Rotatable magnetic anisotropy in a $Fe_{0.8}Ga_{0.2}$ thin film with stripe domains: Dynamics versus statics

S. Tacchi,[1] S. Fin,[2] G. Carlotti,[3,4] G. Gubbiotti,[1] M. Madami,[3] M. Barturen,[5,6,*] M. Marangolo,[5,6] M. Eddrief,[5,6] D. Bisero,[2] A. Rettori,[4,7] and M. G. Pini[8]

[1]*Istituto Officina dei Materiali del CNR (CNR-IOM), Unità di Perugia, c/o Dipartimento di Fisica, Università di Perugia, I-06123 Perugia, Italy*

[2]*CNISM and Dipartimento di Fisica, Università di Ferrara, Ferrara, Italy*

[3]*Dipartimento di Fisica, Università di Perugia, I-06123 Perugia, Italy*

[4]*Centro S3, c/o Istituto Nanoscienze del CNR (CNR-NANO), I-41125 Modena, Italy*

[5]*Sorbonne Universites, UPMC Univ Paris 06, UMR 7588, INSP, 4 place Jussieu, F-75005, Paris, France*

[6]*CNRS, UMR 7588, Institut des NanoSciences de Paris, 4 place Jussieu, F-75005, Paris, France*

[7]*Dipartimento di Fisica e Astronomia, Università di Firenze, I-50019 Sesto Fiorentino (FI), Italy*

[8]*Istituto dei Sistemi Complessi del CNR (CNR-ISC), Unità di Firenze, I-50019 Sesto Fiorentino (FI), Italy*

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A comprehensive investigation of rotatable anisotropy in a $Fe_{0.8}Ga_{0.2}$ thin film with a stripe domain structure has been performed comparing static and dynamic measurements. The stripes' domain formation and their rotation under a transverse magnetic field have been imaged by magnetic force microscopy. The rotatable anisotropy field H_{rot} was determined by fitting the frequency evolution of the dipole-dominated magnetostatic spin-wave mode versus the in-plane orientation of the stripe domains, measured by Brillouin light scattering in the absence of any dc or ac magnetic field. We obtained $H_{rot} \approx 1.35$ kOe, which is nearly ten times larger than the crystallographic in-plane anisotropy field. By applying a dc magnetic field along the stripes' axis, H_{rot} decreases, and eventually vanishes for saturated in-plane magnetization. At remanence, we established a quantitative relationship between static and dynamic properties, that is, the stripes' rotation angle and the in-plane angle dependence of spin-wave frequency.

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I. INTRODUCTION

The concept of rotatable (i.e., direction-independent) anisotropy is ubiquitous in the magnetism of thin films. It was introduced decades ago in the study of magnetostrictive Ni, Fe, and NiFe films [1–4] with perpendicular magnetic anisotropy (PMA) and, subsequently, of polycrystalline exchange-biased ferromagnetic-antiferromagnetic bilayers [5,6], polycrystalline NiFe films [7], amorphous CoZrTa films [8], ferrite-doped CoFe films [9], and permalloy films [10]. In all these systems, an unexpected isotropic shift has been observed in the ferromagnetic resonance (FMR) field [5,7–10], or in the Brillouin light scattering (BLS) frequency of spin waves [6]. In order to explain such experimental evidence in the framework of a model with a uniform in-plane magnetization, it was necessary to invoke a rotatable anisotropy field H_{rot}. Some authors provided just an operative definition for H_{rot}, e.g., Refs. [9,10], as the difference between H_k^{dyn}, the uniaxial anisotropy field obtained from dynamic measurements, and H_k^{stat}, the anisotropy field obtained from static measurements. Others [7] tried to reconcile dynamics and statics by conjecturing a transient nature for H_{rot}. However, it is fundamental to note that a uniform, collinear configuration is not the true ground state in any of the above-mentioned systems. The case of thin films with a PMA of magnetoelastic origin [1–4,11] is paradigmatic. In such films, the PMA may take place in the presence of a negative (positive) magnetostriction constant combined with a planar tensile (compressive) stress. The out-of-plane energy density K_n competes against the dipolar one, $2\pi M_s^2$ (M_s saturation magnetization), which favors the film plane. For a moderate value of the quality ratio, $Q = \frac{K_n}{2\pi M_s^2} \leqslant 1$, a ground state with a stripe domain structure [4,11–14] is favored, provided that the film thickness is greater than a critical value. The perpendicular magnetization component has a periodic modulation, alternately upwards and downwards as the stripes are crossed. Even though the magnetization is along the stripes' axis, it does *not* have a preferential in-plane direction (as is the case in a film with conventional uniaxial anisotropy), because the stripe orientation is not fixed, but depends on sample history. In fact, the whole stripe pattern rotates [4] parallel to the last saturation field, hence, the attributes "rotatable" or "pseudouniaxial" [11] given to H_{rot} in such films. An important contribution to the comprehension of the phenomenon came from the authors in Ref. [11], in the study of $Fe_{1-x}Si_x$ thin films, with a PMA which induces a stripe domain pattern. Measuring the transversely biased initial susceptibility via the transverse magneto-optic Kerr effect (TMOKE), they found it independent of frequency for $\nu \leqslant 1$ kHz. Thus, they proposed a quasistatic one-dimensional (1D) model of the stripes by which they obtained an explicit, field-dependent expression for H_{rot} (i.e., H_{rot} decreases with increasing the intensity H of a magnetic field applied along the stripes' axis, and eventually vanishes when H becomes strong enough to set a uniform in-plane magnetization). From this expression [11], it results that the rotatable anisotropy field associated with the stripe domain pattern cannot be measured by means of conventional magnetometry techniques, based on the balance

*Also at CNEA-CONICET, Centro Atómico Bariloche, R8402AGP San Carlos de Bariloche, Río Negro, Argentina.

of torques, leading to an equilibrium magnetization state (e.g., longitudinal MOKE hysteresis loop). Rather, one can use any (quasistatic or dynamic) experimental technique capable of exerting a *nonzero* torque on the film magnetization [e.g., TMOKE [11], rotational MOKE (ROTMOKE) [15], FMR [5,9,10], BLS [6,16]].

In this paper, we aim at performing a comprehensive study of rotatable anisotropy in a $Fe_{0.8}Ga_{0.2}$ film [17–20], 65 nm thick. For such a system, a positive magnetostriction constant, combined with a compressive planar strain, is responsible for the onset of PMA, and consequently of a stripe domain structure [12,13], for film thickness t greater than a critical value $t_{cr} \approx 35$ [17]. Magnetic force microscopy (MFM) and BLS have been simultaneously exploited to achieve a comparison between the static and the dynamic properties of the stripe domain configuration. MFM has been used to analyze both the formation of the stripes as a function of the intensity of the external applied field, and their rotation under a transverse field. The dynamic response of the film has been investigated by BLS, following the evolution of the spin-wave modes during the evolution of the magnetic ground state.

II. RESULTS AND DISCUSSION

The film analyzed here was grown by coevaporation from independent Fe and Ga Knudsen cell sources on ZnSe/GaAs(001) substrates of a molecular beam epitaxy chamber, as explained in previous work [18]. The thickness $t = 65$ nm, and the atomic Ga concentration $x = 0.2$, were determined by means of x-ray reflectivity and x-ray photoelectron spectroscopy, respectively. The longitudinal hysteresis loop, measured both by MOKE and a vibrating sample magnetometer (VSM), is reported in Fig. 1(a). When the magnetic field is reduced from positive saturation, we observed a linear reversible region, starting around $H \approx 1500$ Oe, typical of materials with sizable PMA [17], suggesting the formation of stripe domains. As it can be seen, in the MOKE loop, which is relative to the upper part (about 10 nm) of the film, the remanent magnetization $M_r/M_s \simeq 0.51$ is higher than that measured in the VSM one, $M_r/M_s \simeq 0.31$, suggesting the presence of regions in-plane magnetized at the sample surface. MFM images of domains were recorded by a Digital Instruments Nanoscope IIIa, using the phase detection mode. Commercially available ferromagnetic CoCr tips were used. We observed [Fig. 1(c)] a well-defined pattern of stripe domains aligned along the direction of the last saturation field. From the fast Fourier transform of the images, we found that the period of the pattern is about (90 ± 5) nm, whatever the stripe orientation, and it remains constant when the field is reduced from 800 to 0 Oe. Very interestingly, we found that when an increasing field H_{trans} is applied in plane along the direction perpendicular to the stripes, the whole stripe structure remains unperturbed up to $H_{trans} \approx 400$ Oe. For higher fields, the stripes start to coherently rotate towards the direction of the applied field [Figs. 1(d)–1(g)] until a complete reorientation is attained for $H_{trans} \approx 800$ Oe [Fig. 1(h)]. From Fig. 1(i) one deduces that this reorientation is irreversible since, when the field is removed, the stripe structure is preserved [21]. To understand the mechanism of stripe formation, the hysteresis cycle has been simulated by the LLG micromagnetic package [22]. In the simulations, the saturation magnetization was set to the value obtained from the VSM measurements $4\pi M_s = 17580$ Oe; the exchange constant $A_{ex} = 1.6 \times 10^{-6}$ ergs/cm, the out-of-plane anisotropy constant $K_n = 4.2 \times 10^6$ ergs/cm^3, and the tetragonal anisotropy $K_1 = -0.8 \times 10^5$ ergs/cm^3, favoring the [110] and [1-10] directions, were obtained by the fit of BLS and FMR measurements in a high magnetic field, when the magnetization is saturated in plane (not shown here). A mesh size of $5 \times 5 \times 5$ nm^3 was used in the framework of in-plane periodic boundary conditions. The calculated hysteresis loops for the whole sample and the top 10 nm thick layer are shown in Fig. 1(b), while the magnetization distribution at remanence is sketched in Fig. 1(j). We found basic domains, where the magnetization points alternately up and down with respect to the film surface. This results in a simulated MFM image characterized by domains, with a period of about 100 nm [Fig. 1(k)], in good agreement with the MFM analysis. Due to the moderate value of $Q = 0.34$, the closure domains consist of regions where the magnetization vector rotates around the longitudinal direction of the stripe domains following concentric semicircular paths [23,24]. The adjacent closure domains are coupled through surface regions, where the magnetization is directed in plane along the stripes' axis. Due to the presence of the closure domains that are in-plane magnetized, in the simulated hysteresis loops the remanent magnetization is higher for the surface region $M_r/M_s \simeq 0.41$ than for bulk $M_r/M_s \simeq 0.31$ [Fig. 1(b)], in qualitative agreement with the measured MOKE and VSM loops [Fig. 1(a)].

BLS measurements were performed in backscattering geometry focusing about 200 mW of monochromatic light (532 nm wavelength) onto the sample surface. The backscattered light was analyzed by a Sandercock-type (3 + 3)-pass Fabry-Pérot interferometer [25]. The incidence angle of light was 15°, corresponding to an in-plane transferred wave vector $q_\parallel = 0.61 \times 10^5$ cm^{-1}. A peculiarity of the BLS technique is that the finite in-plane transferred wave vector determines the spin-wave propagation direction with respect to the stripes' axis even in the absence of a dc bias field. An ac sensing field is not required either, because BLS probes the spontaneous, thermally excited transversal fluctuations of the magnetization. In a first set of measurements, the sample was initially saturated applying a strong magnetic field ($H = 3$ kOe) along the [110] axis, and then removing it, in order to obtain a stripe structure along that direction. BLS measurements were performed at remanence rotating the sample around the film normal, i.e., varying the angle ϕ_q between the in-plane transferred wave vector $\mathbf{q}_\parallel$ and the [110] axis (see the inset in Fig. 2). The measured frequencies, together with two typical spectra, are reported in Fig. 2. We observed two modes having a constant frequency (green and blue solid points), which correspond to perpendicular standing spin-wave modes (nPSSW) characterized by n nodes of the magnetization oscillation through the film thickness [26]. In addition, we observed a spin-wave mode (red points), whose frequency is characterized by a 180° symmetry. Since micromagnetic simulations and MOKE measurements indicated that at remanence there is a noticeable component of the in-plane magnetization directed along the stripes' axis, we interpret the above oscillating frequency

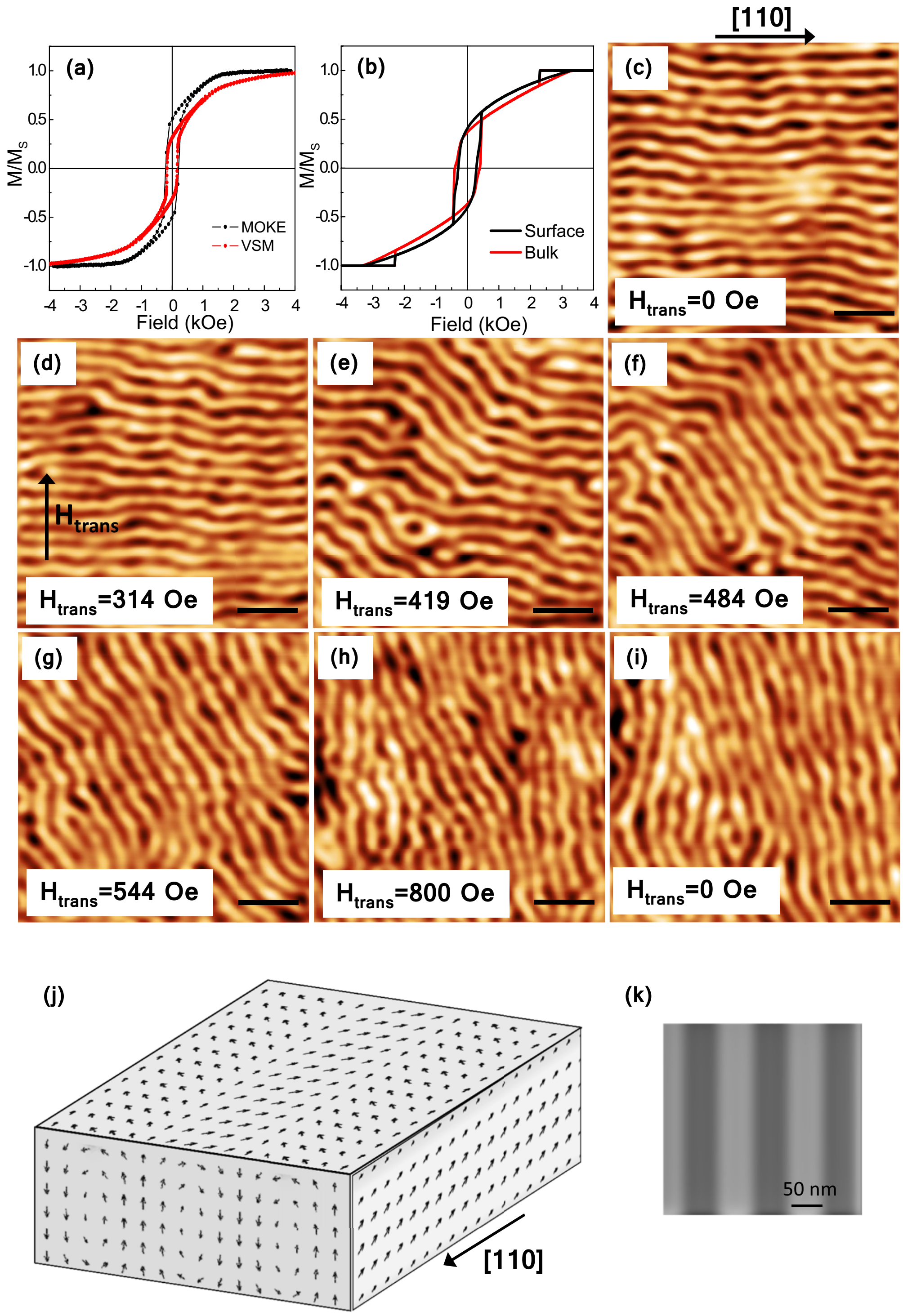

FIG. 1. (Color online) (a) Longitudinal hysteresis loop measured by MOKE and VSM. (b) Simulated hysteresis loop for sample surface (10 nm) and bulk. (c) MFM image taken at remanence, after saturation by a magnetic field along the [110] direction. (d)–(h) Images recorded in the presence of an in-plane transversal magnetic field H_{trans}. (i) When the external field is removed, the stripes conserve the new orientation. Scale bar: 300 nm. (j) Simulated domain configuration at remanence and (k) the corresponding MFM image.

mode as a dipolar-dominated magnetostatic mode (MW), having the maximum spin precession amplitude in the surface region where the magnetization is directed in plane, parallel to the stripes' axis. When $\phi_q = 90^\circ$ ($\phi_q = 0^\circ$), the MW mode propagates with $q_\parallel$ perpendicular (parallel) to the component of the magnetization directed along the stripes' axis, i.e., in the magnetostatic surface wave (MSSW) [backward volume magnetostatic spin wave (BVMSW)] configuration. As expected for a mode mainly localized in the surface region [25], the MW mode exhibits a marked Stokes/anti-Stokes

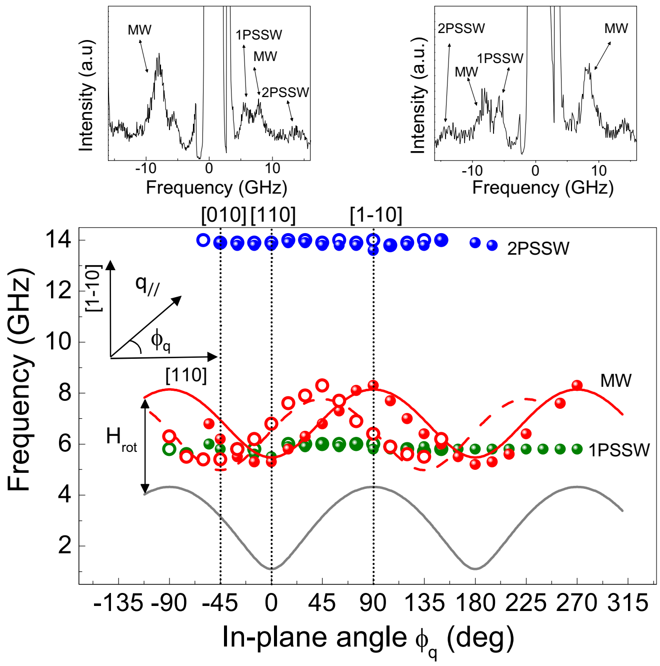

FIG. 2. (Color online) Top: BLS spectra at remanence, showing marked Stokes/anti-Stokes asymmetry when reversing the direction of the magnetic field ($H = 3$ kOe) used to prepare the stripes along [110]. Bottom: Spin-wave frequencies measured by BLS (points) at remanence as a function of the in-plane rotation angle. Red lines are the fit of the MW mode calculated using Eq. (1). The gray line is obtained setting $H_{\rm rot} = 0$ in Eq. (1). Solid (open) points denote BLS data measured by preparing stripes along [110] ([010]).

asymmetry (top of Fig. 2), which can be reversed by simply reversing the direction of the magnetic field used to prepare the stripe structure. When the stripes are prepared along other directions, the frequency of the nPSSW modes remains angle independent, while the MW frequency is rigidly shifted; in Fig. 2 one can see that when the stripes are prepared along the [010] direction (open points), the MW mode has a maximum along the [100] direction ($\phi_q = 45°$), where $q_\parallel$ is perpendicular to the stripes. This proves the rotatable and pseudouniaxial character of the observed anisotropy.

To easily establish a relationship between BLS and MFM measurements, we exploited an approximate analytic expression, valid for thin films with a uniform in-plane magnetization [25,27],

$$\left(\frac{\omega_{\rm MW}}{\gamma}\right)^2 = \left[H_{\rm rot} - \frac{K_1}{M_s}\cos^2 2\phi + 4\pi M_s D\left(1 - \frac{q_\parallel t}{2}\right) - \frac{2K_n}{M_s} + \frac{2A_{\rm ex}}{M_s}q_\parallel^2\right]\left[H_{\rm rot} - \frac{2K_1}{M_s}\cos 4\phi + 2\pi M_s D q_\parallel t \sin^2(\phi - \phi_q) + \frac{2A_{\rm ex}}{M_s}q_\parallel^2\right], \quad (1)$$

where ϕ (ϕ_q) is the angle formed by the in-plane magnetization (the transferred in-plane wave vector $q_\parallel$) with the [110] direction, and D is the demagnetizing factor. Owing to the presence of stripes, $H_{\rm rot}$ and D are phenomenologically introduced as fit parameters [28].

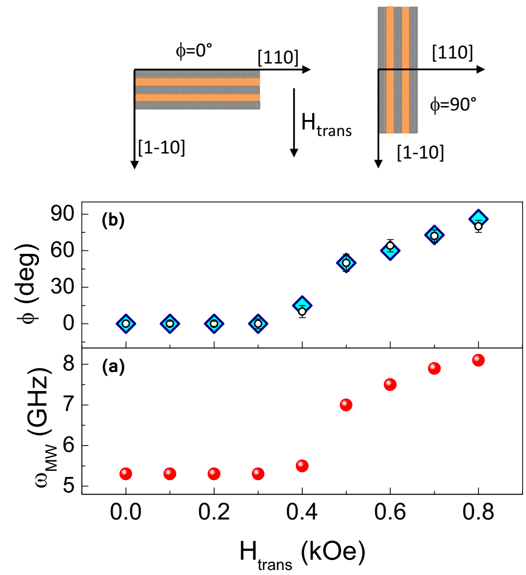

FIG. 3. (Color online) (a) Frequency of the MW mode measured by BLS at remanence after preparing the stripes along [110]. A transversal magnetic field $H_{\rm trans}$ was applied in plane perpendicularly to the stripes' axis, and then removed. (b) Angle between the stripes' axis and the [110] direction, obtained from the fit of $\omega_{\rm MW}$ using Eq. (1) (turquoise symbols) and from MFM data (open points).

Using the same parameters exploited in the micromagnetic simulations, we obtain $H_{\rm rot} = 1350$ Oe, nearly ten times larger than $H_1 = \frac{2|K_1|}{M_s} = 114$ Oe (the effective field associated with in-plane anisotropy), and $D = 0.51$, almost equal to the ratio M_r/M_s measured by MOKE. This substantiates the surface character of the MW mode. Note that $H_{\rm rot} > 0$ is crucial to get a good fit of the MW frequency: The calculated (red) curve of Fig. 2 would be downshifted (gray curve) by more than 3 GHz in the absence of $H_{\rm rot}$, resulting in a relevant disagreement with the experimental data. As a second step of our investigation, we performed BLS measurements which can be directly compared with the rotation of the stripe pattern observed in MFM images. The sample was first saturated by applying a strong magnetic field of 3 kOe along the [110] direction, and then the field was removed in order to nucleate the stripe structure along that direction. Successively, a transverse magnetic field $H_{\rm trans} =$ 100 Oe was applied along the [1-10] axis (i.e., along an in-plane direction perpendicular to the stripes), then the field was removed, and the spin-wave frequency was measured at remanence. The process was repeated, in steps of 100 Oe, up to $H_{\rm trans} = 800$ Oe. As seen in Fig. 3(a), the frequency of the MW mode remains constant up to about 400 Oe. When $H_{\rm trans} >$ 400 Oe the frequency starts to increase, reflecting the rotation imaged by MFM images, and reaches the maximum value for $H_{\rm trans} = 800$ Oe, suggesting that the stripe reorientation is completed. From a best fit of the measured frequency of the MW mode to the values calculated using Eq. (1), we derived the angle between the stripes' axis and the [110] direction. For $H_{\rm trans} > 400$ Oe, the angle exhibits a gradual increase, which is in quantitative agreement with the observed reorientation of the stripes estimated from MFM images [Fig. 3(b)]. Finally, we studied the field dependence of $H_{\rm rot}$, analyzing the dependence of $\omega_{\rm MW}$ (Fig. 4, left axis) as a function of a magnetic field H applied along the [110] direction (parallel to the stripes'

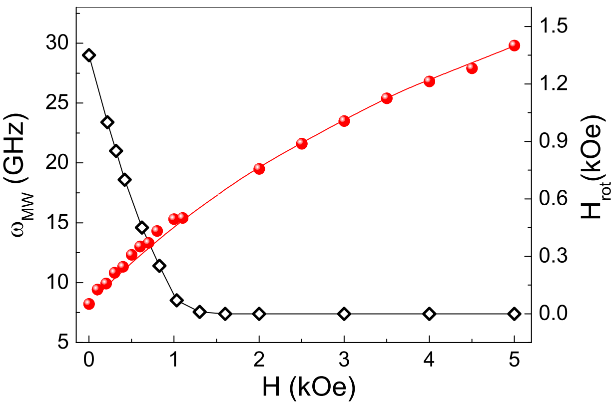

FIG. 4. (Color online) Left axis: Measured (red points) and calculated (curve) frequency of the MW mode as a function of H, applied along the stripes' axis. Right axis: Calculated field dependence of the rotatable field H_{rot} (black open diamonds). The line is a guide for the eyes.

axis). Using the ratio $M(H)/M_s$ measured by MOKE as an approximation of the value of D at different H, we fitted the field dependence of H_{rot} (Fig. 4). H_{rot} presents an almost linear decrease on H, and is found to vanish for fields strong enough to saturate the magnetization in plane [10,11].

III. CONCLUSIONS

In conclusion, we have investigated an epitaxial $Fe_{0.8}Ga_{0.2}$ thin film characterized by a magnetic stripe pattern. The ground state configuration was accounted for by fully three-dimensional micromagnetic simulations, and its evolution with the external magnetic field intensity and direction was studied by MFM. The dynamical properties of the film were analyzed studying spin waves with a finite in-plane wave vector, propagating at different angles with respect to the stripes, by means of the BLS technique, with no need for a sensing field. The rotatable field H_{rot} was determined at remanence (where it has its maximum value) from the BLS data. Moreover, applying an increasing dc magnetic field H along the stripes' axis, H_{rot} was found to decrease and eventually vanish as the magnetization becomes saturated in plane. Lastly, a quantitative relationship between the measured BLS frequency and MFM observations of the stripe rotation angle under a transverse field was established, providing a significant step towards the understanding of the relationship between dynamic and static measurements of the anisotropy in such paradigmatic systems.

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