Asymmetric magneto-optic response in anisotropic thin films

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We report asymmetric hysteresis loops measured using the magneto-optic Kerr effect from epitaxial Fe(110) and Co(110) thin films with in-plane magnetic anisotropy. This asymmetry appears to violate the invariance of the hysteresis loop under the transformation $\mathbf{M}\rightarrow-\mathbf{M}$, $\mathbf{H}\rightarrow-\mathbf{H}$ ($\mathbf{M}$ is the magnetization and $\mathbf{H}$ is the applied field). The *asymmetric magneto-optic response* is due to a product of the longitudinal and transverse components of the magnetization and proves the existence of the transverse component of the magnetization and, therefore, of the coherent rotation model of magnetization reversal. We simulate the observed hysteresis loops with a term in the magneto-optic response second order in the magnetization that is much larger than predicted by the standard magneto-optic theory. [S0163-1829(97)04202-1]

I. INTRODUCTION

The magneto-optic Kerr effect (MOKE) is an important technique for understanding the magnetization reversal process of a ferromagnetic thin film because of its *in situ* capabilities[1] and high sensitivity to the magnetism of thin films.[2] A measurement of magnetization against applied field should be invariant under the transformation $\mathbf{M}\rightarrow-\mathbf{M}$, $\mathbf{H}\rightarrow-\mathbf{H}$ (where $\mathbf{M}$ and $\mathbf{H}$ are the magnetization and applied field, respectively); i.e., should go into itself if reflected through the origin.[3] In this paper, we report the observation and explanation of asymmetric hysteresis loops that appear not to obey the above transformation measured magneto-optically from epitaxial (110)-oriented Fe and Co thin films, and put forth an explanation of these effects that which should apply to the cases of anisotropic Au/Co/GaAs(001) (Ref. 4) and Ni(001)/Fe(001) (Ref. 5) films, as well.

We find that the asymmetry in the magneto-optic response is due to terms in the dielectric tensor second order in the magnetization and is therefore inherent in the magneto-optic response. The second-order term becomes the only important term in the case of perpendicular incidence (the incident light normal to the surface of the sample) and in-plane magnetization, in which case it is called the Cotton-Mouton or Voigt effect. This effect was predicted to be on the order of microradians for ferromagnetic metals by Metzger, Pluvinage, and Torguet,[6] who also showed that, for a metal with in-plane magnetization and $\mathbf{H}$ in the plane of incidence of the light, the second-order term in the magneto-optic response is proportional to the product of the components of the magnetization perpendicular and parallel to $\mathbf{H}$. We show that the calculations by Metzger, Pluvinage, and Torguet predict a second-order effect too small by up to two orders of magnitude. The asymmetric hysteresis loop provides evidence of the existence of the component of $\mathbf{M}$ perpendicular to $\mathbf{H}$, and therefore corroborates the coherent rotation model of magnetization reversal, where the magnetization of the body is assumed to act as a single entity and rotate or jump coherently in an external applied magnetic field. In the absence of a coherently rotating magnetization, there is no component of $\mathbf{M}$ perpendicular to $\mathbf{H}$,[7] and therefore no asymmetry in the magneto-optic response.

Both Fe(110) and Co(110) films were used in this study. The demagnetizing field of such films forces the magnetization to lie in plane, so that the *perpendicular* component of the magnetization is zero. We define the two in-plane components of the normalized magnetization as being *longitudinal* (the component of $\mathbf{M}$ parallel to the plane of incidence formed by the incident and reflected beams) and *transverse* (the component of $\mathbf{M}$ perpendicular to the plane of incidence). The perpendicular, longitudinal, and transverse components of $\mathbf{M}$, normalized to the absolute value of $\mathbf{M}$, are designated, respectively m_p, m_l, and m_t. In our experiment, $\mathbf{H}$ is applied along the longitudinal axis.

The crystalline anisotropy of the (110) surface contributes both uniaxial and biaxial terms to the total anisotropy energy, while strain and surface effects influence the uniaxial component. Therefore, epitaxial (110)-oriented films often have an in-plane anisotropy in addition to in-plane magnetization.[8,9] The energy density of such films is given by

$$E/V=-M_sH\cos(\phi-\theta_2)-K_u\cos2\theta_2 - K_b\cos4(\theta_2+\theta_0), \quad (1)$$

where E/V, M_s, H, K_u, K_b, θ_2, θ_0, and ϕ are, respectively, the energy density, saturation magnetization (magnitude of $\mathbf{M}$), magnitude of $\mathbf{H}$, uniaxial anisotropy constant, biaxial anisotropy constant, angle between $\mathbf{M}$ and the easy axis, angle between the uniaxial and biaxial axes, and angle between $\mathbf{H}$ and the easy axis.[10] The variable θ_2 is used to represent the angle between $\mathbf{M}$ and the easy axis instead of the more traditional θ in order to eliminate confusion with the angle of incidence, defined in the next section. The values of K_u and K_b obtained by torque magnetometry are listed in Table I note that K_u and K_b are all positive. The Fe(110)/Mo(110) samples had an easy in-plane axis along the [001] direction, consistent with previous results on Fe(110)/Mo(110) multilayers,[11] while the Co(110) sample had an easy in-plane axis along the [110] direction.

II. EXPERIMENTAL PROCEDURE

Fe(110) samples were sputter deposited in a UHV system with a base pressure of 10^{-8} torr. [1120]-oriented Al_2O_3 sub-

TABLE I. Orientation, structure, and results of torque magnetometry of three different sputtered, epitaxial (110)-oriented thin films.

Orientation and structure	K_u	K_b	θ_0
811 Å Fe (110)/892 Å Mo(110)/$Al_2O_3(11\bar{2}0)$	9.0×10^4 ergs/cm^3	1.6×10^4 ergs/cm^3	$-117°$
75 Å SiC/811 Å Fe (110)/ 892 Å Mo (110)/$Al_2O_3(11\bar{2}0)$	8.9×10^4 ergs/cm^3	1.3×10^4 ergs/cm^3	$-37°$
15 Å Pt/10 Å Cu/200 Å Co (100)/ 50 Å Cu/50 Å Pt/5 Å Fe/MgO (110)	1.6×10^5 ergs/cm^3	8.4×10^4 ergs/cm^3	$0°$

strates were polished, cleaned, with organics, and heated to at least 640 °C before deposition of a Mo(110) underlayer; the Fe film was sputter deposited after the sample cooled below 100 °C. Such a procedure gives epitaxial Fe(110) thin films with in-plane rocking curve widths of less than 5° and a known strain state.[11] Because the substrate surface is not perfectly lattice matched with the Mo(110) surface, there is some twinning of both the Mo and Fe films about the (110) direction; however, vibrating-sample magnetometry (VSM) and torque measurements indicated that the grains were coupled closely enough to ensure single-domain-type behavior, with the possible exception of the very low-field regime.[12] One of the Fe(110) samples was coated with 75 Å SiC as a barrier to oxidation; the other was left uncapped. Rutherford backscattering spectroscopy was used to verify that the SiC-capped sample was much less oxidized than the uncapped sample.

The Co(110) sample was sputter deposited in a UHV system. A seed layer of 5 Å Fe/50 Å Pt was deposited onto a [110]-oriented MgO at 500 °C, and the sample cooled to 60 ° C in a 3.25 mtorr Ar environment before deposition of a 50 Å Cu/200 Å Co/10 Å Cu/15 Å Pt quadrilayer. Such a procedure resulted in a high-quality, epitaxial Co(110) thin film.

The only light source used in our MOKE system was a He-Ne laser, so that all experiments were done at a photon wavelength of $\lambda'=632.8$ nm. *s*-polarized light (perpendicular to the plane of incidence) was incident onto the sample at angles of between $5°\pm0.5°$ and $32°\pm0.5°$ from the normal. The angle of incidence will be henceforth denoted θ. The sample was set up with the field applied in the plane of incidence of the light (so that the longitudinal component of the magnetization, m_l, lay along the field direction) and within 5° of the in-plane hard axis, except for the SiC-capped Fe(110) sample, where this angle was larger. The transverse component of the magnetization therefore lay along the easy axis of the sample (the in-plane [001] and [110] directions for the Fe(110)/Mo(110) and Co(110) samples, respectively). As explained below, by measuring the total reflectivity and varying the angle of incidence, we were able to determine the optical and magneto-optic parameters of the sample. A lock-in measurement technique utilizing a photoelastic modulator (PEM) was used to measure separately the Kerr rotation and ellipticity (θ_k and ϵ_k, respectively).[13] The analyzer was mounted on the PEM and made an angle of 45° with the axis of the PEM, which was perpendicular to the plane of incidence for measurement of θ_k and ϵ_k. Rotation of the PEM and analyzer by $45°-\beta$, where β is a small angle on the order of 1°, allowed us to measure the transverse component of the magnetization as a function of **H**. The polarizer was rotated by 45° as well during this procedure, which gave the quantity $\theta_k(45°)$, given by the following expression:

$$\theta_k(45°)=2J_2(|r_{pp}|^2\{1+\beta\}^2-|r_{ss}|^2\{1-\beta\}^2+\{1-\beta^2\}\{r_{pp}r_{ps}^*+r_{pp}^*r_{ps}+r_{ss}r_{ps}^*+r_{ss}^*r_{ps}\})/(|r_{pp}|^2\{1+\beta\}^2+|r_{ss}|^2\{1-\beta\}^2 +2|r_{sp}|^2\{1+\beta\}+\{1-\beta^2\}\{r_{pp}r_{ps}^*+r_{pp}^*r_{ps}-r_{ss}r_{ps}^*-r_{ss}^*r_{ps}\}). \quad (2)$$

Here r_{pp}, r_{ss}, r_{sp}, and r_{ps} are reflection coefficients of the sample that give the amplitudes of, respectively, reflecting *p*-polarized light into *p*-polarized light, *s*-polarized light into *s*-polarized light, *p*-polarized light into *s*-polarized light, and *s*-polarized light into *p*-polarized light,[6] the star superscript refers to the complex conjugate operation, and J_2 is the second Bessel function evaluated at 137.8°, the retardation angle used in our experiment ($J_2\cong0.445$).

Although Eq. (2) appears cumbersome, much useful information can be gleaned from it. By carefully adjusting the polarizer, we may set β to eliminate the term in Eq. (2) independent of **M**. To highest order, the $\theta_k(45°)$ signal is then proportional to m_t as a function of H, which gives great insight into the mechanism of magnetization reversal. Displayed in Figs. 1(a) and 1(b) are plots of m_t against H, as derived from the $\theta_k(45°)$ signal, for the capped Fe(110) and Co(110) samples, respectively. These are the raw data [$\theta_k(45°)$ as a function of H] scaled by a constant we calculated based on our determination of the magneto-optic and optical constants; we have assumed the other terms in the $\theta_k(45°)$ signal to be negligible because of the close correspondence between the $\theta_k(45°)$ signal and the m_t measured by vector magnetometry. The m_t signal as a function of H for the uncapped Fe(110) sample was already published in Refs. 14 and 15. These references used slightly different *Q*-values and therefore the maximum value of m_t was slightly different; the same plot of m_t against H was used for all the simulations. These plots show the expected behavior of the component of **M** perpendicular to **H**; m_t goes to zero at high fields and reaches a maximum at zero applied field.

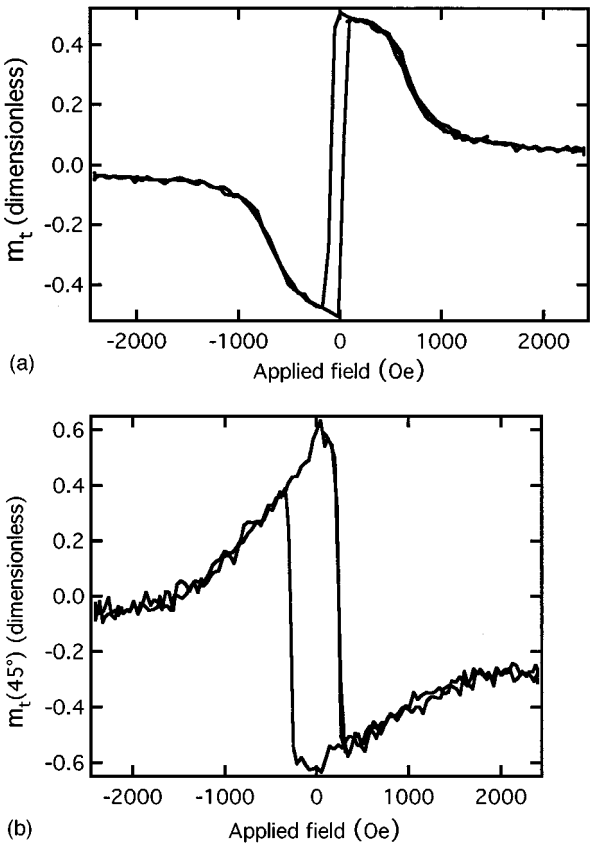

FIG. 1. (a) Measured m_t against H of the SiC-capped Fe(110) film. (b) Measured m_t against H of the Co(110) film. The applied field lies within 5° of the [001] direction.

Note that the maximum value is less than unity, indicating that there is some domain formation. Nevertheless, it is clear that there is coherent rotation of the magnetization.

Hysteresis loops measured in the Fe samples with the field applied along the [001] direction showed square, easy-axis behavior. A measurement of $\theta_k(45°)$ with **H** applied along the [001] direction in these samples gave zero signal [the $\theta_k(45°)$ signal for the Co sample with **H** along the easy axis was nonzero because of the large ratio of biaxial to uniaxial anisotropy in this sample]. The magnetization reverses through domain-wall motion when the field is applied along this axis, and so the transverse component of **M** is nonzero only inside the domain wall, whose contribution to the total signal is negligible.[12]

III. RESULTS

The ellipticities [rotation for the Co(110) sample] measured at an 11° angle of incidence [10° for the SiC-capped Fe(110) sample] are displayed in Figs. 2, 3, and 4 for the capped Fe(110), uncapped Fe(110), and Co(110) samples, respectively. The angle of incidence is defined from the surface normal. These signals appear to break the invariance of the hysteresis loop under the transformation $\mathbf{M}\rightarrow-\mathbf{M}$, $\mathbf{H}\rightarrow-\mathbf{H}$. The asymmetry of the uncapped Fe and Co samples is so large that the hysteresis loop is crossed near its center. We will show that the apparent symmetry breaking

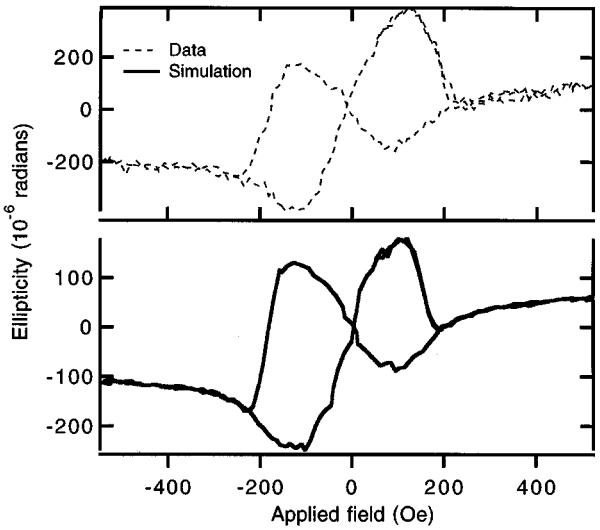

FIG. 2. Simulated and measured ellipticity (θ=11°) of the uncapped Fe(110) film. The applied field lies within 5° of the [1$\bar{1}$0] direction.

comes from a term in the magneto-optic response second order in the magnetization and it is not valid to represent the magneto-optic hysteresis loop as being a plot of **M** vs **H**. The second-order term in the magneto-optic response is proportional to the product of m_l and m_t. Hysteresis loops obtained by torque and vibrating-sample magnetometry (VSM) for all three samples are not asymmetric about the origin and can be reproduced by minimizing Eq. (1) with respect to θ_2 with K_u and K_b given in Table I. Nevertheless, it is not entirely accurate to characterize the asymmetry in the magneto-optic response as being a purely optical effect; the asymmetry is indicative of an interesting *magnetic* effect: a nonzero transverse component of **M**, which is a prediction of the coherent rotation model of magnetization reversal.

Also displayed in Figs. 2, 3, and 4 are fits to the ellipticity [rotation for the Co(110) sample]. The simulations required as input the actual dependence of m_l and m_t on **H**. Measurements of the rotation [ellipticity for the Co(110) sample] at a large angle of incidence gave m_l. These measurements were not asymmetric and therefore were in agreement with the VSM and torque data. The results from Figs. 1(a) and 1(b) and Ref. 15 gave m_t as a function of H.

IV. THEORETICAL BACKGROUND

In order to understand the asymmetry in the hysteresis loops, we have calculated the magneto-optic response for anisotropic thin films. Metzger, Pluvinage, and Torguet[6] have calculated the magneto-optic response based on a dielectric tensor derived from classical electronic equations of motion.[16] Metzger, Pluvinage, and Torguet expanded in the parameter λ which is given by

$$\lambda=\omega_c\tau, \tag{3}$$

where ω_c is the Larmor frequency of the electron in an effective magnetic field proportional to M_s, and τ is a frequency-dependent factor which becomes the relaxation time in the dc limit. These previous calculations assume that *all* the electrons are acted on by the effective magnetic field,

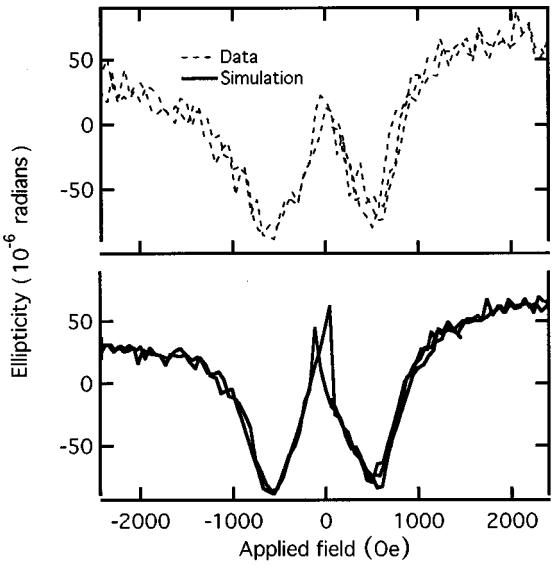

FIG. 3. Simulated and measured ellipticity (θ=11°) of the SiC-capped Fe(110) film.

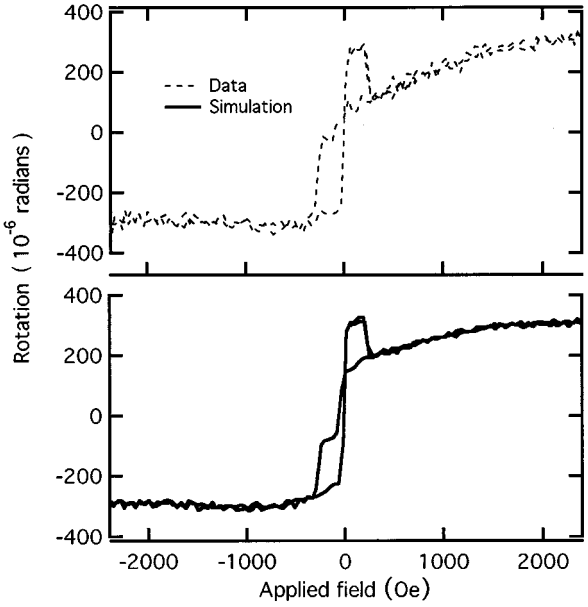

FIG. 4. Simulated and measured ellipticity (θ=11°) of a Co(110) film. The applied field lies within 5° of the [001] direction.

and hence contribute equally to the off-diagonal terms. Metzger, Pluvinage, and Torguet made the assumption that the size of the off-diagonal component of the dielectric tensor was given by the entire on-diagonal component of the dielectric tensor, multiplied by an effective magnetic field given by the magnetization. Argyres, in a full quantum-mechanical calculation of the dielectric tensor, divided his sum over states into an integral over all states occupied by electrons with both spin directions (which yielded the diagonal component of the dielectric tensor) and an integral over states occupied by ''magnetic electrons'' with only one spin direction (which yielded the off-diagonal component of the dielectric tensor).[17] We have made the additional assumption that the size of the off-diagonal component of the dielectric tensor is given by only *part* of the on-diagonal component (the response of the conduction or unbound electrons) multiplied by the effective magnetic field. A similar distinction in the contributions of bound and unbound electrons to the off-diagonal component of the dielectric tensor was made by Bennett and Stern in the case of the Faraday effect.[18]

Extending the arguments in Ref. 17 to our classical approach, we calculate the dielectric tensor ϵ to be

$$\epsilon_{ij}=(1+4\pi\alpha)\,\delta_{ij}+\frac{4\pi\sigma_0}{i\omega}\,\frac{(\delta_{ij}+i\lambda\widehat{\epsilon_{ijk}}m_k-\lambda^2 m_i m_j)}{1-\lambda^2}, \tag{4}$$

where m_i and $\widehat{\epsilon_{ijk}}$ are the ith component of $\mathbf{M}$ and the Levi-Civita density,[19] respectively, σ_0 is the conductivity in the absence of magnetization, and the quantity $4\pi\alpha$ represents the contribution from the electrons whose equations of motion are not affected by the magnetization.[20] This expression becomes identical to that obtained by Bolotin and Sokolov[16] in the limit that $4\pi\alpha\rightarrow 0$.

We now proceed following Metzger, Pluvinage, and Torguet except that we use the above $4\pi\alpha\neq 0$ expression for the dielectric tensor, and we expand in the conventional magneto-optic parameter Q rather than λ. These two parameters are related by

$$\lambda=\frac{n_0'^2 Q}{n_0'^2-1-4\pi\alpha}, \tag{5}$$

which is valid to order Q^2; i.e., to second order in the magnetization. The parameter n_0' is the index of refraction of the magnetic material. In the limit of small angles of incidence, the magneto-optic response is given by

$$\begin{aligned}\theta_k+i\epsilon_k\cong&-\frac{iQ\theta m_l}{(\cos\delta-in_0'\sin\delta/n_0+n_0'-in_0\sin\delta)}\\&\times\frac{1}{(\cos\delta-in_0'\sin\delta/n_0-n_0'\cos\delta+in_0\sin\delta)}\\&-\frac{Q^2 m_l m_t n_0'}{(n_0'+1)^2}\left(\frac{1+4\pi\alpha}{n_0'^2-1-4\pi\alpha}\right)\\&\times\left(\frac{\cos\delta-in_0'\sin\delta/n_0+n_0'\cos\delta-in_0\sin\delta}{\cos\delta-in_0'\sin\delta/n_0-n_0'\cos\delta+in_0\sin\delta}\right),\end{aligned} \tag{6}$$

where θ_k and ϵ_k are the Kerr rotation angle and ellipticity, θ is the angle of incidence, n_0 is the index of refraction of the capping layer, and $\delta=-2\pi n_0 d/\lambda'$ is the phase delay of the wave as it crosses the capping layer. Equation (6) was derived using the 4×4 boundary matrix formalism of Zak *et al.*[21] with two media: a thin non-magnetic material atop an infinitely thick magnetic layer. In Eq. (6), we have kept terms only linear in θ, and assumed that the capping layer had a negligible effect on the Q^2 term (i.e., ignoring terms of order $Q^2\times d$ and higher).

This result is identical to that obtained by Metzger, Pluvinage, and Torguet in the limit that $4\pi\alpha\rightarrow 0$ and $\delta\rightarrow 0$,[6] with the exception of the overall sign, which appears to be due to two errors by Metzger, Pluvinage, and Torguet: in extracting r_{ps} from Eq. (33) of Ref. 6, and in obtaining the term second order in Q from Eq. (34) in Ref. 6. Equation (6) becomes

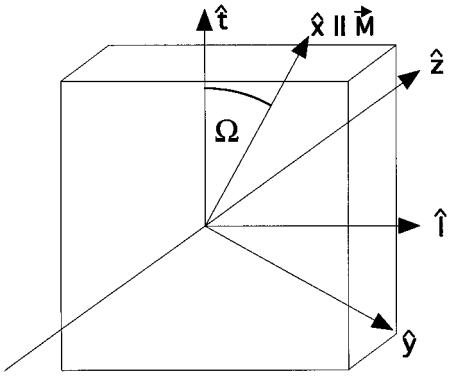

FIG. 5. Schematic illustration of the sample ($\{xyz\}$) and laboratory ($\{tlz\}$) bases. The x axis lies parallel to **M**, and the z axis lies perpendicular to the sample, which is outlined box in the figure. The t and l axes are defined by an external polarizer. The x axis makes an angle Ω with the t axis.

exact in the limiting case of perpendicular incidence, so that the magneto-optic response in the limit $\theta\rightarrow 0$ is entirely proportional to $m_l m_t$. This fact was confirmed experimentally by reducing θ and observing the changes in the plot of magneto-optic response against H with θ. Note that both Metzger's and our results contain a term which is second order in Q. The term proportional to the product $m_l m_t$ does not change sign under the transformation $\mathbf{M}\rightarrow -\mathbf{M}$, $\mathbf{H}\rightarrow -\mathbf{H}$ and therefore causes the asymmetry in the hysteresis loops. The primary difference between our results and Metzger's is that our second-order term contains the parameter $4\pi\alpha$ which justifies decoupling the magnitudes of the first- and second-order terms.

The exact expression for the coefficient multiplying $m_l m_t$ at any arbitrary θ is cumbersome to derive, and yields no great physical insight.[12] Since Eq. (6) becomes exact in the limit $\theta\rightarrow 0$, we can derive the Q^2 term in Eq. (6) in the especially simple yet physically relevant case of normal incidence and in-plane magnetization, where the magneto-optic response is second order in Q. This derivation will illustrate the physics of the problem and will explain how our model, in contrast to the model of Metzger, Pluvinage, and Torguet,[6] can explain a large Q^2 term. This derivation ignores the presence of the capping layer, and we drop the prime on n_0 in order to simplify the notation.

We must first define two reference frames. The $\{xyz\}$ frame can be thought of as the ''sample reference frame'' because it is attached to **M**, which rotates coherently in the case of **H** parallel to the hard axis. The incident light beam propagates along the z axis perpendicular to the sample and the x axis lies parallel to **M**. The ''laboratory reference frame'' has the same z axis as the sample reference frame, but the two other orthogonal directions (labeled **l** and **t**) are, respectively, parallel and perpendicular to the direction of some external polarizer (see Fig. 5). We introduce the reflection coefficients r_{tt}, r_{tl}, r_{lt}, and r_{ll}, exactly analogous to the reflection coefficients already discussed. We can connect the $\{t,l\}$ basis to the traditional $\{p,s\}$ basis at normal incidence: the **t** direction is perpendicular to the plane of incidence that would exist if the angle of incidence were nonzero. Note that unlike the $\{p,s\}$ basis, the $\{t,l\}$ and $\{x,y\}$ bases do not depend on the direction of **k**. Hence, $r_{pp}=-r_{ll}$ and $r_{ps}=-r_{lt}$ (the **p** direction changes upon reflection by 180° if $\theta=0°$).

We will compute the magneto-optic response in terms of the elements of the dielectric tensor. We will then substitute in the elements of the dielectric tensor used by Metzger, Pluvinage, and Torguet[6] [Eq. (4) with $4\pi\alpha$ set to zero] to estimate the size of the Q^2 term for a Fe film. Finally, we will substitute into the expression for the magneto-optic response the elements of the dielectric tensor according to our theory [obtained from Eqs. (4) and (5)]. We will see that the uncoupling of the terms second and first order in the magnetization allows us to fit our experimental observations of a Q^2 term much larger than that predicted by Metzger, Pluvinage, and Torguet,[6] who required that $4\pi\alpha$ equal zero.

We start the derivation of the Q^2 term in Eq. (6) by solving the wave equation for a light wave propagating through a magnetic metal with dielectric tensor $\tilde{\epsilon}$:

$$\mathbf{k}\times\mathbf{k}\times\mathbf{E}+\frac{\omega^2}{c^2}\,\tilde{\epsilon}\,\mathbf{E}=0, \tag{7}$$

where **k**, **E**, ω, and c are, respectively, the wave vector, electric field, frequency of the light wave, and the speed of light. This allows us to calculate the eigenmodes of propagation. As in Yariv[22] (although we use cgs units here), we write

$$\begin{pmatrix} \epsilon_{xx}-n^2 & 0 & 0 \\ 0 & \epsilon_{zz}-n^2 & \epsilon_{yz} \\ 0 & \epsilon_{zy} & \epsilon_{zz} \end{pmatrix}\begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}=\begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix}. \tag{8}$$

This allows us to find the eigenmodes of propagation.
Eigenmode 1:

$$n^2=\epsilon_{zz}+\frac{\epsilon_{yz}^2}{\epsilon_{zz}}, \tag{9}$$

$$\begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}=\begin{pmatrix} 0 \\ \epsilon_{zz} \\ -\epsilon_{zy} \end{pmatrix}. \tag{10}$$

Eigenmode 2:

$$n^2=\epsilon_{xx}=n_0^2, \tag{11}$$

$$\begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}=\begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}. \tag{12}$$

Noting that the eigenmodes of the magnetic metal are polarized parallel and perpendicular to the direction of magnetization, so that we can break down the incoming electric field **E** into components parallel and perpendicular to the magnetization direction, we calculate the reflectivity matrix in the sample reference frame:

$$\begin{pmatrix} r_{yy} & 0 \\ 0 & r_{xx} \end{pmatrix}=\begin{pmatrix} \dfrac{1-n_\perp}{1+n_\perp} & 0 \\ 0 & \dfrac{1-n_\parallel}{1+n_\parallel} \end{pmatrix}. \tag{13}$$

Note that r_{pp} at normal incidence for a homogenous material with index of refraction n is given by $r_{pp}=(n-1)/(n+1)$

because of the change of sign of the **p** direction upon reflection as remarked earlier; neither the **x** nor **y** directions change upon reflection. Since the magnetization is fixed in magnitude, we can rotate the reflectivity matrix an angle Ω about the film normal into the laboratory frame, where the magnetization is at an angle from the transverse direction. The magnetization (which lies along the x axis in the film frame) can be written: $\mathbf{M}=|\mathbf{M}|\hat{\mathbf{x}}=|\mathbf{M}|(\cos\Omega\hat{\mathbf{t}}+\sin\Omega\hat{\boldsymbol{l}})=|\mathbf{M}|(m_t\hat{\mathbf{t}}+m_l\hat{\boldsymbol{l}})$, where $\hat{\mathbf{x}}$, $\hat{\mathbf{t}}$, and $\hat{\boldsymbol{l}}$ are unit vectors along the x axis (in the film frame) and the $\boldsymbol{l}$ and **t** directions as defined previously. In the laboratory frame, the reflectivity matrix can be written

$$\begin{pmatrix} r_{tt} & r_{tl} \\ r_{lt} & r_{ll} \end{pmatrix} = \begin{pmatrix} r_{xx}\cos^2\Omega + r_{yy}\sin^2\Omega & (r_{xx}-r_{yy})\sin\Omega\,\cos\Omega \\ (r_{xx}-r_{yy})\sin\Omega\,\cos\Omega & r_{yy}\cos^2\Omega + r_{xx}\sin^2\Omega \end{pmatrix}. \tag{14}$$

The magneto-optic response at normal incidence $[\theta_k + i\epsilon_k(0°)]$ is

$$\begin{aligned}\theta_k + i\epsilon_k(0°) &= r_{ps}/r_{ss} = -r_{lt}/r_{tt} = \frac{-(r_{xx}-r_{yy})\sin\Omega\,\cos\Omega}{(r_{xx}\cos^2\Omega + r_{yy}\sin^2\Omega)} \\ &= \frac{-(r_{xx}-r_{yy})\sin\Omega\,\cos\Omega}{(r_{xx}\{\cos^2\Omega+\sin^2\Omega\}+\{r_{yy}-r_{xx}\}\sin^2\,\Omega)} \\ &\cong \frac{(r_{yy}-r_{xx})m_l m_t}{r_{xx}},\end{aligned} \tag{15}$$

where we have expanded to second order in Q [since $(r_{yy}-r_{xx})$ is second order in Q] and have equated the reflection coefficients as explained earlier.

Substituting the reflection matrix from Eq. (13) gives the magneto-optic response at normal incidence to second order in Q in terms of the elements of the dielectric tensor:

$$\begin{aligned}\theta_k + i\epsilon_k(0°) &= \frac{[(1-n_\perp)/(1+n_\perp)-(1-n_\parallel)/(1+n_\parallel)]m_l m_t}{(1-n_0)/(1+n_0)} \\ &= \frac{(n_\perp^2-n_\parallel^2)m_l m_t}{n_0(n_0^2-1)} = \frac{(-\epsilon_{xx}+\epsilon_{zz}+\epsilon_{yz}^2/\epsilon_{zz})m_l m_t}{n_0(n_0^2-1)}\end{aligned} \tag{16}$$

(note that a sign error in Osgood, White, and Clemens[14] has been corrected here).

Let us use the dielectric tensor from Eq. (4) with λ given in Eq. (5). In the film frame, with **M** along the x axis, we obtain

$$\begin{pmatrix} \epsilon_{xx} & 0 & 0 \\ 0 & \epsilon_{zz} & \epsilon_{yz} \\ 0 & \epsilon_{zy} & \epsilon_{zz} \end{pmatrix} = \begin{pmatrix} n_0^2 & 0 & 0 \\ 0 & n_0^2 + \dfrac{n_0^4 Q^2}{n_0^2-1-4\pi\alpha} & in_0^2 Q m_x \\ 0 & -in_0^2 Q m_x & n_0^2 + \dfrac{n_0^4 Q^2}{n_0^2-1-4\pi\alpha} \end{pmatrix}, \tag{17}$$

yielding for the magneto-optic response:

$$\theta_k + i\epsilon_k(0°) = + \frac{Q^2 n_0(1+4\pi\alpha)m_l m_t}{(n_0^2-1)(n_0^2-1-4\pi\alpha)}, \tag{18}$$

which agrees with Eq. (6) in the case of $\delta\to 0$, $n_0\to n_0'$, and $\theta\to 0$.

$\theta_k + i\epsilon_k(0°)$ from Eq. (18) can be as much as two orders of magnitude larger than that of Metzger, Pluvinage, and Torguet [Eq. (18) with $4\pi\alpha$ set to zero] because of the factor $(1+4\pi\alpha)/(n_0^2-1-4\pi\alpha)$. The coefficient of $m_l m_t$ can be measured (at small θ) and fitted to obtain $4\pi\alpha$. We now explain how the parameters Q, $4\pi\alpha$, and n_0 were obtained experimentally, and compare the resulting size of the Q^2 term predicted by our model [Eq. (18)] and that of Metzger, Pluvinage, and Torguet [Eq. (18) with $4\pi\alpha$ set to zero].

V. FITTING THE DATA

With the introduction of the parameter $4\pi\alpha$ our expression has six parameters to be fit to the data: the real and imaginary parts of n_0', Q, and $4\pi\alpha$. We note that we have used the exact formula for the magneto-optic response valid at any angle of incidence to fit the data, not Eq. (6), which is valid in the limit $\theta\to 0$. First, we fit the real and imaginary parts of $n_0'^2$ to the measured total s and p reflectivities: $|r_{ss}|^2$ and $|r_{pp}|^2$, which are zeroth order in Q. This gives the values in column 1 of Table II. In the case of the Co(110) and SiC-capped Fe(110) samples, tabulated indices of refraction were used for the capping layers. For the purpose of calculations, the Co(110) layer was approximated as being infinitely thick, and its two capping layers were considered to be a single capping layer with an effective dielectric tensor given by the weighted average of the indices of refraction of the top two layers.

Second, we fit Q and $4\pi\alpha$. To obtain Q, we saturated the magnetization ($m_l=1$, $m_t=0$) and measured the saturation ellipticity and rotation at $\theta=11°$ [10° for the SiC-capped Fe(110) sample]. Because the uncapped Fe(110) sample was

TABLE II. Optical constants of three different sputtered, epitaxial (110)-oriented thin films obtained by fitting the s and p reflectivities (for n_0'), the s rotation and ellipticity vs incidence angle (for Q), and the asymmetry (for $4\pi\alpha$).

Sample	n_0'	Q	$4\pi\alpha$
Fe(110)	$2.3-i\ 2.4$	$0.022+i\ 0.006$	$-1.9-i\ 14.1$
Fe(110) with SiC cap	$2.85-i\ 3.36$	$0.054+i\ 0.004$	$-0.15-i\ 9.46$
Co(110)	$2.21-i\ 4.0$	$0.052-i\ 0.001$	$8.2-i\ 3.4$

coated with an oxide of unknown thickness, the Q-value of this sample has a much larger error bar ($\pm 0.15 \pm 0.15i$) associated with it than for the other samples ($\pm 0.002 \pm 0.002i$). Note that the Q-value for this sample was significantly smaller than the Q-value of the SiC-capped sample, which is presumably due to oxidation of the former sample. The asymmetry was independent of θ, as predicted by the second term above, and so we were able to fit the asymmetry to obtain $4\pi\alpha$. The values of Q and $4\pi\alpha$ obtained by this procedure are listed in columns 2 and 3, respectively, of Table II.

Using values for the Fe(110) sample (see Table II): $Q = 0.022 + i0.006$, $n_0' = 2.3 - i2.4$, we obtain from Eq. (18) with $4\pi\alpha$ set to zero (Metzger's magneto-optic response at perpendicular incidence):

$$\theta_k + i\epsilon_k(0°) \cong (-14 + i0) \times 10^{-6} m_l m_t, \qquad (19)$$

which is at the limits of detection of our system.

Using the values of Q and n_0 given previously, and with $4\pi\alpha = -1.9 - i14.1$, we obtain from Eq. (18): $\theta_k + i\epsilon_k(0°) \cong (-47 + i708) \times 10^{-6} m_l m_t$, which matches well the observed magneto-optic response. Thus, we see that our model can explain a large Q^2 term, while that of Metzger, Pluvinage, and Torguet[6] cannot.

VI. CONCLUSION

We have examined other films [polycrystalline Fe, polycrystalline Fe with ion bombardment induced anisotropy, polycrystalline Ni, Fe(110)/Mo(110) multilayers, and Pt/Co multilayers with out-of-plane anisotropy] and find uniformly that those with in-plane anisotropy exhibit asymmetry in either the rotation or ellipticity and those without in-plane anisotropy exhibit no asymmetry. This is consistent with the in-plane anisotropy causing a coherent magnetization reversal that manifests itself in the asymmetry in the magneto-optic response.

In conclusion, the asymmetric magneto-optic response is due to a term in the magneto-optic response second order in the magnetization which is proportional to the product of the longitudinal and transverse components of **M**. The asymmetry is therefore an indicator of how well the rotation model of magnetization reversal applies to the sample being measured; in the absence of a coherently rotating magnetization, the asymmetry goes to zero. We have extended a previous theory of magneto-optics[6] to separately fit the terms first and second order in the magnetization, the latter of which is much larger than that predicted by Metzger, Pluvinage, and Torguet.[6]

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