

Magnetization Reversals of Fe₈₁Ga₁₉-Based Flexible Thin Films Under Multiaxial Mechanical Stress

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The effect of multiaxial mechanical stress on angle-dependent magnetization reversals is studied on a single 10-nm Ni₈₀Fe₂₀ film, a single 10-nm Fe₈₁Ga₁₉ film, and a Ni₈₀Fe₂₀(10 nm)/Fe₈₁Ga₁₉(10 nm) bilayer. These films are grown on flexible Al “kitchenlike” foils. These flexible foils are bent on two convex optical lenses to apply multiaxial mechanical stresses of different magnitudes. In order to understand the mechanisms driving the physical properties of the flexible systems, the magnetic films are also grown on hard native-oxide-covered Si(100) substrates. By probing the angular dependence of the coercive field and the remanent magnetization, we show that the Ni₈₀Fe₂₀ single layer and the Ni₈₀Fe₂₀/Fe₈₁Ga₁₉ bilayer grown on Si present a uniaxial anisotropy. Along their easy axis, their domain structure at the coercive field exhibits large domains with sawtooth domain walls. However, the Fe₈₁Ga₁₉ layer grown on Si shows a cubic anisotropy and sharp domain walls with a right-angle geometry along its easy axis. All layers grown on Al foils show development or enhancement of a uniaxial anisotropy arising from the Al surface morphology. Multiaxial mechanical stress applied to the flexible Al foils results in very different nonreversible effects as a function of the composition of the layered system. For Ni₈₀Fe₂₀, multiaxial mechanical stress has no effect, as expected for a nonmagnetostrictive system. For Fe₈₁Ga₁₉, it results in a full in-plane randomization of its magnetization-reversal properties. For the bilayer, multiaxial mechanical stress does not fully randomize the in-plane magnetization-reversal properties: the bilayer retains a uniaxial character.

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I. INTRODUCTION

Magnetostrictive thin films have attracted a great deal of interest in the last few decades. Firstly, this is because of a need to understand the fundamental mechanisms driving the strain and magnetic coupling in thin films. Secondly, this interest originates from the fact that magnetostrictive properties are of use in sensing devices, actuators, energy harvesters, and spintronic devices [1,2]. Among the different magnetostrictive alloys, Fe₈₁Ga₁₉, discovered in 2000 by Clark *et al.* [3], has received particular attention as it exhibits remarkable properties such as low hysteresis, large magnetostriction, good tensile strength, and machinability, and there has been recent progress in commercially viable methods of processing [4]. The main advantage of Fe₈₁Ga₁₉ alloys when compared with the rare-earth-alloy family is reduced cost, as well as the fact that these alloys are less brittle.

Magnetostrictive thin films, including those prepared using Fe₈₁Ga₁₉ alloys, can be used for making electronic

circuits and devices on flexible substrates [5–7]. Their magnetic properties are inherently sensitive to mechanical deformation through the inverse magnetostrictive effect (also referred to as the Villari effect). A magnetostrictive material may exhibit reversible magnetic properties that change when it is subjected to elastic strain, whereas a magnetostrictive material subjected to a plastic residual strain may exhibit irreversible changes in its magnetic properties. The interest in flexible devices has its origin in the fact that these devices are significantly lower in cost, as well as lighter and more compact, compared with conventional devices. Also, various flexible devices, displays, logic devices, sensors, and memories are expected to be integrated into multifunctional systems, and to be implemented in wearable technologies [8,9]. Consequently, flexible magnetostrictive systems constitute an essential part of the field named straintronics [10]. In particular, the attributes of Fe₈₁Ga₁₉ introduce opportunities for the use of a smart material in applications that involve stretching, bending, shear, or torsion.

Among the different properties of magnetostrictive flexible thin films to be understood, their magnetization

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reversals (MRevs), which are related to the magnetic-moment direction under external stimuli, including electrical, magnetic, or mechanic stimuli, are of interest for applications and fundamental studies [6]. MRevs are driven by the anisotropic properties, which determine the application areas of the thin film and the associated performance of devices. For instance, the magnetic sensitivity of sensors is related to magnetization reversals and magnetic-anisotropy properties. Consequently, tailoring and understanding MRevs of flexible $\text{Fe}_{81}\text{Ga}_{19}$ thin films would be of prime interest for actuators, transducers, and energy-harvesting devices.

Following this scientific goal, the MRev behavior of $\text{Fe}_{81}\text{Ga}_{19}$ thin films grown on flexible substrates has attracted a great deal of attention in the last decade [11–17]. These previous studies focused on the MRev properties of flexible $\text{Fe}_{81}\text{Ga}_{19}$ under tensile or compressive uniaxial mechanical stress (UMS). The main idea was to achieve different MRev properties in order to identify the potential of flexible $\text{Fe}_{81}\text{Ga}_{19}$ for applications. In addition, these studies revealed large modifications of the MRev properties due to UMS [11,13–15,17–19]. It should be noted that UMS resulted in differences in MRev properties that were found to be irreversible, in the sense that the virgin state before UMS could not be recovered once UMS was applied. This irreversibility property allows stabilization of the MRevs of flexible $\text{Fe}_{81}\text{Ga}_{19}$ and may enlarge the potential applications of this system.

Whereas the use of UMS with flexible $\text{Fe}_{81}\text{Ga}_{19}$ thin films has previously been considered in order to tailor stable MRev properties of $\text{Fe}_{81}\text{Ga}_{19}$, multiaxial mechanical stress (MMS) has not been not studied, despite its key role in applications [20–25]. MMS needs to be considered, as many different combinations and orientations of magnetic fields and mechanical stresses can occur in real devices and applications. Wearable electronics may be subjected to a wide range of directional distributed external stresses. Stretchable sensors capable of detecting different mechanical stimuli are essential in wearable applications involving multiaxial motion [26]. MMS is of interest in the field of soft robotics and smart prosthetics for applications in artificial intelligent skin [27–29]. In order to understand external environments or stimuli in an accurate manner, electronic skin needs to perceive various types of mechanical information (i.e., multiaxial information) so that it can emulate human perception and natural touch. In addition, MMS is purposely used for converting mechanical energy into electrical energy for energy-harvesting purposes [21–23]. Finally, MMS needs to be taken into consideration in transformers, as it may be detrimental when it occurs in electromagnetic devices, where magnetostrictive properties play a key role in generating detrimental noise [24]. Despite all this, the effects of MMS on the MRev properties of $\text{Fe}_{81}\text{Ga}_{19}$ thin films have been overlooked and not yet studied.

In this paper, we report on the effect of MMS on magnetization reversals in flexible $\text{Fe}_{81}\text{Ga}_{19}$ -based thin films. This effect is studied on a single 10-nm $\text{Ni}_{80}\text{Fe}_{20}$ (Py) film, on a single 10-nm $\text{Fe}_{81}\text{Ga}_{19}$ film, and on a $\text{Py}(10\text{ nm})/\text{Fe}_{81}\text{Ga}_{19}(10\text{ nm})$ bilayer. These films are grown on flexible Al “kitchenlike” foils. MMS can take a variety of forms, from biaxial to isotropic. Before approaching an anisotropic MMS geometry such as a biaxial one, the isotropic case is of interest for probing the effect of a fully randomized MMS on MRevs. Consequently, Al foils are bent on two convex optical lenses of different curvature radius to achieve a tensile stress that is isotropic in the film plane with two different magnitudes.

The Al foil substrate is chosen as it is flexible, inexpensive, durable, and greaseproof, and the raw materials required for its manufacture are plentiful. Also, the morphology of Al foils presents a grooved surface due to the rolling process used in its industrial production [30]. This surface may lead to MRev properties of interest with or without MMS. In general, in comparing the properties of flexible films with those of inflexible ones, is of interest to achieve an understanding of the mechanisms driving the physical properties of flexible films. For this reason, in this investigation all magnetic films are also grown on hard native-oxide-covered Si(100) substrates, as these substrates are an archetype of flat and hard substrates.

$\text{Fe}_{81}\text{Ga}_{19}$ is chosen for its magnetostrictive properties of interest. The $\text{Ni}_{80}\text{Fe}_{20}$ film is permalloy. It is chosen as it constitutes an archetype of a soft magnetic material, and as it is considered as a nonmagnetostrictive reference. Beyond the single-film case, a study of a bilayer made of Py and $\text{Fe}_{81}\text{Ga}_{19}$ is performed to probe possible hybridization of the MRev properties, with and without MMS, at the nanoscale. Indeed, interfacing different magnetic thin films is a mechanism for tailoring magnetic properties in general. This mechanism has been shown to be of interest for driving magnetic properties of nonflexible magnetostrictive bilayers [31–35], including nonflexible $\text{Py}/\text{Fe}_{81}\text{Ga}_{19}$ bilayers [33–35]. However, there has been no previous work studying magnetic properties of flexible $\text{Py}/\text{Fe}_{81}\text{Ga}_{19}$ bilayers.

In the first part of this paper, the experimental details and procedures are introduced. In the second part, the MRev properties of films on hard Si substrates are presented. In the third part, the MRev properties of films on flexible Al foils and their dependence on MMS are presented. In the last part of this paper, conclusions are drawn.

II. EXPERIMENTAL DETAILS

In the study presented in this paper, a 10-nm $\text{Fe}_{81}\text{Ga}_{19}$ thin film, a 10-nm Py thin film, and an $\text{Fe}_{81}\text{Ga}_{19}(10\text{ nm})/\text{Py}(10\text{ nm})$ bilayer are grown on Si substrates and Al foils by a sputtering technique. All samples are covered with a 5-nm-thick Ta cap layer to avoid oxidation.

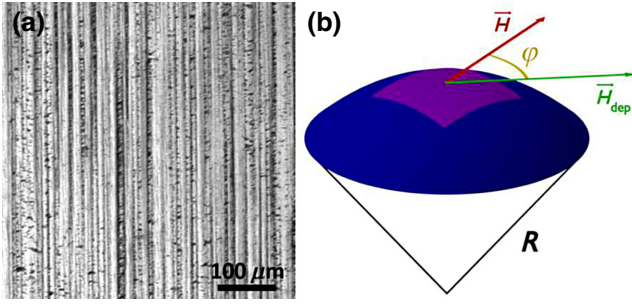


FIG. 1. (a) Scanning-electron-microscope image of the surface morphology of the Al foil. (b) Schematic illustration of an Al foil on top of a convex optical lens. $\varphi = 0$ when the applied field H during MOKE measurements is in the same direction as H_{dep} , the applied magnetic field during growth.

An Fe₈₁Ga₁₉ target and a Ni₈₀Fe₂₀ target are used. Sample compositions are confirmed by utilizing electron-probe microanalysis measurements. The base pressure prior to film deposition is typically 10^{-7} mbar. The rf power used to sputter the 3-in. Fe₈₁Ga₁₉ (Ni₈₀Fe₂₀) target is 100 (100) W, and the argon pressure is 1.5×10^{-2} (10^{-2}) mbar. With these sputtering conditions, the growth rate for the Fe₈₁Ga₁₉ (Ni₈₀Fe₂₀) films is 0.22 (0.20) nm/s. The growth is carried out under an in-plane magnetic field $H_{\text{dep}} \sim 2.4 \text{ kA m}^{-1}$ (300 Oe), in order to favor a preferential magnetic-anisotropy direction.

The thin films are grown on commercially available native-oxide-covered (100) Si substrates [36] and on flexible kitchen-type aluminum foils. The Si substrates are 500- μm -thick rigid substrates and exhibit a root-mean-square roughness below 0.5 nm. The Al foils are 20- μm -thick flexible substrates. Scanning-electron-microscope measurements reveal the typical grooved surface morphology of the Al foils [30], as shown in Fig. 1(a). This grooved surface arises from the rolling mechanism used to achieve the Al foils. The width of the grooves is found to be typically in the order of 10 μm . The Al foils exhibit a root-mean-square roughness of 0.53 μm and an arithmetic average roughness of 0.39 μm . It should be noted here that H_{dep} is systematically applied parallel to the grooves during the growth of the thin films.

Once the magnetic layers have been grown on flat Al foils, magnetization reversals are studied on flat and on spherically curved Al foils. To achieve the bending and the resulting MMS, Al foils are stuck onto convex optical lenses, as shown in Fig. 1(b). Two different lenses are used to allow MMSs of different magnitudes. The radius of curvature of the first convex optical lens is 24.0 cm, and that of the second is 58.0 cm. Because of the dome shape of the convex optical lenses, the bending stresses the substrate; the stress is tensile with axial symmetry, and the film deposited onto the external surface is stressed isotropically in the film plane [37,38]. It should be noted

TABLE I. Samples on Si substrates studied in Sec. III. R is the radius of curvature, as seen in Fig. 1(b).

Sample name	R	Layer structure
Py ^{Si}	∞	Ni ₈₀ Fe ₂₀ (10 nm)/Si
FeGa ^{Si}	∞	Fe ₈₁ Ga ₁₉ (10 nm)/Si
Bilayer ^{Si}	∞	Fe ₈₁ Ga ₁₉ (10 nm)/Ni ₈₀ Fe ₂₀ (10 nm)/Si

here that structural characterizations involving confocal microscopy, scanning electron microscopy, and transmission electron microscopy are performed on the samples before and after bending. These characterizations do not reveal the presence of extended defects (such as cracks) resulting from the bending procedure.

Magnetic properties at room temperature are determined using the magneto-optic Kerr effect (MOKE) in a wide-field Kerr microscope from Evico Magnetics [39,40].

III. MAGNETIZATION REVERSALS ON Si SUBSTRATES

In this section, MRevs of Py, Fe₈₁Ga₁₉, and Py/Fe₈₁Ga₁₉ layers grown on Si substrates are reported. The samples studied in this section and their characteristics are summarized in Table I. In the following, $\varphi = 0$ when the applied field during the MOKE measurements is in the same direction as H_{dep} , the applied magnetic field during growth.

A. Py on Si

Figure 2(a) presents the magnetization-reversal loops for Py^{Si}. The MRev loop along $\varphi = 0^\circ$ exhibits a rectangular shape and a rather small coercive field (H_c) of 2.8 Oe. The loop along $\varphi = 90^\circ$ exhibits very weak hysteresis, with an H_c of 1.0 Oe and a small normalized remanence (M_R/M_s) of 0.13. These MRev-loop properties along and perpendicular to the applied deposition field correspond to a soft magnetic system with a growth-induced uniaxial anisotropy, as expected for a Py thin film grown under an external field [41]. The nonvanishing H_c and M_R/M_s perpendicular to H_{dep} indicate the presence of an anisotropy dispersion.

The domain configuration along the easy axis (EA) is probed in the vicinity of the coercive field, as shown in Fig. 2(b). It exhibits a zigzag (also referred to as “sawtooth”) domain-wall configuration, separating large domains. This particular configuration of domain walls is observed in magnetic thin films with in-plane uniaxial anisotropy [42], and has been previously reported for Py thin films [43,44]. Zigzag domains walls are formed between two areas with opposite head-on magnetization directions; they minimize the magnetostatic energy, thereby reducing the charge density but increasing the

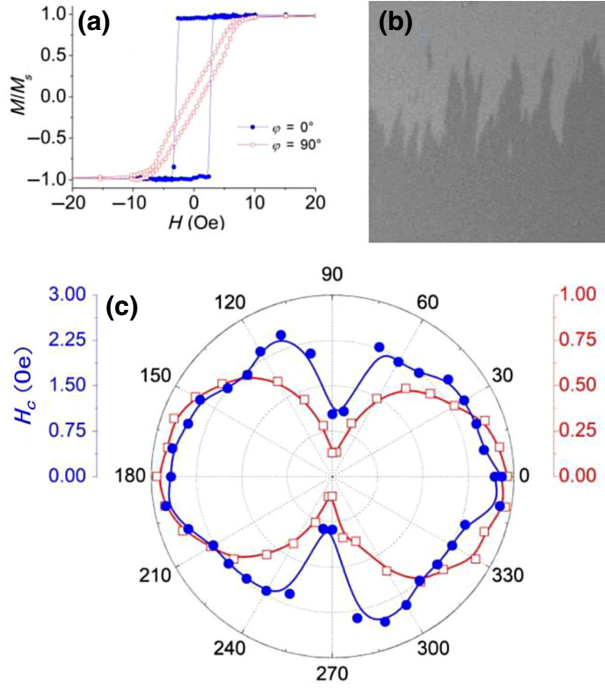


FIG. 2. For PySi , (a) hysteresis loops measured along $\varphi = 0^\circ$ (filled black circles) and along $\varphi = 90^\circ$ (open red circles); (b) 5 mm $\times$ 5 mm MOKE image obtained in the vicinity of the coercive field along $\varphi = 0^\circ$; (c) angular dependence of H_c (filled blue circles) and M_R/M_s (red squares).

length of the wall, with the charge distributed on both sides of the wall [45].

Systematic in-plane angle-dependent measurements are performed on the Py sample to further probe the anisotropic character. For each applied magnetic field direction, the angular dependences of H_c and M_R/M_s are obtained, as shown in Fig. 2(c). Both angular dependences are typical of a uniaxial system, as the H_c polar plot exhibits a butterflylike shape and the M_R/M_s plot exhibits clear symmetrical lobes [46,47]. The angle-dependent study shows that the uniaxial EA is aligned with H_{dep} .

B. $\text{Fe}_{81}\text{Ga}_{19}$ on Si

For FeGaSi , the MRev loop along $\varphi = 0^\circ$ exhibits a rectangular shape with an H_c of 18 Oe, as shown in Fig. 3(a). The loop along $\varphi = 90^\circ$ exhibits the same H_c as that along $\varphi = 0^\circ$. This indicates that FeGaSi is not uniaxial. The domain configuration along $\varphi = 0^\circ$ is probed in the vicinity of the coercive field, as shown in Fig. 3(b). The domain pattern in FeGaSi differs substantially from that in PySi . The domain size is still large, but the transition between two regions is quite sharp and does not exhibit a zigzag shape. A right-angle boundary between two domains is observed, which suggests the presence of a cubic anisotropy (i.e., a fourfold anisotropy).

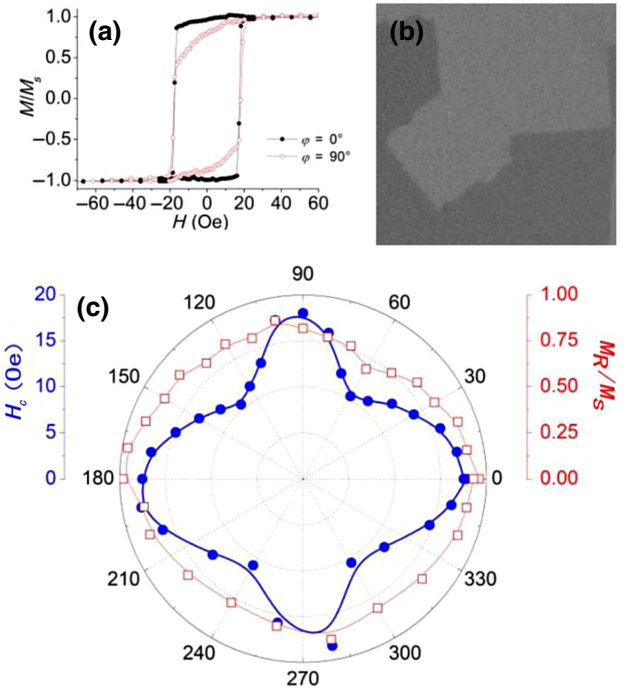


FIG. 3. For FeGaSi , (a) hysteresis loops measured along $\varphi = 0^\circ$ (filled black circles) and along $\varphi = 90^\circ$ (open red circles); (b) 5 mm $\times$ 5 mm MOKE image obtained in the vicinity of the coercive field along $\varphi = 0^\circ$; (c) angular dependence of H_c (filled blue circles) and M_R/M_s (red squares).

Systematic in-plane angle-dependent measurements of H_c and M_R/M_s are performed on FeGaSi to probe further the anisotropic character, as shown in Fig. 3(c). The angular dependence of H_c exhibits four well-defined maxima, and the M_R/M_s polar plot does not exhibit the well-defined symmetrical lobes encountered for PySi . These H_c maxima indicate the presence of a cubic anisotropy component. In fact, we have demonstrated in previous studies that a predominant cubic magnetic anisotropy was present in a 10-nm $\text{Fe}_{81}\text{Ga}_{19}$ film grown on an amorphous glass substrate. It was also shown in that previous study that this cubic character resulted from a (110) crystallographic texture and that it resulted in four maxima in the H_c polar plot [40,48]. Two of the four H_c maxima are along $\varphi = 0^\circ$ and $\varphi = 180^\circ$, showing that one of the cubic easy axes is aligned with H_{dep} . Thus, the angle-dependent study confirms the cubic (i.e., fourfold) anisotropy of FeGaSi . It may be noted that, for an $\text{Fe}_{81}\text{Ga}_{19}$ 20-nm thin film on a GaAs substrate, which exhibited a predominant cubic anisotropy, an H_c value of 25 Oe was previously reported along the easy axis [49].

C. $\text{Py}/\text{Fe}_{81}\text{Ga}_{19}$ on Si

For BilayerSi , the MRev loop shapes along $\varphi = 0^\circ$ and $\varphi = 90^\circ$ exhibit the same characteristics as those for Py, as shown in Fig. 4(a). Indeed, the shape is rectangular

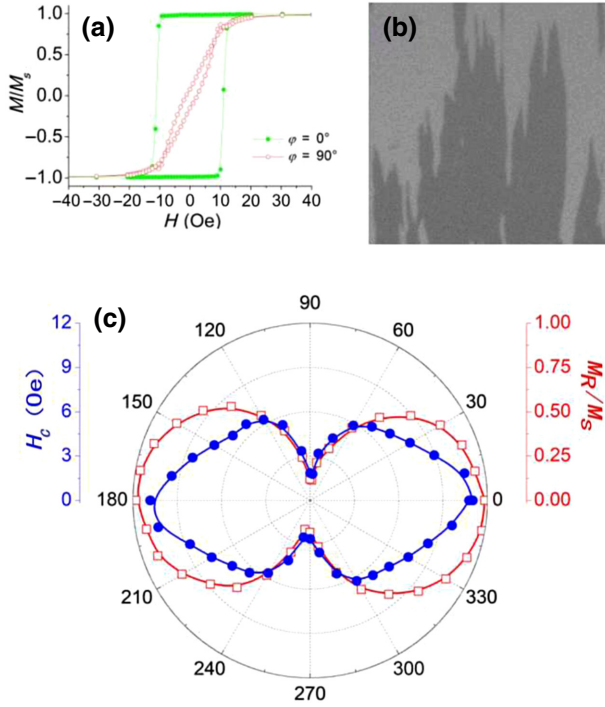


FIG. 4. For Bilayer^{Si}, (a) hysteresis loops measured along $\phi = 0^\circ$ (filled green circles) and along $\phi = 90^\circ$ (open red circles); (b) 5 mm $\times$ 5 mm MOKE image obtained in the vicinity of the coercive field along $\phi = 0^\circ$; (c) angular dependence of H_c (filled blue circles) and M_R/M_s (red squares).

along $\phi = 0^\circ$, and the shape along $\phi = 90^\circ$ exhibits weak values of H_c and M_R/M_s . These results indicate that the hybrid Bilayer^{Si} system shows a uniaxial character in its anisotropy, driven by the Py layer. An H_c value of 11 Oe is found along $\phi = 0^\circ$: it is between the value for Py^{Si} (2.8 Oe) and the value for FeGa^{Si} (18 Oe) obtained at the same angle.

The domain pattern for Bilayer^{Si} along $\phi = 0^\circ$ and in the vicinity of H_c is similar to that for Py^{Si}, as shown in Fig. 4(b). Indeed, it exhibits large domains and zigzag domain walls. The angular dependences of H_c and M_R/M_s are also probed for the bilayer, as shown in Fig. 4(c). The angle-dependent butterfly shape for H_c and the well-defined symmetrical lobes for M_R/M_s confirm the uniaxial anisotropy. The easy axis is along $\phi = 0^\circ$ and is aligned with H_{dep} . The hybrid system is uniaxial but presents a greater anisotropy constant than does Py^{Si}. It may be noted here that an H_c value of 15 Oe was previously reported for an Fe₈₁Ga₁₉(100 nm)/Py(2.5 nm) bilayer grown on Si [35]. This value is in close agreement with the H_c value reported in the present study.

D. Summary of the results for the layers grown on Si substrates

A brief summary is given in this paragraph for the layers grown on Si substrates. Py^{Si} exhibits a uniaxial anisotropy

TABLE II. Samples on Al substrates studied in Sec. IV. R is the radius of curvature.

Sample name	R	Layer structure
Py ^{$R=\infty$}	∞	Ni ₈₀ Fe ₂₀ (10 nm)/Al
Py ^{$R=58$ cm}	58 cm	Ni ₈₀ Fe ₂₀ (10 nm)/Al
Py ^{$R=24$ cm}	24 cm	Ni ₈₀ Fe ₂₀ (10 nm)/Al
FeGa ^{$R=\infty$}	∞	Fe ₈₁ Ga ₁₉ (10 nm)/Al
FeGa ^{$R=58$ cm}	58 cm	Fe ₈₁ Ga ₁₉ (10 nm)/Al
FeGa ^{$R=24$ cm}	24 cm	Fe ₈₁ Ga ₁₉ (10 nm)/Al
Bilayer ^{$R=\infty$}	∞	Fe ₈₁ Ga ₁₉ (10 nm)/Ni ₈₀ Fe ₂₀ (10 nm)/Al
Bilayer ^{$R=58$ cm}	58 cm	Fe ₈₁ Ga ₁₉ (10 nm)/Ni ₈₀ Fe ₂₀ (10 nm)/Al
Bilayer ^{$R=24$ cm}	24 cm	Fe ₈₁ Ga ₁₉ (10 nm)/Ni ₈₀ Fe ₂₀ (10 nm)/Al

with an EA aligned by and with H_{dep} , and a coercive field of 2.8 Oe along its EA. The domain structure in Py^{Si} corresponds to large domains separated by zigzag domain walls. However, FeGa^{Si} exhibits a cubic anisotropy, with one of the EAs aligned with H_{dep} , and a coercivity value of 18 Oe along this EA. The domain structure in FeGa^{Si} is very different from that in Py^{Si} and exhibits sharp domain walls. Finally, the magnetization reversal in Bilayer^{Si} is similar to that in Py^{Si} in the sense of a similar domain structure and a uniaxial character with an EA (aligned with H_{dep}). The difference between Bilayer^{Si} and Py^{Si} is that there is a greater anisotropy constant for the bilayer: Bilayer^{Si} exhibits a coercivity of 11 Oe along the EA. Still, this coercivity is half of that found in FeGa^{Si} along the same direction. Consequently, the coupling between Py and Fe₈₁Ga₁₉ allows a reduction of the coercivity relative to the single Fe₈₁Ga₁₉ film, and a drastic change in the character of the anisotropy of the Fe₈₁Ga₁₉ layer, as it is uniaxial in the bilayer system. These characteristics render the hybrid bilayer system promising for applications that need to combine a magnetostrictive layer, a uniaxial character, and relatively soft magnetic properties.

IV. MAGNETIZATION REVERSALS ON FLEXIBLE Al FOILS UNDER MMS

In this section, MRevs of Py, Fe₈₁Ga₁₉, and Py/Fe₈₁Ga₁₉ layers grown on flexible Al foils are reported. In the following, $\phi = 0$ when the applied field during the MOKE measurements is in the same direction as H_{dep} , the applied magnetic field during growth. It should be noted here that H_{dep} is applied constantly along the grooves of the Al foils during the growth of the thin films. MMS is applied by sticking the Al foils to convex optical lenses. Each sample is described in Table II. Domain imaging using MOKE can not be performed on Al foils, which may be attributed to the large surface roughness of the Al foils [50]. It should be noted that before measurement, MRev loops are measured on different surface regions to ensure that the Al foils are uniformly bent.

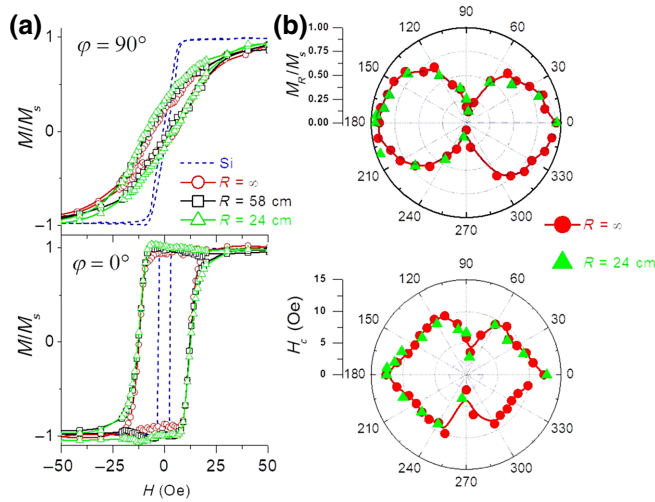


FIG. 5. (a) Hysteresis loops measured along $\varphi = 0^\circ$ (bottom) and along $\varphi = 90^\circ$ (top) for $R = \infty$ (open red circles), $R = 58$ cm (black squares), and $R = 24$ cm (open green triangles) for Py/Al samples. (b) Angular dependence of H_c (bottom) and M_R/M_s (top) for Py on Al foils, for $R = \infty$ (filled red circles) and $R = 24$ cm (filled green triangles).

A. Py on Al

Figure 5(a) shows that the Py MRev loops along $\varphi = 0^\circ$ and $\varphi = 90^\circ$ depend on the nature of the substrate. The coercive and saturation fields have larger values for Al foils than for Py^{Si}. The increase in both fields indicates an increase in the anisotropy constant on Al foils. The shapes of the MRev loops are similar for both substrates, with the presence of a rectangular shape along $\varphi = 0^\circ$. This indicates that the Py layer still exhibits a uniaxial character on flexible Al foils. The MRev loops do not evolve during the application of an isotropic external stress using the two different lenses. This shows the absence of magnetostrictive effects along these two directions at least. It may be noted that the remanence and the coercive field are different from 0 along $\varphi = 90^\circ$. This indicates the presence of an anisotropy dispersion.

The angular dependences of H_c and M_R/M_s are obtained for Py ^{$R=24$ cm} and Py ^{$R=\infty$} , as shown in Fig. 5(b). These angular dependences show an absence of magnetostrictive effects within the plane. Indeed, the H_c and M_R/M_s values with and without MMS are in agreement. The uniaxial anisotropy of Py grown on Al foils is also revealed, as the angular dependence of H_c exhibits a butterflylike shape and the M_R/M_s plot exhibits clear symmetrical lobes. Thus, the Py layers grown on flexible Al are uniaxial and nonmagnetostrictive magnetic thin films, with a greater uniaxial anisotropy constant than for Py^{Si}. This enhancement is likely to arise from the grooved surface morphology of the Al, which would generate a phenomenological shape anisotropy hindering reversal.

B. Fe₈₁Ga₁₉ on Al

In a similar manner to the Py MRev loops, the Fe₈₁Ga₁₉ MRev loops depend on the nature of the substrate, as shown in Fig. 6(a). The coercive and saturation fields are larger for the Al foil than for the Si substrate. It is noted that previous studies have reported H_c values between 20 and 100 Oe for 50-nm Fe₈₁Ga₁₉ films grown on flexible polyethylene terephthalate substrates, depending on the growth conditions [11–19]. However, there are major differences between the MRevs of the Py samples on Al foils and the Fe₈₁Ga₁₉ ones. Indeed, the MRevs of Fe₈₁Ga₁₉ are modified when MMS is applied. The MRev loop shapes, coercivities, and saturation fields are very different for $R = \infty$, $R = 24$ cm, and $R = 58$ cm. This shows that magnetostrictive effects are important for Fe₈₁Ga₁₉ thin films. This is expected from the magnetostrictive properties of Fe₈₁Ga₁₉. In a previous publication [48], the magnetostrictive stress $|b_{\text{eff}}|$ and the magnetostrictive strain λ_{eff} for a 10-nm Fe₈₁Ga₁₉ thin film were reported. At saturation, $|b_{\text{eff}}|$ (λ_{eff}) was found to be 1.2 MPa (16 ppm). These values are close to those previously reported ($\lambda_{\text{eff}} \sim 20$ ppm for similar layer thicknesses) [2,51–53]. Also, some other reported values for $|b_{\text{eff}}|$ (and/or λ_{eff}) in Fe₈₁Ga₁₉ epitaxial thin films were found to be larger [54,55]. In these studies, the larger values were attributed to crystallographic properties driven by the epitaxy. In the present study, it should be noted that the observed effects of MMS on the Fe₈₁Ga₁₉ MRev loops are irreversible: the MRev properties remain constant after the bending has been performed. The sample, after bending and being flattened back again, does not exhibit the initial MRev properties (i.e., those of FeGa ^{$R=\infty$}) but retains the properties observed when curved on the lenses.

To further understand the evolution of the anisotropic properties with the nature of the substrate and with MMS, the angular dependences of H_c and M_R/M_s are probed and are presented in Fig. 6(b). The angular dependence of H_c for FeGa ^{$R=\infty$} exhibits four maxima, revealing that the cubic character of the anisotropy observed in FeGa^{Si} is still present. However, the shape of the angular dependence of H_c for FeGa ^{$R=\infty$} is elongated along $\varphi=0^\circ$ relative to that for FeGa^{Si}.

To understand this elongation phenomenon, simulations are performed using a modified Stoner and Wohlfarth (SW) model [56,57], implemented in a software package named StoneX [46,47,58]. This model can be considered as a modification of the coherent rotational SW model of MRevs, as it includes a random anisotropy, which was not considered in the initial SW model [56,57]. The ferromagnetic material has a magnetic moment per unit volume $M_F \vec{e}_F$, which makes an angle θ with the twofold anisotropy axis [46,47,58]. A value of M_F of 920 kA/m is determined by measurements with a VSM and a superconducting quantum interference device on these samples. The F domain possesses a twofold anisotropy axis along

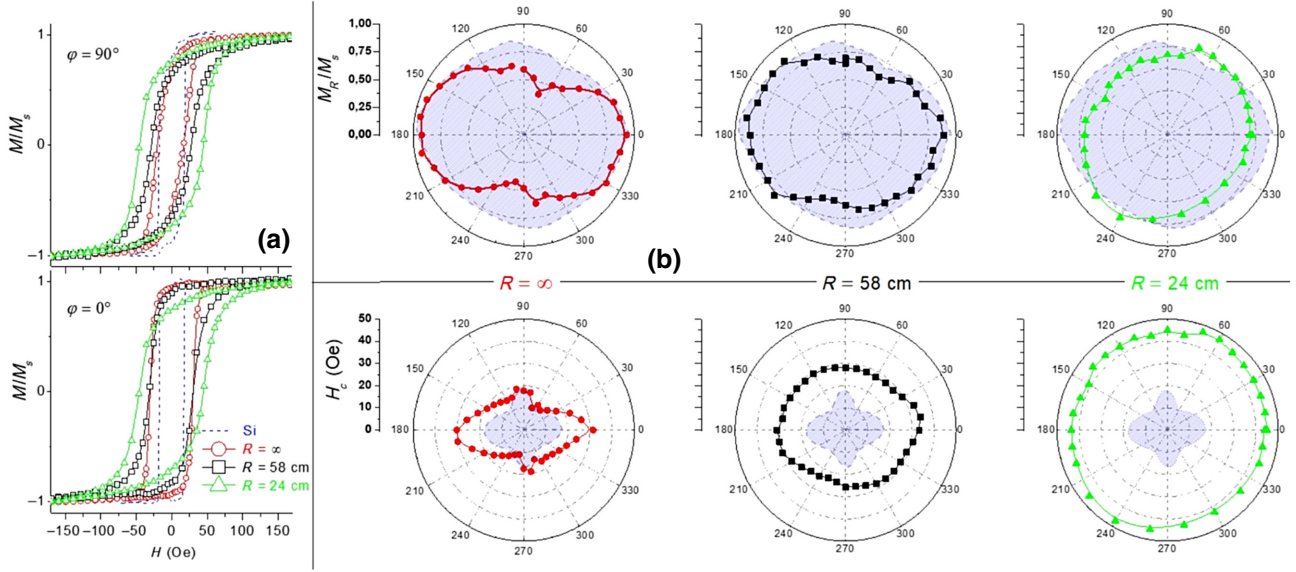


FIG. 6. (a) Hysteresis loops measured along $\varphi = 0^\circ$ (bottom) and along $\varphi = 90^\circ$ (top) for Fe₈₁Ga₁₉/Al samples with $R = \infty$ (open red circles), $R = 58$ cm (black squares), and $R = 24$ cm (green triangles), and also for the Fe₈₁Ga₁₉/Si sample (blue dashed line). (b) Angular dependence of H_c (bottom row) and M_R/M_s (top row) for Fe₈₁Ga₁₉ on Al foils, for $R = \infty$ (left column), $R = 58$ cm (middle column), and $R = 24$ cm (right column). The shaded areas are a guide to the eye to show the physical quantities (i.e., H_c or M_R/M_s) obtained from FeGa^{Si}.

$\vec{e}_{ua}$, which defines the uniaxial EA. The F domain also possesses fourfold (i.e., cubic) anisotropy axes. Finally, it possesses a random anisotropy, to allow one to consider the presence of a randomly distributed anisotropy axis [59–61] or a rotatable anisotropy axis [62–64]. The EA is located at an angle φ relative to the external field $H\vec{e}_x$. The EA is parallel to one of the cubic easy axes. The total free energy $\mathcal{F}$ per unit volume considered in the model is then given by adding the Zeeman and anisotropy energy densities:

$$\mathcal{F}(\theta) = -\mu_0 H M_F \cos(\theta + \varphi) + K_2 \sin^2(\theta) + K_4 \sin^2(\theta) \cos^2(\theta) + K_r \sin^2(\theta + \varphi). \quad (1)$$

Here, K_2 is the uniaxial anisotropy constant, K_4 is the cubic anisotropy constant, and K_r is the random anisotropy constant. Using the parameters presented in Table III, this model reproduces the angular dependence of H_c , including the elongation phenomenon, as shown in Fig. 7. The difference between the parameters for FeGa^{R=∞} and FeGa^{Si} lies in an enhancement of the value of the random anisotropy constant and the appearance of a uniaxial constant for FeGa^{R=∞}. The FeGa^{Si} thin film is a polycrystalline thin film with a (110) preferential orientation [48]. The (110) preferential orientation provides a cubic character to the anisotropy. However, as this orientation is only preferential, it has a random character related to the polycrystalline nature. This crystallographic texture, related to the phenomenon of anisotropy in Fe₈₁Ga₁₉ thin films, was shown and discussed in greater detail in Ref. [48]. Our theoretical model provides a quantification of the anisotropy constant

related to this phenomenon. A random character in the anisotropy cannot induce an elongation, due to symmetry considerations (as it is not angle dependent). Thus, the elongation phenomenon is driven by the development of a uniaxial anisotropy in the samples on Al foils. Also, the angular dependence of M_R/M_s for FeGa^{R=∞} is elongated along $\varphi = 0^\circ$ relative to that for FeGa^{Si}. This confirms the development of a uniaxial character for FeGa^{R=∞}. These modifications of the character of the anisotropy for the Fe₈₁Ga₁₉ layer, such as an enhancement of the uniaxial constant of Py observed on the same surface, support a uniaxial mechanism driven by the substrate morphology, i.e., the grooved morphology of the Al foils. Furthermore, a previous study of Fe₈₁Ga₁₉ thin films grown on a wrinkled surface also identified surface morphology as a driving mechanism for the anisotropic properties [65]. Our study shows that Al foils with grooved surfaces may be used to develop a uniaxial character in an Fe₈₁Ga₁₉ thin film.

It should be noted that the anisotropy-constant calculations presented here are performed with a model assuming a coherent rotational MRev. Such a coherent rotational model is an appropriate approximation when the activation

TABLE III. Physical parameters used in the simulations.

	$M_F = 920 \text{ kA m}^{-1}$		
	$K_2 \text{ (kJ m}^{-3}\text{)}$	$K_4 \text{ (kJ m}^{-3}\text{)}$	$K_r \text{ (kJ m}^{-3}\text{)}$
FeGa ^{Si}	0.000	0.246	0.608
FeGa ^{R=∞}	0.280	0.246	0.950

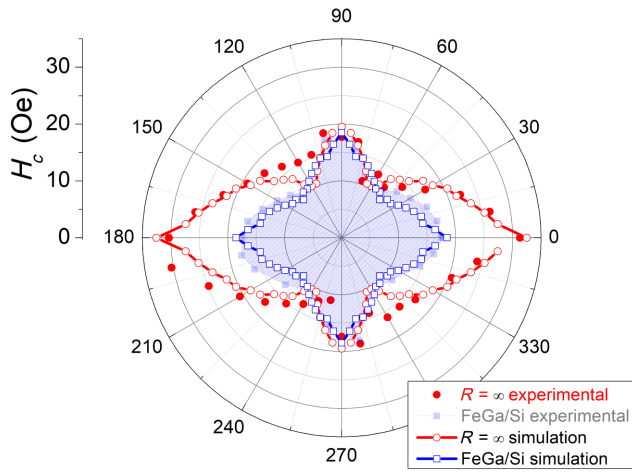


FIG. 7. Experimental (filled red circles) and simulated (open red circles) angular dependences of H_c for $\text{FeGa}^{R=\infty}$, and experimental (filled gray squares) and simulated (blue squares) angular dependences of H_c for FeGa^{Si} . The shaded area is a guide to the eye to indicate the H_c obtained from FeGa^{Si} .

fields of domain walls do not differ strongly from the critical fields estimated by the macrospin model. In Py thin films, it is often observed that the coherent rotational model does not hold, as it overestimates activation fields at and around the easy axis. In our study, we find that a coherent approximation involving uniaxial, cubic, and random anisotropies is not sufficient to describe the angle-dependent MRevs when the samples contain a Py layer.

When MMS is applied and increased in magnitude, the elongations observed in the angular dependence of H_c and M_R/M_s vanish. In particular, the shape of the angular dependence of H_c becomes increasingly more circular, as shown in Fig. 6(b). The combined circular shapes of M_R/M_s and H_c are characteristics of a random distribution of anisotropy axes. Here, these phenomena are observed and indicate that the character of the anisotropy becomes randomized by the MMS. This is confirmed by very similar loops observed along 0° and 90° for $\text{FeGa}^{R=24 \text{ cm}}$, as shown in Fig. 6(a). It should also be noted that the H_c values increase when MMS is applied, indicating an increase in the magnitude of the anisotropy constant. These observed effects of MMS on the angular dependences are irreversible: the properties of the angular dependences remain constant after the bending has been performed. The sample, after bending and being flattened back again, does not exhibit the initial MRev properties (i.e., those of $\text{FeGa}^{R=\infty}$) but retains the properties observed when on the lenses.

To summarize the $\text{Fe}_{81}\text{Ga}_{19}$ results, the 10-nm thin film grown on Si exhibits a cubic anisotropy, which is maintained when it is grown on a flat Al foil. The coercivity observed for $\text{Fe}_{81}\text{Ga}_{19}$ grown on Si is remarkably increased in the direction parallel to the Al grooves. This indicates

a uniaxial anisotropy induced by the striped surface morphology of the Al. A MMS applied to the flexible system results in a progressive increase in the random character of the anisotropy towards complete randomization for the greatest curvature. Also, MMS results in coercivity enhancements. All of these MMS effects are irreversible, which allows long-lasting MRev properties.

C. Py/ $\text{Fe}_{81}\text{Ga}_{19}$ on Al

In a similar manner to the Py and $\text{Fe}_{81}\text{Ga}_{19}$ thin films, the MRevs in Py/ $\text{Fe}_{81}\text{Ga}_{19}$ along and perpendicular to H_{dep} depend on the nature of the substrate. The coercive and saturation fields are larger for the Al foil than for the Si substrate, as shown in Fig. 8(a). The MRevs are modified when MMS is applied. Indeed, the MRev loop shapes, coercivities, and saturation fields are very different between $\text{Bilayer}^{R=\infty}$, $\text{Bilayer}^{R=58 \text{ cm}}$, and $\text{Bilayer}^{R=24 \text{ cm}}$. This indicates that magnetostrictive effects are important for the bilayers. These observed effects of MMS on the angular dependences for the bilayer are irreversible.

To further understand these phenomena, the angular dependences of H_c and M_R/M_s are probed and are shown in Fig. 8(b). The angular dependence of H_c exhibits a butterfly shape, and M_R/M_s exhibits well-defined symmetrical lobes when no MMS is applied (i.e., for $R = \infty$). These characteristics show that the MRev in $\text{Bilayer}^{R=\infty}$ is driven by a uniaxial character, as observed for $\text{Bilayer}^{\text{Si}}$. An enhancement of the uniaxial anisotropy constant of the bilayer is observed when the bilayer is grown on an Al foil, as shown by greater H_c values for $\text{Bilayer}^{R=\infty}$ than for $\text{Bilayer}^{\text{Si}}$. When MMS is applied and increased in magnitude, the angular dependence of H_c evolves from a butterflylike shape to an ellipsoidlike shape. This indicates the development of a random anisotropy. This random-anisotropy effect under MMS is confirmed by the evolution of the angular dependence of M_R/M_s , which exhibits a loss of the two-lobe geometry upon MMS. It should be noted that the coercivity enhancement due to MMS is strongly angle dependent here and shows a minimum along the Al stripes. Also, angle-dependent circular shapes are not observed, whereas they are present in the $\text{Fe}_{81}\text{Ga}_{19}$ case. The observed randomization due to MMS, i.e., the inverse magnetostrictive effect, is not as effective for the bilayer as it is for a single $\text{Fe}_{81}\text{Ga}_{19}$ film. These observed effects of MMS on the angular dependences for the bilayer are irreversible.

To summarize the results for the hybrid system, the 10-nm thin film grown on Si exhibits a uniaxial anisotropy, which is maintained when it is grown on the Al foil. In a similar manner to the single $\text{Fe}_{81}\text{Ga}_{19}$ layer and the single Py layer, the coercive fields observed on Si are increased when the $\text{Fe}_{81}\text{Ga}_{19}$ is grown on flexible Al. When MMS is applied, the hybrid system shows neither the random anisotropy dispersion observed for $\text{Fe}_{81}\text{Ga}_{19}$

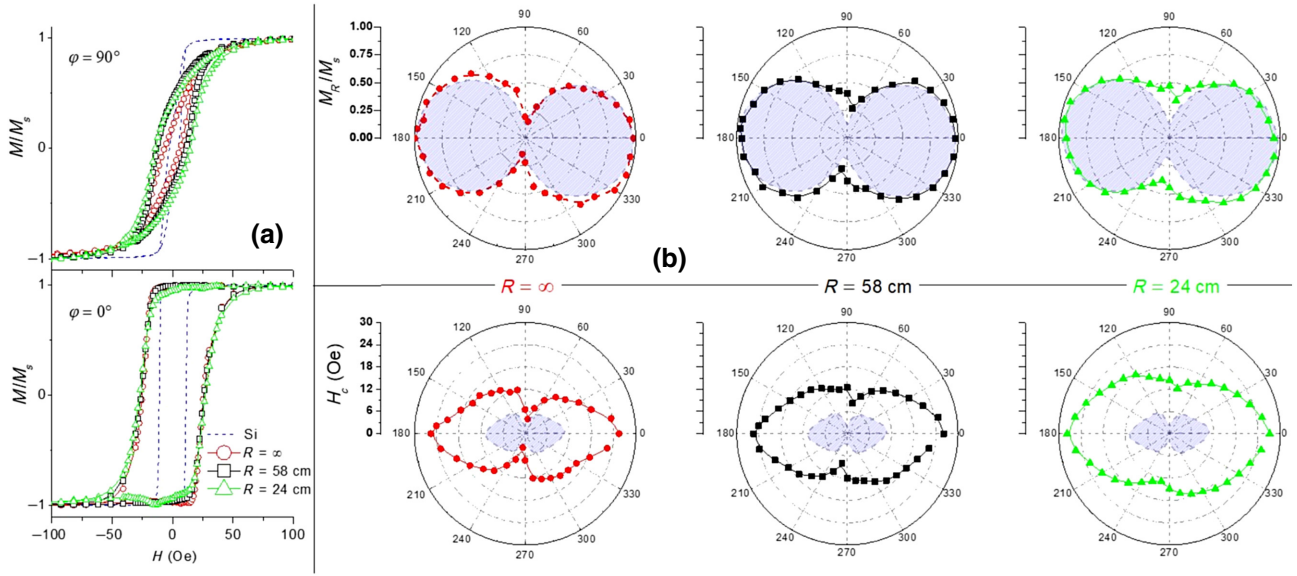


FIG. 8. (a) Hysteresis loops measured along $\varphi = 0^\circ$ (bottom) and along $\varphi = 90^\circ$ (top) for Fe₈₁Ga₁₉/Py/Al samples with $R = \infty$ (open red circles), $R = 58$ cm (black squares), and $R = 24$ cm (green triangles), and also for the Fe₈₁Ga₁₉/Py/Si sample (blue dashed line). (b) Angular dependence of H_c (bottom row) and M_R/M_s (top row) for Fe₈₁Ga₁₉ on Al foils, for $R = \infty$ (left column, filled red circles), $R = 58$ cm (middle column, filled black squares), and $R = 24$ cm (right column, filled green triangles). The shaded areas are a guide to the eye to indicate the physical quantities (i.e., H_c or M_R/M_s) obtained from Bilayer^{Si}.

thin films nor the MMS-independent uniaxial character observed for the nonmagnetostrictive Py, but exhibits a magnetostrictive effect that is strongly angle dependent. The absence of cracks or other structural defects due to the bending and the observed MMS effects presented in this paper should be considered together with the irreversible UMS effects previously reported. These points highlight the key role of permanent plastic residual strain in modifying the magnetization-reversal properties of Fe₈₁Ga₁₉. This is further supported by the unmodified MRevs in the nonmagnetostrictive Py single layer in the presence of bending. Since all these MMS effects are irreversible, this allows long-lasting tailoring of the MRev properties of the bilayer.

V. CONCLUSION

In the study presented in this paper, a 10-nm Fe₈₁Ga₁₉ thin film, a 10-nm Py thin film, and an Fe₈₁Ga₁₉(10 nm)/Py(10 nm) bilayer are grown on Si substrates and on flexible Al foils. Samples grown on flexible Al foils are subjected to MMS through the use of convex optical lenses.

The Py^{Si} sample exhibits a uniaxial anisotropy, with the EA aligned with H_{dep} . Its domain structure around H_c corresponds to large domains separated by sawtooth domain walls. FeGa^{Si} exhibits a cubic anisotropy, with one of the EAs aligned with H_{dep} . Its domain structure around H_c exhibits sharp domain walls with a right-angle geometry. The magnetization reversal in Bilayer^{Si} is similar to

that in Py^{Si} in the sense of a similar domain structure and a uniaxial character, with an EA aligned by and with H_{dep} . Bilayer^{Si} exhibits a greater anisotropy constant than Py^{Si}, and has a reduced coercivity compared with the single Fe₈₁Ga₁₉ layer on Si. The coupling between Py and Fe₈₁Ga₁₉ allows a reduced coercivity and a drastic change in the character of the anisotropy. These characteristics render the hybrid bilayer system promising for applications that need to combine magnetostriction with relatively soft uniaxial magnetic properties.

For all of the layered structures, it is found that the MRevs depend on the nature of the substrate. All layers grown on Al foils show a development or enhancement of the uniaxial character. This effect originates from the surface morphology, with an EA lying along the Al grooves. MMS applied to flexible samples on Al foils results in very different effects as a function of the composition of the layered system. For Py, MMS has no effect, as expected for a nonmagnetostrictive system. For Fe₈₁Ga₁₉, MMS results in a full in-plane randomization of the MRev properties. It increases the anisotropy of the system. For the bilayer, the MMS applied results in a randomization of the anisotropy. However, MMS does not fully randomize the MRev properties, and the bilayer retains a uniaxial character. MMS also results in an increase in the anisotropy. The use of MMS to modify the anisotropic character of Fe₈₁Ga₁₉-based systems is demonstrated here. A wide range of effects can be obtained as a function of the nature of the system and as a function of the MMS strength. All these

effects are irreversible. These MMS effects can be used to tailor and modify the MRev properties of $\text{Fe}_{81}\text{Ga}_{19}$ -based systems in a permanent manner.

The drawback of using Al foils is their metallic nature, which renders some applications difficult, if not impossible, to realize. For instance, the magnetoresistance of nanometric layered systems based on $\text{Fe}_{81}\text{Ga}_{19}$ would be extremely difficult to detect once the system was deposited on a thick metallic layer. To go beyond the findings of our study, further work involving MMS effects with either an insulator as a buffer layer or an insulating flexible substrate could be of interest.

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