Magneto-optic rotation and ellipticity of ultrathin ferromagnetic films

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Magneto-optic rotation ϕ' and ellipticity ϕ'' values are reported as a function of thickness for 0-400 Å of bcc Fe(100) epitaxially deposited on Au(100) for both *s*- and *p*-polarized He-Ne laser light. The values are derived from a formula that connects ϕ' and ϕ'' with our recently reported longitudinal Kerr-effect measurements of $\phi_m = (\phi'^2 + \phi''^2)^{1/2}$ and the parameters of two optical compensators that are used to convert ellipticity to rotation. The measurements were made *in situ* through an ultrahigh-vacuum window; our approach eliminates the effect of the window birefringence. The dominant contribution to ϕ_m reverses from being ϕ'' in the ultrathin limit to being ϕ' in the thick-film limit. Also, in the 0-30 Å range, the rotation remains near zero while the ellipticity increases linearly.

I. INTRODUCTION

The surface magneto-optic Kerr effect is a probe of magnetism on the nanometer scale of film thickness.¹ It provides a means of measuring magnetic hysteresis curves, *in situ*, from monolayer-range samples grown in ultrahigh vacuum (UHV). In such studies, contributions from the birefringence of the UHV window impede straightforward quantification of the results. In our previous work on Fe/Au(100), a method was given of correcting for the window effect by the use of two Soleil-Babinet compensators.² That procedure yields the magnitude of the complex magneto-optic rotation,

$$\phi_m = (\phi'^2 + \phi''^2)^{1/2} , \qquad (1)$$

where ϕ' is the magneto-optic rotation and ϕ'' is the ellipticity. In the present work ϕ' and ϕ'' are derived from the complex rotation measurements using the known compensator settings. The method of analysis is based on a theory derived here that relates the quantities of interest.

In the following section, the necessary experimental details are reviewed, including an operational description of the two-compensator scheme. Section III gives an algebraic analysis of the compensators and the means of separating rotation and ellipticity. Section IV gives the results of the separation, and Sec. V is a discussion of the magneto-optic properties in the ultrathin limit. Section VI gives a brief summary.

II. EXPERIMENTAL BACKGROUND

In order to appreciate the analysis, some critical experimental background details need to be recalled. In the experimental configuration, polarized He-Ne laser light passes through a UHV window, reflects from the sample back through the window, then passes through compensating optics (consisting of two Soleil-Babinet compensators) and an analyzing polarizer to the photodiode detector. The analyzing polarizer is set at a small angle $(\delta=2.35^{\circ})$ from extinction. The intensity of light reaching the photodiode is measured as a function of the applied magnetic field, which lies in both the plane of the film and the plane of incidence. From the resulting hysteresis curve the intensity change in the remanent state normalized by the average total intensity, I_K , is plotted as a function of film thickness.² The magnitude of the complex rotation ϕ_m is determined from I_K and δ . The reason ϕ_m is measured, as opposed to ϕ' , is due to the optical-compensation procedure used. Two Soleil-Babinet compensators are adjusted so as to convert ellipticity to rotation.^{2,3} This has the advantage of optimizing the weak signal associated with the ultrathin regime, and of correcting for the UHV-window birefringence. We show in the present work that the compensator settings can be used to reconstruct the sample rotation and ellipticity values.

The optical-compensation procedure has been previously described with a detailed Poincaré-sphere analysis;² an algebraic analysis will be given in the next section. The operational procedure for setting the two Soleil-Babinet compensators is as follows. (1) Magnetize the sample in the + direction. (2) Set the analyzing polarizer to be crossed with respect to the polarization of the incident beam. (3) With the first compensator (the one nearest the UHV window) in place and the second compensator removed, adjust the axis and retardation of the first compensator to achieve extinction. (4) Introduce the second compensator and set its axis to lie in the plane of incidence. (5) Select and set a retardation for the second compensator. (6) Adjust the analyzer to angle δ from extinction and reverse the magnetization to measure I_{K} . (7) Repeat steps 5 and 6 with different second-compensator retardations until I_K is maximized. The maximum I_K yielded by this procedure corresponds to $2\phi_m$, as will be shown below.

III. ANALYSIS OF THE OPTICAL-COMPENSATION PROCEDURE

The analysis of the optical compensation relies on three types of inputs: (i) the influence of the compensators; (ii) the definition of the Kerr effect, and (iii) the assumption of additivity of the window correction. The influence of a Soleil-Babinet compensator is expressed by a (2×2) transform:⁴

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$$\begin{vmatrix} E_{1x} \\ E_{1y} \end{vmatrix} = \begin{vmatrix} \cos\gamma + i \sin\gamma \cos(2\omega) & i \sin\gamma \sin(2\omega) \\ i \sin\gamma \sin(2\omega) & \cos\gamma - i \sin\gamma \cos(2\omega) \end{vmatrix}$$

where ω is the angle of the principal optical axis of the compensator with respect to the source polarization direction and 2γ is the retardation. The subscripts 0 and 1 correspond to the electric-field components of the light E_x, E_y before and after passing through the first compensator, respectively. The electric-field components also define the rotation ϕ'_{\pm} and ellipticity ϕ''_{\pm} in the small-angle approximation as

$$\frac{E_{0y}^{\pm}}{E_{0x}^{\pm}} = \phi'_{\pm} + i\phi''_{\pm} , \qquad (3)$$

where the + and - denote the sample magnetization directions. The assumption of additivity of the magneto-optic effect and the window birefringence is stated as

$$\phi'_{\pm} = \phi'_{W} \pm \phi', \quad \phi''_{\pm} = \phi''_{W} \pm \phi'', \quad (4)$$

where ϕ' and ϕ'' arise from the film and are of central interest, while ϕ'_{\pm} and ϕ''_{\pm} are the quantities detected. ϕ'_{W} and ϕ''_{W} arise from the window. This assumption will be checked by observing consistency with our experimental results.

With these inputs, the compensation procedure can be described algebraically. The first compensator has its values of ω and 2γ chosen so as to make the light for + sample magnetization linearly polarized in the x direction, i.e., to make $E_{1y} = 0$ in Eq. (2), thereby satisfying

$$\frac{i\sin\gamma\sin(2\omega)}{\cos\gamma - i\sin\gamma\cos(2\omega)} = -\frac{E_{0y}^{+}}{E_{0x}^{+}} = -(\phi'_{+} + i\phi''_{+}) .$$
 (5)

It then follows that γ and ω must satisfy

$$\tan(2\omega) = \frac{1}{\phi'_{+}} (\phi'^{2}_{+} + \phi''^{2}_{+})$$

and

$$\tan\gamma = \frac{-\phi'_+}{\cos(2\omega)\phi''_+} \ .$$

These settings of the first compensator are not changed when the sample magnetization is reversed. For – sample magnetization, the fields E_{1x}^- and E_{1y}^- after the first compensator satisfy [using Eqs. (2)–(5), and keeping terms up to first order in ϕ'_{\pm} and ϕ''_{\pm}]

$$\frac{E_{1y}^{-}}{E_{1x}^{-}} = \left[-(\phi_{+}' + i\phi_{+}'') + \frac{E_{0y}^{-}}{E_{0x}^{-}} \right] \exp(i\alpha)$$
$$= (-2\phi_{-}' - 2i\phi_{-}'')\exp(i\alpha) , \qquad (7)$$

where the phase α is defined by

$$\exp(i\alpha) = \frac{\cos\gamma - i\sin\gamma\cos(2\omega)}{\cos\gamma + i\sin\gamma\cos(2\omega)} . \tag{8}$$

We now define another phase β

$$\begin{bmatrix} E_{0x} \\ E_{0y} \end{bmatrix}, \qquad (2)$$

$$\tan\beta = \frac{\phi^{\prime\prime}}{\phi^{\prime}} \tag{9}$$

and rewrite Eq. (7) as

$$\frac{E_{1y}^{-}}{E_{1x}^{-}} = 2\phi_m \exp[i(\alpha + \beta)] .$$
(10)

It should be pointed out that while β depends on the Kerr rotation and ellipticity of the Fe film only, α depends on the compensator settings which are influenced by both the magneto-optic signal and the window. Equation (10), therefore, shows that the light exiting compensator 1 for — sample magnetization contains a window effect. This effect is corrected by the second compensator, which is used to cancel the phase in Eq. (10), thereby resulting in a maximal signal. This is shown in the following paragraph.

Let $\overline{\omega}$ and $\overline{\gamma}$ be the parameters of the second compensator. The light (E_{1x}, E_{1y}) , after passing the second compensator, will satisfy [using Eq. (2) and remembering that $\overline{\omega}=0$, i.e., that the axis of the compensator is set in the direction of the source polarization]

$$\frac{E_{2y}^{-}}{E_{2x}^{-}} = 2\phi_m \exp[i(\alpha + \beta - 2\overline{\gamma})] .$$
(11)

The maximal signal in the y direction is obtained when $2\overline{\gamma}$ cancels the phase in Eq. (11):

$$\alpha + \beta - 2\overline{\gamma} = 0 . \tag{12}$$

Equations (12) and (8) can be used to find the angle β . In the current experiments, ω for the first compensator is a very small angle (of the order of 1°). It therefore follows from Eqs. (8) and (12) that

$$\exp(i\alpha) \to \exp(-2i\gamma) , \qquad (13)$$

and

(6)

$$\beta = 2\gamma + 2\overline{\gamma} \quad . \tag{14}$$

Thus, measuring the retardation of the first (2γ) and second $(2\overline{\gamma})$ compensators will determine the angle β . This enables one to determine ϕ' and ϕ'' from the previously reported ϕ_m measurements, since Eqs. (1) and (9) yield

$$\phi' = \phi_m \cos\beta$$
 and $\phi'' = \phi_m \sin\beta$. (15)

IV. RESULTS

The results of the measurement of the magneto-optic rotation ϕ_m , taken from Ref. 2, are reproduced in Fig. 1. The values of β , determined according to Eq. (14), are shown in Fig. 2 as a function of Fe thickness. These values of β and ϕ_m were used, as per Eq. (15), to determine ϕ' and ϕ'' , as shown in Fig. 3. Figure 3(a) confirms

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FIG. 1. Magnitude of the complex magneto-optic rotation ϕ_m as a function of thickness of the Fe film. The line serves as a guide to the eye.

the expected opposite-sign rotations for s and p polarizations. Figure 3(b) shows the ellipticity for p polarization; the ellipticity for s polarization is similar in sign and shape, but has been omitted for clarity because its nearzero β values yield noisy results.

The error bars in Fig. 3 include contributions to the error from (i) the uncertainty in β that arises from the uncertainty in $2\overline{\gamma}$ and from (ii) the noise in ϕ_m . The retardations for the second compensator used to maximize I_K for a given film thickness were spaced apart by 11.7°, giving an error in $2\overline{\gamma}$ of $\pm 5.85^{\circ}$. This error in retardation gives an error in ϕ_m of only ~0.5%, which is smaller than the ~2% scatter in ϕ_m due to noise. The first compensator retardation error is much smaller, ~1°, and does not contribute significantly to the error bars in the figure.



FIG. 2. The phase $\beta = \arctan(\phi''/\phi')$ between ellipticity ϕ'' and rotation ϕ' , as a function of Fe film thickness. β is determined from the Soleil-Babinet compensator settings, as described in the text.



FIG. 3. (a) Magneto-optic rotation as a function of Fe film thickness. The results are derived from the measured complex rotation magnitude and the measured phase β , as described in the text. (b) Magneto-optic ellipticity as a function of Fe film thickness, for *p* polarization. The lines through the points are guides to the eye.

To check that the window birefringence was being properly compensated, experiments were carried out for a variety of window treatments. Measurements were made both before and after the removal, replacement, and baking of the window. A vacuum bakeout, in particular, has a marked effect on the window birefringence. The different window birefringences were evidenced by the very different settings required for the first compensator. Nevertheless, the values of β for a thick Fe film were reproduced to within the experimental error. This lends support to the working hypothesis of Eq. (4).

V. DISCUSSION

The results show that it is possible to separate ϕ' and ϕ'' using optical compensators. Methods for performing such separations using polarization modulation techniques and phase-sensitive detection are well known in the literature.⁵⁻⁷ The present approach was motivated by a need to correct for the birefringence of the UHV window.

The results shown in Fig. 3 point out the different behaviors of the magneto-optic quantities in the thick- and ultrathin-film limits. In the ultrathin limit, the ellipticity grows with film thickness, rising linearly from zero ellipticity at zero thickness. The rotation, however, remains near zero for the first few tens of Å and does not begin its linear increase until ~ 30 Å. For thicker films, the ellipticity peaks and decreases whereas the rotation grows and reaches a constant value. Thus, in the ultrathin limit, the linear growth in ϕ_m is due to the growth of the ellipticity, while in the thick limit ϕ_m is dominated by the rotation ϕ' . This change in the relative contributions of the rotation and ellipticity to the total magneto-optic effect is also reflected in the changing of β with film thickness (Fig. 2).

We can speculate on the origin of the different magneto-optic effects in the ultrathin and thick limits. One consideration is that the optical constants may be thickness dependent, as suggested by Kranz and Stremme.⁸ This could be due to intrinsic or extrinsic effects (e.g., purity of the film). An intrinsic consideration is that it is known that in the monolayer range, the electronic and magnetic structure can be radically different than that of thick films.⁹ This would undoubtedly change the magneto-optic response. The change would not persist over the thickness range of the present studies, however. The presence of the substrate, as described in our previous paper, is the more likely origin.² In the ultrathin limit the signal can be regarded as originating from a Faraday effect due to reflection from the substrate. The Faraday component would interfere with the conventional Kerr contribution and contribute to the phase shifts observed in Fig. 3 as a function of thickness. To further elucidate this interesting question, we will model the effect in a forthcoming publication.¹⁰

VI. CONCLUSION

The magneto-optic rotation ϕ' and the ellipticity ϕ'' are reported as a function of thickness, for s- and ppolarized He-Ne light, in the 0 to 400 Å range for a film of Fe grown epitaxially on Au(100). The results were derived from previously reported measurements using an analysis procedure we developed and derived herein. The rotation and ellipticity contributions are found to be strikingly different in the ultrathin region as compared to the thick limit.

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