Influence of exchange-coupled anisotropies on spin-wave frequencies in magnetic layered systems: Application to Co/CoO

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Recent experiments in Co/CoO layered structures have shown a huge effect where the frequency of the lowest spin wave in the ferromagnetic Co has been doubled, apparently due to the interaction with antiferromagnetic CoO. We explore a microscopic model for spin waves in such coupled structures which can explain such an effect. In this model a magnetic material with a significant anisotropy is exchange coupled to a different magnetic material with minimal anisotropy. The shift in the spin-wave frequency increases as either the anisotropy or interface exchange is increased. [S0163-1829(96)08529-3]

I. INTRODUCTION

Layered film structures such as Co/CoO are currently of great interest because of strong coupling between the ferromagnet and antiferromagnet that allows one to introduce effective anisotropies in the ferromagnet.[1–4] This has been studied almost exclusively through measurements of hysteresis. Magnetization studies of the Co/CoO structure have demonstrated enhanced effective in-plane anisotropy with a significant temperature dependence. This is generally attributed to changes of the magnetic properties of the antiferromagnet with temperature.

A recent experimental study[5] suggests an alternate way of investigating effects related to the coupling across the Co and CoO interface by measuring frequencies of long-wavelength spin waves associated with the Co film. This study showed a huge increase in frequency of the low-frequency spin wave in Co as the temperature was reduced below room temperature. Such an effect suggests that it is due to the coupling to the antiferromagnet CoO which has a Néel temperature of 293 K.

This paper provides a theoretical basis for understanding the origin of these large frequency shifts. We use a microscopic spin-wave description[6] of long-wavelength spin waves for exchange coupled Co and CoO films. In this theory the Co film is assumed to have minimal anisotropy, but there are in-plane and out-of-plane anisotropies in the antiferromagnetic film. Because of the exchange coupling at the interface, a spin wave in the Co must drive the spins in the antiferromagnetic CoO also. As a result, the larger anisotropies in the CoO shift the frequency of the low-frequency spin wave significantly. As one approaches the Néel temperature of the antiferromagnet, the effect disappears since effective coupling and the anisotropy in the antiferromagnet are both zero at that point.

The paper is organized as follows. In Sec. II we outline the main theoretical ideas needed to calculate the frequency of the spin-wave modes in the layered structure. In addition we develop an analytic formula for the special case of two ferromagnets coupled together by interfacial exchange. The results of the theoretical calculations are presented in Sec. III. Two cases are treated in detail, two coupled ferromagnetic films and a ferromagnetic film coupled to an antiferromagnetic film. In Sec. IV we summarize our results.

II. THEORY

We consider a layered structure as illustrated schematically in Fig. 1. There are N_a layers of spins in material A and N_b layers for material B. The applied field H_0 is in the z direction. The z axis is also the uniaxis for the in-plane anisotropy and, for simplicity, we consider structures where the equilibrium directions of the magnetic moments are only along the $\pm z$ directions.

The spins in layer i, $\mathbf{S}_i$, see an effective field $\mathbf{H_i}$ which is given by the sum of the the exchange fields, the anisotropy fields, the dipolar field, and the external field. Thus, $\mathbf{H_i}$ can be written as

$$\mathbf{H_i} = \frac{1}{g\mu_B}[J_{i,i-1}\mathbf{S}_{i-1} + J_{i,i+1}\mathbf{S}_{i+1}] + H_i^{\mathrm{in}}(S_i^z/S)\mathbf{z} + (H_i^{\mathrm{out}} + 4\pi M)(S_i^y/S)\mathbf{y} + H_0\mathbf{z}. \tag{1}$$

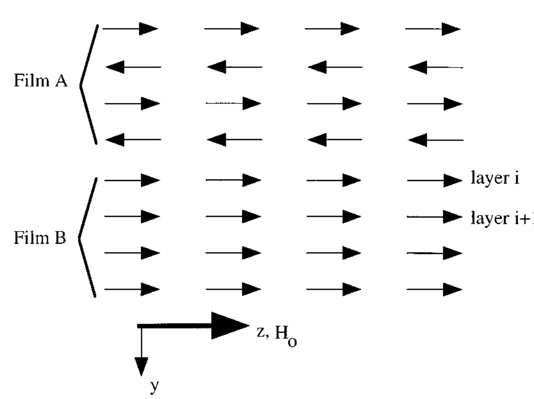

FIG. 1. Geometry used in this paper. The applied field, H_0, is parallel to the surface. There is anisotropy in film A and none in film B. The effective interface exchange coupling between A and B is H_I.

Here $J_{i,i-1}$ is the exchange coupling constant between planes i and $i-1$, $H_i^{\rm in}$ is the in-plane anisotropy field and $H_i^{\rm out}$ is the out of plane anisotropy field. In the limit that the wavelength of the spin wave is long compared to the thickness of the film, the dipolar fields acting on layer i are essentially just the demagnetizing fields ($4\pi M_y$) of a layer, which arise from out-of-plane fluctuations of the magnetization. In this case the dipolar terms may be combined with the out-of-plane anisotropy acting on an individual layer.

The equations of motion for the spins in layer i can then be written

$$\frac{d\mathbf{S}_i}{dt}=\gamma\mathbf{S}_i\times\mathbf{H}_i, \tag{2}$$

where γ is the gyromagnetic ratio. Because of the exchange terms, the spins in layer i are coupled to the spins in the layers above and below. One generally further assumes that the excitation amplitudes (s_x and s_y) are small and the equations are linearized by neglecting terms quadratic in these variables.

When the number of layers is not too large (less than 200) is is a straightforward numerical problem to solve the coupled equations of motion. One assumes a time dependence of the form $e^{-i\omega t}$ and obtains an eigenvalue problem for the allowed frequencies. Larger systems can be treated by using special routines for band diagonal matrices. We will explore the results obtained in this way in the next section.

The theoretical approach outlined above is an extension of earlier work exploring spin-wave modes in ferromagnetic bilayer[7,8] and multilayer[6,9] systems. A key feature of much of this earlier work is the examination of how the spin-wave frequencies change when the ferromagnetic films have magnetization which are canted with respect to each other. In contrast, we concentrate on situations where canting is not expected to occur.

Analytical results may be obtained in some simple cases. We consider the case of two ferromagnets with the exchange coupling at the interface weak compared to the intrafilm exchange. In that case we may assume that the spins within a given material are rigidly coupled together. In effect, this reduces the problem to solving the coupled equations of motion for only two spins, **A** and **B**, where each spin represents an entire film. The first step in finding the equations of motion for the two spins is to write the energy E for the entire system, assuming the spins in each film are coupled rigidly so that $\mathbf{S}_1=\mathbf{S}_2=\ldots\mathbf{S}_{\rm Na}=\mathbf{A}$. A similar assumption is made for spins in film B. An effective field acting on spin **A** is then found by

$$\mathbf{H}_A=-\frac{1}{g\mu_B}\frac{\partial E}{\partial\mathbf{A}} \tag{3}$$

and this field is used in the equations of motion [Eq. (2)]. One finds a similar set of equations for spins **B**, and we obtain a set of four homogenous equations with four unknowns, A_x, A_y, B_x, and B_y. The condition that the determinant of the matrix of coefficients for the four unknowns is zero provides the unknown frequencies. We obtain the following:

$$\Omega^2=\tfrac{1}{2}\,(H_bH_b'+H_aH_a')+e_ae_b\pm\tfrac{1}{2}\,\{(H_bH_b'-H_aH_a')^2 +4e_ae_b[H_a'(H_a+H_b')+H_b(H_b'+H_a)]\}^{1/2}, \tag{4}$$

where

$$H_a=H_0+\frac{1}{g\mu_B}\,J_aB_z+H_a^{\rm in}A_z/A,$$

$$H_b=H_0+\frac{1}{g\mu_B}\,J_bA_z+H_b^{\rm in}B_z/B,$$

$$H_a'=H_a-(H_a^{\rm out}-4\pi M_A)A_z/A,$$

$$H_b'=H_b-(H_b^{\rm out}-4\pi M_B)B_z/B,$$

$$e_a=J_aA_z,$$

$$e_b=J_bB_z.$$

The exchange terms above are the layer number normalized J-interface values written in terms of effective magnetic fields, i.e., $J_a=J_I/(g\mu_BN_a)$ and $J_b=J_I/(g\mu_BN_b)$.

III. RESULTS

In this section we will explore how the spin-wave frequencies depend on the important parameters of the system, anisotropy, interface exchange coupling, and the thicknesses of the different films. Our model parameters are therefore not intended to represent a particular system, but are close to those reported in recent experiments for Co/CoO thin films. Unless otherwise indicated we use the following parameters, all written in terms of effective fields for convenience ($H_{\rm eff}=J/g\mu_B$): For the ferromagnet, the exchange field is $H_{\text{ex-f}}=1000$ kG and $M_s=1.4$ kG. For the antiferromagnet $H_{\text{ex-af}}=500$ kG and $M_s=0.6$ kG. We take $S_a=S_b=1$ and the gyromagnetic ratio $\gamma=2.9$ GHz/kG. The applied field H_0 is 3 kG.

A. Two coupled ferromagnetic films

We consider two ferromagnetic films with film A having a significant anisotropy and film B having no anisotropy. Let film A have two magnetic layers and film B have six magnetic layers. In Fig. 2 we show how the frequency of the allowed spin waves depends on the exchange coupling at the interface. This interfacial coupling is parameterized by H_I an effective exchange field. Both ferromagnetic (positive H_I) and antiferromagnetic (negative H_I) coupling are considered and results are presented for different values of in-plane anisotropy. For the calculations we can use either the full set of coupled equations, or the approximate solution given by Eqs. (4). The results are equivalent for the lowest two modes shown.

For ferromagnetic interface coupling one sees small shifts in frequency which are nearly independent of exchange. In contrast, for antiferromagnetic coupling the shifts are large and depend strongly on the value of the interfacial exchange. In part this is because the assumed ground state for the antiferromagnetic coupling has the spins in film A pointing opposite to the applied field. Such a configuration is unstable

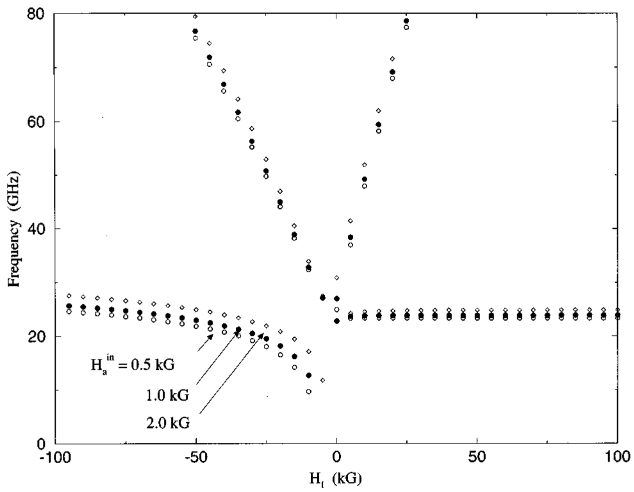

FIG. 2. Frequency as a function of interface exchange for the low-frequency spin waves. We have two ferromagnets, with $N_a=2$, $N_b=6$. There is no anisotropy in B and $H_a^{\text{out}}=0$. Both modes are sensitive to the in-plane anisotropy in A, but the high-frequency mode (optic mode) is also strongly dependent on interface exchange.

at low interfacial exchange or for low values of in-plane anisotropy and the frequency of the mode is driven to zero at this point.

We now explore how the frequency shift introduced by the anisotropy and the exchange coupling depends on the thickness of the film with anisotropy. In Fig. 3, we set $H_a^{\text{in}}=1$ kG in film A and plot frequency as a function of the number of layers in film A,N_a. The number of layers in film B is held constant at $N_b=6$. The results depend dramatically on the sign of the interfacial exchange coupling for the low-frequency mode. For ferromagnetic coupling the frequency of the lowest spin-wave mode increases slightly as the thickness is increased. For antiferromagnetic coupling the frequency decreases significantly. Again this decrease is due to the fact that there is eventually a phase transition because the A spins point opposite to the applied field and as the number of A spins is increased the system must eventually go into a structure where the A spins are parallel to the applied field so that the Zeeman energy is minimized.

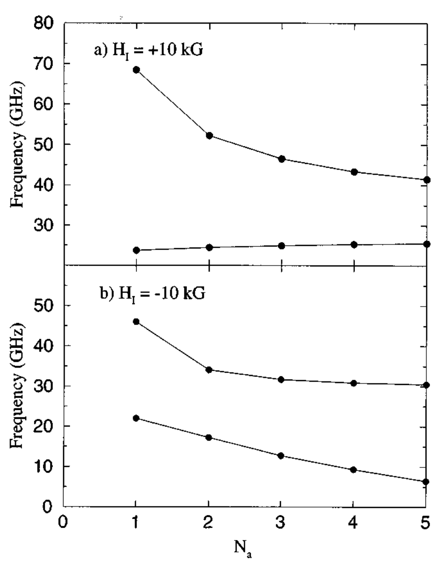

FIG. 3. Frequency as a function of number of layers of material A,N_a. $N_b=6$. A and B are both ferromagnets. All anisotropies are zero except for $H_a^{\text{in}}=2$ kG.

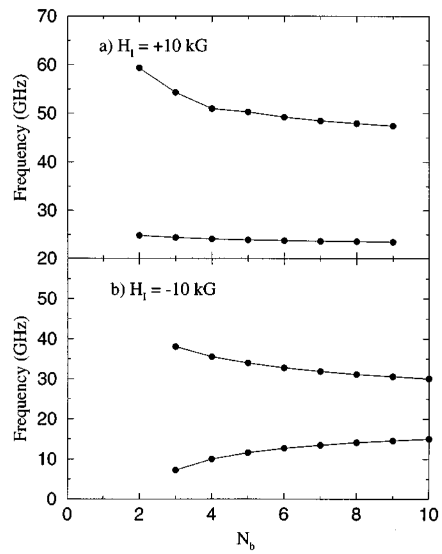

FIG. 4. Frequency as a function of number of layers of material B. A and B are both ferromagnets and $N_a=2$. All anisotropies are zero except for $H_a^{\text{in}}=1$ kG. In (a) the interface exchange is ferromagnetic, in (b) it is antiferromagnetic. Note that in (b) the structure is not stable for $N_b<3$.

In Fig. 4 we study the dependence of the spin-wave frequency of the number of layers of the film with no anisotropy N_b. For ferromagnetic interface exchange the low-frequency mode shows only a slight decrease as N_b is increased. Of course for these parameters the shift in frequency from that for film B alone is small and so one does not expect large changes as the number of layers of film B increases. In Fig. 4(b) we see that there is an increase in frequency for the low-frequency mode as N_b is increased. This arises because as N_b is increased, the equilibrium configuration is farther away from the phase transition.

B. A ferromagnetic film coupled to an antiferromagnetic film

For these calculations we use the microscopic model described earlier. Analytic forms which include the antiferromagnet are difficult to obtain and are likely to be extremely cumbersome. Furthermore it is not clear that the rigid coupling approximation, which was used in obtaining the analytic expressions for the ferromagnetic systems, is appropriate for antiferromagnets.

We now consider a six layer ferromagnetic film coupled to a four layer antiferromagnetic film. In Fig. 5 we plot frequency as a function of interface exchange for different values of the uniaxial in plane anisotropy. We note that anisotropy values in antiferromagnets can be quite large compared to those for typical ferromagnetic metals. For example, the uniaxial in-plane anisotropy in FeF_2 is on the order of 200 kG. As before, the frequency increases with an increasing in-plane anisotropy and with increased interfacial coupling.

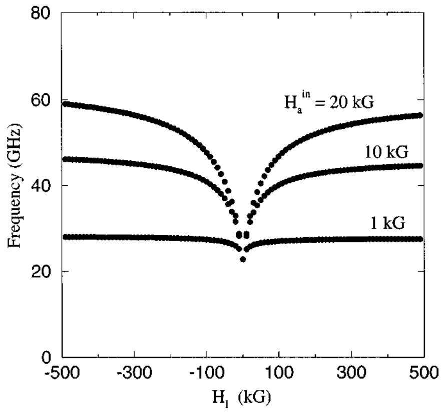

FIG. 5. Frequency as a function of interface exchange for different values of in-plane anisotropy in the antiferromagnet. $N_a=4$ and $N_b=6$. Material A is an antiferromagnet and material B is a ferromagnet.

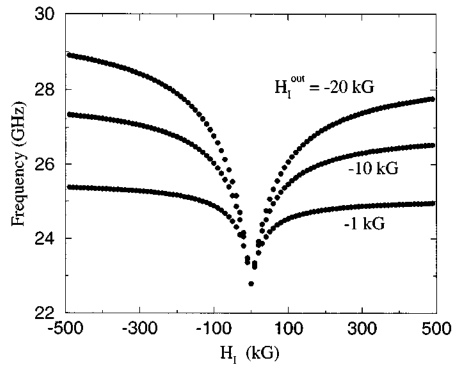

FIG. 7. Frequency as a function of interface exchange for different values of out-of-plane anisotropy in the antiferromagnet. All other anisotropies are zero. $N_a=4$ and $N_b=6$. Material A is an antiferromagnet and material B is a ferromagnet. Note that the out-of-plane anisotropy causes a much smaller shift than the in-plane anisotropy of Fig. 5.

In contrast to the ferromagnet/ferromagnet case, the sign of the interface exchange does not make a significant difference in the frequency. This is because changing the thickness does not significantly change the net magnetization except for the special case of extremely thin antiferromagnetic films.

In the results for the two coupled ferromagnetic films, we pointed out that the higher frequency modes can be very sensitive to interface exchange. This is true for an antiferromagnetic film coupled to a ferromagnetic films as well and is demonstrated in Fig. 6. As can be seen in this figure, the high-frequency modes change much more rapidly as a function of interfacial exchange. These modes might be measured by a number of different techniques. For example Raman scattering has been used to measure spin-wave modes in thin Fe films in the 500 GHz region.[10] Similarly far-infrared spectroscopy has been used to study antiferromagnets in the same frequency range.[11]

In Fig. 7 we examine the influence of out-of-plane anisotropy on the frequency of the lowest mode. We use the convention that H_a^{out} is positive when the normal to the surface is an easy direction, and negative when the normal is a hard direction. This means that a negative H_a^{out} is indicative of an easy-plane system. The result is similar to that found for in-plane anisotropy—the frequency increases with interface coupling or with anisotropy, but the magnitude is significantly smaller. Since out-of-plane anisotropy should not change the hysteresis curve, but does change the frequency, a comparison of spin-wave frequencies and magnetization data might be able to provide a value for the out-of-plane anisotropy.

As with ferromagnetic films, it is of interest to see how the frequency shift caused by the combination of interface coupling and anisotropy can be changed by controllable experimental parameters. In Fig. 8 we plot the frequency of the lowest spin-wave mode as a function of the number of layers of the antiferromagnetic film, N_{AF}. As the number of layers is increased, the frequency also increases. For the parameters used in this example, the increase is substantial, about 50%. There are small oscillations in the frequency depending on whether N_{AF} is odd or even. This is associated with the fact that an odd number of layers has a net magnetic moment and thus has an associated Zeeman energy which changes the frequency slightly. When the interface exchange is ferromagnetic, the curve looks nearly the same. Even the small oscillations due to the odd and even number of layers remain. The

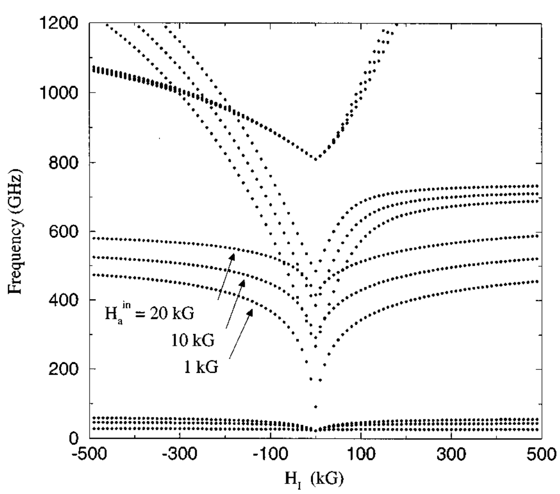

FIG. 6. High-frequency modes for the antiferromagnet/ferromagnet shown in Fig. 5.

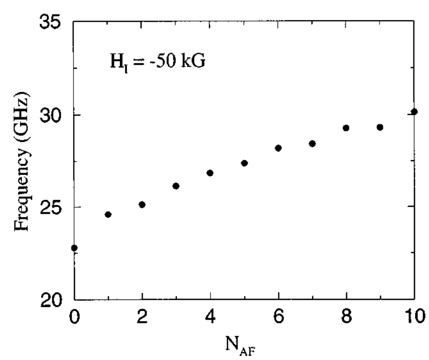

FIG. 8. Frequency as a function of the number of magnetic layers in the antiferromagnet, N_{AF}. The number of layers in the ferromagnet $N_{\mathrm{F}}=6$. $H_a^{\mathrm{in}}=1$ kG and all other anisotropies are zero.

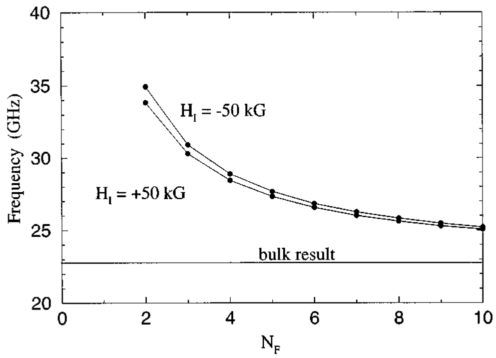

FIG. 9. Frequency as a function of the number of magnetic layers in the ferromagnet, N_F $N_{AF}=4$. $H_a^{in}=1$ kG and all other anisotropies are zero. The bulk result is the frequency of the system in the absence of interface coupling.

higher frequency spin-wave modes (not shown here) are also very sensitive to changes in N_{AF} and show substantial changes in frequency depending on whether N_{AF} is odd or even. As mentioned previously, these high-frequency modes may be most useful for obtaining values of interface exchange.

In Fig. 9 we plot the frequency as a function of the number of layers of the ferromagnet when the number of layers in the antiferromagnet is held fixed at $N_a=4$. The results for positive and negative interfacial exchange are nearly identical. Furthermore the frequency shift from the bulk value can be fit very nicely by a shift which is proportional to $1/N_F$, similarly to what is found in calculations which include surface anisotropies.[12]

All the previous calculations have been at low temperatures where the average spin moments in the antiferromagnet have their full magnitude. As the temperature is increased we may modify our calculations by replacing the spin magnitudes by their thermal averaged values. In fact, it is expected that these values will generally vary from layer to layer in very thin structures as discussed previously. Using a self-consistent local mean-field method, we can calculate the average spin moment in each layer of the antiferromagnet for a given temperature. The results for such a calculation are presented in Fig. 10 for a structure with four layers of antiferromagnet coupled to a six layer ferromagnet. The parameters for this calculation are $H_{ex\text{-}af}=505$ kG and $S_{af}=2$. We have

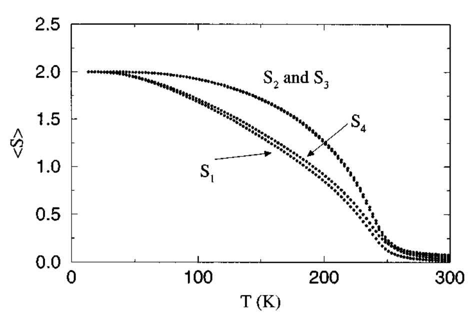

FIG. 10. Thermal averaged magnitudes for the spins in the different layers of CoO in a six layer Co/four layer CoO structure. S_1 is the layer of spins in CoO which is farthest away from the interface.

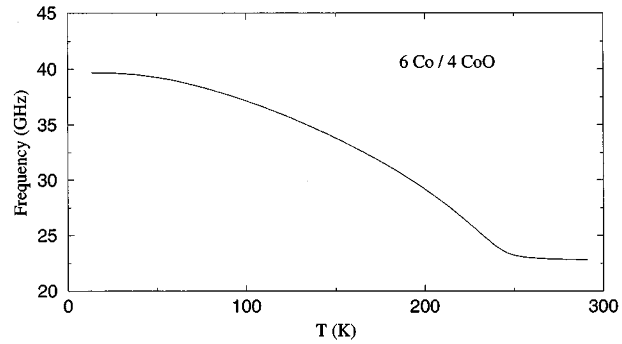

FIG. 11. Frequency of the lowest spin-wave mode in a six layer Co/four layer CoO structure as a function of temperature. $H_I=-50$ kG and $H_a^{in}=2$ kG.

assumed the transition temperature of the ferromagnet is much higher that that of the antiferromagnet, so that the average spin moments in the ferromagnet are at their maximum value, $S=1$, at all temperatures. The interface exchange coupling is 10% of the exchange coupling within the antiferromagnet. The thermal averaged spin magnitude in atomic layer 1 is denoted by S_1, with the other symbols defined similarly.

The key feature to notice in Fig. 10 is that even though the bulk transition temperature for CoO is around 293 K, because of finite-size effects the effective transition temperature is lowered to about 250 K. This is consistent with the experimental data of Ref. 5. In addition we note that the spins in outer layers of the antiferromagnetic film, S_1 and S_4, generally have smaller magnitudes that the spins in the middle. This is because the outer layer spins see a smaller effective field than those in the center and thus are more sensitive to thermally induced fluctuations.

Using the data of Fig. 10 as parameters in the spin-wave calculation, we may calculate the temperature-dependent frequency of the lowest spin-wave mode. The results are presented in Fig. 11. The parameters for this calculation are the same as those for Fig. 10 except that we have also included an in-plane anisotropy field of 2 kG. We see that as the temperature is reduced, the frequency of the spin-wave begins to rise around 250 K and has doubled its value by low temperatures. This is very similar to the recent experimental results in Ref. 5.

IV. SUMMARY AND CONCLUSION

In this paper we have explored a model where the anisotropies in a ferromagnetic film are dynamically coupled to the spin motion in a neighboring magnetic film through interface exchange. We have concentrated on the behavior of the low-frequency spin-wave mode of the ferromagnetic film since this is normally what is measured in Brillouin light-scattering experiments. Our results indicate that the frequency of this mode can be substantially changed by the interface coupling when anisotropy is present in the other material. We show that the shift in frequency depends not only on the interface exchange and anisotropy of the neighboring film, but on its thickness as well. An increase in the thickness leads to an increase in the shift. In contrast to static hysteresis measurements, the spin waves are a dynamic probe of the local coupling between the magnetic materials and may give additional information not available through measurements of magnetization. Finally, we used our theoretical calculations to explain recent Brillouin-scattering experiments on Co/CoO thin films. These experiments showed that the frequency of the low-frequency spin wave doubled as the temperature was reduced. The temperature dependence of the frequency is consistent with the coupling of a thin film of antiferromagnetic CoO to Co.

We emphasize that the true interface structure for Co/CoO is probably not as simple as the one used in our calculation. As emphasized in the literature,[13] the antiferromagnet/ferromagnet interface can be quite complex. Nonetheless, we expect that our results will give a reasonable guide to the behavior in this system. Furthermore our results should also be of interest for studying other structures with Brillouin scattering, for example Fe films with Cr overlayers at low temperatures.

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[1] X. Lin, G. C. Hadjipanayis, and S. I. Shah, J. Appl. Phys. **75**, 6676 (1994).

[2] M. J. Carey and A. E. Berkowitz, J. Appl. Phys. **73**, 6892 (1993).

[3] M. J. Carey and A. E. Berkowitz, Appl. Phys. Lett. **60**, 3060 (1992).

[4] W. H. Meiklejohn and C. P. Bean, Phys. Rev. **105**, 904 (1957).

[5] A. Ercole, T. Fujimoto, M. Patel, C. Daboo, R. J. Hicken, and J. A. C. Bland, J. Magn. Magn. Mater. (to be published).

[6] F. C. Noertemann, R. L. Stamps, and R. E. Camley, Phys. Rev. B **47**, 11 910 (1993).

[7] H. Puszkarski and M. G. Cottam, Acta Phys. Pol. **79**, 549 (1991).

[8] R. L. Stamps, Phys. Rev. B **49**, 339 (1994).

[9] R. W. Wang and D. L. Mills, Phys. Rev. B **50**, 3931 (1994).

[10] S. Blumenröder, E. Zirngiebl, P. Grünberg, and G. Güntherodt, J. Appl. Phys. **57**, 3684 (1985).

[11] M. R. F. Jensen, T. J. Parker, Kamsul Abraha, and D. R. Tilley, Phys. Rev. Lett. **7**, 3756 (1995).

[12] B. Hillebrands, P. Baumgart, and G. Güntherodt, Phys. Rev. B **36**, 2450 (1987).

[13] A. P. Malozemoff, Phys. Rev. B **35**, 3679 (1987).