Soft mode of antiferromagnetic multilayers near the surface spin-flop transition

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We investigate the long wavelength soft mode of antiferromagnetic (AF) multilayers near the field induced instability of the AF order. We show that in the vicinity of the surface spin flop field (H_{SSF}) the frequency of the lowest mode is $\Omega^2/\gamma^2=\alpha(H_{SSF}-H)+\beta(H_{SSF}-H)^2$, where α and β are functions of the magnetic parameters of the multilayer and H is the external field strength. For antiferromagnetic films the first order term is zero (due to the absence of demagnetizing effects) and $\beta=1$.

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The isomorphism between the magnetic structures of artificial transition metal multilayers (TMM) and two-sublattices antiferromagnetic (AF) materials has been often explored in the study of new magnetic phases induced by an external field.[1,2] From the technological applications standpoint, this is an issue of considerable relevance. For instance, the value of the external field strength which turns the AF phase unstable is the onset of the giant magnetoresistance.[3] This is a key parameter for the development of high quality sensors.

The surfaces break the translational symmetry in magnetic multilayers allowing new phases not found in bulk systems. In this regard the use of externally applied fields to induce phase transitions is particularly interesting because the impact of the modifications in the magnetic energy of spins near the surface can be examined. A possible way to investigate surface phase transitions is to look for a surface spin wave whose frequency is driven to zero, as reported for semi-infinite Fe/Gd multilayers.[4] This association of a soft surface spin wave with a surface nucleated phase transition was originally proposed by Saslow and Mills[5] in a study of MnF_2-type antiferromagnets with a (100) surface at low temperatures. They predicted a long wavelength surface spin wave in the frequency gap below the bulk spin wave spectrum, which becomes soft at an external field strength smaller than the bulk spin flop field.[5] This issue was further examined later by Mills,[6] Keffer and Chow,[7] and more recently by Mills.[1] The early studies[5–7] focused on the surface spin wave in an AF film with a single surface (semi-infinite geometry) and were extended to the finite film case using numerical techniques.[1] There is not many experimental reports of surface spin flop (SSF) in AF films, but measurements of the magnetization curves of Fe/Cr superlattices have produced unambiguous evidence of the SSF transition.[2]

In this paper we investigate the long wavelength lowest mode of an AF multilayer, including both cases of AF films, such as a MnF_2(100) film, with noncompensated surfaces, and transition metal AF multilayers. The multilayer consists in the stacking of atomic planes (or thin ferromagnetic layers) with the uniaxial anisotropy easy axis along the $\hat{z}$ direction, and the normal to the surface is along the $\hat{y}$ direction.

The exchange energy within the ferromagnetic (F) layers, in TMM of current interest, is much larger than that coupling adjacent layers. As a result the spins within the thin F layers respond coherently to the external magnetic field. Thus the magnetic structure of the transition metal AF multilayer is isomorphic to that of the AF thin film. The detailed discussion of this isomorphism is found in the work by Wang and Mills.[1] A MnF_2(100) film, for instance, consists in the stacking of atomic planes, each containing spins from one sublattice, and there is a twofold anisotropy axis in the plane. Below the Nèel temperature, and in zero external magnetic field, neighboring planes contain spins pointing in opposite directions ($\hat{z}$ and $-\hat{z}$). Neighboring planes are held in the AF order by the strong exchange energy of nearest neighbor spins. TMM of current interest, such as Fe/Cr AF multilayers,[1] consist of thin sheets of ferromagnetically coupled spins, composing the Fe layers, with antiferromagnetic coupling between the sheets, mediated by the Cr layers. The layers are parallel to the surface and there is a twofold anisotropy axis in the plane. In both systems, the application of a magnetic field parallel to $\hat{z}$ can induce a transition to a SSF phase, where the spins of adjacent layers are canted with respect to $\hat{z}$. The field induced magnetic phases are described using either the spatial arrangement of the magnetic moments per unit cell of each atomic plane of the AF film, or the magnetization of each layer of the TMM.[1,2,8–10] Throughout the paper we use layer to mean either an atomic plane of an AF film or a thin ferromagnetic layer of a TMM.

We use spin variables $\vec{S}_i$ to represent either the magnetic moment, per unit cell, of the *i*th-atomic plane of AF films, or the magnetization of the *i*th-thin ferromagnetic layer of AF transition metal multilayers. The magnetic energy is

$$\begin{aligned} E = \sum_{i=1}^{N} & J[\cos(\theta_i - \theta_{i\pm1})(S_i^{\tilde{x}}S_{i\pm1}^{\tilde{x}} + S_i^{\tilde{z}}S_{i\pm1}^{\tilde{z}}) + S_i^{y}S_{i\pm1}^{y}] \\ & - K[\cos^2\theta_i(S_i^{\tilde{z}})^2 + \sin^2\theta_i(S_i^{\tilde{x}})^2] \\ & - g\mu_B H\cos\theta_i S_i^{\tilde{z}} + 2\pi(g\mu_B)^2(S_i^{y})^2, \end{aligned} \tag{1}$$

where θ_i is the equilibrium value of the in-plane angle between $\vec{S}_i$ and the easy axis ($\hat{z}$ direction). Each term in Eq. (1) represents either the energy per spin for AF films or the

energy per unit volume of F layers for TMM. The first term is the exchange energy coupling adjacent layers, the second term is the uniaxial anisotropy energy, and the third term is the Zeeman energy, for an external field H applied along the easy axis. The last term is the demagnetizing energy, for the case of thin film transition metal AF multilayers.[1] If the magnetization is tilted out of the plane of the layers, the perpendicular component, M_y, produces a demagnetizing field $-4\pi M_y\hat{y}$, confined to each layer, leading to the energy per unit volume of $2\pi M_y^2$, represented in Eq. (1) by the last term.[11]

In the equilibrium configuration all the spins are contained in the layers and local axes are defined from the equilibrium pattern $(\{\theta_n\}, n=1,\ldots,N)$ as $\hat{x}=\cos\theta_i\hat{\tilde{x}}+\sin\theta_i\hat{\tilde{z}}$, $\hat{\tilde{y}}=\hat{y}$, and $\hat{z}=-\sin\theta_i\hat{\tilde{x}}+\cos\theta_i\hat{\tilde{z}}$. The excitation spectrum is obtained from the solutions of the Landau-Lifshitz equations without damping, using the above system of coordinates with the $\hat{\tilde{z}}$-axis along the equilibrium direction of the spin representing each layer. In the long wavelength limit we assume the fluctuations out of equilibrium to be of the form $S_i^{\tilde{x}}(t)=S_i^{\tilde{x}}e^{i\omega t}$ and $S_i^y(t)=S_i^y e^{i\omega t}$, where $S_i^{\tilde{x}}$ and S_i^y are layer dependent amplitudes, and solve the system of $2N$ coupled equations

$$\frac{d\vec{S}_i}{dt}=\gamma\vec{S}_i\times\vec{H}_{eff}(i), \tag{2}$$

for $i=1,\ldots,N$, where the effective field on layer i is given by $g\mu_B\vec{H}_{eff}(i)=-\partial E/\partial\vec{S}_i$. Then, for the ith layer we may write:

$$\Omega S_i^{\tilde{x}}+\left(-\frac{1}{2}\cos(\theta_i-\theta_{i\pm1})+h_A\cos^2\theta_i+h\cos\theta_1+h_D\right)iS_i^y+\frac{1}{2}S_{i\pm1}^y=0,$$

$$i\Omega S_i^y+\left(-\frac{1}{2}\cos(\theta_i-\theta_{i\pm1})+h_A(\cos^2\theta_i-\sin^2\theta_i)+h\cos\theta_i\right)S_i^{\tilde{x}}+\frac{1}{2}S_{i\pm1}^y=0, \tag{3}$$

where we have defined $\Omega=\omega/(\gamma H_E)$, where $H_E=2JS/g\mu_B$ is the interlayer exchange field, and γ is the giromagnetic ratio. In Eqs. (3) we have used the fields in units of H_E, and $h_A=K/J$, $h=H/H_E$, and $h_D=2\pi(g\mu_B)^2/J$.

For antiferromagnetic films, we take $h_D=0$ and write Eqs. (3) in terms of the variables $S_i^{\pm}=S_i^x\pm iS_i^y$. The equations of motion are separable by this transformation, and the resulting system of N coupled equations has the form:

$$\cos\theta_i\Omega S_i^+ +\frac{1}{2}(S_{i+1}^+(1-\delta_{i,N})+S_{i-1}^+(1-\delta_{i,1}))+\left(-\frac{1}{2}(\cos\theta_{i+1}+\cos\theta_{i-1})+h_A\cos\theta_i+h\right)\cos\theta_i S_i^+=0, \tag{4}$$

where $i=1,\ldots,N$, and only one of the exchange terms (either S_{i+1}^+ or S_{i-1}^+) is considered for the surface layers. If N is even the AF order is stable if the external field strength is smaller than the surface spin flop field $[h<(h_A^2+h_A)^{1/2}]$.[9,10] In this case, as seen from Eqs. (4), the equations for all layers $i=1,\ldots,N$ have the same form, except for the fact that the contribution of the Zeeman energy alternate sign ($\pm h$) and the surface layers have half of the exchange contributions. Choosing $\theta_i=0$ for $i=odd$ and $\theta_i=\pi$ for $i=even$, the frequencies of the excitations are obtained from an implicit dispersion relation that has the form $\bar{\mathcal{M}}\cdot\vec{v}=0$. For a film with four atomic planes this eigenvalue equation is written as

$$\begin{pmatrix} a & b & 0 & 0\\ b & c+b & b & 0\\ 0 & b & a+b & b\\ 0 & 0 & b & c\end{pmatrix}\begin{pmatrix} S_1^+\\ S_2^+\\ S_3^+\\ S_4^+\end{pmatrix}=0, \tag{5}$$

where $a=\frac{1}{2}+h_A+(\Omega+h)$, $b=\frac{1}{2}$, and $c=\frac{1}{2}+h_A-(\Omega+h)$. For $i\neq1$ or N the matrix elements of the principal diagonal, m_{ii}, are given by $a+b$ for $i=$odd and by $c+b$ for $i=$even. The matrix elements of the secondary diagonals, $m_{ii\pm1}=b$, are due to the exchange coupling between neighboring planes. For a film with N atomic planes, the excitation spectrum is obtained as an eigenvalue problem involving a $N\times N$ tridiagonal matrix $\bar{\mathcal{M}}_N$ whose nonzero elements are at the principal diagonal and the two secondary diagonals. From the structure of $\bar{\mathcal{M}}_4$, in Eq. (5), it is clear how to obtain $\bar{\mathcal{M}}_2$ and also how to proceed in order to obtain matrices for larger values of N. As in the $N=4$ case in Eq. (5), the elements of the principal diagonal alternate between those corresponding to positive and negative values of the Zeeman energy, and m_{11} and m_{NN} have only half of the exchange contribution.

The matrix $\bar{\mathcal{M}}_N$ can be transformed into an upper triangular form using straightforward algebraic manipulations.[10,12] This procedure allows the calculation of the frequency of the lowest excitation for any finite value of N. For a film with two atomic planes we write the determinant (D_2) of the matrix $\bar{\mathcal{M}}_2$ as

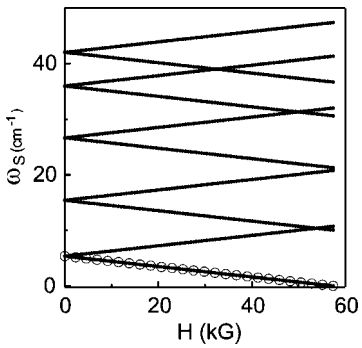

FIG. 1. The spectrum for an AF film with $N=10$ planes and an anisotropy field of $h_A=0.015$ as appropriate to MnF_2. The open symbol curve is the prediction of Eq. (10) and the straight line curves are obtained from the roots of d_{10}.

$$D_2 = \begin{vmatrix} a & b \\ 0 & c - \dfrac{b^2}{a} \end{vmatrix} \tag{6}$$

The determinant (D_4) of the matrix $\bar{\mathcal{M}}_4$ is given by

$$D_4 = \begin{vmatrix} a & b & 0 & 0 \\ 0 & c+b-\dfrac{b^2}{a} & b & 0 \\ 0 & 0 & a+b-\dfrac{b^2}{c+b-\dfrac{b^2}{a}} & b \\ 0 & 0 & 0 & c-\dfrac{b^2}{a+b-\dfrac{b^2}{c+b-\dfrac{b^2}{a}}} \end{vmatrix}. \tag{7}$$

In the upper triangular form, the elements of the diagonal of the matrices $\bar{\mathcal{M}}_N$ display a continued fraction structure, as shown in Eqs. (6) and (7). The determinant of the matrix $\bar{\mathcal{M}}_N$ is given by

$$D_N = \prod_{i=1}^{N} d_i, \tag{8}$$

where d_i is the ith diagonal element of the matrix $\bar{\mathcal{M}}_N$. We have found that the spectrum is obtained from the roots of the last diagonal element d_N, as shown in Fig. 1 for a MnF_2 film with $N=10$ atomic planes. We use[9] $H_E=465$ kG and $H_A=6.97$ kG, leading to a surface spin flop field of $H_{SSF}=57.4$ kG. Notice that the lowest mode becomes soft at the surface spin flop field. This is the surface mode associated with the instability of the AF order, as proposed by Saslow and Mills.[5] We explore the factorization of the secular equation, as expressed by Eq. (8), and the mathematical technique of induction on N, to show analytically the existence of a surface spin wave that softens at the surface spin flop field. We show that the field dependence of the surface soft mode $\Omega_S(H)$ is the same for any even value of N.

For an AF film with $N+2$ atomic planes the last element of the diagonal of $\bar{\mathcal{M}}_{N+2}$, d_{N+2}, is related to the corresponding element of a film with N atomic planes according to

$$d_{N+2} = c - \frac{b^2}{a+b-\dfrac{b^2}{b+d_N}}. \tag{9}$$

By inspection of Eqs. (6) and (7) we find that Eq. (9) is satisfied for $N=2$. If $d_N=0$ for a value of Ω for which $ac=b^2$, then from Eq. (9) $d_{N+2}=0$ at the same value of Ω. From Eq. (6) we find that $d_2=0$ for $ac=b^2$. It follows by induction that the result is valid for any even value of N. The roots of $ac-b^2=0$ are

$$\Omega_S = |h_{SSF} - h|, \tag{10}$$

where $h_{SSF}=(h_A^2+h_A)^{1/2}$. Thus AF films with an even number of atomic planes has a mode that becomes soft at the surface spin flop field. In the limit of large values of N we have $d_{N+2}=d_N$. Using Eq. (9) we find that Eq. (10) describes one of the modes, showing that surface effects are present even for large values of N. In Fig. 1 we show a typical spectrum. The open symbol curve corresponds to the prediction of Eq. (10) and the full lines represent the spectrum for a film with ten atomic planes.

For $h_D \neq 0$ it is not possible to find a simple analytical result valid for any value of N and for any value of the external field strength. We have calculated the spectrum by solving the Eqs. (3) numerically. In Fig. 2 we show $\omega_S(H)=\gamma H_E \Omega_S(H)$, for Fe/Cr multilayers with $N=2$, 10, 20, and 40 ferromagnetic layers. We use[1] $H_E=2$ kG, $H_A=0.5$ kG, and $4\pi M_s=21$ kG, leading to a surface spin flop field of $H_{SSF}=1.11$ kG. Notice that in the neighborhood of the surface spin flop field all curves merge together in a single function $\Omega_S(H)$. Thus it is possible to find an expression for $\Omega_S(H)$ in terms of the magnetic parameters (anisotropy, exchange, and saturation magnetization of the magnetic layers). We have solved the secular equation for a bilayer and have reproduced the numerical results for arbitrary values of N if the external field is close to the critical field ($h \approx h_{SSF}$). Using Eqs. (3) for a bilayer we have found that the frequency of the lowest mode is given by

$$\Omega_-^2 = h^2 + h_{SSF}^2 + h_D\left(\frac{1}{2}+h_A\right) - \sqrt{h^2[h_D^2+2h_D+4(h_Ah_D+h_{SSF}^2)]+\frac{h_D^2}{4}}. \tag{11}$$

In order to find an expression valid in the neighborhood of the critical field, we have expanded Eq. (11) keeping the

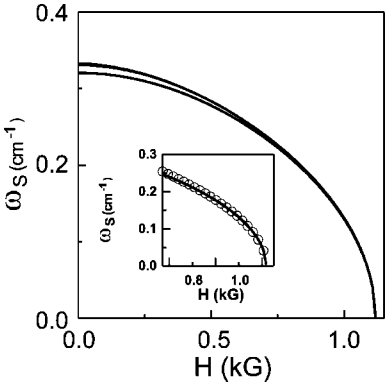

FIG. 2. The frequency of the soft mode, $\omega_S(H)$, for multilayers with $N=2$, 10, 20, and 40 ferromagnetic layers. We use $h_A=0.25$ and $h_D=10.5$. In the inset we show $\omega_S(H)$, for $N=40$, and the prediction of Eq. (15).

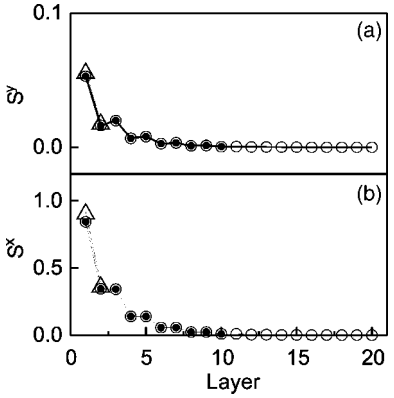

FIG. 3. The absolute values of the amplitudes of fluctuations out of equilibrium, (a) normal to the surface and (b) in the surface, for $h=0.9h_{SSF}$. We use $h_A=0.25$ and $h_D=10.5$. The open symbol curve is for a multilayer with $N=20$ ferromagnetic layers, the full symbol curve is for $N=10$, and the triangles represent a bilayer.

lowest order terms. Using $\epsilon=h_{SSF}-h$ we have found:

$$\Omega_-^2=\alpha\epsilon+\beta\epsilon^2, \tag{12}$$

where

$$\alpha=h_{SSF}\left(2+\frac{h_D^2-4h_{SSF}^2}{2h_{SSF}^2+h_Ah_D+\frac{h_D}{2}}\right) \tag{13}$$

and

$$\beta=1-\frac{Q}{2\sqrt{h_{SSF}^2Q+\frac{h_D^2}{4}}}\left(1-\frac{Qh_{SSF}^2}{Qh_{SSF}^2+\frac{h_D^2}{4}}\right) \tag{14}$$

with $Q=h_D^2+2h_D+4h_Ah_D+4h_{SSF}^2$. For AF films $\alpha=0$ and $\beta=1$, and we recover the expression given in Eq. (10). For $h_D\neq 0$ the dominant term is the first order term. In this case we get

$$\Omega_-=\sqrt{\alpha(h_{SSF}-h)}. \tag{15}$$

As seen in Eq. (15), for $h_D\neq 0$ the slope of the $\Omega(h)$ curve is infinite ($d\Omega/dh=-\infty$) in the close neighborhood of the critical field. Notice, from the inset of Fig. 2, that Eq. (15) reproduces $\Omega_S(H)$ for the chosen multilayers ($N=2$, 10, 20, and 40) in a large interval of external field strength $0.6H_{SSF}<H<H_{SSF}$. We have found that for $h=0$ the lowest mode covers all the layers with a modest enhancement of the fluctuation amplitude near both surfaces. As the external field strength is increased the soft mode gets progressively more localized at the surface where the spins are opposite to the external field. In Fig. 3 we show the absolute values of S_i^y and S_i^x when the external field strength is $h=0.9h_{SSF}$. We show the results for a Fe/Cr bilayer and for two Fe/Cr multilayers, with $N=10$ and $N=20$. The penetration depth of the excitation is around four layers. Thus if $N=6$ or larger there is no relevant change in the energy cost to excite the soft mode. The fluctuations in the equilibrium arrangement of the spins are predominantly in the plane of the layers (S_i^x) because the out of plane fluctuations (S_i^y) involve an extra cost of demagnetizing energy. We show in Fig. 3 that the fluctuations are predominantly at the first two layers, there is almost no difference between the amplitudes of the fluctuations for $N=10$ and $N=20$, and there is not a relevant difference between the results for the bilayer and the two chosen multilayers. This is the reason why near the critical field $\Omega_S(H)$ does not depend on the number of layers. Due to the high degree of localization of the soft mode at the near surface layers, we suggest that measuring $\Omega_S(H)$ may be a promising means of accessing the near surface magnetic parameters of the multilayer. The instability of the AF order is nucleated at the surface where the soft mode is localized. Thus the surface magnetic parameters may be of some technical relevance for development of GMR based devices.

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[1] R. W. Wang and D. L. Mills, Phys. Rev. B **50**, 3931 (1994).
[2] R. W. Wang, D. L. Mills, E. E. Fullerton, J. E. Matson, and S. D. Bader, Phys. Rev. Lett. **72**, 920 (1994).
[3] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Pretoff, P. Etiene, G. Creuzet, A. Friedich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).
[4] J. G. LePage and R. E. Camley, Phys. Rev. Lett. **65**, 1152 (1990).
[5] W. Saslow and D. L. Mills, Phys. Rev. **171**, 488 (1968).
[6] D. L. Mills, Phys. Rev. Lett. **20**, 18 (1968).
[7] F. Keffer and H. Chow, Phys. Rev. Lett. **31**, 1061 (1973).
[8] F. C. Nörteman, R. L. Stamps, A. S. Carriço, and R. E. Camley,

Phys. Rev. B **46**, 10 847 (1992).
[9] A. S. Carriço, R. E. Camley, and R. L. Stamps, Phys. Rev. B **50**, 13 453 (1994).
[10] A. L. Dantas and A. S. Carriço, Phys. Rev. B **59**, 1223 (1999).
[11] Dipolar fields of long wavelength spin waves of TMM may be much larger than the anisotropy field and the exchange field coupling adjacent layers. In Fe/Cr, for instance, typical values are $H_E \approx 2.0$ kG, $H_A \approx 0.5$ kG while $4\pi M \approx 21$ kG. For AF films like MnF_2 the strong exchange coupling between neighboring planes is two orders of magnitude larger than the dipolar fields, and we do not use the demagnetizing energy in Eq. (1).
[12] We use the standard Gauss elimination method. For $N=2$, for instance, this amounts to multiplying the first row of $\bar{\mathcal{M}}_2$ by $-b/a$ and adding the result to the second row, in order to obtain the matrix in Eq. (6).