Interfacial effects in spin-wave resonance of iron films grown by molecular beam epitaxy

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We report theoretical results for iron films prepared by the molecular beam epitaxy technique which give further evidence for the existence of magnetic parameter variations near the film-substrate interface. The ferromagnetic resonance experiments reported earlier complement recent magnetometer measurements. The calculations consist of determining both volume and surface spin-wave dispersions as a function of various assumed variations of magnetic parameters near the interface. This allowed us to predict the excitation of surface modes in addition to the usual volume spin-wave modes. Experimentally, surface modes are excited for thickness $t > 1000$ Å and are displaced about 500—1000 Oe above the main line. In addition, we are able to predict the observed deviation from quadratic behavior for low-order-number spin-wave excitations.

INTRODUCTION

Spin-wave resonance (SWR) data which show evidence for surface-mode excitation in magnetic thin films have been accumulating over the past ten years. These surface spin-wave excitations are localized at the interface between the film and substrate and are contrasted with standing-wave modes or volume spin-wave excitations in which the SWR propagation wave vector is quantized with respect to film thickness. The possibility of such excitations was predicted on theoretical grounds by Wolf in 1965[1] and later by Puszkarski in 1967.[2] Their calculations were based upon a model which assumed a uniform magnetization within the sample, but with a surface anisotropy K_s arising from the interface. The first actual observation of surface waves was reported in 1972 by Brown *et al.*[3] in their work on yttrium iron garnet (YIG) films grown by liquid-phase epitaxy on gadolinium gallium garnet (GGG). They explained their results on the basis of the surface anisotropy model of Puszkarski. The following year Guenzer *et al.*[4] suggested that these results could also arise from a gradient in the magnetization near the interface. Finally, in 1977 a definitive study by Vittoria and Schelleng[5] showed that the surface states located at the YIG/GGG interface arose from a gradient in the magnetization in the YIG due to interfacial diffusion of the constituents.

Previous work on the molecular beam expitaxy (MBE) iron films has indicated that they too have gradients in both the magnetization and magnetic properties near the Fe/Ge interface.[6,7] In this case, however, the gradients are believed to arise from strain at the interface due to lattice mismatch, rather than interfacial interdiffusion, since the MBE iron films are grown at such low temperatures.[8] This paper will concern itself with the development of a theoretical model based upon these interfacial gradients, to explain both the volume and surface-mode SWR excitations observed in these MBE-grown iron films.

We shall consider in turn a step,[9] a linear,[10] and an exponential variation of the magnetization near the interface. Although all three assumptions give rise to qualitatively similar results for the magnetic field dependence of the SWR modes, the exponential variation is easier to justify physically. For example, the linear and step variations introduce artificially abrupt boundary conditions within the film itself, in addition to boundary conditions at the two surfaces. We note that only the exponential variation gives rise to a $\nabla^2 M$ term in the equation of motion used for the analysis. The step and linear variations of M are useful pedagogical exercises, however, in that they reveal the essence of the results in simple mathematical form and allow one to anticipate the numerical results for an exponential variation, since computer numerical analysis was necessary to generate the SWR spectra for that case. We find that the model calculations successfully predict volume and surface-mode excitations, as well as the observed dependence of the resonance position on n_I^2 for the high-mode-order index n_I and the departure from n_I^2 for low n_I, without the need for introducing a surface anisotropy term K_s.

MODELS AND RESULTS

The dynamic equation of motion for a ferromagnet may be written

$$\frac{i\omega}{\gamma}\vec{m}=(\vec{M}_0+\vec{m})\times\left[\vec{H}_i+\frac{2A}{M_0^2}\nabla^2(\vec{M}_0+\vec{m})\right], \tag{1}$$

where A is the exchange-stiffness constant, M_0 the static component of the magnetization, m the dynamic component of the magnetization, ω the angular frequency of applied radiation, $\gamma=g(e/2mc)$, and H_i is the effective static internal magnetic field. For the case of a thin magnetic film saturated normal to the film plane (i.e., parallel to z), $\nabla^2=\partial^2/\partial z^2$. For circularly polarized radiation propagating along z, Eq. (1) reduces to the scalar form

$$\frac{2A}{M_0}\frac{d^2m}{dz^2}+\left[\frac{\omega}{\gamma}-H_i(z)\right]m=0\,. \tag{2}$$

The internal field H_i is now assumed to be composed of

an applied field normal to the film plane H_a, the demagnetizing field from surface poles $4\pi M_0$, an effective anisotropy field H_u, and an effective field arising from gradients in M_0. Since $z=0$ corresponds to the substrate/film interface, the variations in M_0 are assumed to arise from this point and hence will be a maximum there. As $z \rightarrow L$, the free surface of the film, it is assumed that the interfacial effects decrease and the film will approach the properties of bulk material. In the case of a uniform material there is no z dependence, $H_i(z) = H_a - 4\pi M_0 + H_u$, and the separation of the SWR modes in applied field away from the uniform mode obey the well-known n^2 law.[11] One recognizes that Eq. (2) is analogous to Schrödinger's equation and hence we have a one-to-one correspondence to the one-dimensional problem of a particle under the influence of a scalar potential.

The boundary condition on the dynamic magnetization m at the surfaces is[12]

$$A\frac{\partial m}{\partial z} + K_s m = 0 \,. \tag{3}$$

Clearly, if $K_s \gg |Ak|$, then $m \simeq 0$ at the boundary, the well-known result of surface anisotropy pinning. For example, in a film of $d = 1000$ Å the lowest-order mode $n=1$ yields $k = 3\times 10^5$ cm^{-1}. Then the measured value of $A = 2\times 10^{-6}$ erg/cm yields $|Ak| = 0.6$ erg/cm^2. This can be compared to estimated values for $K_s \simeq 1-10$ erg/cm^2,[13,14] and one sees that these two contributions can be estimated to be comparable. Since calculations have already been done on iron assuming $K_s m \gg A\,\partial m/\partial z$,[14] in this paper we shall consider the opposite case of $K_s = 0$, the totally unpinned surface condition. A subsequent paper will treat the general solution for $K_s \neq 0$.

The simplest case to consider[9] is illustrated in Fig. 1 where we assume a step in $H_i(z)$. Here $H_i(0 \leq z \leq L_1) = H_i(0)$ and $H_i(L_1 \leq z \leq L) = H_i(L)$. We then calculate the separation in applied magnetic field between the two lowest-order (highest-field) SWR, i.e.,

$$\delta H = H_a(n_I = 0) - H_a(n_I = 1) \,,$$

as a function of the depth of the surface potential well $[H_i(0) - H_i(L)]$ for various choices of well width, L_1. As can be seen from the figure, significant δH values only occur if the potential does indeed form a well, which is represented on the right-hand side of the figure for which $H_i(L) > H_i(0)$. This may be interpreted by envisioning the well as having a trapped surface state which gradually becomes identical to the uniform SWR mode as the surface potential well disappears. Furthermore, inspection of the form of m for the $n_I = 0$ or surface excitation shows that the amplitude is a maximum for $z=0$ and decays as one proceeds into the film and the spin-wave propagation constant k obeys $k^2(z) < 0$ for $L_1 < z < L$. This describes a nonpropagating spin-wave mode confined to the interface region. For $H_i(L) < H_i(0)$, the well becomes a barrier. There are then no bound surface states and we are on the left-hand side of the figure where we see $\delta H \simeq 0$.

Similar results are obtained[10] for the case of a linear variation in $H_i(z)$. This case is physically more realistic, but the calculation is more tedious. In order to obtain analytical solutions one must actually assume two linear dependences, as illustrated in Fig. 2, where $H_i(z) = H_i(0) + \alpha z$ for $0 \leq z \leq L_1$ and $H_i(z) = H_i(L_1) + \beta z$ for $L_1 \leq z \leq L$ with $H_i(z)$ smoothly varying at $z = L_1$ such that d^2H_i/dz^2 is finite. Again one obtains bound surface states only for $H_i(L) > H_i(0)$, so only the potential-well solutions are displayed in Fig. 2. The solutions in the well are the familiar Airy functions and again one gets a nonpropagating spin-wave mode confined to the surface region. Since all of the other parameters were kept the same in the two cases, comparison of Figs. 1 and 2 shows that the linear gradient must extend much more deeply

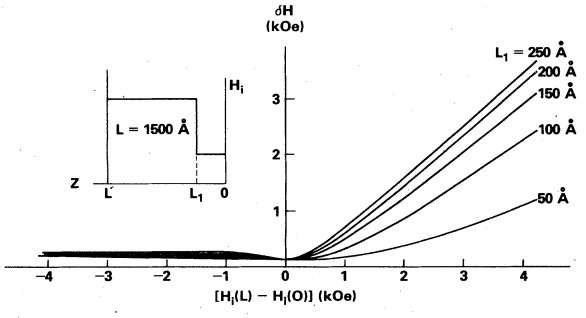

FIG. 1. Difference in magnetic field between the first two (highest-field) excited SWR modes is plotted as a function of $H_i(0) - H_i(L_1)$. The parameter L_i is the width of the step variation. L is the film thickness and is equal to 1500 Å.

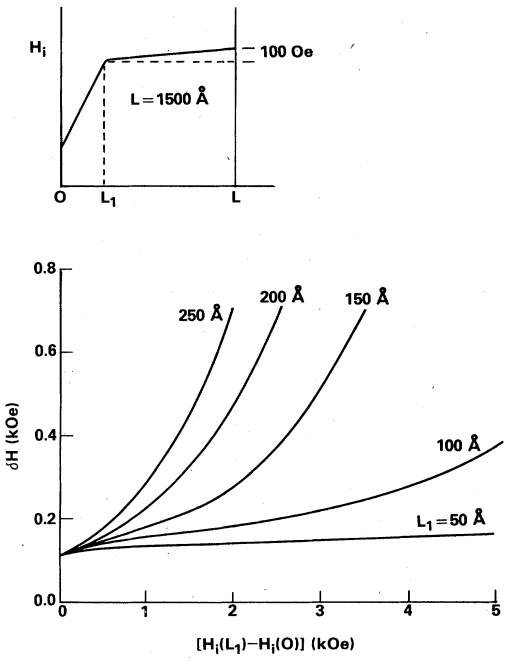

FIG. 2. Difference in magnetic field between the first two excited SWR modes is plotted as a function of $H_i(0) - H_i(L_1)$. The parameter L_1 is the width of the first linear variation, and $L - L_1$ is the width of the second linear variation. $H_i(L) - H_i(L_1) = 100$ Oe for all the calculations shown in the plot. The film thickness is 1500 Å.

into the film (or be larger) to get comparable separations in the first two SWR modes. Or equivalently, for a given depth and width of the potential well, a linear gradient gives several times smaller separation of the first two modes as compared to a step variation.

With these two examples as background, we now assume an exponential dependence of the magnetic properties on the distance from the interface.

Specifically,

$$M(z)=M_0-\Delta M e^{-\Gamma z}\,, \tag{4a}$$

$$H_u(z)=H_u-\Delta H_u e^{-\Gamma z}\,. \tag{4b}$$

Then,

$$\begin{aligned}H_i(z)&=H_a-4\pi M(z)+H_u(z)+H_{\rm ex}(z)\\&=H_a-4\pi M_0+H_u\\&\quad+e^{-\Gamma z}\left[4\pi\Delta M-\Delta H_u-\frac{2A}{M_0}\left[\frac{\Delta M}{M_0}\right]\Gamma^2\right]\,.\end{aligned} \tag{5}$$

The $H_{\rm ex}(z)$ term in Eq. (5) arises from the $(2A/M_0)\nabla^2 M$ term of Eq. (1), the equation of motion. It appears when one has an exponential variation in M, but not for the step or linear variations considered previously.

Equation (1) for the exponential case may be put in the form

$$y^2\frac{d^2m}{dy^2}+y\frac{dm}{dy}+(a+by)m=0\,, \tag{6}$$

where

$$y=e^{-\Gamma z}\,,$$

$$a=\left[\frac{\omega}{\gamma}-H_i(\infty)\right]\Big/\left[\frac{2A}{M_0}\Gamma^2\right]\,,$$

$$b=-[H_i(0)-H_i(\infty)]\Big/\left[\frac{2A}{M_0}\Gamma^2\right]\,.$$

The solution to Eq. (6) is

$$m(y)=\alpha F_+(y)+\beta F_-(y)\,, \tag{7}$$

where F_+, F_-, α, and β are all complex quantities:

$$F_\pm(y)=y^{\pm j\sqrt{a}}\sum_{n=0}^{\infty}c_n^{\pm}y^n\,, \tag{8}$$

where

$$c_n^{\pm}=\frac{(-1)^n b^n}{n!(n\pm 2j\sqrt{a})(n-1\pm 2j\sqrt{a})\cdots(1\pm 2j\sqrt{a})}\,,$$

$$c_0=1,\quad j=\sqrt{-1}$$

and α and β are determined from the boundary conditions and by requiring $m(y)$ to be real. The secular equation is obtained by imposing the free pinning boundary conditions ($K_s=0$) at the two surfaces. Then

$$F'_+(y_0)F'_-(y_L)-F'_+(y_L)F'_-(y_0)=0\,, \tag{9}$$

where

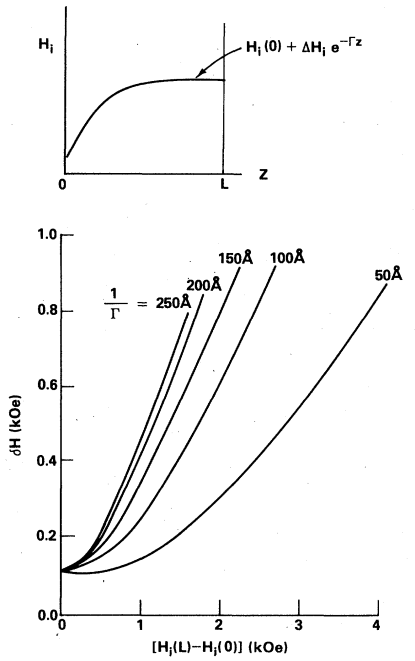

FIG. 3. Difference in magnetic field between the first two excited SWR modes is plotted as a function of $H_i(L)-H_i(0)$. The exponential variation is of the form $H_i(0)+\Delta H_i\exp(-\Gamma z)$. The film thickness is 1500 Å.

$$\begin{aligned}&y_0=1\,,\\&y_L=e^{-\Gamma L}\,,\\&F'_\pm=\frac{d}{dy}F_\pm\,.\end{aligned} \tag{10}$$

The SWR field positions may be calculated by varying a or H_a so that Eq. (9) is satisfied. In Fig. 3 the difference in the first two excited SWR-mode field positions are plotted as a function of $H_i(0)-H_i(L)$. The exponential decay parameter $1/\Gamma$ is varied between 50 and 250 Å.

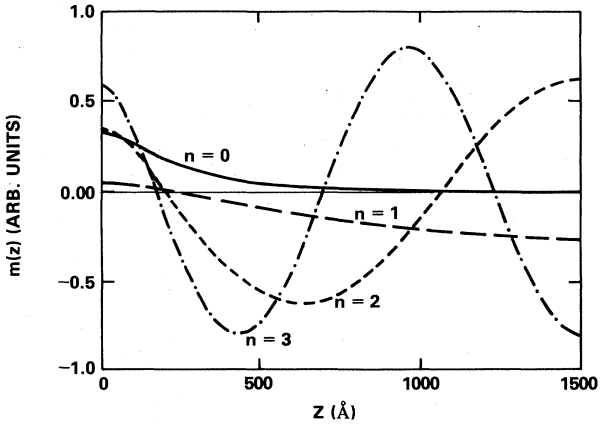

FIG. 4. Solutions of m for both the surface and volume spin-wave excitations for the exponential case with $M=1500$ G, $H_i(0)-H_i(L)=-2$ kOe, $L=1500$ Å, $1/\Gamma=100$ Å.

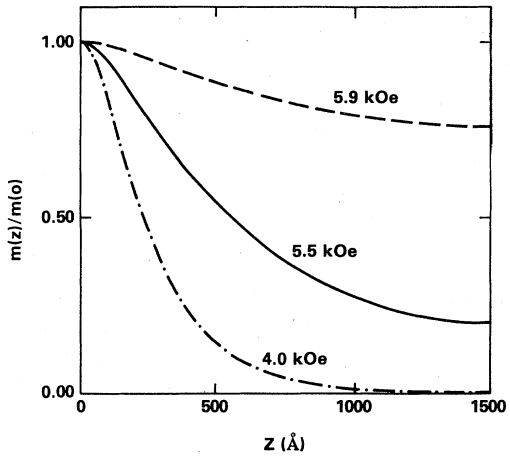

FIG. 5. Surface-mode solutions of m are plotted as a function of distance. For the three curves $1/\Gamma$ is fixed ($=100$ Å), $H_i(0)=6$ kOe, and $H_i(0)-H_i(L)=-0.1$, -0.5, and -2.0 kOe with $H_i(L)$ as the parameter.

The measured[7] value of $2A/M_0$ is 2.55×10^{-9} Oe cm^2 and was used in the calculation. The film thickness is 1500 Å. The curves exhibit the same characteristics as Figs. 1 and 2. Again the first (highest-field) mode of excitation is a surface SWR mode localized near $z=0$, as shown in Figs. 4 and 5.

Any one of the curves in Fig. 3 can give rise to a reasonable fit to experiment. However, we take $M=1500$ G, $1/\Gamma=50$ Å, and $\Delta M/M_0=0.5$, since previous ferromagnetic resonance (FMR) and magnetic measurements[7,15] give us reasonable estimates of these magnetic parameters. Only ΔH_u is taken as adjustable when we fit the calculated SWR spectra to the measured one of Ref. 7 for a 1500-Å film. In Fig. 6 we plot the difference between H_s (surface SWR mode) and successive SWR field positions as a function of an index number squared, n_I^2 taking $n_I=0$ for the surface SWR mode, and $n_I=1$ for the first bulk mode. For high index number, n_I may be regarded as the ordinary spin-wave order number, n. For low n_I the index number merely indicates the progression or the order in which each calculated SWR mode occurs in the spectra. In the absence of any gradients in the material properties and of surface pinning of the magnetization, $n=0$ corresponds to the uniform mode. Symmetric pinning at both surfaces would allow only the odd bulk modes to be excited, and the lowest of these ($n=1$) corresponds closely to the lowest bulk mode seen here. The agreement between theory and experiment is reasonable. In fitting to the experimental data[7] we find that the best result is obtained for

$$H_i(0)-H_i(L)=-2000 \text{ Oe},$$

which implies $\Delta H_u=6.3$ kOe.

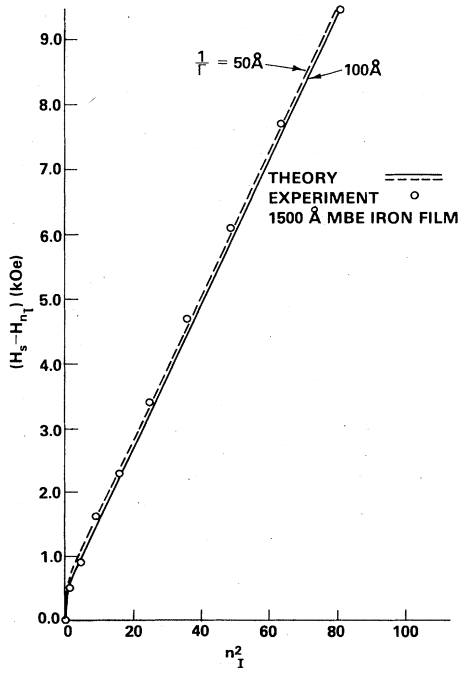

FIG. 6. Comparison between calculated and measured SWR-mode field positions for a 1500-Å-thick film of MBE iron.

DISCUSSION

Variations in M have been implied and measured in MBE iron films.[6,7,15] We now suggest, in addition, that there are variations in H_u in these films. This is not surprising in view of the fact that these MBE iron films may be strained from the lattice mismatch between substrate and film. Furthermore, the strain relaxation length of 50 Å is in reasonable agreement with stress-release mechanisms applicable to metal films evaporated on GaAs.[16] Since the specific values used for Γ and $\Delta M/M_0$ are from iron films grown epitaxially on GaAs, rather than on Ge, the numerical results for ΔH_u are only representative.

It may be possible to improve the fit between theory and experiment by invoking[17] surface anisotropy parameters at the two surfaces other than the $K_s=0$ we have assumed in this calculation. Nevertheless, it is clear that previous[14] work which analyzed FMR data on MBE Fe films in terms of only surface parameters may be subject to numerical error, since it ignores the large contributions to the FMR which we show can arise from gradient effects. This study shows that in order to understand the resonance results for magnetic metal films grown by MBE, as for YIG films[5] grown by Liquid-phase epitaxy, the static magnetic properties must first be determined and their effect upon the resonance modes calculated. Only after this is done can one determine the magnitude of any effects arising from K_s. Future work will include effects of K_s as well as gradients of M on the FMR properties of iron films prepared by the MBE technique.

In summary, we have carried out spin-wave calculations for thin-film magnetic materials which have a z-dependent magnetization and/or uniaxial anisotropy near the substrate interface. The calculation for an exponential variation in the material parameters with distance, the

physically most relevant case, was accomplished for the first time and the results were shown to be in qualitative agreement with the cases involving a step or a linear parameter variation. With an increasingly strong variation near the interface one sees an evolution of the lowest-order spin-wave mode into a surface mode if the variation has the proper sign. The calculated spectrum of spin-wave modes is in good agreement with experiment.

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