## **Experimental evidence for an unidirectional Gilbert damping parameter**

Chantal Le Graët,<sup>1</sup> David Spenato,<sup>1</sup> Souren P. Pogossian,<sup>1</sup> David T. Dekadjevi,<sup>1,2</sup> and Jamal Ben Youssef<sup>1[,\\*](#page-3-0)</sup>

*Laboratoire de Magnétisme de Bretagne, CNRS–Université de Bretagne Occidentale, 6 Avenue le Gorgeu, 29285 Brest Cedex, France*

2 *Department of Physics, University of Johannesburg, P.O. Box 524, Auckland Park, Johannesburg 2006, South Africa*

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In magnetization dynamics, the relaxation is driven by the damping. Here, we demonstrate that damping in exchange-coupled systems is not only anisotropic but also unidirectional evidenced by an asymmetry in the damping by inversion of the magnetic field polarity. Our study reveals that this asymmetry in the damping is enhanced by the increase in the exchange bias field. Finally, we introduce a modified relaxation term in the equation of motion.

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Among the huge variety of scientific fields involving dynamical processes, spin dynamics is one of the most popular because of its key role in high volume information-storage or processing devices based on metallic, insulators, or semiconductors materials. $<sup>1</sup>$  In magnetic storage devices, the key point</sup> is the speed at which magnetic data-storage elements can be manipulated with either a magnetic field, a spin-polarized electric current<sup>2</sup> or with an electric field.<sup>3</sup> In these research fields, commonly accepted theoretical approaches are based on a well-known phenomenological model applied to magnetic field-induced precession motion of the magnetization<sup>4</sup> or current-induced motion[.5](#page-3-5) In this vast amount of research, the relaxation mechanism is introduced through the use of the so-called damping parameter,  $\alpha$ . Thus, it governs the relaxation of the magnetization toward equilibrium. Despite the general and widespread use of these equations of motion, the physics concerning the phenomenological damping parameter is a matter of controversy. Indeed, the experimental results are *often* interpreted in terms of a scalar isotropic damping parameter<sup>6</sup> whereas some authors claim that it should be of a more complicated form that would rely on a damping matrix. This matrix description would then implement the breathing Fermi-surface model<sup>7</sup> or crystalline symmetry considerations.<sup>8</sup> Then, the damping scalar  $\alpha$  should be replaced by an anisotropic term that depends on the orientation of the magnetization. Up to now, the experimental studies that demonstrate the anisotropy of the damping, focus on the role of the magnetocristalline anisotropy $9$  and have not focused on exchange-coupled systems despite the fact that they seem to be promising for high-frequency applications.<sup>10</sup> In this Rapid Communication, experimental evidence for an unidirectional damping parameter is provided for exchangecoupled ferromagnetic (F)/antiferromagnetic (AF) bilayers. The exchange coupling induces a strong anisotropic relaxation process which depends on the direction of magnetization. A frequency-dependent study of the relaxation demonstrates the anisotropic nature of the intrinsic part of the relaxation, i.e., *of the damping*. The anisotropy of the damping parameter is found to be unidirectional and directly related to the exchange field amplitude. Finally, a modified relaxation term in the equation of motion is implemented to reproduce these experimental evidences.

The investigated  $NiO(t_{AF})/Py(20 \text{ nm})$  (where Py denotes  $Ni<sub>81</sub>Fe<sub>19</sub>$ ) bilayers have been grown on Si(100) substrate by conventional rf diode sputtering, under a static magnetic field

 $(H<sub>D</sub>)$  of 300 Oe to induce an uniaxial anisotropy in the Py layer. The base pressure prior to the film deposition was typically  $10^{-7}$  mbar. The NiO thicknesses were  $t_{AF}=0$ , 20, 30, 50, 67.5, and 75 nm. A shifted hysteresis loop, at room temperature, along the depositing field axis, is observed for  $t_{AF}$  higher than 20 nm. In order to precisely determine the relaxation parameters, the frequency dependence of the ferromagnetic resonance (FMR) spectra was investigated within a range of microwave frequencies between 6 and 12.5 GHz using a wideband resonance spectrometer with a nonresonant microstrip line.<sup>11</sup> The FMR is measured via the derivative of the microwave power absorption  $(dP/dH)$  using a small rf exciting field. Resonance spectra were recorded with the applied static magnetic field oriented in plane at an angle  $\varphi$ relative to the depositing field. Moreover our experimental device allows the use of positive and negative polarities of field sweeps,  $(H<sup>+</sup>, H<sup>−</sup>)$ . Static magnetic properties were measured at room temperature by vibrating sample magnetometer (VSM).

Figure [1](#page-0-0) shows the FMR absorption derivative and absorption profiles for the Py thin film and the  $NiO(67.5)$ nm)/Py bilayer measured at 9 and 5 GHz. The presence of

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FIG. 1. (Color online) An example of (a) FMR absorption derivative spectrum and (b) integrated spectrum for a  $Si(100)/Py$  $(20nm)$  film [green (light) line] and a Si $(100)/NiO(67.5$  nm)/Py  $(20$ nm) bilayer [blue (dark) line]. Measurements are performed with positive  $(H^+)$  and negative  $(H^-)$  electromagnet polarity.



FIG. 2. (Color online) Azimuthal angular dependencies for positive (red open circles) and negative (black open squares) applied field polarity of (a) the experimental and simulated (solid line) FMR resonance field (b) the measured FMR linewidth and (c) the measured magnitude of the resonance curve with the integer form, at 9.5 GHz for sample Si(100)/NiO(67.5 nm)/Py. (d) Experimental measurements of the asymmetry ratio, AS, defined in Eq. ([1](#page-1-1)) for a Si(100)/NiO(67.5 nm)/Py bilayer (open triangles). Solid line simulation of AS with  $\xi = 0.18$  [Eq. ([2](#page-2-0))] (e) Simulation of the asymmetry ration AS for  $\xi=0$  (dash line) and  $\xi \neq 0$  (solid line).

the unidirectional anisotropy induces displacement of the resonance field  $(H_{res})$  and an enhancement of the linewidth  $(\Delta H_{pp})$ , as expected in exchange-coupled systems.<sup>12</sup> What is more unusual is that the width and intensity of the bilayer resonance spectra depend on the field polarity, i.e., there is a strong asymmetry of the resonance line with respect to *H*  $=0$  Oe. Such an asymmetry with respect to the field sweeping polarity has previously been reported but not discussed.<sup>13</sup> To achieve an understanding of this pronounced asymmetry, we have systematically measured the FMR spectra for different applied field directions  $(\varphi)$ . For each angular position within the plane and for positive and negative field polarities, the following fundamental FMR quantities were extracted:  $H_{res}$ ,  $\Delta H_{pp}$  and the maximum of the absorption intensity, *I*, as shown in [Figs.  $2(a)-2(c)$  $2(a)-2(c)$ ]. The  $H_{res}$  angular dependence reflects the symmetry of our exchange bias system. For each polarity, the shapes are symmetric with respect to the exchange bias field direction (i.e.,  $\varphi = 180^{\circ}$ ) with two minima and two maxima, as previously expected in unidirectional systems<sup>14</sup> [Fig. [2](#page-1-0)(a)]. They present a similar angular dependence, i.e., symmetry axes along the exchange bias field direction. In order to understand *I* and  $\Delta H_{pp}$  properties, one

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should consider the origin of the absorbed microwave power in a RFM experiment. For a given azimuthal angle, the absorbed microwave power is proportional to the imaginary part of the susceptibility tensor component along the exciting field:  $P \sim \chi_h^{n.15}$  $P \sim \chi_h^{n.15}$  $P \sim \chi_h^{n.15}$  We quantitatively examine the asymmetry with respect to the field polarity using an asymmetry parameter (AS) defined as follow:

$$
AS = \frac{I^+ - I^-}{I^+ + I^-} = \frac{\chi_{h \max}^{\prime\prime +} - \chi_{h \max}^{\prime\prime -}}{\chi_{h \max}^{\prime\prime +} + \chi_{h \max}^{\prime\prime -}},
$$
(1)

<span id="page-1-1"></span>where  $I^+$  and  $I^-$  are the microwave power absorption maximal values at resonance fields for positive and negative values of resonance fields as shown in Fig. [1.](#page-0-0) For a single Py film and for all azimuthal angles, the FMR spectra are symmetric with respect to the applied field polarity, i.e.,  $I^+=I^$ and  $AS = 0$  (not shown in Fig. [2](#page-1-0)). In the presence of the exchange bias, the angular dependence of AS presents a complex behavior as shown in Fig.  $2(d)$  $2(d)$ . In the following, we study this behavior considering the magnetic susceptibility which may be calculated with Landau-Lifshitz-Gilbert  $(LLG)$  equation [Eq.  $(5.3)$  in Ref.  $6$ ] where the effective field is defined as  $H_{eff} = \partial F / \partial M$ . The free-energy functional  $F(M)$ consists, in our case, of the Zeeman energy, the unidirectional anisotropy of the AF layer, the demagnetizing field energy, the saturation magnetization, and the uniaxial anisotropy of the F layer.<sup>14</sup> In addition one can assume that the precession around the *Heff* is of small amplitude so the LLG equation may be linearized.<sup>16</sup> This allows us to calculate the theoretical FMR spectra and thus the theoretical AS parameter. In a first step we have fitted the resonance field angular dependence as shown in Fig.  $2(a)$  $2(a)$ , it allows us to determine the values of the exchange field  $(H_e)$ , F anisotropy field  $(H_K)$ , and the saturation magnetization  $(M_S)$  (i.e.,  $H_e$  $=30$  Oe,  $H_K = 34$  Oe,  $M_S = 10900$  G, and the gyromagnetic ratio  $\gamma = 1.843 \times 10^7 \text{ s}^{-1} \text{ Oe}^{-1}$ . The value of  $H_e$  obtained from the fit is about 30% lower than the one obtained by VSM for all our exchange biased samples as previously observed in such systems.<sup>14</sup> In second step, we have simulated the angular dependence of AS directly by supposing that the Gilbert damping coefficient  $\alpha$  has a constant value and does not depend on the angle between the magnetization and the exciting field as generally admitted in azimuthal measurements[.17](#page-3-17) The simulated curves reproduce the general shape of AS  $[Fig. 2(e)]$  $[Fig. 2(e)]$  $[Fig. 2(e)]$  but not the experimental features along the exchange bias field axis, i.e., for  $\varphi=0^{\circ}$  and  $\varphi$ =180°. Following the linearized LLG model including an isotropic damping, the asymmetry should not be present along this axis. Therefore, we have considered a nonisotropic damping for reproducing the AS in the following manner.

According to Steiauf and Fähnle, $\frac{7}{1}$  in the presence of spinorbit interaction the damping parameter is described by a matrix and will depend on the direction of the magnetization. Thus, the spin-orbit coupling makes the spin degree of freedom respond to its orbital environment. The authors show that there are at most two eigenvalues for the damping matrix:  $\alpha(M)$ , which depends rather sensitively on the orientation of *M* in the crystal. In our case the exchange bias breaks the symmetry of uniaxial anisotropy with respect to the an-

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FIG. 3. Evolution of the peak-to-peak linewidth  $\Delta H_{pp}$  as a function of frequency for sample  $NiO(67.5 \text{ nm})$ /Py with the static field is applied with angle  $\varphi = 90^\circ$  (circles) and  $0^\circ$  (squares) with positive (full symbols) and negative polarity (open symbols).

isotropy axis, and therefore our measurements show different damping in directions of  $\varphi=0^{\circ}$  and  $\varphi=180^{\circ}$  thus supporting the theoretical prediction of the authors mentioned above. To reproduce the experimental features of the resonance absorption, we modify the LLG equation by assuming a magnetization direction-dependent damping parameter that lies between two eigenvalues of damping parameter given by  $\alpha^+$  $=\alpha(1-\xi)$  and  $\alpha^-=\alpha(1+\xi)$ . Thus,

$$
\frac{d\mathbf{M}}{dt} = -\gamma (\mathbf{M} \times H_{eff}) + \frac{1}{M} \mathbf{M} \times \alpha^* \frac{d\mathbf{M}}{dt},
$$
 (2)

<span id="page-2-1"></span><span id="page-2-0"></span>where

$$
\alpha^* = \alpha \left[ 1 - \xi \cdot \frac{He\mathbf{M}}{HeM} \right].
$$
 (3)

The first term on the right-hand side of Eq.  $(2)$  $(2)$  $(2)$  describes the precession of  $M$  around  $H_{eff}$  and the second term represents the damping torque and is the key parameter that governs the relaxation toward equilibrium. The term  $\alpha^*$  represents the anisotropic damping that depends on the direction of the magnetization retained (withheld) by AF spins via exchange coupling  $(H_e)$ . In Eq. ([3](#page-2-1))  $\alpha$  is the mean Gilbert phenomenological damping parameter and the term  $\xi$  is the difference between the measured damping parameters along the unidirectional anisotropy for the positive  $(\alpha^+)$  and negative  $(\alpha^-)$ polarities [i.e.,  $\xi = (\alpha^- - \alpha^+) / (\alpha^- + \alpha^+)$ ]. The best fit to the experimental data was obtained for  $\xi = 0.18$ . The good agreement between theoretical and experimental values [Fig.  $2(d)$  $2(d)$ ] confirms our assumption, given by Eq.  $(3)$  $(3)$  $(3)$ , reflecting the damping parameter dependence on the magnetization direction.

As relaxation mechanisms may include intrinsic and extrinsic contributions, $18$  one needs to separate these effects to show unambiguously that the origin of the AS is intrinsic. In the parallel FMR configuration, with the magnetization parallel to the applied field, the time derivative  $\partial M / \partial t$  Gilbert term in the equation of motion produces a FMR linewidth linear with the microwave frequency *f*. However, in many magnetic systems, while a linear behavior is observed, the linewidth fails to extrapolate to zero with vanishing frequency. This zero-frequency contribution  $\Delta H_0$  to the line-

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FIG. 4. (Color online) Exchange bias dependence, in  $Si(100)/NiO(t_{AF})/Py(20 \text{ nm})$  with  $t_{AF}=20, 30, 50, 67.5,$  and 75 nm, of (a) the asymmetry AS of the susceptibility intensity (b) the zero-frequency peak-to-peak linewidth  $\Delta H_0$  with negative (full black squares) and positive (red open circles) polarity (c) the parameter  $\xi$  [see text and Eq. ([3](#page-2-1))]. All the parameters were measured along the F easy axis  $(\varphi = 0^{\circ})$ 

width is related to extrinsic mechanism and reflects the effect of inhomogeneity on the linewidth. Thus, frequencydependent studies provide intrinsic and extrinsic contributions to the relaxation. The field-swept linewidth, in a given direction, may be written as  $8$ 

$$
\Delta H_{pp} = \Delta H_0 + \frac{2}{\sqrt{3}} \frac{\alpha}{\gamma} 2 \pi f,\tag{4}
$$

<span id="page-2-2"></span>where  $\Delta H_0$  is the inhomogeneous broadening and f is the microwave frequency. The second term is the Gilbert contribution that represents intrinsic contribution. In our study, it should be noted the damping parameter  $\alpha$  should be replaced by  $\alpha^*$  in Eq. ([4](#page-2-2)).

Figure [3](#page-2-3) shows the dependence of  $\Delta H_{pp}$  on the microwave frequency for both angle  $\varphi$  values  $0^{\circ}$  and  $90^{\circ}$  with positive and negative polarities. The FMR linewidth is linearly dependent on the microwave frequency for both orientations, as previously observed in such systems $^{19}$  and as ex-pected from Eq. ([4](#page-2-2)). The inversion of the field polarity along the exchange field axis ( $\varphi = 0^{\circ}$ ) induces a change in the slope of the FMR linewidth. It confirms the dependence of the intrinsic contribution toward the field polarity as well as the intrinsic origin of the AS. Furthermore, the intercept at the origin of the FMR linewidth slopes, i.e.,  $\Delta H_0$  give the same value by inversion of the polarity for  $\varphi = 0^{\circ}$ . This shows unambiguously that the polarity inversion of the applied magnetic field only modifies the intrinsic contribution to the linewidth. It should also be noted that the  $\Delta H_{pp}$  frequency dependence does not depend on the polarity along  $\varphi = 90^\circ$ .

To illuminate the influence of the exchange coupling on the anisotropic nature of the damping, Py films were deposited on NiO underlayers with different thicknesses resulting in a large variety of exchange bias field values from 0 to 60 Oe. Figure  $4(a)$  $4(a)$  shows the influence of the exchange field value on the asymmetry of the linewidth measured along the F easy axis. It shows clearly that the condition for the appearance of AS requires a NiO film thicker than a critical thickness, above which  $H_e \neq 0.^{20}$  The asymmetry is then proportional to  $H_e$ . To confirm the intrinsic origin of AS, intrinsic and extrinsic contributions were obtained for each samples. Figure  $4(b)$  $4(b)$  shows the dependence of  $\Delta H_0$  on the  $H_e$ and for both polarities. The presence of a thin NiO layer increases  $\Delta H_0$  relatively to the value obtained for the Py single layer. However,  $\Delta H_0$  shows no significant dependence on *He* and does not depend on field polarity. Therefore, the extrinsic contribution does not contribute to the AS mechanism. The influence of  $H_e$  on the intrinsic damping anisotropy is depicted in Fig.  $4(c)$  $4(c)$ . Under the critical thickness, the value of  $\alpha$  (i.e.,  $\xi = 0$ ) remains unchanged by inversion of the

polarity. However,  $\xi$  increases with  $H_e$ , for a greater NiO thickness. It shows unambiguously that the anisotropy in the intrinsic Gilbert damping originates from the pinned AF spins and is proportional to *He*.

In conclusion, we report on experimental evidence for an anisotropic unidirectional intrinsic Gilbert damping term in exchange-coupled systems. A modified damping term in the LLG equation of motion is introduced to reproduce this anisotropy. It depends on the direction between the magnetization and the unidirectional anisotropy. We also show that the magnitude of the damping anisotropy is directly related to the *He* magnitude. This anisotropy should be considered in the fundamental understanding of the dynamic of exchangecoupled systems and would be great importance in lowenergy storage applications or high-frequencies applications.

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<span id="page-3-0"></span>\*jamal.ben-youssef@univ-brest.fr

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