlattices, very much as it does in $MnFe_2$ or FeF_2. Recent studies[2] of finite Fe/Cr (211) structures show that these materials display not only the spin-flop transition, but in addition to the surface spin-flop transition discussed many years ago in the literature on antiferromagnetism at surfaces.[3,4] Here we present an investigation of dynamic effects around both the bulk and surface spin-flop transitions.

Magnetic superlattices possess collective excitations, whose dispersion relation and frequency spectrum are controlled by the combination of interfilm exchange, intrafilm anisotropy, and Zeeman interaction with an external magnetic field. As noted many years ago,[5] interfilm coupling via macroscopic dipolar fields generated by spin motions within the film must also be included. It has been well known for decades that in ferromagnetic resonance, the frequency of the uniform mode of the film is affected by internal demagnetizing fields which also have their microscopic origin in dipolar interactions.[6] We will deal here with collective modes whose wave vector $\mathbf{k}_\parallel$ parallel to the film surface is nonzero. In this circumstance, the dipolar fields leak out of a film within which they are generated, to provide interfilm coupling.[5] For the particular case of Fe, for which $4\pi M_s \cong 21$ kG, the interfilm dipolar fields can be very substantial in strength. Recently, two of the present authors have developed the theory of the collective modes of the Fe/Cr (211) structures, with attention to the issues just mentioned.[7]

The purpose of this paper is to present an experimental Brillouin-scattering study of the collective spin-wave modes in an Fe/Cr (211) structure. In these experiments, the modes excited have $k_\parallel \cong 10^5$ cm^{-1}. In this regime, the modes are influenced very importantly by the interfilm dipolar coupling, in addition to interfilm anisotropy and exchange.[7] In order to interpret the experimental spectra we have generated, within an approximation scheme described below, theo-

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retical light-scattering spectra which compare very favorably with the data. Our investigation establishes that the rich spectrum of collective modes in these structures, which can be easily altered by application of a modest magnetic field, is well described by the theory developed in Ref. 7.

II. EXPERIMENTAL DETAILS

The sample employed in this study is an Fe/Cr (211) superlattice. The sample, prepared and characterized as described in Ref. 1, consists of 22 double layers of Fe(40 Å)/Cr(11 Å). The static magnetization properties of the sample used in this investigation are described in Ref. 2.

The Brillouin spectra were recorded with a 5+4 pass tandem interferometer,[8] using 300 mW of 515 nm radiation. The polarization of the scattered light was analyzed at 90° relative to that of the incident light. This isolates the purely magnetic scattering, and greatly reduces any scattering from surface phonons (Rayleigh waves), and bulk phonons with appreciable amplitude within the skin depth. The magnetic field was applied perpendicular to the scattering plane, (which contains both the incident and scattered photon wave vectors) and parallel to the easy axis of the sample: the spin waves observed in the Brillouin spectra therefore have wave vectors perpendicular to the easy axis. The scattering geometry is a backscattering configuration, where the incident and scattered wave vectors of the light make angles of 45° and 0° with respect to the surface normal, respectively. Typical data acquisition times were one-half to 2 h per spectrum.

All experimental spectra show a discontinuity at around $\pm 0.2\ \mathrm{cm}^{-1}$. This artifact is caused by filter which must be inserted into the beam as the spectrometer scans through the laser frequency. All spectral features which appear close to these frequencies must be treated with caution.

III. THEORETICAL DETAILS

If a sample with an even number of Fe layers is placed in a weak magnetic field H (i.e., H lies below the field required to initiate the surface spin-flop transition) parallel to the easy axis, then the structure resides in the simple antiferromagnetic ground state.[2] Necessarily the magnetization in one outermost Fe film will be parallel to the applied field, while that on the opposite side of the finite structure will be antiparallel. As H is increased the surface spin-flop transition is initiated through interaction of H with the outer film whose magnetization is antiparallel to the magnetic field.

The theoretical model we have employed is based on the theory of collective modes developed in Ref. 7, extended to make contact with the Brillouin data. Quite generally, when a spin wave is excited, the magnetization within a given film has the form

$$\mathbf{M}(x,y,z;t)=M_s\hat{n}_0+\delta\mathbf{m}(z)\exp[i\mathbf{k}_\parallel\cdot x_\parallel-i\Omega t], \tag{1}$$

where $\hat{n}_0$ describes the equilibrium orientation, and $\delta\mathbf{m}(z)$ (perpendicular to $\hat{n}_0$) is the fluctuation in magnetization associated with the spin wave. Here the z axis is normal to the film surface. The picture utilized here applies to the case where the ferromagnetic films have thickness d small compared to an effective exchange length $l_{\mathrm{ex}}=(D/\Omega)^{1/2}$, where D is the intrafilm exchange stiffness, and Ω a typical spin-wave frequency. For bulk Fe, $D\cong 2.5\times 10^{-9}$ Oe cm^2. If we assume this value applies to the Fe films in the superlattice, then $l_{\mathrm{ex}}\cong 100$ A. When $d\ll l_{\mathrm{ex}}$, we may ignore the variation of $\delta m(z)$. This remains true so long as $k_\parallel$ is sufficiently small, compared to $1/d$, a condition amply satisfied for modes excited in either Brillouin-scattering experiments such as those discussed here, or ferromagnetic resonance. Ignoring interfilm spin-wave dipolar coupling, the theory of the collective excitations of the superlattices is very simple: it is isomorphic to that of a suitable one-dimensional line of spins with exchange coupling and single site anisotropy. In Ref. 7, we introduced a simple procedure for appending the spin-wave generated interfilm dipolar coupling to this picture. The calculation of both the frequencies and eigenvectors of the superlattice collective excitations is calculated rather straightforwardly in this picture.

We wish to elaborate further on the remarks of the paragraph above. Quite a number of years ago, a continuum theory of spin waves in thin ferromagnetic films was developed, with focus on the regime where exchange, dipolar, and Zeeman energies are all comparable in magnitude.[9] The theory was applied to early light-scattering studies of thin films, and a remarkable quantitative account of the relative intensities observed for various modes emerged from the theory. Those experiments explored films with thickness $d\gg l_{\mathrm{ex}}$, so a full theory which accounts for the full spatial variation of $\delta m(z)$ was essential for a successful account of the data. The theory is rather complex to implement, since for each frequency, the response of the film is accounted for by superimposing six waves, each with complex wave vector in the direction normal to the surface. Boundary conditions which account for the possible presence (or absence) of spin pinning at each interface are required by the mathematical structure of the theory. Fortunately, the data that motivated the original work[10] was accounted for nicely without invoking spin pinning; only bulk parameters were required.

The theoretical structure described in the preceding paragraph, supplemented by anisotropy terms appropriate to ultrathin films, has been extended to superlattice structures by Stamps and Hillebrands.[11] To apply this theory, one requires the value of the intrafilm exchange stiffness D, along with information on the degree of spin pinning at each interface. The magnetic response of each film is described again by superimposing six waves. It would be extremely involved to use this full theory to account for the excitations of a superlattice with complex ground states such as those we encounter in the present analysis.

In the end, at least so long as $d\ll l_{\mathrm{ex}}$, on physical grounds one knows that $\delta m(z)$ must be quite uniform across a given film. Therefore, both the excitation spectrum and light-scattering spectrum will be insensitive to many details included in the full theory of Ref. 11 and we may use the simplified theory of Ref. 7 with quantitative accuracy. Once the orientation of the ground-state moments in the superlattice is determined (this is a challenging numerical task for the examples discussed here and in Ref. 2), the spin-wave eigenvectors and frequencies are generated very straightforwardly within the simple theory.

Once the spin-wave frequencies and eigenvectors are known, we have calculated the light-scattering spectra as follows. The superlattice is approximated as an optically homogeneous material, i.e., we did not consider its microstructure and its influence on both the incident and scattered radiation. This is, in effect, an effective-medium approach appropriate to the case where all films have a thickness small compared to the length scale of the optical field. The light scattering process is described by the formalism developed earlier,[5,9] with the incident and scattered phonon treated as plane waves.

There are two sources of coupling between the photon and fluctuations in the spin system. The first has origin in terms in the Hamiltonian quadratic in the electric field, and linear in the magnetization. These interactions also are responsible for the phenomenon of the Faraday rotation. There are, in addition, terms quadratic in both the electric-field and magnetic-field components. In magneto-optics, these lead to the Cotton-Mouton effect. Noting that earlier analyses based on only the linear terms provide a good account of the relative intensities of features in the Brillouin spectra of Fe-based ferromagnets,[9] we have retained only the Faraday terms. If we assume also that the Faraday tensor for the superlattice can be approximated by the form appropriate to bulk Fe, there is then only one nonzero coupling parameter. This parameter controls only the overall intensity of the spectra.

The above picture allows us to generate theoretical Brillouin spectra by introducing no parameters beyond those employed to describe the magnetic ground state of the superlattice, and its spectrum of collective excitations. In our view, there are two approximations invoked above which are potentially troublesome. The first is our use of only the Faraday term to couple light to the fluctuations in the spin system. If we were to extend the treatment to a more realistic picture which recognizes the influence of the film geometry on the Faraday tensor, and also which includes the Cotton-Mouton terms, we would encounter several additional parameters in the Brillouin matrix element whose values are unknown for these structures. We shall see however that the calculations with the simple model account nicely for most features in the measured spectra, and it is doubtful if we could obtain deeper insight into the data with a very complex multiparameter matrix element.

Modeling of the magnetic ground state and the spin-wave spectrum follows the approach employed in Refs. 2 and 7. Based on previous experiments on this sample[2] the principal parameters which describe its magnetic behavior are a uniaxial in-plane anisotropy and a bilinear interfilm exchange coupling. In calculating the spin-wave frequencies, we have, of course, included dipolar coupling whose strength depends on $4\pi M$. Some studies of Fe/Cr have indicated that biquadratic exchange and cubic anisotropy may be appreciable in some of these structures.[12] However, since for the sample investigated here the strength of the biquadric coupling and cubic anisotropy are not accurately known, their influence would have to be evaluated by introducing an additional parameter. We have not attempted this approach here.

Although it may be concluded from the arguments in the preceding paragraph that there are two adjustable parameters in the calculation of Brillouin spectra, the numerical value of the parameters is constrained by the need to reproduce the fields at which the surface and bulk spin flops occur. The parameters, in the notation of Ref. 2, used in the present calculations are $H_E = 1.9$ kG, and $H_A = 0.1$ kG, and $4\pi M = 21$ kG from bulk Fe. The calculated Brillouin spectra should therefore be considered to contain no adjustable parameter except for a phenomenological "width" of the spin-wave modes. This "width" is introduced because, since no lifetime effects are considered, the calculated spin-wave spectra are δ functions in frequency. In order to mimic the experimental spectra we have broadened all δ functions by an experimental instrumental function modeled as a Lorenzian.

As a final point we mention that the sample on which the measurements were made had 22 Fe layers. However because of the difficulty of calculating the spin arrangements in the finite superlattice reliably and accurately becomes increasingly difficult as the number of ferromagnetic layers is made larger, particularly in the asymmetric surface spin-flop state, we limit the total number of layers to 16.

IV. RESULTS AND DISCUSSION

We begin by reminding the reader of the principal features of the magnetic phase diagram of an Fe/Cr (211) superlattice with antiferromagnetic coupling between adjacent films. In these materials, the Fe magnetization lies in plane, with an easy uniaxial axis in plane. For very small applied fields H, the system remains in the antiferromagnetic state. If there are an even number of layers, upon increasing the external field, a field-induced phase transition into the surface spin-flop

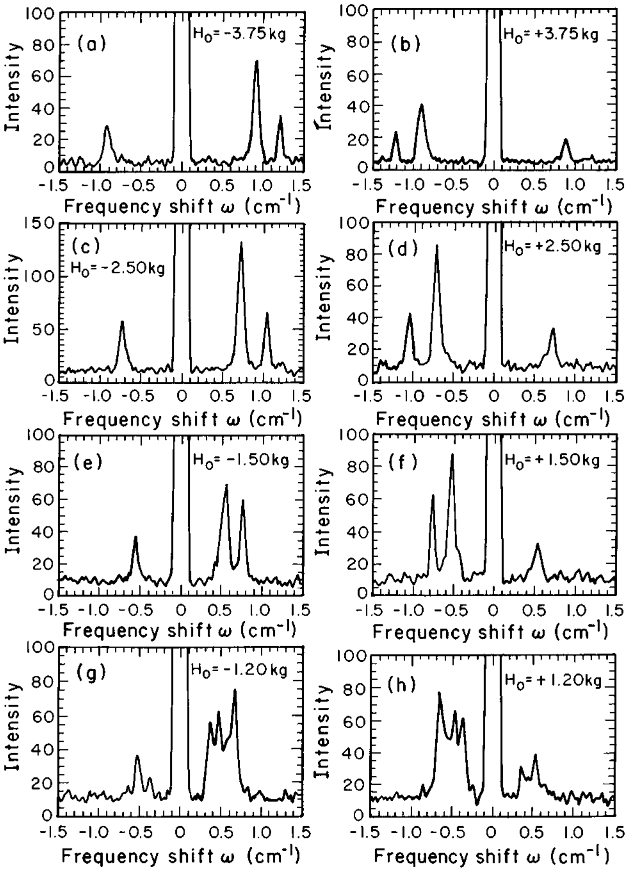

FIG. 1. Brillouin spectra for an Fe/Cr (211) superlattice with 22 Fe films for fields in the range 1.2 to 3.75 kG.

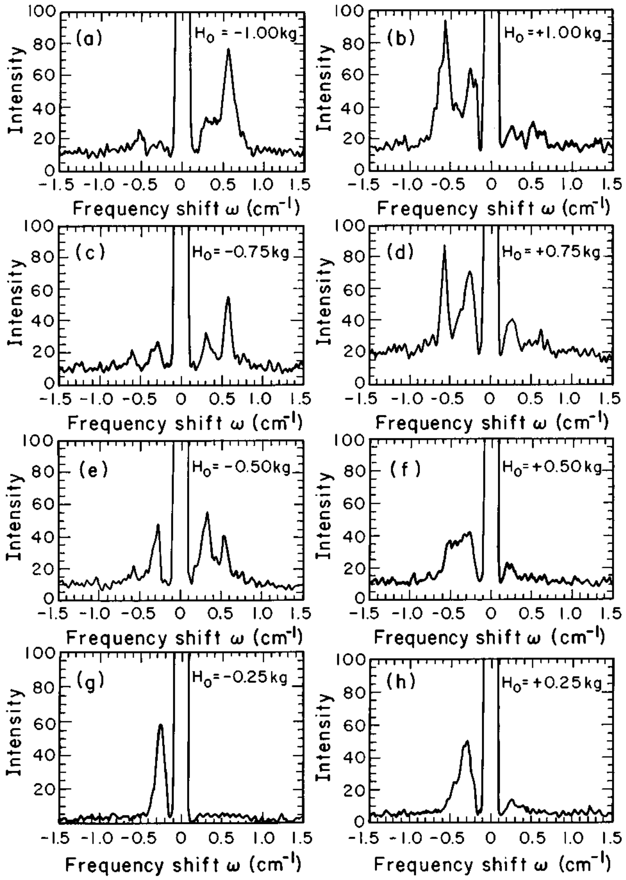

FIG. 2. The same as Fig. 1, for external magnetic fields in the range 0.25 to 1.0 kG.

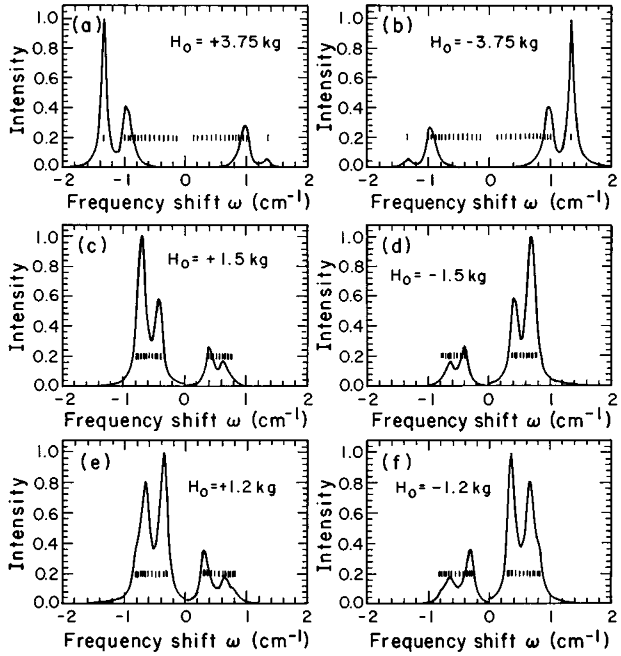

FIG. 3. Theoretical light-scattering spectra calculated as described in the text, for the six applied fields indicated. The small vertical lines are placed at the frequencies of the collective modes of our model 16-layer slab.

phase is initiated. By the mechanism described in Ref. 2, this state evolves into a symmetric configuration which may be identified with the bulk spin-flop phase, modified near the surface by virtue of the missing "exchange bonds." At high fields the symmetric spin-flop configuration reaches saturation, where all Fe film moments are aligned parallel to the external field. Experimentally the surface and bulk spin flops are found at 0.5 and 1.1 kG, the calculations yield 0.45 and 0.61 kG, respectively.

Because it is not possible from the light-scattering spectra alone to decide whether the magnetization at the illuminated end is parallel or antiparallel to the field, one must rely on the past history of the sample to do this. Based on the Kerr loops in Ref. 2 we do not know that the two surfaces switch at plus and minus fields, respectively. To guarantee that the regime around the surface spin flop would be explored in the experiments, we swept the field through zero, from a value well above the surface spin-flop field, to a value well below. Even then, it is not possible to tell from the light-scattering data alone if the transition at the illuminated surface will occur for positive or negative field.

Spectra were recorded as a function of monotonically changing field from $+3.75$ down to -3.75 kG. Figures 1 and 2 show spectra in the high- and low-field regions, respectively. At high fields the magnetization of all layers are roughly aligned with the field and hence it is not surprising that the observed spectra are very similar to those in conventional (uncoupled) superlattices.[5,13–15] The spectra are "typical" in two respects: there is a dramatic Stokes–anti-Stokes intensity asymmetry, and there are two salient peaks. The higher-frequency peak is expected to be due to a surface (Damon-Eshbach) type magnon and it only appears on one side of the spectrum. The lower frequency mode is due to a "bulk" or standing spin-wave resonance of the structure; it appears on both sides of the spectra but with unequal intensity. As expected, on reversal of the magnetic field the spectral features are found to change from the Stokes to anti-Stokes side of the spectrum. Experimentally the two-mode behavior persists down to fields close to 1.2 kG at which point additional features appear in the spectrum.

Figure 3 contains the high-field calculated spectra which are to be compared with the experimental spectra in Fig. 1. The small vertical lines in the figure indicate the position of the individual collective modes. In agreement with experiment, the calculations show spectra with two strong features, the frequencies agree to within around 10% (recall that there are no adjustable parameters to fit the frequencies), and the features change side on reversal of the field.

The split-off mode is the Damon-Eshbach surface spin-wave mode mentioned earlier, which for one sense of $k_{\parallel}$ is localized on the upper surface of the structure, and for the other is localized on the bottom surface. This mode is well known to provide a dramatic asymmetry in the Stokes–anti-Stokes ratio. If D is the thickness of the structure, and if also $k_{\parallel}D \gg 1$ then the origin of the Stokes–anti-Stokes asymmetry resides in the fact that for one sign of $k_{\parallel}$, the mode is localized on the surface exposed to the laser beam, and for the other, it is localized on the "dark side" (recall that in a Brillouin experiment, the Stokes and anti-Stokes sides of the spectrum sample opposite values of $k_{\parallel}$). In the present experiments, however, we are in the regime $k_{\parallel}D \sim 1$, and the surface mode is thus not completely localized at a single surface. In the system under study here, the large Stokes–anti-Stokes asymmetry thus has its origin in the mechanism discussed some years ago by Camely and Grimsditch.[16]

The second, lower frequency feature is not produced by a single mode, but rather by modes that cluster together near the maximum frequency of the standing spin-wave band. If Ω_M is this frequency, then for a semi-infinite ferromagnet, one may show that the density of modes diverges as $(\Omega_M-\Omega)^{-1/2}$ as Ω approaches Ω_M from below. In the finite structure, the modes cluster near the top of the band, and the matrix element which couples light to these modes is large. We remark that the spectra here are similar to earlier spectra taken on Fe/Pd superlattices, by Hillebrands *et al.*[17] Of course, in the present case, the frequencies and eigenvectors of the modes are influenced strongly by the antiferromagnetic interfilm coupling, ignored in earlier theories.[9]

The "two-mode" structure present at 3.75 kG extends down to lower fields, into the spin-flop regime. We see this from the data and theory at $H=\pm1.5$ kG, where the system is in the symmetric spin-flop configuration. We no longer have the Damon-Eshbach mode split off from the bulk continuum at this field, as one sees from the array of vertical dashes. There is a structure at both the maximum frequency Ω_M, and the minimum frequency Ω_m of the collective spin-wave band. Once again, in the semi-infinite limit, we have van Hove singularities at both Ω_m and Ω_M, and the two mode structure here is a reflection of these two singularities.

In the theory, this apparent "two-mode" structure continues down to lower fields, as we see from the calculations for $H=\pm1.2$ kG. However, by the time we reach 1.2 kG, structure not present in theory is evident in the data. We see no evidence, in the calculations, of the three mode spectrum displayed in 1(g) and 1(h).

Below 1.2 but above 0.5 kG (i.e., the region between the bulk and surface spin flops) the experimental spectra in Figs. 1 and 2 are less well defined and noisier. The reason for this can be found in Figs. 4(c) and 4(d) which show the calculated spectra at ±0.5 kG. For fields in the range between the surface and the bulk spin flop, the orientation of the magnetization of the films forms very complicated arrangements similar to the one labeled $H=1.492$ kG shown in Fig. 1(b) of Ref. 2. The resulting spin-wave spectrum is also quite complicated and similar to the one at $H=1.5$ kG in Fig. 6 of Ref. 7; where the surfacelike modes are localized in the gap between low-lying spin waves of acoustic character and higher frequency resonances of optical character.

Just below $+0.5$ kG we know (Kerr loops Ref. 2) that the sample undergoes a surface spin flop in which one of the surface layers aligns antiparallel to the field. However, as mentioned earlier, we do not know at which surface it occurs. Since the experimental spectra at $+0.5$ and $+0.25$ kG in Fig. 2 show no major changes and, what is more, the peaks do not change from the Stokes to the anti-Stokes side, it is a clear indication that it is the back layer which has switched. As the field is decreased, changed in sign, and brought to -0.25 kG we still observe no major change: this is consistent with the spins remaining in the antiferromagnetic alignment with the outermost layer being now antiparallel to the field as inferred from the Kerr data. A further decrease in field to -0.5 kG produces the surface spin flop on the outermost surface and leads to the clear changes in the spectrum most notably the Stokes–anti-Stokes switch in the high-frequency peak.

In Figs. 4(a) and 4(b), we show the theoretical spectra for the two cases $H=-0.25$ kG, and $H=+0.25$ kG, respectively. While the theoretical spectra contain more detailed structure than found in the experimental data, the theory is in rather good accord with experiment. Note that the dominant contributions to the Brillouin spectrum are on the anti-Stokes side of the laser line, for both field directions. It is also the case in both theory and experiment that the spectrum is broader at $H=+0.25$ kG, than for $H=-0.25$ kG. In the theoretical plots, the small vertical marks indicate the frequency of a collective spin-wave mode of the 16-layer superlattice. We see that the feature in the experimental spectrum comes not from one particular mode, but is a structure formed from contributions of a spectrum of modes near the bottom of the spin-wave band. These have the character of standing spin-wave resonances of the whole structure. We see a high-frequency mode, split off from the band of standing spin-wave resonances, near 1.5 cm^{-1}. This is a surface mode, which contributes only a weak line to the theoretical spectra. There is no evidence of this mode in the experimental data.

We show theoretical spectra for $H=\pm0.5$ kG in Figs. 4(c) and 4(d). In contrast to the theory and experiments at ±0.25 kG, we see that now reversal of the direction of the magnetic field shifts the dominant part of the scattering from the Stokes to the anti-Stokes side. The prominent asymmetric peak with maximum near 0.2 cm^{-1} is produced by scattering from low-lying acoustic spin-wave modes, and three rather weak higher frequency peaks have their origin in the optical modes. The sharp peak near 0.6 cm^{-1} is a surface mode, bound relatively weakly to the surface. Here again there is more structure in the theory than found in the data, but the principal features displayed by theory are evident in the experimental spectra. For example, in the experimental spectrum labeled $H=-0.50$ kG, we see the surface mode peak near 0.6 cm^{-1} on the high-frequency side, and the lower-frequency feature from the acoustic spin-wave band on both sides. We do not perceive the structures from the optical mode region in the data; the intensities of these are also

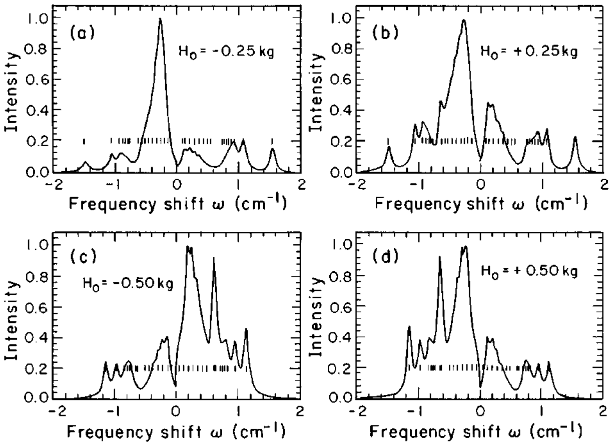

FIG. 4. Theoretical light-scattering spectra calculated as described in the text, for the four applied fields indicated. The small vertical lines are placed at the frequencies of the collective modes of our model 16-layer slab.

rather weak in the theory. One sees a peak near 0.6 cm^{-1} in the spectrum for $H=+0.50$ kG in the data, but the peak is not fully resolved in this case. A more complete and quantitative description of the matrix element which couples the evanescent light field with the collective spin wave may improve the correspondence between theory and experiment.

Except for the difficulty mentioned in the last paragraph, in our view the overall accord between theory and experiment is quite satisfactory. The principal trends are reproduced nicely by theory, and in fact we have quantitative agreement for many features of the data.

V. CONCLUDING REMARKS

This paper compares experimental and theoretical Brillouin spectra for Fe/Cr (211) superlattices and shows that the principal features in the data agree with theory. A few minor differences as discussed above can be understood on the basis of the simplifications in the theoretical model we have used. Improvements in the model would require introduction of new fitting parameters (higher-order anisotropies, biquadratic interlayer coupling, quadratic magneto-optic coupling) whose values may not be determined uniquely by the data: for this reason, we have limited the complexity of the model.

We are intrigued by this fascinating class of materials. Previous studies have shown that the Fe/Cr (211) structures have a rich magnetic phase diagram which includes a surface spin-flop phase, which with increasing external field evolves into a symmetric ‘‘bulk’’ spin-flop phase. There is a dramatic even-odd effect, in that the surface spin-flop phase is absent from samples with an odd number of Fe layers. The present paper shows that there is a rich range of dynamical responses displayed by these structures, as one traverses the various field regimes.

Both the phase diagram, and the dynamic response characteristics can be controlled and varied by the superlattice geometry. We thus have a new class of magnetic material, whose rich phase diagram can be scanned by very modest excursions in the magnetic field.

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