

Simultaneous existence of two spin-wave modes in ultrathin Fe/GaAs(001) films studied by Brillouin light scattering: Experiment and theory

M. G. Pini,^{1,2,*} P. Politi,^{1,2} A. Rettori,^{2,3} G. Carlotti,^{4,5} G. Gubbiotti,⁵ M. Madami,^{4,5} and S. Tacchi^{4,5}

¹Istituto di Fisica Applicata "N. Carrara", CNR, Via Madonna del Piano, I-50019 Sesto Fiorentino, Italy

²INFN, Unità di Firenze, Via G. Sansone 1, I-50019 Sesto Fiorentino, Italy

³Dipartimento di Fisica, Università di Firenze, Via G. Sansone 1, I-50019 Sesto Fiorentino, Italy

⁴Dipartimento di Fisica, Università di Perugia, Via Pascoli, I-06123 Perugia, Italy

⁵INFN, Unità di Perugia, Via Pascoli, I-06123 Perugia, Italy

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A double-peaked structure was observed in the *in situ* Brillouin Light Scattering (BLS) spectra of a 6 Å thick epitaxial Fe/GaAs(001) film for values of an external magnetic field H , applied along the hard in plane direction, lower than a critical value $H_c \approx 0.9$ kOe. This experimental finding is theoretically interpreted in terms of a model which assumes a nonhomogeneous magnetic ground state characterized by the presence of perpendicular up/down stripe domains. For such a ground state, two spin-wave modes, namely an acoustic and an optic mode, can exist. Upon increasing the field the magnetization tilts in the film plane, and for $H \geq H_c$ the ground state is homogeneous, thus allowing the existence of just a single spin-wave mode. The frequencies of the two spin-wave modes were calculated and successfully compared with the experimental data. The field dependence of the intensities of the corresponding two peaks that are present in the BLS spectra was also estimated, providing further support to the above-mentioned interpretation.

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I. INTRODUCTION

Ferromagnetic-semiconductor heterostructures, like ultrathin Fe/GaAs(001) films, have received considerable attention¹⁻⁴ for their potential technological applications in new magnetoelectronic devices.^{5,6} A sharp and well ordered interface, without any dead magnetic layer, can be obtained, as bcc Fe grows epitaxially on GaAs thanks to the small lattice mismatch (1.4%). Together with the expected cubic anisotropy of bulk bcc Fe, a strong in-plane uniaxial anisotropy has been found in ultrathin Fe/GaAs(001) films, resulting from the atoming bonding at the interface.⁷ The evolution of the latter anisotropy with film thickness has been quantitatively analyzed by some of us⁸ in a thorough *in situ* Brillouin scattering study of the dynamical magnetic properties of such films.

The same system Fe/GaAs(001) has now been found to display a very interesting phenomenon, for small iron thickness ($t_{\text{Fe}}=6$ Å): below a critical field $H_c \approx 0.9$ kOe *in situ* BLS spectra show a "double-peak" structure, therefore revealing the existence of two spin-wave modes for $H < H_c$. This feature is not completely new: it was already observed⁹ in Co/Au(111) films, for $t_{\text{Co}} \geq 6$ ML and $H < H_c \sim 3$ kOe. However, the novelty of our contribution is twofold. (i) The experimental observation of the double-peak structure has been done in two different samples and both upon increasing and decreasing the magnetic field. This confirms that the phenomenon can be well reproduced and rules out the possibility it may be due to metastability effects. (ii) We develop a theory which explains the field dependence of the observed spin-wave frequencies as well as the intensities of the corresponding BLS peaks.

The starting point of our theory is that the observed splitting of the spin-wave spectrum into two modes is not com-

patible with a homogeneous ground state. For this kind of system, the simplest and most natural explanation is to assume a perpendicularly magnetized up/down domain structure (for such low values of film thickness, in-plane magnetized domains are *not* energetically favored¹⁰⁻¹²). Another possibility, a two sublattice spin arrangement, can be readily disregarded because the splitting is only observable below a critical value of the field and because it is hardly applicable to an epitaxial iron film. Our assumption is therefore an up/down domain ground state. With increasing the magnetic field H , the magnetization of each domain gradually tilts in the plane and, for H greater than a critical value H_c , the ground state is homogeneously magnetized in-plane. Two branches are found for $H < H_c$ and a single (uniform) mode for $H \geq H_c$.

At present an *in situ* high resolution mapping of the magnetization is outside the reach of conventional microscopic techniques. In the absence of detailed information on the actual domain structure and for the sake of generality, we are going to assume a stripe domains structure. Such a one-dimensional model has the advantage that the frequencies of the spin-wave excitations can be easily evaluated and their field dependence can be reproduced for different in-plane directions.

The format of the paper is as follows: In Sec. II the experimental details concerning the sample preparation and the BLS technique are summarized. In Sec. III the frequencies of the spin-wave excitations with respect to a nonhomogeneous ground state with up/down stripe domains are calculated using the Landau-Lifshitz equations of motion. We also estimate the field dependence of the spin-wave intensity of the two modes in the framework of a classical macroscopic model which relates the scattered intensity to the magnetization-dependent permittivity tensor. In Sec. IV the

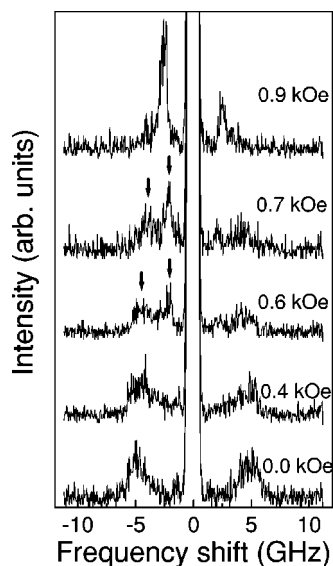


FIG. 1. *In situ* Brillouin spectra taken from a $t_{\text{Fe}}=6 \text{ \AA}$ thick epitaxial Fe/GaAs(001) film for different values of the external magnetic field H , applied within the film plane along $[-110]$, the hard in-plane direction. Two peaks, indicated by arrows, are simultaneously observed for $H=0.6$ and 0.7 kOe.

experimental results are presented and compared with the predictions of the theoretical model. Finally, in Sec. V the conclusions are drawn. Details about the calculation of the spin-wave frequencies can be found in the Appendix.

II. EXPERIMENT

Following the previous investigation of Fe/GaAs(100) films with different thickness,⁸ in this work we focus our attention on 6 Å thick Fe films, because for this particular thickness BLS spectra exhibit the double-peak structure described in the following. The reproducibility of the results was checked studying two different samples with the same nominal thickness. The two iron films were grown on a Si-doped GaAs(001) single crystal in an ultrahigh-vacuum (UHV) chamber, specially designed to allow *in situ* BLS measurements^{13,14} (background pressure 3×10^{-10} mbar) at GHOST laboratory, Perugia University.¹⁵ Details about sample preparation and structural characterization can be found in Ref. 8. About 200 mW of monochromatic p -polarized light, from a solid state laser (532 nm line), was focused onto the sample surface using a camera objective of numerical aperture 2 and focal length 50 mm. The backscattered light was analyzed by a Sandercock-type (3+3)-pass tandem Fabry-Pérot interferometer.¹⁶ The external dc magnetic field was applied parallel to the surface of the film and perpendicular to the plane of incidence of light i.e., in the so-called Damon-Eshbach geometry. BLS measurements of the spin-wave frequency were performed *in situ* at room temperature as a function of both the intensity and the in-plane direction of the applied magnetic field. Typical BLS spectra for such a film, taken with the magnetic field applied along the hard in-plane direction $[-110]$, are shown in Fig. 1. The

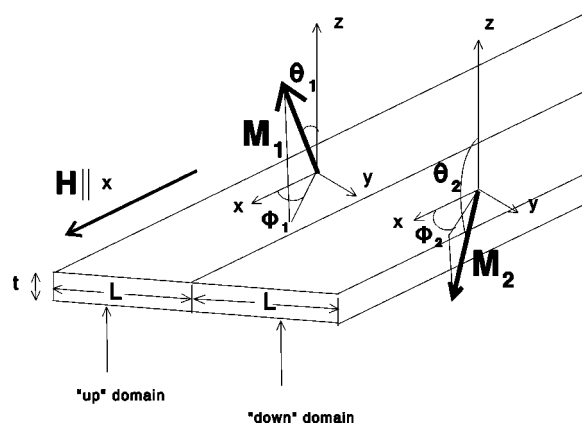


FIG. 2. One-dimensional up/down stripe domain structure and coordinate system used to define the domain variables: z denotes the normal to the film plane.

presence of a double-peak structure is evident for applied field values $0.4 < H < 0.9$ kOe. In contrast, a single peak was observed in the BLS spectra measured for magnetic field applied along both the $[100]$ and the $[110]$ directions (not shown).

III. THEORY

As discussed in the Introduction, the observed splitting of the spin-wave spectrum into two modes is not compatible with a homogeneous ground state. In the absence of experimental data about the actual spin configuration, we assume a simplified model with perpendicularly magnetized up/down domains in the shape of parallel stripes of infinite length along the in-plane field direction and with vanishing thickness of the wall between opposite domains (see Fig. 2). While more complicated up/down (or possibly canted) domain patterns cannot in principle be ruled out, the former one-dimensional model for the nonhomogeneous ground state presents the advantage that the frequencies of the spin-wave excitations can be easily evaluated using the theory of domain mode ferromagnetic resonance (DMFMR),^{17–20} and analytical results can be obtained for field applied in plane along high symmetry directions. Such a simple model turns out to be a useful tool to reproduce the spin-wave behavior; however, one should not expect it to be entirely realistic or able to account for other properties of the system, like the domain wall structure, the spatial dependence of the demagnetization factors, as well as the domain width and its field dependence.

The free energy per unit volume of the system in Fig. 2 is given by the sum of various contributions

$$G = G_H + G_{\text{dip}} + G_{2\perp} + G_{2\parallel}, \quad (1)$$

where G_H is the Zeeman energy term due to the external field, G_{dip} is the term due to demagnetization, while $G_{2\perp}$ and $G_{2\parallel}$ are the terms due to uniaxial out-of-plane and in-plane anisotropies, respectively. One has¹⁷

$$G_H = -\frac{HM_s}{2}[\sin \theta_1 \cos \phi_1 + \sin \theta_2 \cos \phi_2],$$

$$G_{\text{dip}} = \frac{\pi}{2}M_s^2[\cos \theta_1 + \cos \theta_2]^2 + \frac{\pi}{2}M_s^2N_{zz}[\cos \theta_1 - \cos \theta_2]^2 \\ + \frac{\pi}{2}M_s^2N_{yy}[\sin \theta_1 \sin \phi_1 - \sin \theta_2 \sin \phi_2]^2,$$

$$G_{2\perp} = -\frac{K_{2\perp}}{2}[\cos^2 \theta_1 + \cos^2 \theta_2], \quad (2)$$

where M_s is the saturation magnetization; H is the external field applied within the film plane (xy) along the x direction; $K_{2\perp} > 0$ is a uniaxial out-of-plane anisotropy that favors the up/down spin alignment along z , the normal to the film plane; N_{zz} is the demagnetization factor associated to the up/down domain structure: since the stripes are assumed to be parallel to the field direction (x axis), one has $N_{xx}=0$ and $N_{yy}=1-N_{zz}$. In general, the static demagnetization factor N_{zz} is a function of the domain aspect ratio L/t (where L is the domain width and t the film thickness) and of the rotational permeability μ .^{17,18,21} For magnetic field applied along the hard in-plane direction, the in-plane anisotropy contribution to the free energy is written as

$$G_{2\parallel}^{(i)} = -\frac{K_{2\parallel}}{2}[\sin^2 \theta_1 \sin^2 \phi_1 + \sin^2 \theta_2 \sin^2 \phi_2] \quad (3)$$

while for field applied along the easy in-plane direction

$$G_{2\parallel}^{(ii)} = -\frac{K_{2\parallel}}{2}[\sin^2 \theta_1 \cos^2 \phi_1 + \sin^2 \theta_2 \cos^2 \phi_2]. \quad (4)$$

In the former case the uniaxial in-plane anisotropy $K_{2\parallel} > 0$ favors the y direction (perpendicular to the field and to the stripes), and in the latter case it favors the x direction (parallel to the field and to the stripes).

In terms of the free energy parameters, we define the out-of-plane anisotropy field $H_{2\perp} = 2K_{2\perp}/M_s$, the in-plane field $H_{2\parallel} = 2K_{2\parallel}/M_s$, and the dipolar field $H_{\text{dip}} = 4\pi M_s$. It is customary to introduce the quality factor $Q = H_{2\perp}/H_{\text{dip}}$ as the ratio between the out-of-plane anisotropy field $H_{2\perp}$, favoring the perpendicular direction (z), and the dipolar field H_{dip} , favoring the film plane (xy). In the case under study, we have $Q < 1$. Finally, the saturation field is defined as $H_{\text{sat}} = H_{2\perp} - H_{\text{dip}}N_{zz}$.^{17,18}

In the following, the equilibrium values of the polar and azimuthal angles, obtained by minimizing the free energy Eq. (1), will be denoted by the suffix “ e .” The frequencies of the spin-wave excitations are evaluated^{17,18} by the Landau-Lifshitz equations of motion (see the Appendix for details). Two modes, denoted by ω^+ (acoustic mode) and ω^- (optic mode), are found for $H < H_c$ and a single (uniform²²) mode for $H \geq H_c$. It is worth noticing that, despite their names, neither of the two modes is fully in-phase or fully out-of-phase: their peculiar character is disclosed by the analysis of the eigenvectors associated to the two modes.¹⁹ In fact, assuming the external magnetic field to be parallel to the x direction (along which the domains are infinitely long), one

finds that the acoustic mode with frequency ω^+ is characterized by dynamic fluctuations such that $m_1^x(t) + m_2^x(t) = 0$ and $m_1^y(t) + m_2^y(t) \neq 0$. This corresponds to an out-of-phase precession of the dynamic moments \mathbf{M}_1 and \mathbf{M}_2 in the direction parallel to the domain wall and an in-phase precession perpendicular to the domain wall. In contrast, for the optic mode the precession parallel to the domain wall is in-phase and the precession perpendicular to the domain wall is out-of-phase.¹⁹

Hereafter we give the expressions of the spin-wave frequencies²³ when the field is applied in-plane along the hard axis, see Eq. (3), or the easy axis, see Eq. (4). In both cases one has $\phi_{1e} = \phi_{2e} = 0$.²⁴

Case (i): H is along the hard in-plane axis. For $H < H_c = H_{\text{sat}}$, the minimum free energy is obtained for $\theta_{2e} = \pi - \theta_{1e}$, where $\sin \theta_{1e} = H/H_{\text{sat}}$. The frequencies of the acoustic and optic modes are

$$(\omega^+/\gamma)^2 = \left[H_{\text{sat}}^2 - H^2 \left(1 - \frac{H_{\text{dip}}N_{yy}}{H_{\text{sat}}} \right) \right] \left[1 - \frac{H_{2\parallel}}{H_{\text{sat}}} \right], \\ (\omega^-/\gamma)^2 = [H_{\text{sat}}^2 - H^2] \left[1 + \frac{H_{\text{dip}}N_{yy} - H_{2\parallel}}{H_{\text{sat}}} \right], \quad (5)$$

respectively (γ is the gyromagnetic factor). For $H \geq H_c = H_{\text{sat}}$, one has $\theta_{1e} = \theta_{2e} = \pi/2$: the stripe domain structure is wiped out, and the film is homogeneously in-plane magnetized. The optic mode (ω^-) disappears and the acoustic one (ω^+) becomes the saturated, in-plane, uniform²² mode with frequency

$$(\omega/\gamma)^2 = [H - H_{2\parallel}][H - (H_{2\perp} - H_{\text{dip}})]. \quad (6)$$

Case (ii): H is along the easy in-plane axis. For $H < H_c = H_{\text{sat}} - H_{2\parallel}$, the ground state has $\theta_{2e} = \pi - \theta_{1e}$, where $\sin \theta_{1e} = H/H_c$ and the frequencies of the acoustic and optic spin-wave excitations are

$$(\omega^+/\gamma)^2 = H_{\text{sat}}H_c \left[1 - \left(\frac{H}{H_c} \right)^2 \left(1 - \frac{H_{\text{dip}}N_{yy}}{H_c} \right) \right], \\ (\omega^-/\gamma)^2 = H_{\text{sat}}H_c \left[1 - \left(\frac{H}{H_c} \right)^2 \right] \left[1 + \frac{H_{\text{dip}}N_{yy}}{H_c} \right]. \quad (7)$$

For $H \geq H_c = H_{\text{sat}} - H_{2\parallel}$, one has $\theta_{1e} = \theta_{2e} = \pi/2$ and the frequency of the uniform mode is

$$(\omega/\gamma)^2 = [H + H_{2\parallel}][H - (H_{2\perp} - H_{\text{dip}}) + H_{2\parallel}]. \quad (8)$$

IV. RESULTS AND DISCUSSION

A. Spin-wave frequencies

In Fig. 3 the measured spin-wave frequencies are plotted as a function of the intensity of the in-plane applied magnetic field H . When the field is parallel to the hard in-plane direction $[-110]$ [Fig. 3(a)], two spin-wave modes are observed for $0.4 < H < 0.9$ kOe. No differences in frequency, within experimental error, are found upon increasing or decreasing the field intensity, thus ruling out the possibility of metastable

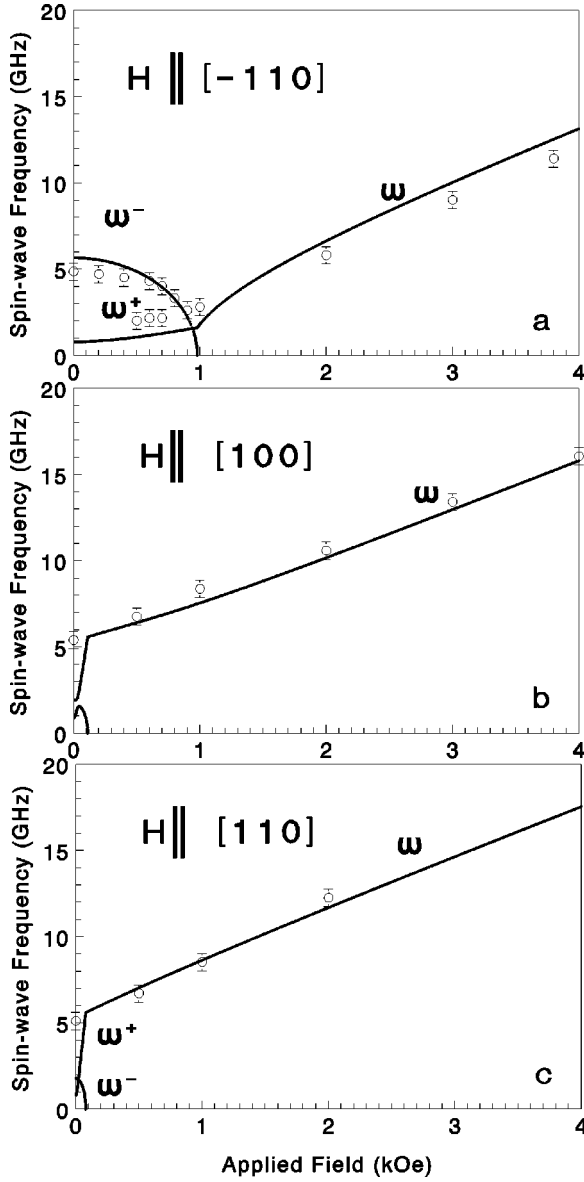


FIG. 3. Full-line curves: frequencies of the acoustic (ω^+), optic (ω^-), and uniform (ω) mode, as a function of the in-plane field H , calculated using $H_{2||}=0.9$ kOe, $H_{2\perp}=13.9$ kOe, $H_{\text{dip}}=17$ kOe, $N_{zz}=0.76$ ($H_{\text{sat}}=0.98$ kOe), for three different cases: (a) field applied parallel to the stripes and along $[-110]$, the hard in-plane direction (see Eqs. (5) and (6)); (b) field applied parallel to the stripes and along $[100]$, the intermediate in-plane direction; (c) field applied parallel to the stripes and along $[110]$, the easy in-plane direction (see Eqs. (7) and (8)). Open circles: experimental data.

bility effects. In contrast, a single spin-wave mode is observed for $H \geq 0.9$ kOe. When the field is applied along either the intermediate $[100]$ or the easy $[110]$ in-plane directions [see Figs. 3(b) and 3(c), respectively], a single spin-wave mode is experimentally observed at any investigated field intensity. The full-line curves in Fig. 3 are the theoretical spin-wave frequencies. The Hamiltonian parameters for the calculations were deduced from previous experimental work on Fe/GaAs(001) films,⁸ where a detailed fit of the BLS data was made for a whole set of samples with different values of the iron thickness, ranging between t_{Fe}

$=4$ Å and 100 Å. As t_{Fe} increases, the out-of-plane single-ion anisotropy $K_{2\perp}$ strongly decreases, while the dipolar field $H_{\text{dip}}=4\pi M_s$, favoring in-plane magnetization, increases; moreover, a biaxial in-plane anisotropy, favoring the $[100]$ and $[010]$ crystallographic axes, gradually develops. For the sample with $t_{\text{Fe}}=6$ Å, only the data at fields high enough for the magnetization to be homogeneous and in-plane, were used to obtain the fit. The dipolar field was estimated to be $H_{\text{dip}}=17$ kOe,²⁵ the out-of-plane anisotropy field $H_{2\perp}=13.9$ kOe and the in-plane anisotropy field $H_{2||}=0.9$ kOe.²⁶ Using these parameters, the demagnetization factor N_{zz} was self-consistently calculated²¹ for different values of the domain aspect ratio L/t . The variation of the domain size with the applied field intensity, and thus the corrections to the frequency due to such variations, were neglected as being of a higher order.^{17,18}

The best overall agreement between theory and experiment was found assuming for the static demagnetization factor the value $N_{zz}=0.76$. Such an assumption, although it corresponds to a probably too low aspect ratio,²⁷ is nevertheless able to justify the presence of domains in the system in spite of the fact that, for the considered iron thickness $t_{\text{Fe}}=6$ Å, one has $H_{2\perp} < H_{\text{dip}}$ (quality factor $Q < 1$). In fact, for the onset of up/down stripe domains, the condition $H_{2\perp} - H_{\text{dip}}N_{zz} > 0$ has to be satisfied. As a further support to the domain hypothesis, it is worth observing that in epitaxial Co/Pt multilayers a perpendicular (up/down) stripe domain structure was indeed experimentally observed at remanence by magnetic force microscopy, while torque magnetometry measurements, providing $Q < 1$, had suggested a preference for in-plane orientation of the magnetization.²⁸

Note that the two modes are well observable only for the case of field applied along the hard in-plane direction [Fig. 3(a)]. Otherwise [see Figs. 3(b) and 3(c)] one has a considerable shrinking of the coexistence region of the two modes and moreover metastability phenomena are likely to occur since the energy of the stripe domain ground state is close to that of a homogeneous in-plane configuration.

B. Light scattering intensities

For the experimental backscattering geometry (H in-plane parallel to the x axis and scattering plane perpendicular to H) the incident light has p -polarization (the optical wave vector and the optical incident electric field \mathbf{E}_I have only y and z components) while the scattered light has s -polarization. Then its intensity is proportional to the square modulus of the x component of the polarization \mathbf{P} induced in the film^{29–33}

$$\begin{aligned}
 4\pi P_x = & m^x(t) [-KE_I^y \sin \theta_e \cos \theta_e - 2G_{44}M_s E_I^z \cos \theta_e (\sin^2 \theta_e \\
 & - \cos^2 \theta_e)] + m^y(t) [2G_{44}M_s E_I^y \sin \theta_e - KE_I^z] + m^z(t) \\
 & \times [KE_I^z \sin^2 \theta_e + 2G_{44}M_s E_I^z \sin \theta_e (\sin^2 \theta_e - \cos^2 \theta_e)],
 \end{aligned} \tag{9}$$

where K and G_{44} denote the first- and second-order (complex) magneto-optic coupling coefficients, respectively. For the film with up/down domains, we assume that $m^\alpha(t) = m_1^\alpha(t) + m_2^\alpha(t)$ ($\alpha=x, y, z$) since the size of the laser spot is

much greater than the lateral size of the domains.³³ Taking into account that $\phi_{1e}=0$ and $\sin \theta_{1e}=H/H_c=h$ and expressing the dynamic fluctuations of the magnetization in terms of the fluctuations of the angle coordinates $\Delta\theta^\pm(t)$, $\Delta\phi^\pm(t)$ defined in the Appendix, we obtain

$$4\pi P_x = \Delta\theta^-(t)[-KE_1^y h(1-h^2) - 2G_{44}M_s E_1^z(1-h^2)(2h^2-1)] \\ + \Delta\phi^+(t)[2G_{44}M_s E_1^y h^2 - KE_1^z h] + \Delta\theta^+(t)[-KE_1^y h^3 \\ - 2G_{44}M_s E_1^z h^2(2h^2-1)]. \quad (10)$$

The field dependence of the intensities of the two modes can now be estimated taking into account that the eigenvector associated with the acoustic mode is characterized by $\Delta\theta^-(t)=\Delta\phi^-(t)=0$ while the optic mode has $\Delta\theta^+(t)=\Delta\phi^+(t)=0$.

Acoustic mode with frequency ω^+ . The intensity $I^+(H)$ of the light scattered by the acoustic mode has a maximum for $H \rightarrow H_c$ since the fluctuations become very large. For zero field the intensity is zero, $I^+(0)=0$, since $\Delta\theta^-(t)=0$ and the coefficients of the “+” angle fluctuations are zero for $h=0$. For $H \rightarrow +\infty$, the intensity vanishes, $I^+(H) \rightarrow 0$, since, upon increasing H above H_c , the ω^+ mode evolves into the uniform mode ω and the fluctuations progressively decrease. This behavior for $I^+(H)$ is similar to that of a perpendicularly magnetized uniform film.³⁴

Optic mode with frequency ω^- . For $H=H_c$, the intensity of the light scattered by the optic mode is zero $I^-(H_c)=0$, since $\Delta\theta^+(t)=\Delta\phi^+(t)=0$ and the coefficient of the $\Delta\theta^-(t)$ angle fluctuation is zero for $h=1$. For zero field, the intensity $I^-(0)$ can be finite provided that the second order magneto-optic coupling coefficient is nonzero, $G_{44} \neq 0$.

The field dependence of the intensity of both the acoustic and the optic modes, as deduced from Eq. (10), is qualitatively confirmed by the experimental spectra in Fig. 1. The intensity of the former mode exhibits a neat maximum for field values slightly lower than $H_c \approx 0.9$ kOe and then it vanishes as the field is reduced below about 0.5 kOe. The optic mode intensity, instead, shows a minimum approaching H_c , in agreement with the theoretical predictions.

V. CONCLUSIONS

In conclusion, we have shown that a double-peaked structure is displayed by the Brillouin Light Scattering spectra of Fe/GaAs(001) films with $t_{Fe}=6$ Å when the field is applied in-plane along the hard axis and is smaller than a critical value $H_c=0.9$ kOe. The existence of two peaks in the BLS spectrum should be the general feature of a film with a perpendicular domain structure (it is irrelevant whether the magnetization is canted or not). This feature disappears when $H \geq H_c$ and the magnetization lies in the film plane. The reason why the unravelling of such a two-peaked structure in the Brillouin light scattering spectra of ultrathin magnetic films is so rare might well be that many conditions have to be simultaneously satisfied. In fact, the optic mode, with frequency ω^- , has enough intensity in an appreciable range of fields only if $G_{44} \neq 0$ and H_c are not too small. In contrast, the acoustic mode, with frequency ω^+ , has more chances to

be observed since its intensity, though always vanishing in the $H \rightarrow 0^+$ limit, is expected to increase as H increases and to reach a maximum just at H_c . Another stringent requirement for the simultaneous observation of two modes is that the competing out-of-plane anisotropy field $H_{2\perp}$ and easy-plane dipolar anisotropy field H_{dip} are of comparable magnitude and that $H_{2\perp} - H_{dip} N_{zz} > 0$, so that a perpendicularly magnetized up/down domain structure is energetically favored for $H \rightarrow 0^+$. This seems just to be the case of the Fe/GaAs films with $t_{Fe}=6$ Å. In fact, for higher Fe thickness, one has $H_{dip} \gg H_{2\perp}$, so that a homogeneous in-plane magnetized ground state is realized, while, upon reducing the Fe thickness, one would expect $H_{dip} \ll H_{2\perp}$ and a single spin-wave mode to be excited with respect to a homogeneous, perpendicularly magnetized metastable state.

We hope that the results of this paper can stimulate other experimental groups to directly visualize the domain pattern, e.g. using magnetic microscopy techniques as a function of the external magnetic field intensity.³⁵ This should be done *in situ*, because the magnetic anisotropy is strongly affected by the presence of a protective overlayer, so that formation of magnetic domains can be prevented.⁸

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APPENDIX: SPIN-WAVE FREQUENCIES

The static equilibrium configuration of the system is obtained by minimizing the free energy G , Eq. (1), with respect to the polar and azimuthal variables while the frequencies of the spin-wave excitations are evaluated¹⁷ by the Landau-Lifshitz equations of motion

$$\frac{d\theta_{1,2}}{dt} = -\frac{\gamma}{\sin \theta_{1,2}} \frac{\partial \mathcal{G}}{\partial \phi_{1,2}}, \quad \frac{d\phi_{1,2}}{dt} = \frac{\gamma}{\sin \theta_{1,2}} \frac{\partial \mathcal{G}}{\partial \theta_{1,2}} \quad (A1)$$

where $\mathcal{G}=2G/M_s$. The small oscillations of the system in response to an external perturbation are obtained by expanding \mathcal{G} in a Taylor series about its equilibrium value \mathcal{G}_e up to the second order. Next, assuming for the set of variables a harmonic time dependence with frequency ω and introducing the normal coordinates $\Delta\theta^\pm = \Delta\theta_1 \pm \Delta\theta_2$ and $\Delta\phi^\pm = \Delta\phi_1 \pm \Delta\phi_2$ (where Δ denotes a small variation), the equations of motion can be rewritten in matrix form as

$$\begin{bmatrix} A^+ & -iz & 0 & B^+ \\ iz & C^+ & B^- & 0 \\ 0 & B^- & A^- & -iz \\ B^+ & 0 & iz & C^- \end{bmatrix} \begin{bmatrix} \Delta\theta^+ \\ \Delta\phi^+ \\ \Delta\theta^- \\ \Delta\phi^- \end{bmatrix} = 0, \quad (A2)$$

where $z = \omega \sin \theta_{1e}$ and $A^\pm = \mathcal{G}_{11} \pm \mathcal{G}_{13}$, $B^\pm = \mathcal{G}_{12} \pm \mathcal{G}_{23}$, $C^\pm = \mathcal{G}_{22} \pm \mathcal{G}_{24}$. By $\mathcal{G}_{ij} = \partial^2 \mathcal{G} / \partial X_i \partial X_j|_e$ we denote the second partial derivatives of the free energy with respect to the angular

variables ($X_1=\theta_1$, $X_2=\phi_1$, $X_3=\theta_2$, $X_4=\phi_2$). The frequencies of the normal modes are obtained by imposing the condition for nontriviality of the solutions of Eq. (A2), i.e., the vanishing of the matrix determinant.

For $H \geq H_c$ the ground state is homogeneously in-plane magnetized, ($\theta_{1e}=\theta_{2e}=\pi/2$ and $N_{zz}=1$) and it results that $A^+=A^-=\mathcal{G}_{11}$, $B^+=B^-=0$, and $C^+=C^-=\mathcal{G}_{22}$, so that a single, uniform mode²² is obtained, with frequency $(\omega/\gamma)^2=\mathcal{G}_{11}\mathcal{G}_{22}$.

*Electronic address: mgpini@ifac.cnr.it

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