

Magnetoelasticity in Fe/GaAs(110) films: Depth profile of magnetic anisotropy

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Despite extensive studies of epitaxial Fe(110) films on GaAs(110), the magnetic properties of films thicker than 20 nm remain largely unexplored, even though this thickness range exhibits rich and unconventional magnetic behavior. In this work, we investigate epitaxial, Ag-capped bcc Fe(110) films with thicknesses ranging from 5 to 100 nm grown by molecular beam epitaxy. Their magnetic and structural properties are characterized using ferromagnetic resonance, vibrating sample magnetometry, x-ray diffraction, and microfocused Brillouin light scattering. Our measurements reveal the coexistence of cubic and uniaxial magnetic anisotropies, that in combination generate an easy axis aligned along the in-plane $\langle 001 \rangle$ directions, which persists throughout the film volume even for thicknesses as large as 100 nm, having a magnitude comparable to that of the cubic anisotropy. To quantify the uniaxial volume anisotropy in thicker films, we fit the experimental data using a layer-resolved magnetic model that allows for a depth-dependent variation of magnetic properties across the film thickness. The precession-amplitude profiles of the perpendicular standing spin-wave modes across the film thickness lead to a depth-selective sensitivity of different modes, enabling the sensing of the local anisotropy landscape, from which a depth profile of anisotropy can be retrieved. X-ray diffraction measurements demonstrate that anisotropic strain—characterized by lateral anisotropic expansion accompanied by vertical compression—is the microscopic origin of the observed in-plane uniaxial anisotropy, persisting even in films with thicknesses up to 100 nm.

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I. INTRODUCTION

Single-crystal magnetic iron films grown by molecular beam epitaxy (MBE) provide an attractive model system and playground for exploring intriguing spin-related phenomena, ranging from anisotropic damping to spintronic effects, with significance for both fundamental studies and technological applications [1–7]. Due to the favorable lattice match of bcc Fe ($a_{\text{Fe}} = 2.87 \text{ \AA}$) with half the lattice parameter of fcc GaAs ($a_{\text{GaAs}} = 5.65 \text{ \AA}$), GaAs is frequently used as a substrate for the growth of high-quality iron films.

Most extensively studied, and best structurally and magnetically characterized, are Fe films grown on the (100) GaAs facet [8–10], even though its polar nature—arising from the truncation of the bulk lattice along a polar crystallographic orientation and the associated net surface charge—promotes various surface reconstructions and terminations, resulting in complex surface structures.

In contrast, the intrinsically nonpolar (110) surface of GaAs is much more stable and not prone to reconstructions. It consists of 50% Ga atoms and 50% As atoms arranged in zigzag chains of alternating Ga and As atoms along the in-plane $\langle 110 \rangle$ directions, with gaps between the ridges along the $\langle 001 \rangle$ directions [11,12]. The first MBE growth of single-crystal Fe films on GaAs(110) was reported in 1981 by Prinz *et al.* [13].

The crystal structures of both GaAs and Fe are shown in Fig. 1. Figure 1(c) shows a sketch of the topmost atomic layer of the (110) plane of the clean GaAs(110) substrate and the Fe film grown on it. The relevant crystallographic directions of the (110) plane are indicated. Note that throughout the paper we denote the family of structurally equivalent crystallographic directions within the (110) plane by $\langle hkl \rangle_{\text{ip}}$, i.e., $\langle 110 \rangle_{\text{ip}}$ includes the $[1\bar{1}0]$ as well as the $[\bar{1}10]$ directions, as shown in Fig. 1.

Early ferromagnetic resonance (FMR) studies [14] on the in-plane magnetic anisotropy of Fe(110) films (terminated either by a native oxide layer or an epitaxial Al cap) revealed a significant uniaxial in-plane anisotropy component $K_{2\parallel}$ that exists in addition to the expected cubic magnetocrystalline anisotropy contribution K_4 . The cubic anisotropy itself promotes three distinguished magnetic axes: the $\langle 001 \rangle$ easy axes (e.a.), the $\langle 110 \rangle$ intermediate axes (i.a.), and the $\langle 111 \rangle$ hard

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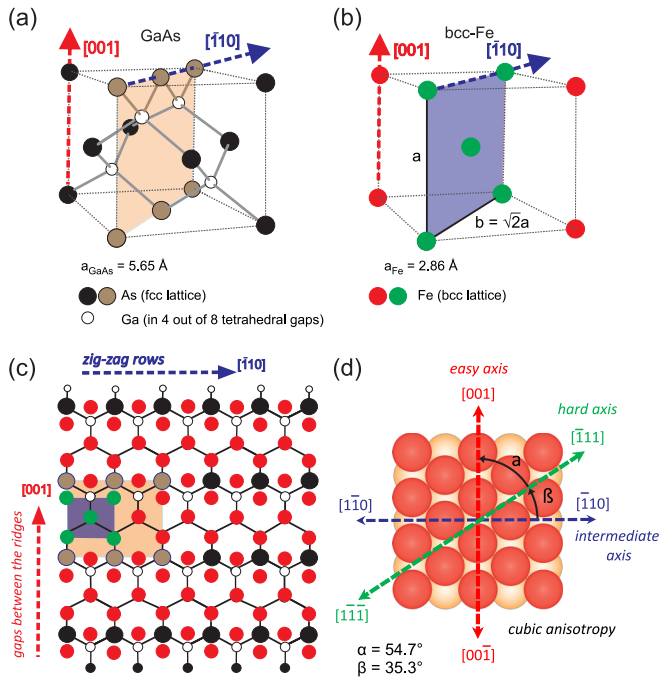


FIG. 1. Unit cells of (a) GaAs and (b) Fe, with the (110) plane highlighted in orange for GaAs and blue for Fe as well as the main crystallographic directions indicated by arrows (red: [001], blue: $[\bar{1}10]$). (c) Atomic arrangement of the (110) surface showing the topmost GaAs layer (black, brown, and white) and the first Fe layer (red and green). (d) Hard-sphere model of the bcc Fe(110) surface, illustrating the cubic axes that define the symmetry of the magnetic anisotropy.

axis (h.a.), cf., Fig. 1(d). Thus, an advantage of using (110)-oriented films is that all three low-indexed orientations are accessible within this plane: $\langle 100 \rangle_{ip}$, $\langle 110 \rangle_{ip}$, and $\langle 111 \rangle_{ip}$.

The presence of a thickness-dependent in-plane uniaxial anisotropy in Fe(110) films was confirmed by subsequent studies [15,16], all of which investigated films no thicker than 20 nm. Remarkably, even for films in the 10–20 nm range, the reported saturation magnetization and cubic anisotropy constants had values below the bulk Fe ones, raising questions about the structural quality of the studied films. Consequently, the absolute values of the reported anisotropy constants should be treated with caution. Nonetheless, the trend of increasing in-plane uniaxial anisotropy with film thickness was consistently observed, indicating that it rather is an intrinsic feature of the Fe(110)/GaAs(110) system.

More than a decade later, the thickness-dependent behavior of in-plane uniaxial and cubic magnetic anisotropies in the Fe/GaAs(110) system was revisited using longitudinal magneto-optic Kerr effect (MOKE) and Brillouin light scattering (BLS) for Fe films in the 2–20 nm thickness range [17]. Magnetization curves along different in-plane crystallographic directions were monitored *in situ* during deposition, revealing a continuous evolution of the net anisotropy axes with increasing film thickness. *Ex-situ* BLS measurements on Fe(110) films with a protective Cr-cap layer confirmed the presence of both cubic and uniaxial contributions to the overall in-plane magnetic anisotropy. The two anisotropies were found to be of similar magnitude but

exhibited distinct thickness dependencies. While the cubic anisotropy approached the bulk Fe value for $d_{Fe} \gtrsim 20$ nm, the uniaxial anisotropy changed sign from negative in thin films to positive in thicker films and continued to increase for Fe thicknesses beyond 20 nm [17].

Reports based on magnetization curves indicated that the hard axis orientation evolves continuously from $\langle 001 \rangle_{ip}$ in the thin-film regime towards $\langle 111 \rangle_{ip}$, and possibly $\langle 110 \rangle_{ip}$ in thicker films [9]. Simultaneously, the easy axis switches abruptly from $\langle 110 \rangle_{ip}$ in thin films to $\langle 001 \rangle_{ip}$, once the films exceed a critical thickness.

The observed thickness dependence of magnetization and cubic anisotropy was attributed to the incorporation of As atoms into the Fe matrix, which modifies the electronic structure of Fe and induces additional strain through occupation of the face-centered sites in the bcc lattice [17]. However, the continuous shift of the hard axis from $\langle 111 \rangle_{ip}$ towards $\langle 110 \rangle_{ip}$ could not be satisfactorily explained [17]. Notably, even for Fe films as thick as 20 nm, the in-plane uniaxial anisotropy did not reach saturation—a surprising finding given the previously proposed interfacial/surface origin of the uniaxial anisotropy in the work of Prinz *et al.* [14] and the reported absence of strain in 20-nm-thick Fe(110) films [13].

No subsequent studies were carried out to investigate this unexpected phenomenon or to elucidate the origin of the thickness-dependent uniaxial anisotropy and its implications on the magnetic properties of even thicker epitaxial Fe(110)/GaAs(110).

A few years later, Höllinger *et al.* observed a similar thickness-dependent magnetic behavior in Au-capped epitaxial Fe(110) films with $d_{Fe} = 0.8$ –12.8 nm using alternating gradient magnetometry. For films thinner than 24 monolayers (ML), i.e., below 4.8 nm, the $\langle 110 \rangle_{ip}$ directions were found to become the easy axes [18].

From the thickness dependence of the effective in-plane anisotropy constants—the cubic contribution $K_{4||}^{eff}$ and the uniaxial contribution $K_{2||}^{eff}$ —the authors extracted volume (K^{vol}) and interface (K^{int}) contributions to each anisotropy via the, by that time widely used, relation:

$$K^{eff} = K^{vol} + \frac{K^{int}}{d_{Fe}}, \quad (1)$$

where d_{Fe} denotes the Fe(110) film thickness. Interestingly, the resulting volume contribution to the effective uniaxial anisotropy was relatively high ($K_{2||}^{vol} = 2.7 \times 10^4$ J/m³) in comparison to its interface contribution ($K_{2||}^{int} = 1.4 \times 10^{-4}$ J/m²) [18]. Moreover, even for films as thick as 12.8 nm, the value of the effective in-plane uniaxial anisotropy constant was only slightly lower than the effective cubic anisotropy constant. Thus, the presence of a significant uniaxial anisotropy in thick films reported already by Gester *et al.* [17] was confirmed by Höllinger *et al.* [18]. However, they again did not explore this observation further towards larger film thicknesses, focusing once more only on the re-orientation of the easy axis in the thin-film regime.

Interestingly, the presence of in-plane uniaxial anisotropy was also reported for epitaxial Fe(110) films grown on a number of substrates other than GaAs(110), such as Cu(110) for Fe film thicknesses of up to 10 nm [19], W(110) for Fe

thicknesses of up to 22 nm [20], and a Mo(110) buffer layer for Fe film thicknesses of up to 37 nm [21]. For the latter system, i.e., Fe/Mo(110), the magnitude of in-plane uniaxial anisotropy decreased with increasing Fe thickness, leveling off but not vanishing even for the thickest films. This behavior was explained in terms of anisotropic strain relaxation in as-grown Fe films [21]. These results are in good agreement with the findings of Clemens *et al.* [22], who directly measured a strong thickness dependence of the in-plane anisotropic strain (the out-of-plane strain was not measured) in up to 40-nm-thick Fe films grown on Mo(110). Notably, the calculated magnetostrictive anisotropy constants derived from the measured strain exhibited a linear dependence on the reciprocal Fe film thickness. As a result, they could easily be mistaken for a surface anisotropy [22].

In summary, the literature on the Fe/GaAs(110) system indicates the following:

- (1) Only a very limited number of studies exist, particularly when compared to the Fe/GaAs(100) system.
- (2) A sizable in-plane uniaxial anisotropy has been consistently observed [15–18], which is surprising even at the maximum investigated film thickness of ~ 20 nm. Such uniaxial anisotropies in Fe films are often interfacial in nature and would therefore be expected to vanish in thicker films. The presence of this in-plane anisotropy in Fe(110) films appears to be a more general phenomenon, not restricted to GaAs(110), as indicated in the previous paragraph [19–21]. Our work focuses exactly on how and why the unexpected in-plane uniaxial anisotropy develops in thicker Fe films.

In this work, we report on FMR, vibrating sample magnetometry (VSM), and microfocused Brillouin light scattering (μ BLS) [23] measurements on epitaxial Fe(110) films with thicknesses ranging from 5 to 100 nm grown on GaAs(110). We determined the effective magnetization, in-plane cubic anisotropy, and the in-plane as well as out-of-plane uniaxial anisotropy fields. To elucidate the origin of the unusual in-plane uniaxial anisotropy, which according to our results contributes significantly to the magnetic behavior even in 100-nm-thick films, all samples were thoroughly characterized using x-ray diffraction (XRD) to extract the lattice strain. All experimental details are explained in Sec. II followed by the introduction of the two fit models in Sec. III. Section IV presents the experimental results with the anisotropy modeling and analysis, and discusses the microscopic origins. The two Appendixes contain further theoretical details about our layer-resolved magnetic properties (LRMP) model and the derivation of the magnetoelastic anisotropy in the macrospin approximation for a cubic (110) thin film.

II. EXPERIMENTAL DETAILS

The epitaxial Fe(110) films studied in this paper were prepared on GaAs(110) wafers based on recipes of [24] and references therein. After cleaning them with acetone and then isopropanol in an ultrasonic bath, the substrates were transferred into an ultrahigh vacuum chamber with a base pressure of 5×10^{-11} mbar. Subsequently, they were heated for 2 hours at 100 °C, followed by 1 hour of Ar sputtering with an energy of 500 eV. Afterwards, the substrates were annealed for 30

minutes at 577 °C and then Ar-sputtered for 30 minutes with 500 eV. The final annealing took 45 minutes at 627 °C.

Low-energy electron diffraction (LEED) imaging of the GaAs substrate after the whole cleaning process revealed a well-ordered unreconstructed (110) surface—see Supplemental Material Fig. S1(a) at [25]. The Fe films were deposited with rates of 0.21 Å/s on the as-prepared substrates held at 200 °C to achieve epitaxial growth. The pressure during the evaporation was 1×10^{-8} mbar. The Fe film thickness was monitored using a quartz crystal balance. It was calibrated by the help of reference films, whose thicknesses were determined accurately by x-ray reflectometry (XRR) measurements.

LEED images of the Fe films show a rectangular structure of four spots with an aspect ratio of $\sqrt{2}$, reflecting the elementary cell of a bcc(110) surface, see Supplemental Material Fig. S1(b) [25]. To protect the Fe films from oxidation they were capped with a 5-nm-thick Ag layer (immiscible with Fe) deposited at the rate of 0.16 Å/s at 100 °C.

We probed the magnetic properties of the Fe films using FMR, μ BLS, and VSM. The FMR was measured using a broadband vector-network-analyzer (VNA), where the sample was placed flip-chip on top of a coplanar waveguide [26]. The absolute value of the microwave transmission parameter S_{21} was recorded as the FMR signal, while the magnetic field $\mathbf{B}_0$ was swept at constant frequency. Such field-sweeps were either measured for frequencies between 0.5 and 40 GHz with the field along the [001] or $[\bar{1}10]$, or at 20 GHz while varying the in-plane field angle φ_B from 0° to 360°, respectively. Afterwards, the spectra were fitted with complex Lorentzian functions to retrieve the resonance fields B_{res} . Finally, the magnetic anisotropy parameters were determined by fitting the resonance equation to the angle- and frequency-dependent FMR data sets.

We determined the saturation magnetization M_s using a commercial VSM from Microsense.

The μ BLS allowed for detecting several higher order PSSW modes that were not visible in FMR and was used to extract the exchange constant values A_{ex} . Note that PSSW modes are exchange dominated. The higher the mode number, the more important the exchange becomes.

The XRD measurements were carried out to elucidate the crystal structure of the epitaxial Fe(110) films and determine their strain state. We employed a Rigaku SmartLab diffractometer with a rotating Cu anode operated at 45 kV and 200 mA. It is equipped with a two-bounce Ge(220) monochromator to select the $K\alpha_1$ line ($\lambda = 0.15406$ nm) and a five-circle goniometer, which allows the HyPix-3000 detector to move horizontally (within the sample surface plane) along the so-called $2\theta_\chi$ axis. The $2\theta/\omega$ and $2\theta_\chi/\varphi$ scans were fitted with Rigaku's analysis software SMARTLAB STUDIO II to determine the predominant lattice plane spacing and thus the strain along three orthogonal directions.

III. THEORETICAL BACKGROUND

To extract the magnetic parameters from the FMR results, we need to model the high-frequency response due to magnetic resonance of the thin film samples. This was done in two ways:

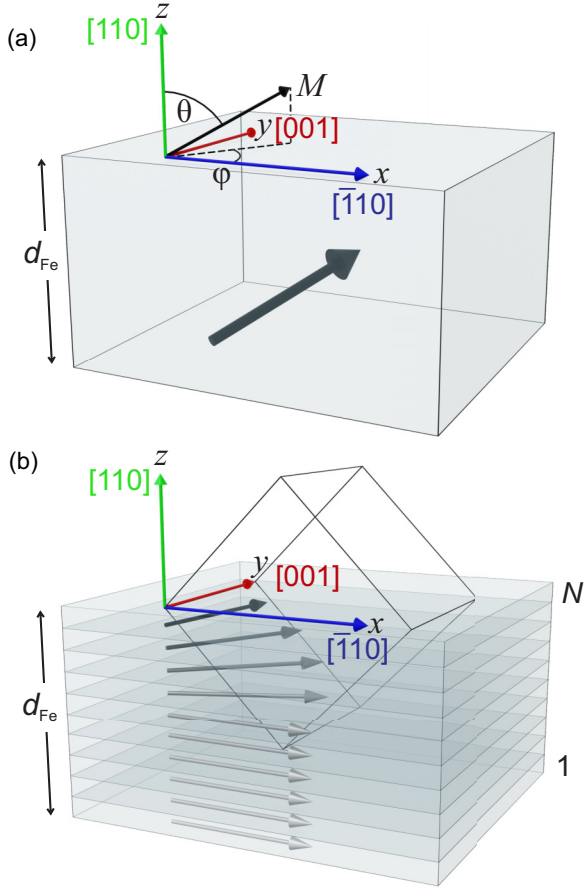


FIG. 2. Sketch of the two model systems with the (x, y, z) reference frame, that is defined by the main crystallographic directions of the (110)-oriented film. (a) Macrospin model with θ and ϕ defining the direction of $\vec{M}$. (b) LRMP model, for which the film is divided into N sublayers. The gray arrows represent each layer's equilibrium magnetization orientation, which varies along the film thickness d_{Fe} . The cube drawn by thin black lines illustrates the cubic unit cell, which is rotated towards the (110)-orientation.

(1) By assuming that the thin film's magnetization reacts as a “macrospin”, i.e., in a manner that all magnetic moments are rigidly locked and respond in-phase to the radio frequency drive. Due to the $80\ \mu\text{m}$ width of the signal line of the coplanar waveguide used in our FMR setup, it is a good approximation to assume that only modes with an in-plane wave vector close to zero are excited. Along the film's normal direction, due to the finite thickness of the Fe(110) film in a range well below the spin-wave propagation length, the wave vector perpendicular to the film surface becomes discrete and PSSW can form. Thus, the assumption of a macrospin is limited to the thicknesses, for which the exchange interaction dominates, holding all the spins rigidly locked [as sketched in Fig. 2(a)]. This is the case for films not much thicker than the so-called exchange length given by

$$d_{\text{ex}} = \sqrt{\frac{2A_{\text{ex}}}{\mu_0 M_s^2}}, \quad (2)$$

with the exchange constant A_{ex} in units of energy/length and the saturation magnetization M_s taken from Table I. For our Fe

films we find $d_{\text{ex}} = 3.1(2)\ \text{nm}$, which matches the literature value of $3.0\ \text{nm}$ [27] within error. In fact, if the film is thicker than a few times the exchange length, the magnetization strictly cannot be approximated anymore by a macrospin. For Fe that means that films thicker than about $10\ \text{nm}$ might deviate already from a macrospin approximation. At this thickness nonuniform exchange modes become in principle possible. However, they become spectrally accessible and experimentally observable in films only for much higher thicknesses, where the first PSSW modes fall into the measurable frequency range. For Fe we observe the first PSSW mode at a thickness above $30\ \text{nm}$, and so it is expected that a macrospin approximation renders useful up to this thickness limit only.

(2) By considering layer-resolved variations of the in-plane and out-of-plane uniaxial anisotropies along the Fe film thickness, which become increasingly pronounced for thicker Fe samples ($50, 75$, and $100\ \text{nm}$), we extend the theoretical description beyond the uniform-anisotropy approximation (macrospin model). In our LRMP model, based on a dynamic matrix approach [28,29], the Fe film is discretized into $N = 40$ laterally infinite sublayers, each characterized by its own local anisotropy parameters, thereby accounting for depth-dependent variations across the thickness [as sketched in Fig. 2(b)]. The detailed description of this model with all the relevant equations is provided in Appendix A. To reproduce the experimentally observed FMR results, we consider anisotropy profiles with a pronounced surface character, as discussed below. These surfacelike profiles are found to capture the essential features of the experimental data, particularly the shift of the resonance field observed in the thicker samples.

A. Macrospin description of magnetization dynamics for a cubic (110)-oriented thin film with uniaxial anisotropy

The free energy density of a cubic thin film in (110) orientation in an external magnetic field and including uniaxial in- and out-of-plane as well as cubic anisotropies is given by

$$\begin{aligned} F = & -M_s B_0 (\sin \theta \sin \theta_B \cos(\varphi - \varphi_B) + \cos \theta \cos \theta_B) \\ & - \left(\frac{\mu_0}{2} M_s^2 - K_{2\perp} \right) \sin^2 \theta - K_{2\parallel} \sin^2 \theta \cos^2(\varphi - \varphi_u) \\ & + \frac{K_4}{4} \left(\cos^4 \theta + \sin^4 \theta \cdot [\cos^4 \varphi + \sin^2(2\varphi)] \right. \\ & \left. + \sin^2(2\theta) \cdot \left[\sin^2 \varphi - \frac{1}{2} \cos^2 \varphi \right] \right). \end{aligned} \quad (3)$$

Here, the orientation of the magnetization (external magnetic field) vector, with magnitude M_s (B_0), is specified by the polar angle θ (θ_B) and by the azimuthal angle φ (φ_B). The former two angles are measured with respect to the film normal ([110] direction) and the latter angles are measured with respect to the in-plane $[\bar{1}10]$ direction. The term proportional to $K_{2\parallel} = K_{2\parallel}^{\text{me}} + K_{2\parallel}^{\text{other}}$ describes the uniaxial in-plane anisotropy, while the term proportional to $K_{2\perp} = K_{2\perp}^{\text{me}} + K_{2\perp}^{\text{other}}$ describes the uniaxial out-of-plane anisotropy. Both terms are split into a potential magnetoelastic contribution due to film strain (‘me’) and terms having different origins (‘other’), such as surface or magnetocrystalline anisotropy. Since the classical surface—or in general

TABLE I. Magnetic properties of the Fe(110) films. For the 5–30-nm-thick films the FMR data was analyzed using the macrospin model, i.e., uniform magnetic parameters across the films' thickness. For the 30–100-nm-thick films the LRMP model having a profile of the uniaxial in-plane anisotropy along the film thickness was used (values in italics). For comparison, the 30-nm-thick Fe film was fitted with both models. $\mu_0 M_{\text{eff}}$ is the effective magnetization, $2K_{2\parallel}/M_s$ is the in-plane uniaxial anisotropy field, $2K_{4\parallel}/M_s$ is the in-plane cubic anisotropy field, $2K_{2\perp}/M_s$ is the out-of-plane uniaxial anisotropy field, A_{ex} the exchange constant, and $\mu_0 M_s$ the saturation magnetization in units of mT. The g factor for all films is 2.09 within experimental error. Note, the anisotropy field $2K_{4\parallel}/M_s$ changes as $\mu_0 M_s$ varies, but the anisotropy constant $K_{4\parallel}$ is constant.

d_{Fe} (nm)	$\mu_0 M_{\text{eff}}$ (mT)	$2K_{2\parallel}/M_s$ (mT)	$2K_{4\parallel}/M_s$ (mT)	$2K_{2\perp}/M_s$ (mT)	A_{ex} (pJ/m)	$\mu_0 M_s^a$ (mT)
5	1660(10)	1.0(1.0)	44(1)	560(10)		2220(80)
20 ^b	1910(10)	58.2(1.6)	50(2)	180(10)	16.5 ^c	2130(100)
30	1900(10)	62.2(1.2)	47(1)	260(10)		2160(110)
30	1900	59/62.7 ^d	48.9	260		2160(110)
50	2020	45/66.4 ^d	48.9	140/210 ^d	19.0 ^c	2160(110)
75	2050	52/75.4 ^d	49.7	90/130 ^d	17.5	2140(110)
100	2020	48/69.6 ^d	48.7	150/220 ^d	17.5 ^c	2170(120)

^afrom VSM measurements.

^bvalues obtained from averaging over four 20-nm-Fe samples.

^cfrom μ BLS measurements.

^dfirst value denotes the bulk (volume) anisotropy, the second value is an average when a considerable interface anisotropy is considered.

interface anisotropy—scales with the inverse film thickness, it is considered rather small in our films, except for the thinnest one with thickness of 5 nm (see details in the Supplemental Material [25]). Likewise, magnetocrystalline contributions of uniaxial symmetry are expected to be weak in the bulk of a cubic system, such as Fe. As we will show, uniaxial anisotropy indeed results mainly from anisotropic film strain, so that its major contribution is of magnetoelastic nature. In what follows, we will therefore drop the superscript ‘me’ and use $K_{2\parallel}$ and $K_{2\perp}$ to denote the *experimental* magnetoelastic uniaxial anisotropies. In Appendix A it is shown that magnetoelastic contributions enter both uniaxial anisotropies, the in- as well as out-of-plane ones. Moreover, we give explicit expressions for the magnetoelastic anisotropy as function of film strain. Due to the low symmetry of film strain we do not expect magnetoelastic anisotropy of cubic symmetry.

The term $\frac{\mu_0}{2} M_s^2 \sin^2 \theta$ in (3) denotes the thin film's shape anisotropy energy density. It always favors an energy minimum in the film plane (i.e., for $\theta = \pi/2$). The easy axes resulting from the terms proportional to $K_{2\perp}$ and K_4 depend on the sign of the respective anisotropy constant. For $K_{2\perp} > 0$ (< 0) the film normal (film plane) becomes the energy minimum. For $K_4 > 0$ (< 0) the $(001)_{\text{ip}}$ ($(111)_{\text{ip}}$) directions are the easy ones, respectively. We note that the term proportional to $K_{2\parallel}$ describing uniaxial in-plane anisotropy is somewhat specific in the sense that its energetic minima are in principle defined by *two* parameters, $K_{2\parallel}$ and φ_u . While the angle φ_u defines the in-plane *direction* of the uniaxial axis with respect to the cubic $[110]$ direction, the sign of $K_{2\parallel}$ determines, whether this is an uniaxial easy or hard axis. The energy related to the orientation of the magnetization vector within the plane being *perpendicular* to the uniaxial is isotropic, i.e., the uniaxial in-plane term results in an easy plane (for $K_{2\parallel} < 0$) or hard plane ($K_{2\parallel} > 0$) behavior for the plane perpendicular to the uniaxial axis. To avoid confusion and following other work in literature, we define the anisotropy constant to be limited to $K_{2\parallel} \geq 0$ and set the easy in-plane directions by φ_u . Once the equilibrium angles of the magnetization φ_0 and θ_0 are de-

termined by minimizing the free energy density, the resonance condition for a magnetization vector confined to the film plane can be calculated following the approaches developed by Smit and Beljers [30] or the one proposed by Gilbert, who formulated a Lagrange equation for the motion of $\vec{M} = M_s \cdot \hat{e}_M$ [31], where $\hat{e}_M$ is the unit vector describing the magnetization's actual position. The generalized resonance equation based on F derived from those formalisms is given by

$$\left(\frac{\omega}{\gamma}\right)^2 = \frac{1}{M_s^2 \sin^2 \theta_0} (F_{\theta\theta} F_{\phi\phi} - F_{\theta\phi}^2). \quad (4)$$

Here, $\omega = 2\pi f$ with the microwave frequency f , $\gamma = g\mu_B/\hbar$ is the absolute value of the gyromagnetic ratio given by the g factor and the F_{ij} ($i, j \in \{\theta, \phi\}$) are the partial derivatives of F with respect to θ and ϕ , taken at the equilibrium angles θ_0 and φ_0 , respectively. In our case and under the assumption of a magnetization vector confined to the film plane ($\theta_0 = \pi/2$), this equation leads to the following functional dependence between microwave frequency and resonance field B_{res} :

$$\left(\frac{\omega}{\gamma}\right)^2 = [B_{\text{res}} \cos \Delta\varphi + \mu_0 M_{\text{eff}} + c_1 + u_1] \times [B_{\text{res}} \cos \Delta\varphi + c_2 + u_2], \quad (5)$$

with $\Delta\varphi = \varphi_0 - \varphi_B$ and the effective magnetization

$$\mu_0 M_{\text{eff}} = \mu_0 M_s - \frac{2K_{2\perp}}{M_s}. \quad (6)$$

c_1 and c_2 are the so-called stiffness fields due to the cubic anisotropy:

$$c_1 = \frac{K_{4\parallel}}{M_s} (3 \sin^4 \varphi_0 + \sin^2 \varphi_0 - 2) \quad (7)$$

$$c_2 = \frac{K_{4\parallel}}{M_s} (12 \sin^4 \varphi_0 - 11 \sin^2 \varphi_0 + 1). \quad (8)$$

Note that the subscript of K_4 was changed to $K_{4\parallel}$, since all measurements were solely done within the film plane. The

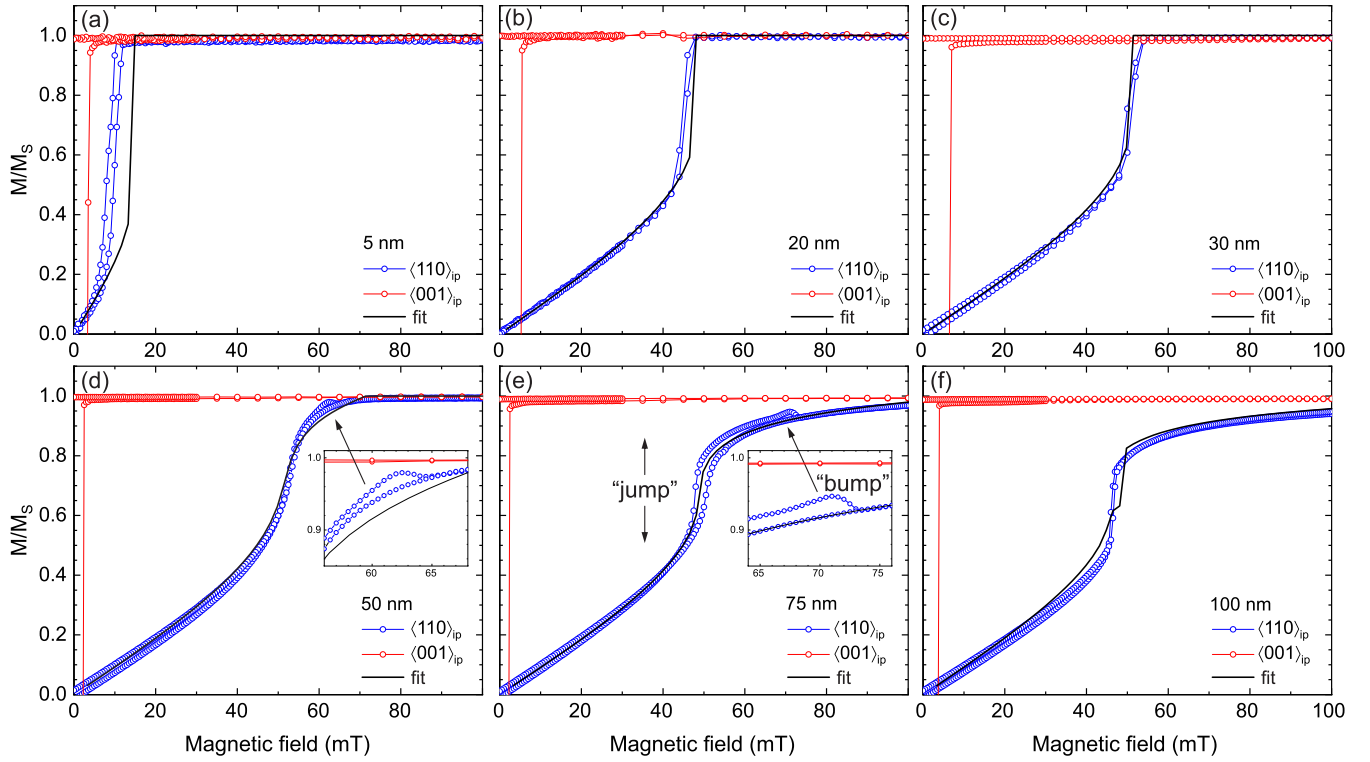


FIG. 3. First quadrant of the in-plane magnetic reversal curves of epitaxial Fe(110) films for various thicknesses measured by VSM. The magnetic field was applied along the $\langle 001 \rangle_{ip}$ (red) and along the $\langle 110 \rangle_{ip}$ direction (blue). Black solid lines show the fits using the LRMP model described in the text and the Appendix A. Note that the sudden field increase of the fit for the 100-nm-thick film is due to a numeric error. The insets on graphs (d) and (e) show a magnification of the “bump”-like part of the magnetization curve along $\langle 110 \rangle_{ip}$.

stiffness fields u_1, u_2 due to uniaxial anisotropy are given by

$$u_1 = \frac{2K_{2||}}{M_s} \cos^2(\varphi_0 - \varphi_u) \quad (9)$$

$$u_2 = \frac{2K_{2||}}{M_s} \cos 2(\varphi_0 - \varphi_u). \quad (10)$$

B. Layer-resolved magnetic properties model

To allow for magnetic modes with nonhomogeneous profile of magnetization during spin precession, we model the system as a set of exchange-coupled magnetic slabs. The individual slabs exhibit homogeneity with respect to their magnetic parameters, however, those parameters are allowed to differ between different slabs. Such an approach enables us to introduce a vertical profile of magnetic parameters into our model. While the detailed mathematical derivation of the LRMP model is given in Appendix A, we will describe its main features and properties in the following. In the model, the Landau-Lifshitz (LL) equation of motion is used to study the FMR response, which is $\dot{\vec{M}}(z, t) = -\mu_0 \gamma \vec{M}(z, t) \times \vec{H}^e(z, t)$, where $\vec{H}^e(z, t)$ denotes the effective field. To account for variations in the magnetic properties along the film normal (z direction), the dynamic matrix method, which partitions the magnetic medium into N ultrathin slabs, is applied [28,29,32–35]. For each slab, the LL equation of motion is formulated and solved for the case of strong exchange coupling between neighboring sublayers, whose magnetic parameters can vary along the film normal. As we will see from our experimental results, the parameters, which vary across

the film thickness, are the in-plane and out-of-plane uniaxial anisotropies. We therefore allow both to vary in the different sublayers. Assuming small oscillations of the magnetization around its equilibrium orientation (dynamic magnetization) and solving the linearized LL equation of motion of the system of coupled sublayers for the dynamic magnetization components of each slab, yields the FMR modes of the system. For details, see Appendix A. In the Appendix, we also verify that the LRMP model and the macrospin model converge to the same result, when the magnetic parameters of all slabs are assumed to be identical.

IV. RESULTS AND DISCUSSION

A. Magnetization reversal loops

The first quadrant of the in-plane magnetic reversal loops measured for the epitaxial Fe(110)/GaAs(110) films is shown in Fig. 3. For the films with thicknesses equal to or above 5 nm, the in-plane easy axis is oriented along the $\langle 001 \rangle_{ip}$ directions, as expected for bulk iron [36]. For the same films, the intermediate axis is oriented along the $\langle 110 \rangle_{ip}$ directions, and the hard axis along the $\langle 111 \rangle_{ip}$ directions. Even for films as thin as 5 nm, the measured saturation magnetization $\mu_0 M_s$ is very close to bulk iron, i.e., $\mu_0 M_s^{\text{Fe bulk}} = 2.15 \text{ T}$ [37] [see also Table I and Fig. 5(b)], indicative of a good quality of the epitaxially grown films.

We found that for a 3-nm-thick Fe(110) film, the anisotropy axes reoriented, i.e., the easy axis became parallel to the $\langle 110 \rangle_{ip}$ directions and the intermediate axis parallel to the

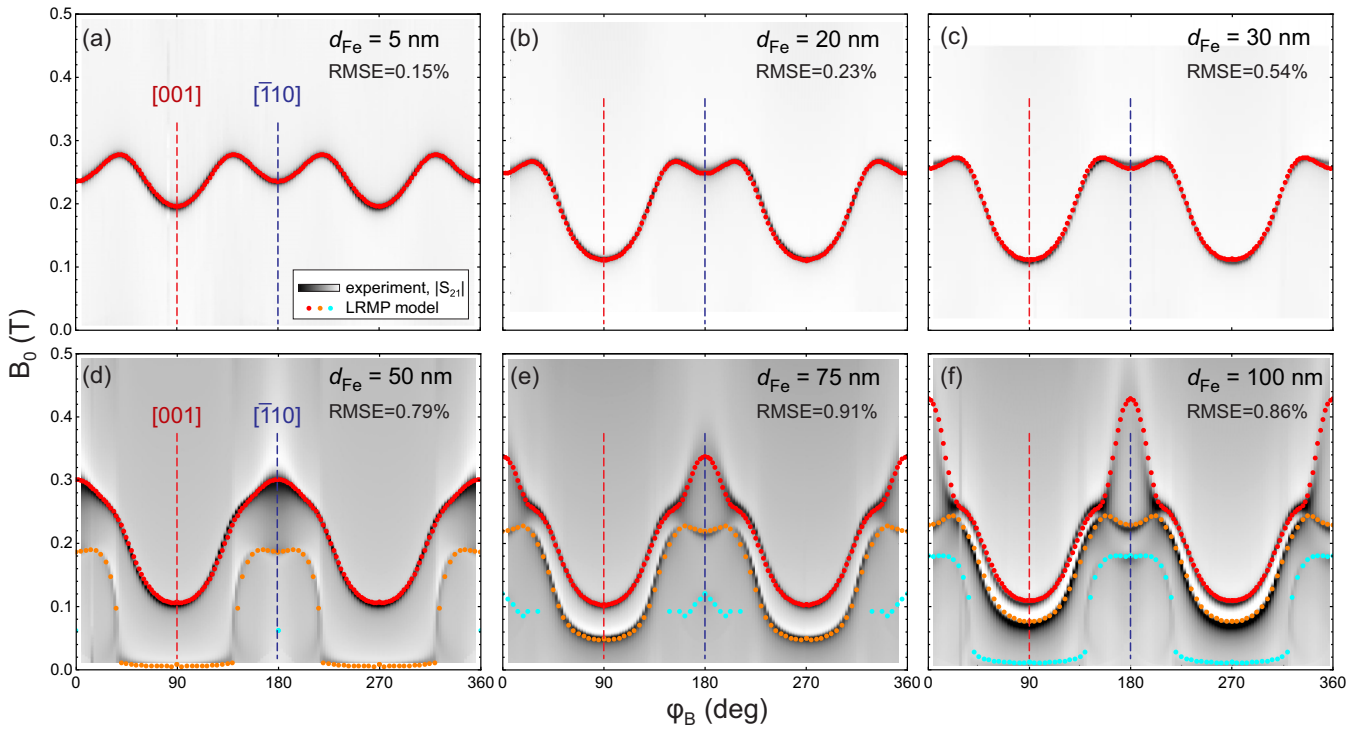


FIG. 4. Azimuthal angular dependence of the resonance field B_0 for the Fe(110) films of various thicknesses: (a) 5 nm, (b) 20 nm, (c) 30 nm, (d) 50 nm, (e) 75 nm, and (f) 100 nm measured at 20 GHz. Experimental results are presented as gray-scale plots. The fits using the LRMP model are given as dotted lines where the various colors are assigned to the various PSSW modes. For thinner films depicted in (a)–(c) the fits of the macrospin model and layer-resolved model are identical. Note: Minima correspond to the directions of magnetically easy axis, maxima to the directions of magnetically hard axis.

$\langle 001 \rangle_{ip}$ directions. This behavior has already been discussed in the literature [14,18,38,39], where this critical reorientation transition thickness was observed between 3 and 5 nm. Accordingly, our FMR results document a sign change of the in-plane uniaxial anisotropy constant $K_{2||}$ from positive values for the 5-nm-thick film towards negative values for the 3 nm film, see the Supplemental Material [25].

Furthermore, a steep distinct step at around 10 mT for the 5-nm-thick film and at 40–55 mT for all other film thicknesses (called further a “jump”) is visible in the magnetization reversal loops in Fig. 3 when the external magnetic field is applied along the intermediate $\langle 110 \rangle_{ip}$ directions. At zero field, the magnetization is oriented parallel to the easy $\langle 001 \rangle_{ip}$ directions, so that no magnetization component is detected parallel to $\langle 110 \rangle_{ip}$. Then, with increasing field parallel to $\langle 110 \rangle_{ip}$ the magnetization coherently rotates from $\langle 001 \rangle_{ip}$ towards the field direction, as reflected by the linear increase of the magnetization component parallel to $\langle 110 \rangle_{ip}$, (see the low field behavior of the blue curves in Fig. 3). At a given field value, which depends on the film thickness, the magnetization component suddenly jumps to a higher value. That “jump” occurs, when the external field provides enough energy to overcome the energy barrier between the $\langle 001 \rangle_{ip}$ and $\langle 110 \rangle_{ip}$ directions, which is given by the cubic anisotropy that has its hard axes parallel to $\langle 111 \rangle_{ip}$ [see Fig. 1(d)]. As can be seen in Figs. 3(a)–3(c), the “jump” appears to be hysteretic, i.e., the sudden increase or decrease of magnetization component parallel to $\langle 110 \rangle_{ip}$ is accompanied by the formation of magnetic domains. Irreversible effect, such as domain formation, cannot

be described by our LRMP model, that assumes a reversible (coherent) magnetization process. Consequently, the fit of the magnetization curve deviates from the experimental behavior towards larger external magnetic field values. This discrepancy becomes less important for thicker films, for which the hysteretic magnetization reversal around the “jump” field reduces. While up to film thicknesses of 30 nm the “jump” ends at saturation, thicker films still require higher fields to gradually saturate the sample magnetization. As we will explain below, the film thickness of 30 nm comprises the limit at which the film can still be described within the macrospin assumption. Once this condition is no longer fulfilled in thicker films, the saturation process becomes increasingly gradual. This gradual approach to saturation, accompanied by a rising saturation field, provides a first indication of a noncollinear magnetization profile across the film thickness.

We note that for the 50- and 75-nm-thick Fe films, the reversal loops measured along the $\langle 110 \rangle_{ip}$ direction exhibit a small “bump” at around 62.5 and 71 mT respectively, manifested as a sudden steep increase in magnetization followed by a decrease upon further increasing the magnetic field. This unusual decrease in magnetization with increasing magnetic field can be best seen in the insets of Figs. 3(d) and 3(e). The mentioned feature is accompanied by an additional hysteresis between the ascending and descending branches of the reversal loop. Notably, the discussed “bump” in the magnetization curves correlates with a characteristic field dependence of the resonance frequency along the $\langle 110 \rangle_{ip}$ direction (intermediate axis) for the 50- and 75-nm-thick films (see Supplemental

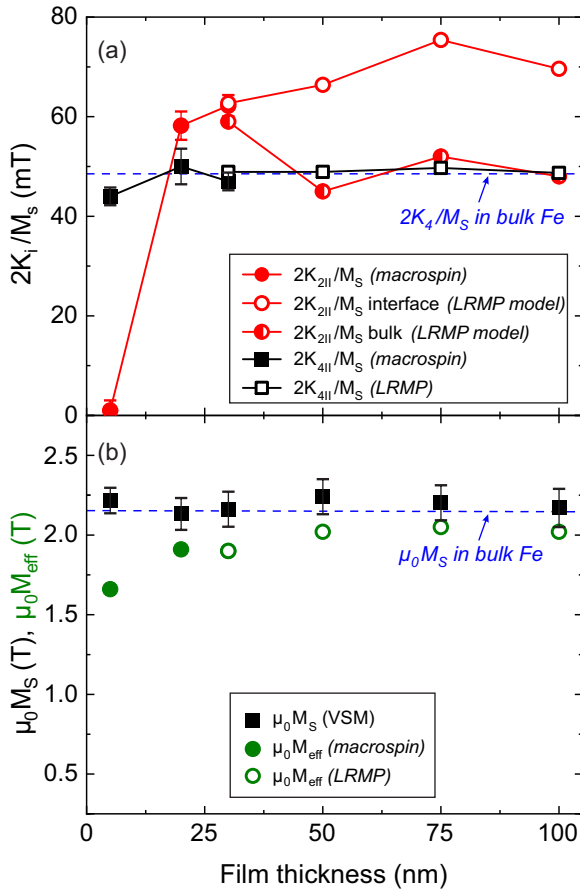


FIG. 5. Dependence of (a) the in-plane uniaxial and cubic anisotropy fields, and (b) the effective magnetization and saturation magnetization on the Fe film thickness, respectively. Error bars are within symbol size. The blue dashed lines mark the Fe-bulk values.

Material Figs. S2(h) and S2(j) at [25]), with both phenomena occurring in the same field range.

At lower fields of about 45 mT, another unusual feature is observed in these two loops. There, the ascending and descending branches of the hysteresis loop cross each other and only later—after the steep part of the loop and the bump—merge again. This implies that the hysteretic area associated with the steep part of the loop (jump) is negative, with the ascending branch lying above the descending branch [40,41]. While in the provided references the negative hysteresis is due to a temperature dependent phase transition from an antiferromagnetic to a ferromagnetic phase in synthetic antiferromagnets [40] and due to domain frustration effects in stripe domain systems [41], in our case the negative hysteresis originates most likely from the complex vertical profile of magnetic anisotropy.

B. Determination of the magnetic anisotropy contributions by FMR

The in-plane dynamic magnetic response of the Fe(110) films was further probed by VNA-FMR. For all thicknesses the azimuthal (in-plane) angular dependence of the ferromagnetic resonance field $B_{\text{res}} = \mu_0 H_{\text{res}}$ at 20 GHz was measured. The results are shown in Fig. 4. The FMR frequency-field

dependencies for the two in-plane crystallographic directions $\langle 001 \rangle_{\text{ip}}$ and $\langle 110 \rangle_{\text{ip}}$ are given in the Supplemental Material Fig. S2 [25]. Figure 4 contains not only the representative set of gray-scale plots of the azimuthal angular dependence of the resonance fields for Fe(110) films of varying thicknesses, but also their corresponding fits (discussed further in the text). Note that the azimuth angle of the applied magnetic field φ_B is defined with respect to the $\langle \bar{1}10 \rangle$ in-plane crystallographic direction.

The thinner films ($d_{\text{Fe}} = 5\text{--}30$ nm) exhibit only one resonance mode (the uniform macrospinlike or Kittel mode), whereas films thicker than 30 nm show additional modes. Those are perpendicular standing spin-wave modes (PSSW modes), for which the dynamic magnetization during the collective precession exhibits vertical variations along the film thickness.

Within the (110) plane we observe the presence of a fourfold anisotropy (magnetocrystalline cubic anisotropy, characteristic of the bcc Fe) as well as a coexistence with a uniaxial twofold in-plane anisotropy. Both have their easy axes along the $\langle 001 \rangle_{\text{ip}}$ directions, but differ with respect to their hard magnetization axes (i.e., $\langle 111 \rangle_{\text{ip}}$ for the cubic and $\langle 110 \rangle_{\text{ip}}$ for in-plane uniaxial anisotropies), in accordance with the literature data [16–18].

The angular dependence of the resonance field *plotted individually* for each anisotropy contribution is illustrated in the Supplemental Material Fig. S3 in [25] using the macrospin approach. As shown in Figs. 4(d)–4(f), the number of detectable PSSW modes increases with increasing Fe thickness. Despite this increased complexity, the first mode (red dotted line) always exhibits a very clear maximum along the $\langle 110 \rangle_{\text{ip}}$, indicating the hard magnetic axis. In contrast, the second mode in this spectral region exhibits a local minimum, which is characteristic of an intermediate magnetic axis. The twofold symmetry (180° periodicity) observed in the angular dependence—manifested as maxima for the first PSSW mode and local minima for the second PSSW mode along $\langle 110 \rangle_{\text{ip}}$ —suggests that the depth-dependent nonuniformity predominantly originates from uniaxial anisotropy contributions. The first mode is influenced more strongly by the uniaxial in-plane anisotropy than the PSSW modes at smaller resonance fields, which are dominated by the cubic crystal anisotropy. This rather peculiar behavior, in turn, indicates that the first mode at highest resonance fields “senses” a different part of the film than the others. As we will explain below with the help of the theoretical model, the high-field PSSW mode has the largest amplitude at the Fe(110)/Ag interface. This means that it is confined to the upper interface of the films (see Fig. 8). Whereas in contrast, the lower-field PSSW modes are confined to the volume region of the films. This scenario in fact allows for employing the PSSW modes as “depth sensors”, from which the vertical profile of magnetic anisotropy can be inferred using the LRMP model.

For the thin Fe(110) films ($d_{\text{Fe}} = 5\text{--}30$ nm), the crystalline cubic in-plane anisotropy constant $K_{4\parallel}$ and uniaxial in-plane anisotropy constant $K_{2\parallel}$ were determined by fitting the macrospin model approximation, see Sec. III A) to the azimuthal angular dependence of the resonance field (Fig. 4) and the frequency-field dependencies for the $\langle 001 \rangle_{\text{ip}}$ and $\langle 110 \rangle_{\text{ip}}$ directions (Supplemental Material Fig. S2 at [25]). The

parameters resulting from this macrospin fit procedure are listed in Table I with nonitalic numbers.

For the thicker films ($d_{\text{Fe}} = 50\text{--}100\text{ nm}$) a fitting procedure based on the LRMP model (see Sec. III B) was employed. This way the whole mode spectra detected by FMR (Fig. 4 and Supplemental Material Fig. S2) as well as by μBLS (Supplemental Material Fig. S5) [25] were perfectly reproduced. The root-mean-square error (RMSE) between experimental data and LRMP model fit is below 1% for all thicknesses and given in Fig. 4. Please note, that in order to test the consistency of both models, the experimental data of the 30-nm-thick Fe(110) film were fitted with both of them, yielding quite similar parameters. The values characterizing the Fe(110) films can be found in Table I and their thickness dependence is visualized in Fig. 5.

Even for the thinner films, the measured saturation magnetization only slightly differs from the saturation magnetization of bulk iron, see Fig. 5(b). This confirms the high quality of the epitaxially grown films. At the same time, the effective magnetization $\mu_0 M_{\text{eff}}$, especially for the thinner films, is lower than $\mu_0 M_s$. This indicates a contribution of a uniaxial out-of-plane anisotropy $2K_{2\perp}/M_s$ contributing to the magnetic landscape of the studied system, according to the definition given in Eq. (6). For the 5-nm-thin films this uniaxial out-of-plane anisotropy field reaches a value as high as 0.6 T, whereas for 100-nm-thick films it is reduced but still not vanishing, as can be seen in Fig. 5(b) from that $\mu_0 M_{\text{eff}}$ is still lower than $\mu_0 M_s$.

The cubic in-plane anisotropy fields $2K_{4\parallel}/M_s$ of thinner Fe films are slightly lower than those of bulk iron. However, they increase rapidly with increasing Fe thickness and reach an almost constant value for films 20 nm thick and thicker. This value deviates only slightly from the bulk value (~ 50 vs 48 mT for bulk Fe [18]). Interestingly, the $2K_{2\parallel}/M_s$ anisotropy field does not saturate even for films as thick as 30 nm and remains nonzero (both bulk and interface contributions) for all thicker films up to the maximum thickness studied. Its magnitude is comparable to that of the cubic anisotropy, whereas no such uniaxial contribution is expected in bulk Fe.

This unusual behavior was previously reported in literature for Fe films grown in (110)-orientation [16–18,42]. Only Rupp *et al.* [43] investigated 150-nm-thick Fe films using BLS and FMR. However, due to the fact that the studied films exhibited cubic anisotropy constant and saturation magnetization well below bulk values, results reported therein have to be treated with caution and cannot be used as reference. The observation of uniaxial in-plane anisotropy for such thick films indicates that it cannot originate from classical interface anisotropy, as it would scale with reciprocal film thickness, and thus, be expected to vanish for thick films.

In contrast, for films with thicknesses below 5 nm [18], it is the classical anisotropy originating from the GaAs/Fe interface that drives the reorientation of the easy axis. It indeed scales inversely with film thickness. As shown in the Supplemental Material [25], the behavior reported in [18] can be reproduced for our film series. The uniaxial in-plane anisotropy located at the GaAs/Fe interface has—consistent with the literature—a negative sign, i.e., it favors the in-plane $\langle 110 \rangle_{\text{ip}}$ axes as easy directions. Consequently, in very thin

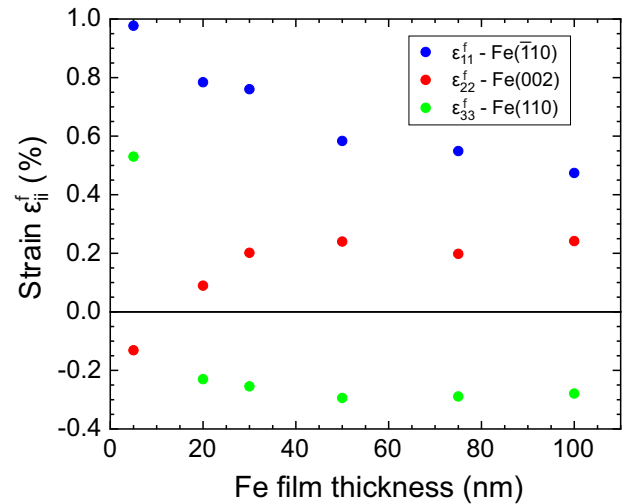


FIG. 6. Thickness dependence of the in-plane and out-of-plane lattice strain of the Fe(110) films deduced from the XRD measurements. The reference value for the lattice constant of bcc Fe taken from [44] is 2.8664 Å. The error bar of the determined strain is $\pm 0.05\%$.

Fe-films, where this contribution becomes dominant, the easy axis reorients to $\langle 110 \rangle_{\text{ip}}$.

Unlike the uniaxial in-plane anisotropy originating from the GaAs/Fe interface, the uniaxial in-plane anisotropy observed in the very thick films is *positive* in sign, i.e., favoring the $\langle 001 \rangle_{\text{ip}}$ axes. Furthermore, it shows a relatively weak dependence on the film thickness (see Table I). Taken together, these observations indicate that its physical origin is distinct from that of the GaAs/Fe interface-related anisotropy.

C. Strain and magnetoelastic anisotropy analysis

Motivated by the studies of Fraune *et al.* [21] and Clemens *et al.* [22], who tried to correlate the strain in Fe films grown on Mo(110) buffers with the in-plane uniaxial anisotropy observed therein, we have undertaken an x-ray diffraction study to quantify the strain along *all three* principal directions, i.e., along the two in-plane directions $[\bar{1}10]_{\text{ip}}$ and $[002]_{\text{ip}}$ as well as along the perpendicular out-of-plane $[110]$ direction. Our measurements focus on the $(\bar{1}10)$, (002) , and (220) planes of Fe, the first two of which are perpendicular to the film plane and were measured at an incidence angle ω of 0.3° to 0.4° . Our reference for the relaxed state is ICDD's PDF-5+ database card 00-006-0696 with a lattice parameter of 2.8664 Å [44]. The results of the XRD study are summarized in Fig. 6, where the data in three different colors show the thickness dependence of the lattice strain along the $\langle 110 \rangle_{\text{ip}}$ (blue circles), $\langle 002 \rangle_{\text{ip}}$ (red circles), and the out-of-plane $[110]$ directions (green circles).

For the smallest film thickness of 5 nm the $\langle 110 \rangle_{\text{ip}}$ directions exhibits an expansion, whereas the $\langle 002 \rangle_{\text{ip}}$ directions become compressed relative to bulk Fe, which is accompanied by an expansion along the perpendicular $[110]$ direction. At the same time, for films with thickness of 20 nm or larger, the two in-plane directions of the Fe-films are expanded with respect to the bulk lattice, while the perpendicular direction

TABLE II. Uniaxial in-plane and out-of-plane anisotropies calculated from the magnetoelastic model (“me”) in the macrospin approximation for the 5-, 20-, and 30-nm-thick films in comparison to their experimental values (third and fifth column).

d_{Fe} (nm)	$2K_{2\parallel}^{\text{me}}/M_s$ (mT)	$2K_{2\parallel}/M_s$ (mT)	$2K_{2\perp}^{\text{me}}/M_s$ (mT)	$2K_{2\perp}/M_s$ (mT)
5	-16.5	1.0(1.0)	57	560(10)
20	41.9	58.2(1.6)	54	180(10)
30	45.2	62.2(1.2)	56	260(10)

is compressed relative to its bulk value. Remarkably, the film strain persists up to the largest thickness of 100 nm.

To verify our hypothesis that the observed uniaxial anisotropy in the thick films is a direct result of the anisotropic strain detected by XRD, we developed a macrospin model that allows us to calculate the magnetoelastic anisotropies, which are expected in case of nonvanishing strain. The derivation of the corresponding expressions is nontrivial, as (i) the (110) orientation of our thin films deviates from the more often studied (100) orientation, and (ii) our model must account for anisotropic strain with distinct values along all three principal directions. A detailed formulation of the model, together with the derivation of the magnetic anisotropy expressions arising from the anisotropic film strain, can be found in Appendix B.

We start by discussing the magnetoelastic anisotropy *within* the film plane. The anisotropic strain with different values along the two in-plane directions $\langle 110 \rangle_{\text{ip}}$ and $\langle 001 \rangle_{\text{ip}}$ leads to uniaxial symmetry. All three strain components ϵ_{11} , ϵ_{22} , and ϵ_{33} , influence the value of this anisotropy. Its value is given by

$$\text{MAE}_{\text{ip}}^{[\bar{1}10]-[001]} = K_{2\parallel}^{\text{me}} = -B_1 \epsilon_{22}^f - B_2 \left(-\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) + B_1 \left(\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right), \quad (11)$$

with $B_1 = -0.343 \times 10^7 \text{ J m}^{-3}$ and $B_2 = 0.783 \times 10^7 \text{ J m}^{-3}$ being the tabulated values of the magnetoelastic coupling constants for bcc Fe (see [45]). From Fig. 6 it follows that for Fe-thicknesses of 20 nm and above $\epsilon_{11}^f, \epsilon_{22}^f > 0$, $\epsilon_{33}^f < 0$ and $\epsilon_{22}^f > (\epsilon_{11}^f + \epsilon_{33}^f)$. These observations lead to the conclusion that $K_{2\parallel}^{\text{me}}$ is a *positive* quantity, and thus, able to explain the observed uniaxial anisotropy with easy axes parallel to $\langle 001 \rangle_{\text{ip}}$ for thick films. The values of $K_{2\parallel}^{\text{me}}$ according to Eq. (11) are summarized in Table II together with the respective experimental values taken from Table I. The calculated values match the experimental ones surprisingly well. Deviations can be explained by the fact that the magnetoelastic coupling constants B_i in our strained thin films differ somewhat from the tabulated Fe-bulk values that were used in the calculation or that there are other contributions to the overall measured anisotropy $K_{2\parallel}$. Nevertheless, the conclusion can be made that the main contribution to the uniaxial in-plane anisotropy for the thick films is of magnetoelastic origin. The magnetoelastic model predicts a reduction of $K_{2\parallel}^{\text{me}}$ towards the thinnest film of 5 nm thickness. This behavior is a direct consequence of the strain components ϵ_{11} and ϵ_{33} . This means, the film strain along the in-plane $\langle 001 \rangle_{\text{ip}}$ and the orthogonal $\langle 110 \rangle_{\text{ip}}$

directions change their signs. In the very thin film limit the Fe lattice becomes compressed along the $\langle 001 \rangle_{\text{ip}}$ directions and expands along the film normal parallel to [110]. One may speculate that this trend continues for even thinner films, which would consequently change the sign of $K_{2\parallel}^{\text{me}}$. This scenario could indeed play a key role to induce the in-plane reorientation of the easy axis towards the $\langle 110 \rangle_{\text{ip}}$ for very thin films. The fact that the experimental value for $K_{2\parallel}$ has already changed sign for the 5-nm-thick film, can be seen as an indication that on top of the sign change of the magnetoelastic contribution $K_{2\parallel}^{\text{me}}$, a classical (not of magnetoelastic origin) interface anisotropy at the GaAs/Fe interface contributes to the overall $K_{2\parallel}$.

From the film strain evaluated by XRD, one also can calculate the expected uniaxial out-of-plane anisotropy according to (see Appendix B)

$$\text{MAE}_{\text{oop}}^{[001]-[110]} = K_{2\perp}^{\text{me}} = B_1 \epsilon_{22}^f - B_2 \left(-\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) - B_1 \left(\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right). \quad (12)$$

The results of the calculation for thicknesses in the macrospin regime are listed in Table II together with the experimental values. Again, the magnetoelastic model is able to predict the sign of the measured anisotropy correctly. The positive sign implies that the uniaxial out-of-plane anisotropy given by $K_{2\perp}^{\text{me}}$ favors the film normal as easy axis. In contrast to the uniaxial in-plane anisotropy, there is a significant deviation between the calculated and measured value for its out-of-plane counterpart. The measured one is much larger. An indication for the reason of this discrepancy can be inferred from looking to the experimental value $2K_{2\perp}/M_s$ of the thinnest film of 5 nm thickness (see Table II). It is about three times larger than that of the 20-nm-thick film. This in turn indicates a large surface contribution to the overall uniaxial out-of-plane anisotropy, which is diminishing with thickness but still influences the 20- and 30-nm-thick films.

D. Anisotropy profiles and analysis of the PSSW mode profiles using the LRMP model

To elucidate the origin of the resonance field maxima in the azimuthal angular dependence of the PSSW mode at the highest fields [red curve in Figs. 4(d)–4(f)]—or, equivalently, frequency-field dependence of the PSSW mode at the lowest frequencies (see the frequency-field dependence in the Supplemental Material [25]) for magnetic fields applied along or close to the $\langle 110 \rangle_{\text{ip}}$ directions, we need to go beyond the macrospin approach. Specifically, we model the thick Fe films using the LRMP model, which was introduced in Sec. III B and derived in Appendix A, to include a vertical profile of magnetic parameters.

As can be seen from Figs. 4(d)–4(f), the maxima of the FMR resonance field of the PSSW mode at highest fields (first PSSW mode, red curve) exhibit a twofold, i.e., uniaxial symmetry. Thus, it is plausible to expect uniaxial anisotropy fields to be predominantly responsible for its shape. Cubic anisotropy exhibits fourfold symmetry whereas shape anisotropy is isotropic within the film plane. They cannot

account for the observed angular dependencies. As we will show in the following, the angular dependent behavior of all modes can be well explained by considering a vertical profile of uniaxial anisotropy. Focusing in the following on the 100-nm-thick sample, we start by considering different anisotropy profiles of the uniaxial in-plane anisotropy $K_{2\parallel}$ along the film thickness, including smoothly graded and interfacial-like distributions. For a meaningful comparison, all profiles are constrained to have the same thickness-averaged anisotropy value of

$$\left\langle \frac{2K_{2\parallel}}{M_s} \right\rangle = \frac{1}{d} \int_0^d \frac{2K_{2\parallel}(z)}{M_s} dz = 48 \text{ mT}, \quad (13)$$

equal to the experimental value. Note that the dependence of the anisotropy constant with the normal coordinate $K_{2\parallel}(z)$ is accounted through the ν parameter, namely $K_{2\parallel}^{(\nu)} = K_{2\parallel}(z)$, as shown in Appendix A.

Figure 7 summarizes the results for the different vertical profiles of the uniaxial in-plane anisotropy. Figure 7(a) illustrates the anisotropy profiles fulfilling Eq. (13). Figure 7(b) compares the computed angular dependence of the resonances using these vertical anisotropy profiles with the experimental FMR dataset of the 100 nm sample. The key result is that only profiles with a pronounced *interfacial character*, i.e., with a large anisotropy concentrated close to the Fe/Ag interface, and a much weaker one in the inner volume, reproduce the experimentally observed upward shift of the FMR field at the corresponding angular positions (around 0° and 180°) for the first PSSW mode. The smallest RMS deviation of 0.96% between the calculated resonance fields of all three modes and the experimental values was found for the profile shown in violet. However, the profile in red color with a just slightly higher RMSE of 1.02% matches closer on the resonance maxima at 0° and 180° . In contrast to that, the profiles that are close to uniform or smoothly graded fail to account for the magnitude and/or shape of the measured angular dependence of the first PSSW mode, despite having the same average anisotropy $\langle 2K_{2\parallel}/M_s \rangle$ (see, e.g., the greenish profile in Fig. 7 having an RMSE of 1.8%).

Importantly, the comparison also reveals a mode selectivity. The resonances excited at low fields in the angular dependence (referred to as bulk modes) are not notoriously affected by the specific choice of $K_{2\parallel}(z)$, as can be seen in Fig. 7. Their resonance fields and overall angular trends remain nearly unchanged, regardless which $2K_{2\parallel}(z)/M_s$ anisotropy profile was used. This low sensitivity is expected, because bulk modes extend across the entire film thickness, as shown in Fig. 8. In contrast, the resonance occurring at high fields is strongly influenced by the vertical anisotropy profile. This mode is preferentially localized near the film surface/interface, therefore its frequency and/or resonant field is strongly influenced by the local values of $K_{2\parallel}(z)$. Hence, the experimentally observed enhancement of the resonance field is explained by an interfacial-like anisotropy distribution, which increases the effective anisotropy experienced by the surface-localized precession without significantly altering the bulklike excitations.

To fit the experimental frequency-field dependence (see results in the Supplemental Material [25]), the perpendicular

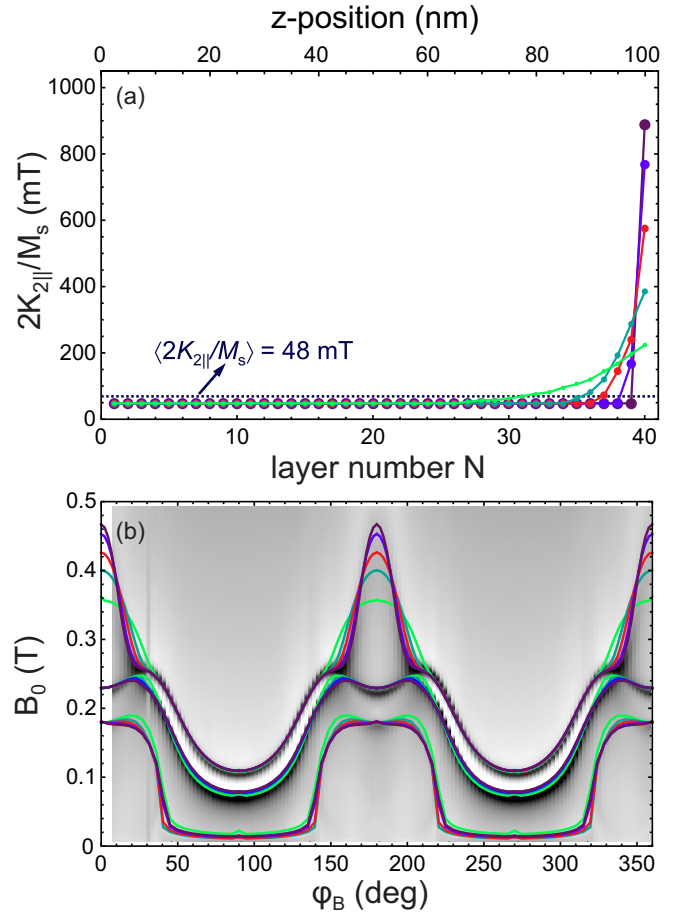


FIG. 7. (a) Representative anisotropy profiles $2K_{2\parallel}(z)/M_s$ along the normal coordinate ranging from a smooth gradual incline over several Fe layers (light green) to a very steep single-step profile at just the uppermost Fe layer (dark violet). They are all constrained to have the same average value $\langle 2K_{2\parallel}(z)/M_s \rangle = 48 \text{ mT}$. (b) Corresponding calculated angular dependence [lines, colors correspond the profiles in (a)] compared to the experimental FMR data (gray scale) of the 100-nm-thick film. Bulk modes (low-field resonances) are weakly affected by the anisotropy profiles. In contrast, the surface mode (high-field resonance) is strongly shifted at 0° and 180° and is best reproduced by interfacial-like profiles (violet, blue, and red curves). The best fit is given by the red curve.

magnetic anisotropy (PMA) field $2K_{2\perp}/M_s$ was also allowed to vary along the film thickness. In particular, its depth dependence was assumed to follow the same spatial profile as the in-plane uniaxial anisotropy, a common physical magnetoelastic origin for both anisotropic inhomogeneities. The fact that magnetoelasticity enters *both* uniaxial anisotropy terms, is expected as can be shown on a theoretical basis (see Appendix B).

E. Microscopic origin of the anisotropic lattice strain

Uniaxial magnetic anisotropies in our iron films and their dependency on film thickness can be to large extent explained by magnetostriction based on the measured lattice parameters along the three fundamental orientations (Fig. 6). Our explanation builds on the pioneering work of Clemens

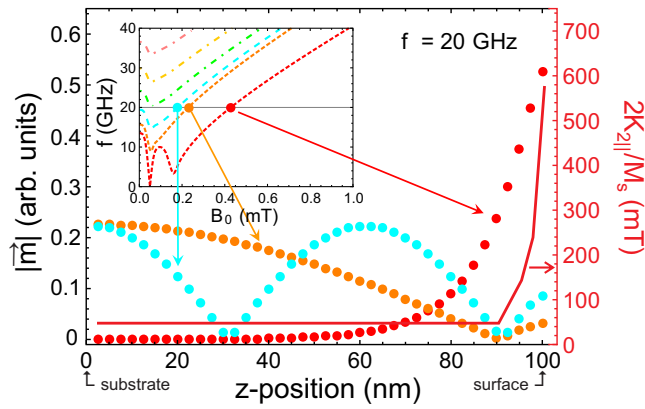


FIG. 8. Representation of the anisotropy profile (red solid line) and three mode profiles (circles) across the Fe(110) film thickness observed for the 100-nm-thick Fe film to describe the magnetization dynamics in such systems. The inset shows the $\langle 110 \rangle_{\text{ip}}$ hard-axis frequency-field dependence.

et al. [22], who investigated Fe(110) films on Mo(110). They likewise observed strong difference in strain between the two in-plane orientations, which persisted up to the maximum examined film thickness of about 40 nm. Our magnetoelastic macrospin model, being valid up to film thicknesses of about 30 nm, demonstrates that the positive sign of the uniaxial anisotropy in these films can be directly related to the experimentally determined anisotropic film strain. It therefore appears natural to assume that the enhanced magnitude of this anisotropy, required to explain the peculiar behavior of the PSSW modes, has the same physical origin. Significantly larger anisotropic strain at the *upper* interface region of the iron films would straightforwardly account for the enhanced uniaxial anisotropy employed in the anisotropy profiles discussed above [see Fig. 7(a)].

In the following we discuss the microscopic origin of the measured lattice strain given in Fig. 6. Our films were deposited at 200 °C and cooled to room temperature for all measurements. Therefore, we need to consider thermal stress in all films. As Fe has a higher thermal expansion coefficient than GaAs [46,47], the iron lattice would contract more strongly during cooling, if this contraction was not partially constrained by the GaAs substrate. This mismatch in thermal expansion can explain an isotropic in-plane tensile strain of approximately 0.1%–0.2%. Such tensile strain should be independent of film thickness, which can partly explain, why all in-plane strain curves are shifted to higher values compared to bulk Fe.

Obviously, at small thicknesses, the strain state is governed primarily by interface effects and elastic anisotropy. The GaAs(110) surface exhibits pronounced in-plane anisotropy, giving rise to direction-dependent interface bonding and surface stress. In particular, Fe-As and Fe-Ga bonds at the interface favor strain accommodation along specific crystallographic directions rather than isotropic in-plane compression [48]. Since even for the low film thickness of 5 nm the saturation magnetization is close to bulk (see Table I), we can rather rule out other effects, e.g., intermixing and oxidation.

For thin films, we additionally consider the lattice misfit with the substrate. A simple comparison of the bulk lattice constants of iron and GaAs reveals an almost 2:1 ratio. The remaining small lattice mismatch corresponds to an in-plane compressive strain of approximately 1.4% along both the $\langle 001 \rangle_{\text{ip}}$ and $\langle 110 \rangle_{\text{ip}}$ directions. In fact, for the thinnest film of 5 nm, we observe such compressive strain along $\langle 001 \rangle_{\text{ip}}$, but a tensile strain along $\langle 110 \rangle_{\text{ip}}$.

All measured strain values are lower in magnitude than the calculated misfit. This indicates that already in the 5-nm-thick films some relaxation occurred, and coherent epitaxial growth is only possible in much thinner films. Indeed, Clemens *et al.* [22], who used a Mo buffer with a much larger misfit of 9.8% and accordingly found much higher strains, observed that relaxation starts already in the subnanometer range. In case of thicker films even both in-plane directions exhibit tensile strain, which remains anisotropic. This is in agreement with the observations of Clemens *et al.* [22], who attributed this to the elastic anisotropy within the Fe(110) plane and the particular orientations of the slip plane and slip orientations, required for misfit dislocations in Fe in this particular orientation. For a more detailed explanation, why a substantial strain can still persist at these large thicknesses, we propose to consider also the interaction between the dislocation networks, which may hinder relaxation due to the reduced symmetry of (110) films.

So far the discussion focused on in-plane strain. In contrast to the in-plane directions, there is no constraint along the out-of-plane one, thus, this direction usually compensates the in-plane stresses. Typically, this occurs by preserving the lattice volume. Indeed, for the 100-nm-thick film the lattice volume is only expanded by about 0.5%. However, this approximation often fails for very thin films, where the electronic influence of the substrate and the free surfaces become significant. Such non-volume-conserving strain states have been reported for example for ultrathin Fe(110) films grown on W(110), where anisotropic interface and surface stresses dominate the elastic response [45,49].

Having said this, we finally return to our hypothesis that the strain is significantly larger at the *upper* interface region of the iron film, providing a direct reason for the enhanced uniaxial anisotropy employed in our anisotropy profiles. As Fe and Ag are also immiscible within the liquid state, we can exclude intermixing between both layers, but the cover layer may still strain the Fe lattice below. Indeed, this could be the reason why even for the maximum thickness we still observe a difference in strain between both in-plane directions. To test this scenario, we varied the angle of incidence in our in-plane x-ray diffraction measurements (not shown here). With decreasing incidence angle, the measurements become increasingly sensitive to the topmost regions of the film. Although this approach did not yield conclusive results for all samples, we indeed observed an increased lattice plane spacing for the 30-nm-thick film along the $[001]_{\text{ip}}$ direction, consistent with an enhanced strain of $\epsilon_{22}^f \approx 0.8\%$ near the film surface (compared to 0.2% otherwise). This local enhancement was observed within the topmost 5–10 nm of the iron film. For a final proof of this explanation, however, further studies are required.

V. CONCLUSION

We studied the thickness-dependent evolution of magnetic anisotropy and magnetization in Ag-capped epitaxial Fe films on GaAs(110), with film thicknesses between 5 and 100 nm. The films grow epitaxially with the [110] aligned parallel to the film normal, while the cubic $\langle 001 \rangle_{\text{ip}}$ and $\langle \bar{1}10 \rangle_{\text{ip}}$ directions lie in the film plane. Lattice parameters along both in-plane and out-of-plane directions were determined by x-ray diffraction.

Magnetic characterization by FMR and VSM reveals that all films, even as thick as 100 nm, exhibit both in-plane and out-of-plane uniaxial anisotropies of comparable or even larger magnitude than the cubic anisotropy. This behavior is explained by anisotropic strain in the films, with expansion along the $\langle 001 \rangle_{\text{ip}}$ and $\langle 110 \rangle_{\text{ip}}$ directions and compression along the film's normal [110].

A magnetoelastic macrospin model reproduces the observed uniaxial anisotropy for intermediate thicknesses (20–30 nm). For the thinnest 5 nm film, a change of sign in the in-plane strain along the $\langle 001 \rangle_{\text{ip}}$ indicates the onset of the reorientation of the in-plane easy axis, occurring for even smaller Fe thicknesses in accordance with previous literature.

Thick films exhibit multiple higher-order PSSW modes, which display a characteristic in-plane angular dependence of the FMR resonance field. This indicates a vertical profile of both in-plane and out-of-plane uniaxial anisotropies, with their enhancement near the topmost film surface (Fe/Ag interface). Theoretical modeling reproduces this profile, linking the increased surface anisotropy to enhanced local strain. This anisotropy gradient (profile) causes a noncollinear spin orientation across the film thickness. Analysis of the PSSW modes allows us to probe the depth-dependent magnetic anisotropy and to quantify the thickness of the surface region with enhanced anisotropy.

In other words, our results demonstrate that VNA-FMR can probe local magnetic properties within the film depth, in contrast to VSM, which only provides access to volume-averaged quantities (see Fig. 3). In contrast to the commonly held assumption that strain relaxation at large thicknesses fully propagates to the film-substrate interface, we find that the magnetoelastic contribution remains active even at 100 nm, similar to observations in epitaxial and textured Co films [50,51]. In the present Fe films, this magnetoelastic contribution varies with depth across the thickness of the epitaxial layers.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

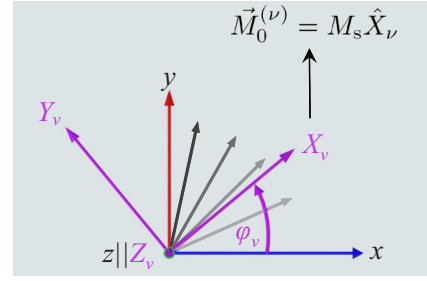


FIG. 9. A top view of the LRMP model system, defining the (X_v, Y_v, Z_v) reference system. In this frame, X_v aligns with the equilibrium magnetization of the ν -th sublayer, while Y_v lies in the film's plane and $z = Z_v$.

APPENDIX A: THEORETICAL TREATMENT OF THICK FILMS: THE LAYER-RESOLVED MAGNETIC PROPERTIES MODEL

In the following we provide the detailed derivation of the layer-resolved magnetic properties (LRMP) model used to describe the FMR response of films thicker than the exchange length. In such cases and unlike the macrospin approximation, the amplitude of the spin precession is not uniform across the film thickness, and thus, can be significantly influenced by a vertical profile of magnetic parameters. In what follows, we consider a cubic (110)-oriented thin film with uniaxial in-plane anisotropy and magnetization confined to the film plane, see Fig. 2(b). The in- as well as out-of-plane uniaxial anisotropy fields can have a nonuniform distribution across the film thickness. Perpendicular standing SW modes were calculated using free boundary conditions, applied at the level of each discretized sublayer within the dynamic matrix method, with any effective pinning arising from the local magnetic energy terms.

As described in the main text, we start from the Landau-Lifshitz equation of motion, which is $\dot{\vec{M}}(z, t) = -\mu_0 \gamma \vec{M}(z, t) \times \vec{H}^e(z, t)$, where $\vec{H}^e(z, t)$ denotes the effective field. The vertical variation of magnetic parameters along the z direction [see Fig. 2(b)] is accounted for by employing a dynamic matrix method, which partitions the magnetic medium into N small ultrathin sublayers. [28,29,32–35]. Therefore, the time evolution of the ν th sublayer is given by $\dot{\vec{M}}^{(\nu)}(t) = -\mu_0 \gamma \vec{M}^{(\nu)}(t) \times \vec{H}^{e(\nu)}(t)$. Thus, the normal dependence is given by the parameter ν , i.e., $\vec{M}^{(\nu)}(t) = \vec{M}(z, t)$. Then, assuming small oscillations of the magnetization around the equilibrium state, the magnetization vector and the effective field are

$$\vec{M}^{(\nu)}(t) = M_{s_\nu} \hat{X}_\nu + \vec{m}(t) \quad (\text{A1})$$

$$\vec{H}^{e(\nu)}(t) = H_{X_\nu}^{e0} \hat{X}_\nu + \vec{h}^e(t), \quad (\text{A2})$$

where

$$\vec{m}(t) = m_{Y_\nu}(t) \hat{Y}_\nu + m_{Z_\nu}(t) \hat{Z}_\nu \quad (\text{A3})$$

represents the dynamic magnetization, M_{s_ν} is the saturation magnetization of the ν th sublayer, and $\vec{h}^e(t)$ is the dynamic field that is directly proportional to $\vec{m}(t)$ in a linearized approach. In the reference coordinate system (X_v, Y_v, Z_v) (see Fig. 9), the X_v axis aligns with the ν th equilibrium

magnetization, the Z_v axis is perpendicular to the magnetic film, and the Y_v axis lies within the film's plane. Thus, assuming that $\vec{m}(t) = \vec{m}e^{i\omega t}$ with ω being the angular frequency, and neglecting second-order terms in $\vec{m}$, the Landau-Lifshitz equation of motion can be expressed as

$$i(\omega/\mu_0\gamma)m_{Y_v} = -m_{Z_v}H_{X_v}^{e0} + M_{s_v}h_{Y_v}^e \quad (\text{A4})$$

and

$$i(\omega/\mu_0\gamma)m_{Z_v} = -m_{Y_v}H_{X_v}^{e0} + M_{s_v}h_{Z_v}^e, \quad (\text{A5})$$

where the equilibrium conditions are given by $H_{Y_v}^{e0} = H_{Z_v}^{e0} = 0$.

As mentioned above, the variation of the dynamic magnetization along the z direction arises from the inhomogeneity of the in-plane and out-of-plane uniaxial anisotropies across the film thickness. Accordingly, we assume that $K_{2\parallel}(z) = K_{2\parallel}^{(v)}$ and $K_{2\perp}(z) = K_{2\perp}^{(v)}$, while the saturation magnetization is taken as uniform, $M_{s_v} = M_s$. By accounting for the different magnetic interactions in the Fe system, the dynamic effective fields can then be written in the general form as

$$h_{Z_v}^e = \sum_{\eta} \Gamma_{Z_{\eta}}^{Z_v} m_{Z_{\eta}} + \sum_{\eta} \Gamma_{Y_{\eta}}^{Z_v} m_{Y_{\eta}}, \quad (\text{A6})$$

$$h_{Y_v}^e = \sum_{\eta} \Gamma_{Z_{\eta}}^{Y_v} m_{Z_{\eta}} + \sum_{\eta} \Gamma_{Y_{\eta}}^{Y_v} m_{Y_{\eta}}. \quad (\text{A7})$$

The effective fields can be obtained from the energy density of each sublayer, which is given by

$$F_v = -\vec{M}^{(v)} \cdot \vec{B}_0 + \frac{\mu_0}{2} \vec{M}^{(v)} \cdot \vec{H}_d - \sum_{\eta} \frac{J_1 \vec{M}^{(v)} \cdot \vec{M}^{(\eta)}}{d_v M_s^2} - \frac{K_{2\parallel}^{(v)}}{M_s^2} (\vec{M}^{(v)} \cdot \hat{u})^2 - \frac{K_{2\perp}^{(v)}}{M_s^2} (\vec{M}^{(v)} \cdot \hat{n})^2 + F_c, \quad (\text{A8})$$

where $\vec{B}_0$ is the external field, $\vec{H}_d$ is the demagnetizing field, d_v is the thickness of a sublayer, and $\hat{u}$ ($\hat{n}$) corresponds to the in-plane (out-of-plane) uniaxial anisotropy field direction. For the system shown in Fig. 2(b) the easy directions are $\hat{u} = \hat{y}$ and $\hat{n} = \hat{z}$, with $K_{2\perp,2\parallel}^{(v)} > 0$. The term proportional to J_1 corresponds to the interlayer exchange interaction between the sublayers and F_c is the cubic anisotropy energy density.

Then, the effective fields can be calculated from $\mu_0 \vec{H}^{e(v)} = -\delta \epsilon_v / \delta \vec{M}^{(v)}$ and, thus, the in-plane static components of the effective field are

$$\mu_0 H_{X_v}^{e0} = B_0 \cos(\varphi_v - \varphi_B) + \frac{2K_{2\parallel}^{(v)}}{M_s} \cos^2(\varphi_v - \varphi_u) + \frac{K_{4\parallel}}{2M_s} \cos^2 \varphi_v [3 \cos(2\varphi_v) - 5] + \mu_0 H_{X_1}, \quad (\text{A9})$$

$$\mu_0 H_{Y_v}^{e0} = B_0 \sin(\varphi_B - \varphi_v) - \frac{K_{2\parallel}^{(v)}}{M_s} \sin[2(\varphi_v - \varphi_u)] + \frac{K_{4\parallel}}{8M_s} [2 \sin(2\varphi_v) - 3 \sin(4\varphi_v)] + \mu_0 H_{Y_1}, \quad (\text{A10})$$

with

$$\mu_0 H_{X_1} = \sum_{\eta} \frac{J_1}{d_v M_s} \cos(\varphi_v - \varphi_{\eta}) (\delta_{v,\eta-1} + \delta_{v,\eta+1}), \quad (\text{A11})$$

$$\mu_0 H_{Y_1} = \sum_{\eta} \frac{J_1}{d_v M_s} \sin(\varphi_{\eta} - \varphi_v) (\delta_{v,\eta-1} + \delta_{v,\eta+1}). \quad (\text{A12})$$

Note that in the last expressions, $\delta_{i,j}$ is the Kronecker delta function, which captures the exchange interaction of the nearest neighbor sublayers. On the other hand, for $v = \eta$ (intralayer interaction terms), the dynamic effective fields are obtained from $\Gamma_{Y_v}^{Z_v} = \Gamma_{Z_v}^{Y_v} = 0$, and

$$\begin{aligned} \mu_0 M_s \Gamma_{Y_v}^{Y_v} &= \frac{K_{4\parallel}}{8M_s} \cos^2 \varphi_v [9 \cos(4\varphi_v) + 7] \\ &\quad - \frac{2K_{2\parallel}^{(v)}}{M_s} \sin^2(\varphi_v - \varphi_u), \\ \mu_0 M_s \Gamma_{Z_v}^{Z_v} &= \mu_0 M_s - \frac{2K_{2\perp}^{(v)}}{M_s} + \frac{K_{4\parallel}}{2M_s} [3 \cos(2\varphi_v) - 1]. \end{aligned} \quad (\text{A13})$$

The case $v \neq \eta$ considers the interaction between sublayers, which are given by $\Gamma_{Y_{\eta}}^{Z_v} = \Gamma_{Z_{\eta}}^{Y_v} = 0$, and

$$\begin{aligned} \mu_0 M_s \Gamma_{Y_{\eta}}^{Y_v} &= \frac{J_1}{d_v M_s} \cos(\varphi_v - \varphi_{\eta}) (\delta_{v,\eta-1} + \delta_{v,\eta+1}), \\ \mu_0 M_s \Gamma_{Z_{\eta}}^{Z_v} &= \frac{J_1}{d_v M_s} (\delta_{v,\eta-1} + \delta_{v,\eta+1}). \end{aligned} \quad (\text{A14})$$

The previous expressions allow obtaining the FMR modes of the system. In the case of a macrospin approach, where the equilibrium magnetization is uniform along the thickness, $v = 1$ and $K_{2\perp,2\parallel}^{(v)} = K_{2\perp,2\parallel}$; the FMR frequency is simply

$$\left(\frac{\omega}{\gamma}\right)^2 = (\mu_0 H_X^{e0} - \mu_0 M_s \Gamma_Y^Y) (\mu_0 H_X^{e0} - \mu_0 M_s \Gamma_Z^Z), \quad (\text{A15})$$

with

$$\begin{aligned} \mu_0 H_X^{e0} &= B_0 \cos(\varphi_0 - \varphi_B) + \frac{2K_{2\parallel}}{M_s} \cos^2(\varphi_0 - \varphi_u) \\ &\quad + \frac{K_{4\parallel}}{2M_s} \cos^2 \varphi_0 [3 \cos(2\varphi_0) - 5], \end{aligned} \quad (\text{A16})$$

$$\mu_0 M_s \Gamma_Y^Y = -\frac{K_{4\parallel}}{8M_s} [9 \cos(4\varphi_0) + 7] + \frac{2K_{2\parallel}}{M_s} \sin^2(\varphi_0 - \varphi_u), \quad (\text{A17})$$

$$\mu_0 M_s \Gamma_Z^Z = -\mu_0 M_s + \frac{2K_{2\perp}}{M_s} + \frac{K_{4\parallel}}{2M_s} [3 \cos(2\varphi_0) - 1], \quad (\text{A18})$$

where we have assumed that $\varphi_1 = \varphi_0$, with φ_0 being the equilibrium angle. Thus, by taking into account that $\mu_0 M_{\text{eff}} = \mu_0 M_s - 2K_{2\perp}/M_s$, it can be shown that Eq. (A15) is equivalent to Eq. (5).

APPENDIX B: MAGNETOELASTIC ANISOTROPY OF A CUBIC (110)-ORIENTED THIN FILM IN MACROSPIN APPROXIMATION

We start from the equation for the magnetoelastic energy density for a system in macrospin approximation with cubic

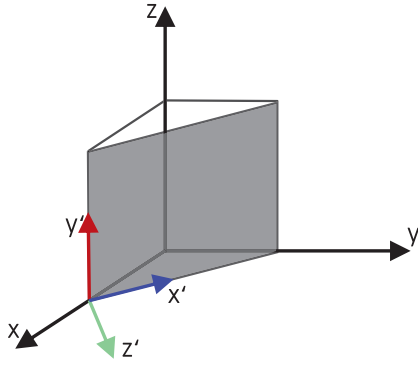


FIG. 10. Schematic representation of the coordinate transformation from the unprimed (100) crystal coordinate system to the primed (110) thin film system. The sketch is redesigned from [45].

symmetry and (100)-orientation [52,53]:

$$F_{me} = B_1(\epsilon_{11}\alpha_1^2 + \epsilon_{22}\alpha_2^2 + \epsilon_{33}\alpha_3^2) + 2B_2(\epsilon_{23}\alpha_2\alpha_3 + \epsilon_{13}\alpha_1\alpha_3 + \epsilon_{12}\alpha_1\alpha_2) + \dots \quad (B1)$$

The direction cosines of the magnetization $\vec{M}$ with respect to the cubic axes are given by $\alpha_i = \frac{\vec{M}_i \cdot \hat{e}_i}{|\vec{M}|}$, where the $\hat{e}_i$ ($i = 1, 2, 3$) are the Cartesian unit vectors along x , y , and z direction, which respectively correspond to the cubic [100], [010] and [001] directions. The strains ϵ_{ij} are measured along the cubic axes. Positive (negative) $\epsilon_{ij} = (a_{\text{film}} - a_{\text{bulk}})/a_{\text{bulk}}$ means that the thin film lattice exhibits larger (smaller) atomic distances a_{film} as compared to the distance of the volume material a_{bulk} . The equation describes how the magnetization direction and the strains are connected to result in a magnetoelastic energy density that is given by the so-called magnetoelastic coupling coefficients B_1 and B_2 . The latter have the values $B_1 = -0.343 \times 10^7$ and $B_2 = 0.783 \times 10^7 \text{ MJ m}^{-3}$.

In order to apply Eq. (B1) to the present case of (110)-oriented films, one needs to express the film strains ϵ_{ij} along the cubic axes in terms of the film strains ϵ_{ij}^f of the (110) plane. Similarly, the directions cosines of the magnetization in the (110)-film system α_i^f need to be evaluated in terms of the direction cosines α_i . We follow Ref. [45] to derive the respective expressions, thereby allowing the strain components along the three Cartesian coordinates to be different from each other, whereas in [45] isotropic strain has been assumed within the film plane. Choosing a possible {110} plane which is defined by the points (1,0,0), (0,1,0), and (1,0,1) (see Fig. 10) allows for visualizing the coordinate transformation from the original (unprimed) Cartesian (100)-system to the (primed) (110)-system of the films. In the primed system, the direction cosine α_1^f relates to the in-plane x' direction, pointing along the $[\bar{1}10]$, α_2^f relates to the in-plane y' direction, pointing along the [001] direction, while α_3^f relates to the out-of-plane z' direction, being oriented along the [110] direction.

From Fig. 10 it can be verified that the transformation matrix between the coordinate systems is given by

$$\underline{a} = \begin{pmatrix} -\frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 \\ 0 & 0 & 1 \\ \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 \end{pmatrix}. \quad (B2)$$

With the film strain

$$\underline{\epsilon}^f = \begin{pmatrix} \epsilon_{11}^f & 0 & 0 \\ 0 & \epsilon_{22}^f & 0 \\ 0 & 0 & \epsilon_{33}^f \end{pmatrix} \quad (B3)$$

and the direction cosines of the film system

$$\vec{\alpha}^f = \begin{pmatrix} \alpha_1^f \\ \alpha_2^f \\ \alpha_3^f \end{pmatrix} \quad (B4)$$

one obtains the expressions for the strain tensor and directions cosines in the unprimed coordinate system in terms of the film parameters:

$$\underline{\epsilon} = \begin{pmatrix} \epsilon_{11} & \epsilon_{12} & \epsilon_{13} \\ \epsilon_{21} & \epsilon_{22} & \epsilon_{23} \\ \epsilon_{31} & \epsilon_{32} & \epsilon_{33} \end{pmatrix} = \underline{a}^T \underline{\epsilon}^f \underline{a} = \begin{pmatrix} \frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} & -\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} & 0 \\ -\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} & \frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} & 0 \\ 0 & 0 & \epsilon_{22}^f \end{pmatrix}, \quad (B5)$$

and

$$\vec{\alpha} = \begin{pmatrix} \alpha_1 \\ \alpha_2 \\ \alpha_3 \end{pmatrix} = \underline{a}^T \vec{\alpha}^f = \begin{pmatrix} -\frac{\alpha_1^f}{\sqrt{2}} + \frac{\alpha_3^f}{\sqrt{2}} \\ \frac{\alpha_1^f}{\sqrt{2}} + \frac{\alpha_3^f}{\sqrt{2}} \\ \alpha_2^f \end{pmatrix}. \quad (B6)$$

Applying these relations to Eq. (B1) provides the magnetoelastic energy density for the (110)-plane of a cubic crystal:

$$F_{me} = B_1 \left[\epsilon_{22}^f (\alpha_2^f)^2 + \left(\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) \left(-\frac{\alpha_1^f}{\sqrt{2}} + \frac{\alpha_3^f}{\sqrt{2}} \right)^2 + \left(\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) \left(\frac{\alpha_1^f}{\sqrt{2}} + \frac{\alpha_3^f}{\sqrt{2}} \right)^2 \right] + 2B_2 \left(-\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) \left(\frac{\alpha_1^f}{\sqrt{2}} + \frac{\alpha_3^f}{\sqrt{2}} \right) \left(-\frac{\alpha_1^f}{\sqrt{2}} + \frac{\alpha_3^f}{\sqrt{2}} \right). \quad (B7)$$

From Eq. (B7), the magnetoelastic energies along the principal crystallographic direction of the (110)-oriented film can be calculated to be

$$F_{me}^{[\bar{1}10]} = B_1 \left(\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) - B_2 \left(-\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) \quad \text{for } \alpha_1^f \rightarrow 1, \alpha_2^f \rightarrow 0, \alpha_3^f \rightarrow 0 \quad (B8)$$

$$F_{me}^{[001]} = B_1 \epsilon_{22}^f \quad \text{for } \alpha_1^f \rightarrow 0, \alpha_2^f \rightarrow 1, \alpha_3^f \rightarrow 0 \quad (B9)$$

$$F_{me}^{[110]} = B_1 \left(\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) + B_2 \left(-\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) \quad \text{for } \alpha_1^f \rightarrow 0, \alpha_2^f \rightarrow 0, \alpha_3^f \rightarrow 1. \quad (B10)$$

The magnetoelastic anisotropy then follows from evaluating the differences between the respective expressions for two crystallographic directions:

$$\begin{aligned} \text{MAE}_{\text{ip}}^{[\bar{1}10]-[001]} &= -B_1 \epsilon_{22}^f - B_2 \left(-\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) \\ &\quad + B_1 \left(\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) \end{aligned} \quad (\text{B11})$$

$$\begin{aligned} \text{MAE}_{\text{oop}}^{[001]-[110]} &= B_1 \epsilon_{22}^f - B_2 \left(-\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) \\ &\quad - B_1 \left(\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) \end{aligned} \quad (\text{B12})$$

$$\text{MAE}_{\text{oop}}^{[\bar{1}10]-[110]} = -2B_2 \left(-\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right). \quad (\text{B13})$$

Note that a positive MAE corresponds to the second crystallographic direction of the difference being the easy one, e.g., for a positive $\text{MAE}_{\text{ip}}^{[\bar{1}10]-[001]}$ the $[001]$ direction is the easy one, whereas for negative $\text{MAE}_{\text{ip}}^{[\bar{1}10]-[001]}$ the $[\bar{1}10]$ is the easy direction. Inserting the tabulated values for bulk Fe, the three equations, Eqs. (B11)–(B13), can be used to calculate the magnetoelastic anisotropy for given strain ϵ_{ij}^f parallel to the three principal axes of the (110)-oriented thin film.

Using the explicit expression for the direction cosines in the film system, i.e., $\alpha_1 = \sin \theta \cos \varphi$, $\alpha_2 = \sin \theta \sin \varphi$, and $\alpha_3 = \cos \theta$, where θ is the angle of the magnetization with respect to the film normal ($[110]$ direction) and φ the angle with respect to the in-plane $[\bar{1}10]$ direction, Eq. (B7) yields

$$\begin{aligned} F_{\text{me}} &= B_1 \left[\epsilon_{22}^f (\sin \theta \sin \varphi)^2 + \left(\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) \left(-\frac{\sin \theta \cos \varphi}{\sqrt{2}} + \frac{\cos \theta}{\sqrt{2}} \right)^2 + \left(\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right) \left(\frac{\sin \theta \cos \varphi}{\sqrt{2}} + \frac{\cos \theta}{\sqrt{2}} \right)^2 \right] \\ &\quad + 2B_2 \left(-\frac{\sin \theta \cos \varphi}{\sqrt{2}} + \frac{\cos \theta}{\sqrt{2}} \right) \left(\frac{\sin \theta \cos \varphi}{\sqrt{2}} + \frac{\cos \theta}{\sqrt{2}} \right) \left(-\frac{\epsilon_{11}^f}{2} + \frac{\epsilon_{33}^f}{2} \right), \end{aligned}$$

which, after some algebraic manipulations, is equal to

$$\begin{aligned} F_{\text{me}} &= \left[B_1 \left(\frac{\epsilon_{11}^f}{2} - \epsilon_{22}^f + \frac{\epsilon_{33}^f}{2} \right) + B_2 \left(\frac{\epsilon_{11}^f}{2} - \frac{\epsilon_{33}^f}{2} \right) \right] \sin^2 \theta \cos^2 \varphi \\ &\quad + \left[B_1 \left(\frac{\epsilon_{11}^f}{2} - \epsilon_{22}^f + \frac{\epsilon_{33}^f}{2} \right) - B_2 \left(\frac{\epsilon_{11}^f}{2} - \frac{\epsilon_{33}^f}{2} \right) \right] \cos^2 \theta + B_1 \epsilon_{22}^f. \end{aligned} \quad (\text{B14})$$

Then, by defining the constants

$$K_{2\parallel}^{\text{me}} := \frac{1}{2} [B_1 (\epsilon_{11}^f - 2\epsilon_{22}^f + \epsilon_{33}^f) + B_2 (\epsilon_{11}^f - \epsilon_{33}^f)],$$

$$K_{2\perp}^{\text{me}} := B_2 (\epsilon_{11}^f - \epsilon_{33}^f),$$

$$c_1 := B_1 \epsilon_{22}^f + K_{2\parallel}^{\text{me}} - K_{2\perp}^{\text{me}},$$

one obtains

$$F_{\text{me}} = K_{2\parallel}^{\text{me}} \sin^2 \theta \cos^2 \varphi + (K_{2\perp}^{\text{me}} - K_{2\parallel}^{\text{me}}) \sin^2 \theta + c_1. \quad (\text{B15})$$

Using

$$\cos^2 \varphi = 1 - \cos^2 \left(\varphi - \frac{\pi}{2} \right),$$

Eq. (B15) can equivalently be written as

$$F_{\text{me}} = -K_{2\parallel}^{\text{me}} \sin^2 \theta \cos \left(\varphi - \frac{\pi}{2} \right)^2 + K_{2\perp}^{\text{me}} \sin^2 \theta + c_2 \quad (\text{B16})$$

with

$$c_2 = c_1 = B_1 \epsilon_{22}^f + K_{2\parallel}^{\text{me}} - K_{2\perp}^{\text{me}}.$$

This expression of the free magnetoelastic energy density has the same mathematical form as the anisotropy terms proportional to $K_{2\perp}$ and $K_{2\parallel}$ in Eq. (3), respectively, when setting $\varphi_u = \frac{\pi}{2}$. Thus, adding the magnetoelastic contribution to *both* terms is justified. Note that the angle-independent, constant terms c_1 and c_2 do not alter the resonance equation, for which solely the derivatives of the free energy density with respect to the angles θ and φ have to be inserted. From Eqs. (B11)–(B13) one notices that

$$\text{MAE}_{\text{ip}}^{[\bar{1}10]-[001]} = K_{2\parallel}^{\text{me}}, \quad (\text{B17})$$

$$\text{MAE}_{\text{oop}}^{[001]-[110]} = K_{2\perp}^{\text{me}} - K_{2\parallel}^{\text{me}}, \quad (\text{B18})$$

$$\text{MAE}_{\text{oop}}^{[\bar{1}10]-[110]} = K_{2\perp}^{\text{me}}. \quad (\text{B19})$$

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