Induced anisotropies in a ferromagnet coupled to a polycrystalline antiferromagnet

Jyotirmoy Saha,¹ Kevin Garello,^{2,3,4} B. Viala,^{2,3} A. Marty,¹ and N. Vukadinovic⁵

1 *CEA, INAC/SP2M/NM, 38054 Grenoble, Cedex 9, France*

2 *CEA, LETI, MINATEC, 38054 Grenoble, France*

³*SPINTEC, CEA, CNRS, INPG, CEA/INAC, UJF, 38054 Grenoble, France*

⁴*XLIM, CNRS, 87060 Limoges, France*

5 *Dassault Aviation, 92552 Saint-Cloud, France*

Received 22 December 2009; published 22 January 2010-

The effect of thermal activation of a polycrystalline antiferromagnet (AF) coupled to a ferromagnet (FM) on the unidirectional anisotropy responsible for positive shift of the resonant frequency (f_R) of the FM is presented. Hysteresis loops and ferromagnetic resonance plots of $\text{Co}_{35}\text{Fe}_{65}/\text{Ni}_{50}\text{Mn}_{50}$ are evaluated and matched with those obtained from experiments. The contribution of the AF magnetic structure to f_R is highlighted. An inverse relation between f_R and AF grain size is predicted.

DOI: [10.1103/PhysRevB.81.024417](http://dx.doi.org/10.1103/PhysRevB.81.024417)

 $: 75.30$.Gw, 75.50.Ee, 75.60. $-d$, 76.50. $+g$

I. INTRODUCTION

When a ferromagnet (FM) is coupled to a polycrystalline antiferromagnet (AF), the magnetocrystalline anisotropy of the AF induces various forms of anisotropies in the FM me-diated by uncompensated spins^{1[,2](#page-3-1)} at the AF-FM interface (henceforth, interface). The tuning of these anisotropies has been an important endeavor for industries in the development of spin valve and microwave devices.³ The two basic yet cardinal experimental outcomes that quantify these anisotropies are: (1) hysteresis loops (HLs) yielding exchange bias (H_E) and enhanced coercivity (H_{EC}) and (2) ferromagnetic resonance (FMR) plots yielding the resonant frequency (f_R) . The important distinction between these two classes of experiments is that H_E is a measure of the induced unidirectional anisotropy (henceforth, UA) in the FM as the FM switches between the two magnetic states whereas f_R is a perturbative measure of the UA about the ground state of the system[.4](#page-3-3)[,5](#page-3-4) In either case thermal activation of the AF primarily regulates the UA albeit in disparate ways. A strong research focus has been to formulate a canonical connection between these two classes of UA but an overall consensus in this matter is still at large. To achieve this, most (prior) endeavors have estimated f_R utilizing information from experimental HLs and tailoring techniques using the single domain particle (Kittel) approach.^{4[,6,](#page-3-5)[7](#page-3-6)} These techniques are system specific and moreover do not present a treatment of the thermal activation of the large AF degrees of freedom. Further progress in this field necessitates a theoretical model that can independently evaluate the FMR profiles and HLs in a multitude of systems based on system (material) attributes. An earlier work reported a model that evaluates HLs and also studied this aspect of AF/FM systems extensively.⁸ Utilizing building blocks of this model, in this paper we present a technique to evaluate the FMR response of AF/FM systems. Here, we investigate two metallic systems that are of current technological relevance: (1) $\text{Co}_{35}\text{Fe}_{65}/\text{Ni}_{50}\text{Mn}_{50}$ (henceforth CoFe/NiMn) and (2) $\text{Ni}_{80}\text{Fe}_{20}/\text{Ni}_{50}\text{Mn}_{50}$ (henceforth Py/ NiMn). Experiments are carried out for the CoFe/NiMn system and the model predictions for the Py/NiMn system are compared to existing reports.

II. EXPERIMENTAL DETAILS AND MATERIAL CHARACTERISTICS

The experimental magnetic bilayer structure consists of: $\text{Si}(525 \ \mu\text{m})/\text{SiO}_2(2 \ \mu\text{m})/\text{Ru}(5 \ \text{nm})/\text{Ni}_{50}\text{Mn}_{50}(t_{\text{AF}})$ $= 50 \text{ nm}$)/Co₃₅Fe₆₅(t_{FM})/Ru(5nm) with $t_{\text{FM}} = 10 \text{ nm}$, 15 nm, 20 nm and 25 nm; where, t_{AF} (t_{FM}) is the AF (FM) thickness. The composite system has an area of 4×4 mm². They are grown by RF-magnetron sputtering on thermally oxidized silicon. The deposition is done at room temperature in the absence of a magnetic field with a base pressure of 9×10⁻⁸ mbar and an Argon plasma pressure of 2×10^{-3} mbar. Following deposition, the composite system is annealed in vacuum for an hour at 573 K in an applied field of 500 Oe to induce the undirectional anisotropy in the system. The HLs are obtained using a vibrating sample magnetometer (VSM). Post annealing, the $Co₃₅Fe₆₅$ free layer has a saturation magnetization (M_S) of 1.87×10^3 emu/cc and the easy axis loop yields a coercivity of 17.5 Oe. The complex permeability spectra of the samples are measured from 0.1–10 GHz using a refined single-loop coil technique, based on the model of Bekker *et al.*[9](#page-3-8) The small amplitude radio frequency (rf) field is applied perpendicular to the field-cooling direction. The magnetic characteristics of CoFe (Py) used in the model are: $M_S=1.8\times10³$ emu/cc (8.6) $\times 10^2$ emu/cc),^{[8](#page-3-7)[,10](#page-3-9)} uniaxial magnetocrystalline anisotropy $(K_{\text{FM}}) = 1.57 \times 10^4 \text{ ergs/cc}$ $(K_{\text{FM}}) = 1.57 \times 10^4 \text{ ergs/cc}$ $(K_{\text{FM}}) = 1.57 \times 10^4 \text{ ergs/cc}$ $(4.3 \times 10^3 \text{ ergs/cc})^{8,10,11}$ Curie temperature $(T_C) = 1299 \text{ K}$ ([8](#page-3-7)50 K),^{8[,12](#page-3-11)} exchange constant $(A_{\text{exchange}}) = 1.75 \times 10^{-6} \text{ ergs/cm}$ $(1.0 \times 10^{-6} \text{ ergs/cm})^{8,10}$ $(1.0 \times 10^{-6} \text{ ergs/cm})^{8,10}$ $(1.0 \times 10^{-6} \text{ ergs/cm})^{8,10}$ $(1.0 \times 10^{-6} \text{ ergs/cm})^{8,10}$ and damping parameter (α) =0.005 (0.02).^{[8](#page-3-7)[,10](#page-3-9)} The magnetic structure of NiMn incorporates a biaxial in-plane (easy plane) magnetocrystalline anisotropy (K_{AF}) of 5 $\times 10^5$ ergs/cc in the (001) plane and an out-of-plane anisotropy of 5.8×10^6 ergs/cc and grows with the $\langle 111 \rangle$ texturing[.13](#page-3-12) The interfacial AF moments are restricted to this (easy) plane due to the absence of an energy scale that can match the out-of-plane anisotropy. The density of lattice sites $(D_{lattice})$ at the interface is 8.64×10^{14} sites/cm² and the Néel temperature (T_N) of NiMn is 1070 K.¹³

III. THEORETICAL MODEL

In the model the FM layer is treated micromagnetically, a thin sample of FM is discretized into uniformly magnetized

FIG. 1. (a) Discretization of the AF and FM layers; (b) easy plane (001) of an AF grain with respect to its texturing $\langle 111 \rangle$, the X', Y', Z' axes are denoted by their spherical angles with respect to the **X-Y-Z** coordinate system, $\theta_Z = 54.7^\circ$; and (c) easy plane of an AF grain. $H_{\text{ex-FM}}$ is the exchange field on an AF grain due to the interaction with the FM cell $[Eq. (1)]$ $[Eq. (1)]$ $[Eq. (1)]$.

rectangular parallelepipeds with a square interface that couples with an AF grain [Fig. $1(a)$ $1(a)$]. The AF grains are considered noninteracting. The coupling energy¹⁴ between each FM cell and the corresponding AF grain is

$$
E_{\text{AF-FM}} = -\mathbf{J}_{\text{int}} \widetilde{N} \hat{\boldsymbol{\mu}}_{\text{AF}} \cdot \hat{\boldsymbol{\mu}}_{\text{FM}}, \qquad (1)
$$

where, $J_{int} = 6\sqrt{J_{F/M}J_{AF}}/(N_{int} + 1)$; \tilde{N} is the net imbalance of the moment direction at the interface due to uncompensated spins; $\hat{\mu}_{AF}$ is the direction of the net AF moment $(\tilde{N}\mu_{AF})$ induced at the interface; and $\mu_{FM} = M_S V_{FM} \hat{\mu}_{FM}$, where V_{FM} is the volume of the FM cell. J_{FM} and J_{AF} are estimated in the mean field sense from the knowledge of T_c and T_N . N_{int} is the number of interdiffused interfacial monolayers (chosen to be 1) which corresponds to an AF-FM interface thickness of 4.2 \AA .^{[13](#page-3-12)} The time evolution of each FM degree of freedom is evaluated by numerically integrating the Landau-Lifshitz-Gilbert equation: $\frac{d\mu_{FM}}{dt} = -\left(\frac{\gamma}{1+\alpha^2}\right)\mu_{FM} \times H - \left(\frac{\alpha\gamma}{1+\alpha^2}\right)\hat{\mu}_{FM} \times (\mu_{FM})$ \times **H**); where γ is the gyromagnetic ratio of the free electron and **H** is the net field experienced by a FM cell.¹⁵

The energy (E_{AF}) of each AF grain of volume V_{AF} is considered within the framework of a uniform rotation model [Fig. $1(c)$ $1(c)$]

$$
E_{\rm AF} = 2K_{\rm AF}V_{\rm AF} \left[\frac{\sin^2 2\lambda}{8} - \text{h}\cos(\delta - \lambda) \right],\tag{2}
$$

where $h \sim \frac{J_{\text{int}}\tilde{N}}{2K_{\text{AF}}V_{\text{AF}}} \cos(\beta)$. The time evolution of $\tilde{N}\mu_{\text{AF}}$ of each AF grain is determined by its ability to switch over energy barriers (E_{Barrier}) to a lower energy state.⁸

The switching occurs when the Néel-Arrhenius condition is satisfied

$$
E_{\text{Thermal}} \ge E_{\text{Barrier}} \tag{3}
$$

where $E_{\text{Thermal}} = k_B T \ln(2 f_O t_{SW})$; where, k_B is the Boltzmann constant, $f_{\text{O}}(=1 \text{ GHz})$ is the attempt frequency, t_{SW} is the time available for the AF grain to switch and is estimated from typical VSM sweep rates. To incorporate the polycrystalline nature of the AF, φ_Z , and φ_X [Fig. [1](#page-1-0)(b)] are uniformly distributed in the range $[0, 2\pi]$. $\hat{\mu}_{FM}$ and λ [Fig. [1](#page-1-0)(c)] are initialized randomly. To set the UA, a field of 1000 Oe along X (Fig. [1](#page-1-0)) is selected and a temperature of 1000 K is chosen. To compute the HL, the applied field is swept along the $\pm X$ direction. To evaluate the frequency response of the system, the zero-applied-field equilibrium value of the average FM magnetization (M_0) is noted. An impulse¹⁶ along the **Y** di-rection (Fig. [1](#page-1-0)) is applied and the system is then allowed to relax back to M_O . As the system relaxes, the quantity M_O $-\mathbf{M}(t)$ is recorded where $\mathbf{M}(t)$ is the average FM magnetization as a function of time. The discrete Fourier transform of the **Y** component of $M_0 - M(t)$ yields the real and imaginary (absorption) part of the response of the system. All model calculations are evaluated at $T=300$ K with 64×64 elements. This size yields a system average good enough to be compared to experimental results. $8,14$ $8,14$

IV. RESULTS AND DISCUSSION

In the CoFe/NiMn system, we estimate the average AF grain size (GS_{AF}) in the experimental sample by comparing the experimentally obtained HL to those obtained from model calculations with typical GS_{AF} values^{17[,18](#page-4-0)} as observed in metallic systems. For $t_{\text{FM}}=20$ nm and $t_{\text{AF}}=50$ nm and using GS_{AF} =40, 50, and 60 nm in the model calculations the values obtained are: H_E = 105, 115, and 110 Oe; H_{EC} = 71, 50, and 25 Oe, respectively; the experimental values are: H_E = 105 Oe and H_{EC} = 40 Oe. A good agreement in both H_E and H_{EC} is obtained for $GS_{AF} = 50$ nm and this value of GSAF is retained in the evaluation of FMR response for a similar system. The HLs [Figs. $2(a)$ $2(a)$ and $2(b)$] and FMR responses [Figs. $2(c)$ $2(c)$] obtained from the model and experiment for t_{FM} = 20 and 25 nm are presented. The ability of the model to predict experimental outcomes with this degree of accuracy is a point of merit especially in the light of absence of adjustable parameters. Table [I](#page-2-1) summarizes the results for t_{FM} = 25, 20, 15, and 10 nm. The model values (for f_{R}) exceed those of the experimental ones by 16% as t_{FM} is decreased to 10 nm. We attribute this to a possible change in the microstructure of the experimental sample, for example: the average AF grain size, 19 effective grain volume of the FM and one-to-one correspondence between AF-FM grains. We repeat the model calculation for t_{FM} = 10 nm with a bigger grain size of 70 nm. The results restore the numerical agreement of H_E and f_R to a certain extent between experimental and model outcomes. In the $Py(t_{FM})$ $= 25$ nm)/NiMn($t_{AF} = 50$ nm) system the model predicts f_R ranging from 2.9 to 5.3 GHz for a GS_{AF} variation of 70 to 25 nm, respectively. This brackets the experimentally observed value⁷ of 3.5 GHz. We now estimate the typical exchange field $(H_{\text{ex-AF}})$ experienced by an FM cell due to its coupling with the AF grain to examine the t_{FM} dependence of f_R . Using Eq. (1) (1) (1) , we get

FIG. 2. (Color online) For the CoFe (t_{FM}) /NiMn $(t_{AF}=50 \text{ nm})$ system: (a) HLs for t_{FM} = 20 nm; (b) HLs for t_{FM} = 25 nm; and (c) experimental (dashed lines) and model evaluated (solid lines) FMR response. There exists only one peak in the frequency range 0–10 GHz. The model plots are evaluated with $GS_{AF} = 50$ nm.

$$
H_{\text{ex-AF}} = \frac{J_{\text{int}} \tilde{N}}{M_{\text{S}} V_{\text{FM}}} \langle \hat{\boldsymbol{\mu}}_{\text{AF}} \cdot \mathbf{X} \rangle, \tag{4}
$$

where **X** denotes the UA direction [Figs. $1(b)$ $1(b)$ and $1(c)$]. The $\langle \rangle$ brackets denote a time average as well as the ensemble average over the polycrystalline degrees of freedom of the AF, \tilde{N} is chosen to be $\sqrt{2N_{\text{total}}/\pi}$ where, $N_{\text{total}} = D_{\text{lattice}}$ GS_{AF}. The system average UA field (H_{UA}) takes the form

$$
H_{\text{UA}} = H_{\text{C}} + \frac{J_{\text{int}} \sqrt{2D_{\text{lattice}}/\pi}}{M_{\text{S}} \text{GS}_{\text{AF}} t_{\text{FM}}} \langle \hat{\mu}_{\text{AF}} \cdot \mathbf{X} \rangle, \tag{5}
$$

where $H_C = 2K_{FM}/M_S$. The Kittel equation for our film geometry is

$$
f_{\rm R} = (\gamma/2\pi)\sqrt{H_{\rm UA}(H_{\rm UA} + 4\pi M_{\rm S})}.
$$
 (6)

As $\langle \hat{\mu}_{AF} \cdot X \rangle$ has no explicit t_{FM} dependence, its ensemble average $(\langle \hat{\boldsymbol{\mu}}_{AF} \cdot \mathbf{X} \rangle_{EAV})$ is adequate to investigate f_R as a

FIG. 3. f_R vs t_{FM} plots for CoFe/NiMn $(t_{AF} = 50 \text{ nm})$ system. The theoretical plots are evaluated with $GS_{AF} = 50$ nm. Lines are a guide to the eye.

function of t_{FM} . Figure [3](#page-2-2) shows three plots of f_R vs t_{FM} which were obtained: (1) experimentally, (2) from the model, and (3) using Eqs. ([5](#page-2-3)) and ([6](#page-2-4)) with parameter values: GS_{AF} = 50 nm, J_{int} = 18 × 10⁻¹⁴ ergs, H_C = 17.5 Oe. To estimate $\langle \hat{\boldsymbol{\mu}}_{AF} \cdot X \rangle_{EAV}$, φ_Z is uniformly distributed in the range $[0, 2\pi]$, φ_X is chosen to be zero (without any loss of generality) and λ [Fig. [1](#page-1-0)(c)] is uniformly distributed in the range $[-\pi/4, \pi/4]$. For a quarter million averaging points, $\langle \hat{\boldsymbol{\mu}}_{AF} \cdot X \rangle_{EAV}$ yields 0.74. Experiments have demonstrated a similar trend. $20,21$ $20,21$ The deviation of the single domain particle prediction from the model prediction at low t_{FM} (Fig. [3](#page-2-2)) is not intuitively straightforward but can be loosely put as: for low t_{FM} , $\langle \hat{\mu}_{AF} \cdot X \rangle_{EAV}$ decreases with the finiteness of *A*exchange.

The model evaluated f_R vs t_{AF} plot is shown in Fig. [4](#page-3-17)(a). We will discuss below the case: $GS_{AF} = 50$ nm as a similar discussion holds for $GS_{AF} = 30$ nm. A decrease in t_{AF} below 45 nm increases the population of the AF grains satisfying the condition: $K_{AF}V_{AF} \ll 4J_{int}\tilde{N}$. Such grains are devoid of energy barriers [Eq. ([2](#page-1-2))] thereby turning superparamagnetic. These grains readily trail (align with) the FM moments responding to the rf field. Their inability to consolidate the UA direction results in the reduction in H_{UA} . An increase in t_{AF} from 45 to 75 nm stabilizes the AF grains and pins them in their field-cooling configuration resulting in an increase in H_{UA} . This increase reaches a maximum value denoting an optimum number of AF grains consolidating the UA direc-

TABLE I. Exchange bias (H_E) , enhanced coercivity (H_{EC}) , and resonance frequency (f_R) obtained from experiments (exp.) and model calculations for $\text{CoFe}(t_{\text{FM}})/\text{NiMn}(t_{\text{AF}}=50 \text{ nm})$. Model calculations are done with $GS_{AF} = 50$ nm. (* denotes the model calculations with $GS_{AF} = 70$ nm.)

| t_{FM} (nm) | $H_{\rm E}$ (model) (Oe) | $H_{\rm E}$ (exp.) (Oe) | $H_{\rm EC}$ (model) (Oe) | H_{EC} (exp.) (Oe) | $f_{\rm R}$ (model) (GHz) | $f_{\rm R}$ (exp.) (GHz) |
|-------------------------|--------------------------------|-------------------------------|---------------------------------|-----------------------------------|---------------------------------|--------------------------------|
| 25 | 94 | 80 | 41 | 36 | 4.9 | 5.0 |
| 20 | 115 | 105 | 50 | 40 | 5.4 | 5.4 |
| 15 | 148 | 119 | 59 | 39 | 6.2 | 5.9 |
| 10 | 226 190 [*] | 140 | 78 23* | 56 | $7.4, 6.0^*$ | 6.4 |

FIG. 4. Model evaluated: (a) f_R vs t_{AF} for CoFe $(t_{\text{FM}}=20 \text{ nm})$ /NiMn with $GS_{AF}=30 \text{ nm}$ (open circle symbols), 50 nm $[(+)$ symbols] and (b) f_R vs GS_{AF} for CoFe $(t_{\text{FM}}=20 \text{ nm})/\text{NiMn}(t_{\text{AF}}=50 \text{nm})$. Lines are a guide to the eye.

tion. Beyond this maximum $(t_{AF} > 75 \text{ nm})$, a further increase in t_{AF} results in an additional number of AF grains attaining thermal stability but the net angular dispersion in $\hat{\mu}_{AF}$ reduce H_{UA} . This reduction in H_{UA} is mainly dictated by the magnetic structure of the AF $\langle \hat{\mu}_{AF} \cdot X \rangle_{EAV}$. A similar trend of f_R vs t_{AF} has also been experimentally observed.^{4[,22](#page-4-4)} Next we discuss the GS_{AF} dependence of f_R [Fig. [4](#page-3-17)(b)]. An increase in GS_{AF} enhances the locking of the AF grains along the UA

direction [Eq. ([2](#page-1-2))] simultaneously decreasing the UA field [Eq. (5) (5) (5)]. The net effect of these two conflicting contributions is a monotonic decrease in f_R as a function of GS_{AF} [Fig. [4](#page-3-17)(b)]. This implies that $(J_{int} \sqrt{2D_{lattice}/\pi} / M_S G S_{AF} f_{FM})$ has a dominant contribution over $\langle \hat{\mu}_{AF} \cdot X \rangle$ in the case of FMR measurements. The nontrivial behaviors of f_R vs t_{AF} and f_R vs GS_{AF} attributable to the thermal activation of the large AF degree of freedom have not been the natural outcome of any previous theoretical work. To estimate the (theoretical) maximum f_R of any particular system from HLs, we (model) compute HLs at $T=0$ K and measure the field (H_{SW}) at which the FM magnetization reverses starting from the field-cooled state. For the $\text{CoFe}(t_{\text{FM}}=20 \text{ nm})/\text{NiMn}(t_{\text{AF}})$ $= 50$ nm) system with $GS_{AF} = 50$ nm, H_{SW} is 155 Oe which corresponds to a f_R value of 5.3 GHz using $H_{\text{UA}} = H_{\text{SW}}$ in Eq. (6) (6) (6) . This fits well with the values obtained from both the model and the experiment [Fig. $4(a)$ $4(a)$].

V. CONCLUSIONS

We have established the tunability of f_R by altering thermal stability of the AF (dictated by t_{AF} and GS_{AF}) in an AF/FM system. As the UAs measured by H_E and f_R have different physical origin, a general formula may not be achievable to predict f_R based on information from experimental HLs for a wide range of either t_{AF} or GS_{AF} or temperature. Nevertheless, the maximum attainable f_R in such a system can be estimated from a (experimental) HL evaluated at a temperature (low enough) such that the coercivity of the system is mainly due to the magnetocrystalline anisotropy of the FM.

ACKNOWLEDGMENTS

This work is supported by the French National Research Agency ANR—INSTITUTS CARNOT. The support of J. C. Toussaint and L. Buda-Prejbeanu in this project is acknowledged.

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- ¹⁵**H** includes the applied field, the demagnetizing field, the exchange field due to *A*exchange, the FM MCA field and the exchange field from the corresponding AF grain as outlined in Ref. [8.](#page-3-7)

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¹⁶The impulse is chosen such that the system response is linear.

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