Longitudinal magneto-optical Kerr effect in subwavelength thick ferromagnetic films investigated by Mueller matrix ellipsometry

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ABSTRACT

Accurately characterizing the magneto-optical (MO) Kerr and optical responses of subwavelength thick ferromagnetic films is of great significance in the field of MO devices. In this paper, an in-situ characterization method based on the Mueller-matrix ellipsometry in a longitudinal MOKE configuration has been proposed, with which the longitudinal MO Kerr effects phenomenon occurring in the sub-wavelength thick ferromagnetic films can be thoroughly investigated. We establish an analytical representation of the MO Kerr responses by simplifying the transfer-matrix model using a more appropriate constraint about the film thickness. By virtue of the high repeatability precision of the measured Mueller matrix better than 0.0002, the MO Kerr response signal of the ferromagnetic films with thicknesses far from the ultra-thin limit can be reliably captured, despite the well-known weak signal amplitudes. The MO coupling constant, the Kerr ellipticities and rotations can be further determined from the measured Mueller matrix. Experimental results on the Ni and NiFe 2O 4 films with thicknesses less than 100 nm demonstrate the proposed characterization approach based on the Mueller matrix ellipsometry as a practical and precise technique. As the film thickness increases to larger than the ultrathin film threshold, the proposed method can provide more reasonable characterization results, and the influence of film thickness on the MO responses can be more accurately evaluated.

1. Introduction

Magneto-optical Kerr effect (MOKE) is a typical physical phenomenon that occurs in a ferromagnetic material, which can be manifested by the magnetization-induced additional changes in the polarization state, amplitude, and phase of the light beam reflected from the material surface [1–3]. Benefiting from the high sensitivity to magnetization, ease of implementation, and in-situ deployment, MOKE has been widely used as a powerful probing tool for magnetic domains [4], 2D materials' ferromagnetism [5–7], the antiferromagnetic state [8,9], the magnetic order and spin reorientation transition [10,11]. Meanwhile, it also serves as a theoretical basis for various exciting applications such as plasmonic magneto-optical devices [12–14], magneto-optical Kerr switching [15], spintronics devices [16], bio- and gas-sensing [17–19], optical isolation [20], optical filter [21], light modulation [22], and optical nonreciprocity [23]. Among these applications [24], it remains worthy of precisely evaluating or thoroughly understanding the magneto-optical Kerr response of various magnetic film materials, especially ferromagnetic films with the actual or equivalent thickness located between optically ultra-thin and ultra-thick scales.

According to the spatial topology relationship between the magnetization direction, the material surface, and the incidence plane, the MOKE can be manifested in three types: transverse, longitudinal, and polar [25,26]. Take the longitudinal MOKE in the magneto-optically isotropic case as an example [27], which corresponds to the magnetization direction parallel to both the incident plane and the material surface. With the longitudinal magnetic field loading on the isotropic and ferromagnetic material, the oscillating dipoles polarized by the incident electric field will experience a Lorentz force [28], which is directly perpendicular to both the magnetization direction and the dipole motion direction. Then, the Lorentz force will cause an out-of-plane anisotropy of the magnetized material, manifested as a dielectric tensor with non-zero off-diagonal elements [29], which is responsible for the existence of the macroscopic magneto-optical (MO)
responses. Considering the inherent weak magnitude of the MO interaction [28], the characterization of the MOKE above-mentioned poses significant challenges to the previous MO measurement instrument based on polarization optics regarding measurement sensitivity, anisotropy-capturing ability, and measurement robustness. Meanwhile, owing to the symmetry-breaking effect triggered by the surface or interface of the stratified film structure [30,31], a notable disparity is expected to emerge in the MO responses between the bulk materials and the thin films. When the thickness of ferromagnetic films varies at a subwavelength scale, their magneto-optical and optical responses will manifest a dependency on the thickness, stemming from the size effect [32]. As for the bulk ferromagnetic materials with optically thick thickness, ellipsometry combined with the surface magneto-optical Kerr theory provides reliable and widely accepted results [33–35]. While for the ultra-thin ferromagnetic films, their magneto-optic Kerr responses are often represented by the analytical method, originating from the simplification of 4 × 4 transfer matrix formalism under the ultra-thin approximation assumption [35–38]. However, the ultrathin approximation, which corresponds to 2π|N|d/λ<1 for the single-layer ferromagnetic film or 2π|N|dN/d<1 for the multi-layer ferromagnetic stack [35,37], usually limits the thickness of the typical ferromagnetic film to single-digit nanometers. Therefore, these methods used for modeling and characterizing the bulk or ultrathin ferromagnetic materials are challenging to determine the MOKE for the ferromagnetic films with thickness located between optically ultra-thin and ultra-thick scales and investigate their thickness dependency.

During the past two decades, the mainstream MOKE measurement method for ferromagnetic films is the magneto-optical Kerr ellipsometry [39–45], which has attracted widespread attention owing to its ability to provide non-destructive, non-contact, and rapid measurements, its high precision, as well as its capacity to capture spectral feature information. Given that ellipsometry is a model-based technique, it is not surprising that the modeling and extraction methods for the MOKE have also gained significant attention [35,39–41,46–48]. Among them, the MOKE modeling method proposed by Zak has been used until now [46,37], in which the 4 × 4 transfer matrix has been used to describe the electromagnetic field transfer characteristics of the magnetized materials. However, heavy numerical work and multi-parameter regression are inevitable in the complicated modeling approach to achieve accurate results [50,51], which is urgently avoided by ellipsometry analysis, especially when exploring the MOKE in configurations such as multi-wavelength or multi-incidence. Moreover, the complicated model based on the 4 × 4 transfer matrix might hinder intuitive physical insight into the thickness-dependent MOKE phenomenon [35,36]. Therefore, Zak’s model is usually simplified to a theoretical tool for studying ferromagnetic ultrathin films with thicknesses much less than the light wavelength [35,37], which is widely adopted in most ellipsometric studies of the MOKE to date [52,53]. Oppeneer et al. [54], Gasche et al. [55], and Delin et al. [56], successively evaluated the theoretical MOKE signal of ferromagnetic metallic ultrathin films under saturated magnetization conditions. Mok et al. also proposed a MO generalized ellipsometry to measure the dielectric tensor εMO of ultrathin Co film with the thickness satisfying the thickness limit condition 2π|N|d/λ<1 [2]. However, in many practical magneto-optical devices [1–3,12–21], the actual and equivalent ferromagnetic film thickness cannot afford such a limitation of less than 10 nm but often lies in the range of several nanometers to subwavelength. Therefore, there is an urgent need to reveal the MO behaviors of magnetic films with thickness located between the ultra-thin and sub-wavelength scales to answer the questions such as, “are the MO properties of thicker films the same as that of widely studied ultra-thin films?”, “how can we efficiently decouple the magneto-optical and optical properties of subwavelength thick ferromagnetic films”, and “is there a thickness dependency of the magneto-optical coupling constant?”, etc.

To tackle the issue mentioned above, an analytic model for MOKEs has been proposed to describe the MO responses of the ferromagnetic films with thickness beyond the limit condition 2π|N|d/λ<1 under the longitudinal magnetization, in which the effects of thickness have been sufficiently considered. On this basis, an in-situ characterization method based on the Mueller-matrix ellipsometry in a longitudinal MOKE configuration is established to investigate the MO properties of ferromagnetic films with different thicknesses. Then the experimental measurements are carried out on the Ni and NiFe2O4 films with thicknesses less than 100 nm to demonstrate the validity of the approach.

2. Methods

2.1. Modeling method of the longitudinal MOKE

As depicted in Fig. 1(a), with the magnetization direction parallel to both the sample surface and the incidence plane, the MO interaction between the magnetized film and the probing light is manifested as the longitudinal MOKE [57]. The physical essence of the longitudinal MOKE is the modification of the isotropic film’s dielectric properties from isotropic to anisotropic, which can be described by the occurrence of the MO coupling constant Q in the non-zero off-diagonal elements of the dielectric tensor εMO [51], as shown in follows,

$$\begin{bmatrix}
1 & 0 & iQ \\
0 & 1 & 0 \\
-iQ & 0 & 1 \\
\end{bmatrix}$$  

Where N2 is the complex refractive index of the ferromagnetic film without magnetization. In the monolayer film structure shown in Fig. 1(b), the ferromagnetic film exhibits the anisotropic behavior, while the ambient medium and the substrate are still homogeneously isotropic. Correspondingly, the multiple interference of the beam in the sample needs to be described by using the medium boundary matrix A and the medium transmission matrix D simultaneously,

$$T = A_{d1}^\dagger A_{d} A_{1}^\dagger A_{1}$$  

Where A0, A1, and A2 are the medium boundary matrices of the ambient medium, the film layer, and the substrate, respectively. Moreover, the expressions of these medium boundary matrices can be found in the literature [35,46]. T is transmission matrix of the sample. D is the medium transmission matrix of film layer [58], which has the following form.

$$D_i = \begin{bmatrix}
U\cos\theta & U\sin\theta & 0 & 0 \\
-U\sin\theta & U\cos\theta & 0 & 0 \\
0 & 0 & U^{-1}\cos\theta & -U^{-1}\sin\theta \\
0 & 0 & U^{-1}\sin\theta & U^{-1}\cos\theta \\
\end{bmatrix}$$  

Where $U = \exp[-i2\pi N \cos(\theta_2)/\lambda]$ and $\sigma = \pi N \sin(\theta_2)/Q[\pi \cos(\theta_2)]$. Since the MO coupling constant Q is usually on the order of 0.01, it is a reasonable approximation that $\sigma << 1$ when the thickness d is in the range of [0, 2]. Correspondingly, we can approximate $\sin(\sigma) \approx \sigma$ and $\cos(\sigma) \approx 1$, and the terms containing $\sigma^2$ or $\sigma Q$ can be omitted.

In fact, the $4 \times 4$ transmission matrix T can be expressed as a $2 \times 2$ block matrices, like $T = [G, H; I J]$. Accordingly, the complex reflection coefficients can be calculated as follows [45],

$$\begin{bmatrix}
r_x & r_y \\
r_x' & r_y' \\
\end{bmatrix} = IG^{-1}$$  

Where r with the subscript index i, j = s or p quantifies the transformation from the incident i-polarized light to the reflective j-polarized light.

Since the thickness d2 of the ferromagnetic film lies in the range of several nanometers to subwavelength, i.e. $d_{\text{threshold}} \leq d_2 \leq \lambda/2$ with $d_{\text{threshold}}$ representing the thickness threshold for the ultrathin film, the thickness d2 does not satisfy the ultrathin condition $2\pi|N_2|d_2/\lambda << 1$ [30,46]. Correspondingly, the approximations $c = \cos(2\pi|N_2|\cos(\theta_2)/\lambda)$
\( s = \sin[2\pi d_N H \cos(\theta_2)/\lambda] \approx 2\pi d_N H \cos(\theta_2)/\lambda \) adopted in the case of the ultrathin ferromagnetic films are no longer valid. Instead of, much more rigid expressions for the complex reflection coefficients \( r_{ss} \), \( r_{pp} \), \( r_{sp} \) and \( r_{ps} \) has been derived without using the approximations \( \approx 1 \) and \( \approx \pi \), as shown in follows,

\[
\begin{align*}
    r_s &= \frac{N_1 N_s \cos \theta_1 \cos \theta_2 - N_2 N_s \cos \theta_1 \sin \theta_2}{N_1 N_s \cos \theta_1 \sin \theta_2 + N_2 N_s \cos \theta_1 \cos \theta_2 - i (N_1 N_s \cos \theta_1 \cos \theta_2)} \\
    r_p &= \frac{N_1 N_p \cos \theta_1 \cos \theta_2 - N_2 N_p \cos \theta_1 \sin \theta_2}{N_1 N_p \cos \theta_1 \sin \theta_2 + N_2 N_p \cos \theta_1 \cos \theta_2 - i (N_1 N_p \cos \theta_1 \cos \theta_2)} \\
    r_{mp} &= -r_{pm} = 2 \frac{d_N N_1 \cos \theta_1 (\cos^2 \theta_2 - \cos^2 \theta_1) + N_2 N_1 \cos \theta_1 (\cos^2 \theta_1 + \cos^2 \theta_2) - i (N_1^2 + N_2^2) \cos \theta_1 \cos \theta_2 - i (N_1^2 + N_2^2) \cos \theta_1 \cos \theta_2) \cos \theta_1}{N_1 N_p \cos \theta_1 \sin \theta_2 + N_2 N_p \cos \theta_1 \cos \theta_2 - i (N_1 N_p \cos \theta_1 \cos \theta_2)}
\end{align*}
\]

From Eq. (5a) to (5c), it can be seen that the longitudinal MOKE only affects the off-diagonal elements \( r_{mp} \) and \( r_{pm} \) of the sample’s Jones matrix rather than the main diagonal elements \( r_s \) and \( r_p \). That is to say, no matter whether the longitudinal magnetic field magnetizes the ferromagnetic film, its complex refractive index \( N_2 \) remains unchanged, which is the basis for us to decouple the optical properties and the MO coupling constant \( Q \) in the characterization of ferromagnetic films with thickness-dependent magnetization response. To facilitate the extraction of the optical constants \( N_2 \) and thickness \( d_2 \) of the ferromagnetic film from the ellipsometry, it is necessary to use the Jones matrix’s main diagonal elements to calculate the sample’s theoretical ellipsometric parameters. The corresponding formula is shown as follows,

\[
\tan(\psi_{\text{calc}}) \exp(i \Delta_{\text{calc}}) = r_{mp} / r_{pm} = f(N_2, d_2),
\]

where \( \psi_{\text{calc}} \) and \( \Delta_{\text{calc}} \) are the theoretical ellipsometric parameters, \( f \) represents the mapping relation between the ellipsometric parameters and material’s properties \((N_2, d_2)\). It should be noted that, both \( \psi_{\text{calc}} \) and \( \Delta_{\text{calc}} \) are transcendental functions of a ferromagnetic film’s complex refractive index \( N_2 \) and thickness \( d_2 \), corresponding to the Fresnel interference model of isotropic films [59]. The complex refractive index \( N_2 \) of Ni film is described by the combination of four Tauc-Lorentz oscillators and an infrared pole term [60], while the complex refractive index \( N_2 \) of NiFe\textsubscript{2}O\textsubscript{4} film is defined by the combination of two Tauc-Lorentz oscillators and an infrared pole term. Further, the \( 4 \times 4 \) Mueller matrix can be calculated from the Jones matrix in the case of minimal depolarization, as shown in follows,

\[
M = A \left( \begin{array}{cccc}
    r_{pp} & r_{pp} & r_{pp} & r_{pp} \\
    r_{pp} & r_{pp} & r_{pp} & r_{pp} \\
    r_{pp} & r_{pp} & r_{pp} & r_{pp} \\
    r_{pp} & r_{pp} & r_{pp} & r_{pp}
\end{array} \right) A^{-1},
\]

Where \( \otimes \) represents the direct product. A is a \( 4 \times 4 \) transform matrix, which can be found in the literature about ellipsometry [61]. Due to the algebraic equivalence of the transformation shown in Eq. (7), the Mueller matrix can inherit typical advantages of the Jones matrix, like estimating the magnitude and orientation of the LMOKE by their non-trivial off-diagonal elements with symmetry. Meanwhile, by combination with the matrix’s differential decomposition [61], the Mueller matrix also provides a unique capability of revealing which one among some typical anisotropic features, such as linear birefringence, linear dichroism, or circular birefringence, is directly correlated with the longitudinal MOKE.

2.2. Measurement method of longitudinal MOKE

According to the schematic diagram in Fig. 1(a), the in-situ measurement system for characterizing the longitudinal MOKE can be developed by combing a micro-spot Muller matrix ellipsometer with a longitudinal magnetic field loading module. Fig. 2(a) shows the detailed system, while Fig. 2(b) depict the corresponding prototype system. In the magnetic field loading module, two NdFe\textsubscript{B} permanent magnets placed opposite each other in the horizontal plane were used to generate a roughly uniform magnetic field, and correspondingly the sample was placed at the midpoint of the line connecting the two magnets. Meanwhile, by rotating the whole loading module, the two permanent magnets can be located in the ellipsometer’s incident plane to realize the longitudinal loading of the magnetic field. It should be noted that the two permanent magnets were fixed on the slider of the screw-nut transmission mechanism by the aluminum alloy casing. Then, the
The distance between the two magnets could be manually adjusted using the screw-nut transmission mechanism, which allows for achieving a magnetic field with a variable strength covering a range of 0–210 mT. The magnetic field strength at each different spacing between the magnets can be calibrated using a commercial Teslameter (HT20, Shanghai Hengtong Inc.) with a measurement precision of 0.01 mT, and the corresponding results are shown in the insert illustration of Fig. 2(b). Through setting the magnets’ spacing according to the magnetic field strength curve, the MO response of Ni and NiFe$_2$O$_4$ films under the loading of longitudinal magnetic field with different magnitudes could be captured by the measurement system. Both Mueller matrix and ellipsometric parameters were measured by an ellipsometry module (RC2, J. A. Woollam) used in the in-situ measurement system, in which a measurement spectrum covering a range of 193–1690 nm and variable incidence angles covering a range of 45–75° could be accessed.

Benefiting from the fact that the entire magnetic field loading module is fixed on a rotatable sample stage, the direction of the magnetic field loading can be precisely adjusted relative to the incident plane of the ellipsometer. Correspondingly, using the azimuth calibration technology based on the one-dimensional standard Si grating [62], the parallel relationship between the magnetic field loading direction and the incident plane can be reliably guaranteed to realize the longitudinal magnetic field loading configuration.

According to the established model, both $r_{ss}$ and $r_{pp}$ are independent of the MO coupling constant $Q$, that is, the longitudinal MOKE will not change the principal diagonal elements in the Jones matrix or the principal diagonal block in the Mueller matrix. Therefore, the complex refractive index $N_2$ and the thickness $d$ of the ferromagnetic film can be determined by the regression analysis of the ellipsometric parameters captured by the in-situ measurement system, in which the procedure is similar to the ellipsometric analysis of typical isotropic films [63]. Then, the MO coupling constant $Q$ can be obtained by analyzing the off-diagonal elements of the Mueller matrix with the combination of the pre-determined $N_2$ and $d$, according to Eq. (5c). To more conveniently use the self-built in-situ measurement system to simultaneously determine the optical constants $N_2$, the thickness $d$, and the MO parameters of the ferromagnetic film, we proposed a two-step extraction method for both MO Kerr and optical parameters based on the measured Mueller matrix, as shown in Fig. 3. Firstly, the thickness $d$ and optical constants $N_2$ can be determined by fitting the principal diagonal block elements in the Mueller matrix measured at the incident angle of 55° with the theoretical values calculated by a parametrized forward optical model [60]. The merit function $\chi^2$ used in the least-squares regression analysis is shown as follows.

$$\chi^2 = \sum \frac{(M_{\text{meas}} - M_{\text{calc}})^2}{\sigma^2}$$
\[ x^2 = \frac{1}{2N_i} P \sum_{i=1}^{N_i} \left[ \left( m_{12,i}^{\text{meas}} + m_{21,i}^{\text{meas}} + 2\cos 2\psi_i^{\text{calc}} \right)^2 + \left( m_{33,i}^{\text{meas}} + m_{44,i}^{\text{meas}} - 2\sin\psi_i^{\text{calc}} \cos \Delta_i^{\text{calc}} \right)^2 \right], \]

where \( I \) represents the \( i \)-th wavelength point from the total wavelength number \( N_i \), \( P \) is the number of the measurands, \( m_{12,i}^{\text{meas}} \), \( m_{21,i}^{\text{meas}} \), \( m_{33,i}^{\text{meas}} \), \( m_{44,i}^{\text{meas}} \), \( m_{31,i}^{\text{meas}} \), and \( m_{42,i}^{\text{meas}} \) are the principal diagonal block elements in the measured Mueller matrix, while \( \psi_i^{\text{calc}} \) and \( \Delta_i^{\text{calc}} \) are the calculated ellipsometric parameters reported by the forward optical model.

Subsequently, a parameter-extraction method based on the analytical representations of longitudinal MOKE parameters will be adopted to determine the MO coupling constant \( Q \), the Kerr rotations \( \varphi \), and the Kerr ellipticities \( \varsigma \), in which the fine fitting of the off-diagonal elements in the Mueller matrix can be avoided. Since the values of the off-diagonal elements in the Mueller matrix are minute, nearly on the order of \( 10^{-2} \), the accuracy for fitting the off-diagonal elements is difficult to guarantee. This is the fundamental reason for using the analytical expression based on the Mueller matrix elements to extract the magneto-optical response parameters. Owing to the measurement errors in the off-diagonal elements of the measured Mueller matrix, the anisotropy caused by the longitudinal MOKE will be evaluated using the difference between the off-diagonal elements of the samples’ Mueller matrix before and after the magnetic field loading. Thus, the Mueller matrix \( \Delta M_{\text{mag}} \) of a magnetized sample will be calculated by the following formula.

\[
\Delta M_{\text{mag}} = \begin{bmatrix} m_{22}^{\text{norm}} & m_{23}^{\text{norm}} & m_{24}^{\text{norm}} & m_{21}^{\text{norm}} \\ m_{32}^{\text{norm}} & m_{33}^{\text{norm}} & m_{34}^{\text{norm}} & m_{31}^{\text{norm}} \\ m_{42}^{\text{norm}} & m_{43}^{\text{norm}} & m_{44}^{\text{norm}} & m_{41}^{\text{norm}} \end{bmatrix},
\]

Further, in order to quantify the additional changes in the polarization state of reflection light induced by the magnetization, the Kerr rotations \( \varphi \) and the Kerr ellipticities \( \varsigma \) are usually adopted in the MOKE characterization experiments, which can be determined from the Mueller matrix \( \Delta M_{\text{mag}} \).

\[
\begin{align*}
\varphi_p + i\varsigma_p &= \frac{-r_{pp} - \Delta m_{13}^{\text{norm}} + \Delta m_{22}^{\text{norm}} - i(\Delta m_{14}^{\text{norm}} + \Delta m_{24}^{\text{norm}})}{2r_{pp}} \quad (13.a) \\
\varphi_s + i\varsigma_s &= \frac{-r_{ss} - \Delta m_{33}^{\text{norm}} + \Delta m_{44}^{\text{norm}} + i(\Delta m_{31}^{\text{norm}} + \Delta m_{42}^{\text{norm}})}{2r_{ss}} \quad (13.b)
\end{align*}
\]

where the subscript \( p \) and \( s \) denote \( p \)- and \( s \)-polarized light, respectively.

2.3. Descriptions of samples and experiments

Given the benefits of electron beam evaporation, such as compatibility with magnetic targets, high film purity, uniform density distribution, and precise thickness control, all the measured nickel thin films were prepared using the technique (OHMIKER-508 E-Beam Vapor System, Cello Technology Co., Ltd.). Before the film growth process, a single-sided polished single-crystal Si (100) wafer was cut into some substrates of \( 20 \times 20 \text{ mm}^2 \) along the crystal direction using a diamond knife. To ensure the film quality, these Si substrates need to be successively cleaned in the ultrasonic ambient by the cleaning agents such as ethanol, acetone, and deionized water, enabling the removal of possible pollutants on the wafer surface. By appropriately adjusting the evaporation rate and time and using a quartz-crystal film-thickness monitor for real-time monitoring, it is possible to accurately deposit Ni films with 10 and...
40 nm nominal thicknesses. The NiFe\textsubscript{2}O\textsubscript{4} films with nominal thicknesses of 25, 40, and 80 nm were prepared by the laser pulse deposition method (Customized Instrument, TSST), in which the thicknesses were controlled by adjusting the pulse number and the pulse energy. To suppress the natural oxidation of the Ni films and the pollution of NiFe\textsubscript{2}O\textsubscript{4} films by the atmospheric atmosphere as much as possible, the prepared Ni and NiFe\textsubscript{2}O\textsubscript{4} films were quickly put into vacuum bags for storage and transportation. Moreover, the interval between the sample preparation and the characterization experiments was shortened as much as possible to further reduce the influence of natural oxidation on the measurement results. Then, the initial remanence of the Ni and NiFe\textsubscript{2}O\textsubscript{4} films was checked by measuring the magnetic field strength on the samples’ surface with the commercial Teslameter. The measurement results close to zero indicate that the samples’ remanence in the initial state can be negligible.

The surface topography of each Ni film was characterized by the AFM (MultiMode 8, Bruker Corporation), and the thickness of the 10nm-thick Ni film was also checked by measuring the height difference of stairs located on the film edge via the AFM. Meanwhile, the thickness of 100nm-thick NiFe\textsubscript{2}O\textsubscript{4} films was checked through observing the cross-sectional structure by SEM (Helios NanoLab G3 CX, FEI Corporation), and its surface roughness was characterized by the white light interferometer (NewView 9000, Zygo Corporation Co., USA). Subsequently, these Ni and NiFe\textsubscript{2}O\textsubscript{4} films in their initial states were carefully characterized by the original Mueller matrix ellipsometer at an incident angle of 55°, in which their optical constants, thicknesses, and surface roughness were estimated. It is worth emphasizing that these physical parameters in the initial state will be the reference benchmarks for the measured results in the subsequent longitudinal magneto field loading experiments. After the above experiments, the Mueller matrices of the magnetized samples were captured by the in-situ measurement system, in which the magnitude of the magnetic field can be changed by varying the distance between the two magnets. Finally, the in-plane hysteresis loops of Ni and NiFe\textsubscript{2}O\textsubscript{4} films were measured using a vibrating sample magnetometer (VSM 7404, LakeShore), which allows us to determine the saturation magnetization of the two samples. Since the magnetized samples still have residual magnetism even if the loaded magnetic field is removed, the experimental steps should be strictly followed.

3. Results and discussions

Fig. 4 shows the auxiliary experimental characterization results of Ni thin film and NiFe\textsubscript{2}O\textsubscript{4} thin film, including surface morphology, SEM section, and hysteresis loop. From the results reported by AFM shown in Fig. 4(a) and (b), the surface roughness of 10 and 40 nm-nominal-thick Ni films are 0.390 and 0.652 nm, respectively, which reflect the excellent film-growth quality. Since it is challenging to avoid Ni films’ natural oxidation during the characterization experiment and an intertwined mixing between the oxide layer and the surface roughness layer exists, we use the equivalent medium approximate model to describe their effects in the ellipsometric analysis for the initial state. The thicknesses of the equivalent layers on the Ni film surface are 0.25 and 0.47 nm, respectively, reported from the ellipsometry analysis. These results indicate that the natural oxidation on the surface of Ni films is negligible, consistent with metallic film’s expected natural oxidation behavior [65]. The possible reasons for the negligible natural oxidation are that the Ni films were sealed in vacuum bags immediately after the sample preparation, and the interval between the film growth and characterization experiments did not exceed 2 h. Immediately after these initial state characterizations, the samples were also sealed in vacuum bags until the Mueller matrix ellipsometry characterization of the longitudinal magneto-optical Kerr effect.

According to the surface morphology of 80nm-nominal-thick NiFe\textsubscript{2}O\textsubscript{4} film shown in Fig. 4(c), the surface roughness is just 1.869 nm, which indicates good film quality. The cross-sectional view of NiFe\textsubscript{2}O\textsubscript{4} film captured by SEM is shown in Fig. 4(d), which tells a film thickness of 87.7 nm. According to the in-plane hysteresis loop of 10nm-nominal-thick Ni film shown in Fig. 4(e), the external magnetic field of $H = \pm 100$ mT can enable the in-plane saturation magnetization of the Ni film. By setting the magnets’ spacing at 62 mm in the experimental setup, the in-situ measurement system can easily capture the MO responses of Ni films under the saturation magnetization state. As for the apparent difference between $\pm 100$ mT measured in our experiments and the $\pm 30$ mT reported in the literature [2], it might be attributed to the difference in the sample preparation process and material purity. While the external magnetic field of $H = \pm 290$ mT can enable the in-plane saturation magnetization of the 80nm-nominal-thick NiFe\textsubscript{2}O\textsubscript{4} film.
film, according to the in-plane hysteresis loop curves of NiFe$_2$O$_4$ film shown in Fig. 4(f). Limited by the sample stage’s physical size and the probe beam’s spatial layout, only a stable magnetic field of up to 210 mT can be generated by shortening the distance between the two magnets, which is also the magnetic field we adopted when characterizing the MO responses of NiFe$_2$O$_4$ films. Although the magnetic field fails to magnetize the NiFe$_2$O$_4$ completely, it is sufficient for us to study the magneto-optic Kerr properties of the magnetized film under unsaturated magnetization.

Fig. 5. Reliability verification of optical properties measured by ellipsometry. Real (a) and imaginary (b) parts of the dielectric functions of 10 nm-thick Ni film. (c) Absorption coefficients of 80 nm-thick NiFe$_2$O$_4$ film.

Fig. 6. Comparison of off-diagonal elements of Mueller matrix measured at an incidence angle of 55° for the 10nm-thick Ni thin film with and without the magnetic field loading.

Fig. 7. Comparison of the MO coupling constants $Q$ for (a) 10nm- and (b) 40nm-nominal-thick Ni films characterized by the proposed and the previous method.
3.1. Validation of ellipsometric results

Although the ultra-high measurement accuracy of typical ellipsometry for the thickness of nano-film materials has been widely recognized, it is still essential to pre-validate the accuracy of the ellipsometric results for the above ferromagnetic films. The actual thickness of 10nm-nominal-thick Ni film was determined as 11.78 nm from the ellipsometry analysis, which is very close to the step height of 11.2 nm measured by AFM at the edge of the Ni film. Meanwhile, the dielectric functions of the Ni film are highly similar to the results reported in the literatures [66–69], as shown in Fig. 5(a) and (b). The ellipsometric thickness of the 80nm-nominal-thick NiFe$_2$O$_4$ film is determined as 84.33 nm, which is very close to the SEM measurement result of 87.70 nm. The absorption coefficient determined by the ellipsometry analysis is similar to the results in the literature [70,71], as shown in Fig. 5(c). Correspondingly, the band gap of 1.68 eV is very close to Holinsworth’s measurement result of 1.64 eV [70]. Besides, the Ni films’ surface roughnesses of 0.39 and 0.65 nm indicate the high quality of film growth, which lays a good foundation for ensuring the reliability of the ellipsometric results. The magnitude of the depolarization effect occurring in Ni and NiFe$_2$O$_4$ films is smaller than 2.0%, which also indicates the high growth quality. The above comparative analysis results provide conclusive evidence to support the accuracy and reliability of the ellipsometric results.

3.2. MO responses and optical properties of Ni films

Due to the weak nature of the MO coupling interaction between the typical ferromagnetic films and the longitudinal magnetic field [72], the sample will only exhibit a tiny optical anisotropy. Correspondingly, the off-diagonal elements in the measured Mueller matrix of the magnetized Ni film are always relatively small, which can be easily overwhelmed by measurement error. To overcome the limits of the instrument precision and to achieve higher sensitivity to the magnetization-induced changes, the difference of these off-diagonal elements in the Mueller matrices measured before and after the magnetization has been utilized as the signal to be analyzed, just following Eq. (9). Benefiting from the instrument’s high repeatability precision on the order of $10^{-4}$, the optical response caused by the longitudinal MOKE can be identified. Fig. 6(a–h) compares the off-diagonal elements in the Mueller matrix of 10nm-nominal-thick Ni film before and after the magnetization. The dashed lines represent the measurement results before the magnetization, while the solid lines correspond to the measured results under the saturation magnetization caused by the 100 mT magnetic field. As shown in Fig. 6 (a–h), the results achieved under the same external conditions are highly consistent, while a significant difference exists between the results achieved under the two different external conditions. It indicates that the observed difference in the measurement results is indeed induced by the longitudinal MOKE. This provides a reliable basis for establishing the in-situ characterization method based on the difference of off-diagonal elements in the Mueller matrix measured under the two external conditions. Moreover, such a delicate process following Eq. (9) can minimize the influence of instrument precision. The method significantly differs from the classic characterization method based on electromagnet magnetization in forward and reverse directions [2,43].

Further, the MO coupling constant $Q$ can be obtained according to Eq. (12), where the cross-reflection coefficient $r_{sp}$ required in Eq. (12) is obtained from the synthesized Muller matrix $\Delta M_{mag}$. The corresponding values of $Q$, covering a spectral range of 193–1690 nm, are presented in Fig. 7(a). It can be easily found that both the real and imaginary parts of $Q$ are highly similar to that measured by Mok in terms of the magnitude, the variation trend with wavelength, and the line shape [2,26]. As for the difference between the measured results and Mok’s results, it can be attributed to different preparation processes of samples and differences in the dielectric functions. Considering the weak nature of the longitudinal MOKE, the similarity summarized above is enough to explain the reliability and accuracy of the measured results. It is worth emphasizing that the measured near-infrared $Q$ might play a potential role in more comprehensively revealing the effects of net spin polarization and electronic band structure on the magneto-optical properties of Ni films [2].

The comparison results between 10nm- and 40nm-nominal-thick Ni films are shown in Fig. 7(b), while the reference MO coupling constants $Q$ of these two films are also presented in Fig. 7(b). The former reference results were calculated using the conventional ultrathin film model [34], while the latter was calculated using the bulk model [35]. The measured $Q$ for the 10nm-nominal-thick Ni film is very close to that calculated by the conventional ultrathin film model, which is attributed to the rationality of the approximations $c = \cos(2\pi dN_s/\lambda)$ and $s = \sin(2\pi dN_s/\lambda)$ in the case of 10nm-nominal-thick Ni film. However, with the film thickness increasing to 40 nm, the ultrathin film model is no longer valid, and a more rigorous consideration of the thickness effect in the MO response model will be indispensable. According to the results shown in Fig. 7(b), the $Q$ of the 40nm-thick Ni film is significantly different from that of the 10nm-thick Ni film, which may be caused by the enhanced metallicity and the increased optical path difference in the Ni film due to the thickness increase. The former can lead to a significant change in the dielectric function of the Ni film in the spectrum from visible to near infrared light, while the latter causes the appearance of the in-plane Faraday effect, both of which will significantly enhance the MO coupling effect. When comparing the measured $Q$ for the 40nm-thick Ni film with the reference results calculated from the bulk model [35], the high consistency between the two is exceptionally striking. Considering that 40 nm is highly close to the mean free path of conduction electrons in metallic films [73], i.e.,
the critical thickness used to distinguish the metallic bulk and film, it is unsurprising to observe good consistency between the two results of $Q$. It is worth emphasizing that the high agreement further demonstrates the self-consistency and accuracy of the proposed method.

The Kerr ellipticities $\varsigma$ and rotations $\phi$ calculated from the measured Mueller matrix are shown in Fig. 8. With the film thickness increasing, both the Kerr ellipticities and rotations increase nearly in the whole spectrum. Also, it is easy to notice that the effects of increasing the thickness on the Kerr ellipticities of $p$- and $s$-polarized components are similar, manifesting as the downward shift of the ellipticity curves in the spectrum. As for the Kerr rotations, the effects of increasing the thickness will exhibit significant dichroism. Specifically, increasing the

![Fig. 9. Comparison of off-diagonal elements in the Mueller matrix for the 80nm-nominal-thick NiFe$_2$O$_4$ thin film with and without the magnetic field loading.](image1)

![Fig. 10. The MO coupling constant $Q$ of NiFe$_2$O$_4$ films with different thicknesses. (a) The $Q$ of 25nm-nominal-thick NiFe$_2$O$_4$ film characterized by the proposed and ultra-thin-model method. (b) The thickness-dependent MO coupling constant $Q$ of NiFe$_2$O$_4$ films.](image2)

![Fig. 11. The off-diagonal elements $\Delta m_{13}^{norm}$ and $\Delta m_{31}^{norm}$ in the synthesized Muller matrix $\Delta M_{mag}$ of 10nm- (a) and 40nm-nominal-thick (b) Ni films.](image3)
thickness makes the Kerr rotation of the p-polarized light significantly increase toward the negative value direction, while the Kerr rotation of the s-polarized light increases toward the positive value direction with the thickness increasing. Besides, the Kerr rotations of p- and s-polarized components for 10nm-thick Ni film at 632.8 nm are 0.023° and 0.008°, respectively, which are very close to the corresponding results of ferromagnetic Co ultrathin film reported in the literature [35]. This similarity in the Kerr parameters of materials within the same chemical element group also verifies the validity of the above measurement results. The above characterization results will provide a valuable reference for precise angle measurements based on the MOKE in the future.

3.3. MO responses and optical properties of NiFe$_2$O$_4$ films

Similar to the measurement results of Ni films, the off-diagonal elements in the Mueller matrix of NiFe$_2$O$_4$ films with different thicknesses still exhibit discrepancies before and after loading the 210 mT magnetic field. The representative results for the 80nm-nominal-thick NiFe$_2$O$_4$ film are shown in Fig. 9. It can be easily noticed that the unsaturated Kerr rotation of the s-polarized light increases toward the positive value direction, while the Kerr rotation of the p-polarized light significantly dominates the Kerr rotation of the film as the thickness increasing. Besides, the Kerr rotations of p- and s-polarized light shown in Fig. 10. While the disparity observed in the near-infrared spectrum could be attributed to the fact that the thickness of 25 nm does not satisfy the ultra-thin film threshold condition [30, 46]. The ultraviolet spectrum can be attributed to the fact that the thickness of the ultraviolet and near-infrared bands, which shows that the ultrathin-film responses. The relatively significant difference in the magnitude of the relative changes in these Mueller matrix elements is larger than the repeatability precision of the instrument, which enables the reliable confirmation of the longitudinal MOKE phenomena happening in the NiFe$_2$O$_4$ film. The magnitude of the relative changes in $m_{13}$ and $m_{23}$ of NiFe$_2$O$_4$ film is larger than that of Ni film, which indicates that the longitudinal MO Kerr response of NiFe$_2$O$_4$ film is stronger than that of Ni film. This will also be reflected in subsequent MO coupling constant Q.

Correspondingly, the magneto-optical coupling constant Q can be determined from the synthesized Muller matrix $\Delta M_{\text{mag}}$, as shown in Fig. 10. According to the results in Fig. 10(a), the Q of 25nm-nominal-thick NiFe$_2$O$_4$ film measured by the proposed method is similar to that calculated by the conventional ultrathin film model regarding the line shape and the magnitude value. It indicates that the wavelength dependency of the two results is highly consistent. However, the Q determined by the two methods is significantly inconsistent in the ultraviolet and near-infrared bands, which shows that the ultrathin-film model is challenging to calculate the sample’s magneto-optical Kerr responses. The relatively significant difference in the magnitude of Q in the ultraviolet spectrum can be attributed to the fact that the thickness of 25 nm does not satisfy the ultra-thin film threshold condition [30, 46]. While the disparity observed in the near-infrared spectrum could be attributed to the film surface effect rather than the violation of the ultrathin assumption [35].

Fig. 10(b) exhibits the MO coupling constant Q of NiFe$_2$O$_4$ films with different thicknesses. With the film thickness increasing, the MO coupling constant Q increases obviously in the spectrum of 193–900 nm, while the MO coupling constant Q in the near-infrared band only shifts to the negative value direction. The whole line shape of Q does not shift up or down consistently as expected but only increases in amplitude, which may be attributed to the sharp change of the optical-path phase $\Delta = 2\Delta dN$,$\Delta dN = \epsilon_{2}/\lambda$ with the film thickness. As for the Q of the 80nm-nominal-thick NiFe$_2$O$_4$ film, the prominent peak and valley values in the spectrum from 300 to 600 nm may be caused by the sharp increase of the Faraday effect. At the peak or valley wavelength of 500 nm, the optical-path phase $\Delta = 2\Delta dN$,$\Delta dN = \epsilon_{2}/\lambda$ caused by the film thickness d = 80 nm can reach $\pi$ with the complex refractive index $N = 2.93 - i0.41$, which makes the off-diagonal element $r_{sp}$ approaching to a peak and makes the Faraday effect pronounced. Such a vast magneto-optical coupling strength will provide meaningful guidance for developing magneto-optical devices based on NiFe$_2$O$_4$ thin films.

3.4. Magnetization orientation estimation from the Mueller matrix

Another benefit of the in-situ proposed in our work is the ability to discriminate the magnetization direction qualitatively, which can be realized using the symmetry of the off-diagonal elements in the synthesized Muller matrix $\Delta M_{\text{mag}}$. By comparing the off-diagonal elements $\Delta m_{i3}^{\text{norm}}$ and $\Delta m_{i3}^{\text{norm}}$ shown in Fig. 11(a), the rough symmetry of $\Delta m_{13}^{\text{norm}}$ and $\Delta m_{31}^{\text{norm}}$ about $\Delta m_{13}^{\text{norm}} \approx 0$ can be found, which are consistent with the longitudinal MOKE results reported in Mok’s work [2, 43]. Thus, we can identify the magnetization orientation as the longitudinal direction, which is parallel to the incident plane. With the thickness increasing to nearly 40 nm, the symmetry of $\Delta m_{13}^{\text{norm}}$ and $\Delta m_{31}^{\text{norm}}$ about $\Delta m_{13}^{\text{norm}} = -0.00025$ is weakened as shown in Fig. 11(b), which might indicate that the apparent symmetry is just easily obtained only in the case of ultrathin films. Meanwhile, it can be noticed that the intersection of $\Delta m_{13}^{\text{norm}}$ and $\Delta m_{31}^{\text{norm}}$ corresponds to a wavelength of around 900 nm, and the intersection point can shift to a shorter wavelength with the thickness increasing, which are similar to the results in literature [2, 43].

Similarly, we also observed the symmetry of the off-diagonal elements in the synthesized Muller matrix $\Delta M_{\text{mag}}$ of the NiFe$_2$O$_4$ films, as shown in Fig. 12. As for the 25 nm-nominal-thick NiFe$_2$O$_4$ film, the symmetry of $\Delta m_{13}^{\text{norm}}$ and $\Delta m_{31}^{\text{norm}}$ about $\Delta m_{13}^{\text{norm}} = 0 = 0.00013$ can be easily noticed, as shown in Fig. 12(a). While the symmetry of $\Delta m_{13}^{\text{norm}}$ and $\Delta m_{31}^{\text{norm}}$ will gradually weaken with the film thickness increasing, which can be seen in the results for the 40 and 80 nm-nominal-thick NiFe$_2$O$_4$ films, as shown in Fig. 12(b–c). From these results, it is reasonable to infer that the symmetry of $\Delta m_{13}^{\text{norm}}$ and $\Delta m_{31}^{\text{norm}}$ might be more pronounced in the case of ultrathin films. Thus, it is possible to determine the magnetization orientation using the symmetry of the off-diagonal elements in the synthesized Mueller matrix $\Delta M_{\text{mag}}$, especially in the ultrathin film cases.

When checking the symmetry of other off-diagonal elements in the synthetic Muller matrix, it is uneasy to obtain observable results due to the tiny magnitude of other off-diagonal elements. This suggests that the symmetry of $\Delta m_{13}^{\text{norm}}$ and $\Delta m_{31}^{\text{norm}}$ dominates the symmetry between $r_{sp}$ and $r_{sp}$ predicted by the theoretical model. Furthermore, by introducing the Mueller-matrix differential decomposition technique [61], the origin of the anisotropy manifested as the symmetry of $\Delta m_{13}^{\text{norm}}$ and $\Delta m_{31}^{\text{norm}}$ can

Fig. 12. The off-diagonal elements $\Delta m_{13}^{\text{norm}}$ and $\Delta m_{31}^{\text{norm}}$ in the synthesized Muller matrix $\Delta M_{\text{mag}}$ of 25nm- (a) and 40nm- (b) and 80nm-nominal-thick (c) NiFe$_2$O$_4$ films.
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Fig. 13. The linear birefringence (LB) and dichroism (LD) of Ni and NiFe$_2$O$_4$ films. (a) LB and (b) LD of Ni films, (c) LB and (d) LD for NiFe$_2$O$_4$.

be further revealed. Correspondingly, the resultant linear birefringence (LB) and dichroism (LD) along the p-s axes of the reference frame are shown in Fig. 13, which are quite apparent in magnitude. While the linear birefringence (LB') and dichroism (LD') along the ±45° axes, the circular birefringence (CB) and dichroism (CD) are all on the order of $10^{-3}$, much smaller than LB and LD. Thus, these four elementary properties can be negligible and are not presented in Fig. 13.

These results indicates that the symmetry of $\Delta M_{33}^{\text{mag}}$ and $\Delta M_{31}^{\text{norm}}$ might be attributed the LB and LD caused by the longitudinal magnetization. As shown in Fig. 13(a), the LB of Ni films decreases with increasing thickness, corresponding to the decreasing difference in Kerr rotations between the p- and s-polarized light shown in Fig. 8(b). While the reverse and shrink of LD caused by increasing the thickness, as shown in Fig. 13(b), roughly corresponds to the reverse and decreasing difference in Kerr ellipticities between the orthogonal polarizations shown in Fig. 8(a). The self-consistency among these results once again confirms the reliability of the proposed method. Fig. 13(c and d) exhibits the dramatic variations in LB and LD with the increasing of NiFe$_2$O$_4$ films’ thickness, which can be attributed to the thickness-dependent MO coupling strength.

4. Conclusions

In summary, we have proposed an in-situ characterization method based on the Mueller-matrix ellipsometry in the longitudinal MOKE configuration, which enables the investigation of the MO Kerr and optical responses of ferromagnetic films with thicknesses located between optically ultra-thin and subwavelength scales. Meanwhile, a two-step analysis method, which consists of the regression analysis of the principal diagonal block elements in the Mueller matrix and the parameter-extraction method based on the analytical representations of longitudinal MOKE parameters, has been proposed to simultaneously determine the dielectric functions, the MO coupling constant, the Kerr rotations, and the Kerr ellipticities. The core of the method lies in the derivated analytical formulas of the magneto-optical Kerr effect, obtained from simplifying the transfer-matrix model using a more appropriate constraint about the film thickness. The analytical model, emphasizing a more rigorous consideration of the thickness effect, allows us to more accurately describe the magneto-optic Kerr response of ferromagnetic films with subwavelength thickness.

By virtue of the high repeatability precision of the measured Mueller matrix better than 0.0002, the MO Kerr response signal of the ferromagnetic films with thicknesses larger than the ultra-thin limit $2\pi|d|/\lambda<<1$ can be reliably captured, despite the well-known weak MO signal amplitudes. Experimental results on the Ni and NiFe$_2$O$_4$ films with thicknesses in the range of 10–80nm demonstrate the accuracy and reliability of the proposed method. Compared with the existing ultra-thin film model, the proposed method can provide more reliable characterization results for those subwavelength thick ferromagnetic films and more accurately evaluate the thickness dependency of the MO responses. Besides, the magnetization orientation can be roughly identified by checking the symmetry of the off-diagonal elements $\Delta M_{33}^{\text{mag}}$ and $\Delta M_{31}^{\text{norm}}$ in the synthesized Mueller matrix $\Delta M_{\text{mag}}$. Furthermore, by introducing the Mueller-matrix differential decomposition, the origin of the anisotropy manifested as the symmetry of $\Delta M_{33}^{\text{mag}}$ and $\Delta M_{31}^{\text{norm}}$ can be revealed as the linear birefringence and dichroism caused by the longitudinal magnetization. Predictably, the proposed measurement method of the MOKE will provide a more reliable and accurate way for the optimal design and performance evaluation of sub-wavelength thickness magneto-optic devices, even with the non-ideal polarization characteristics case.

CRediT authorship contribution statement

Jiamin Liu: Conceptualization, Methodology, Software, Validation, Writing – original draft. Wenci Gong: Writing – review & editing, Visualization. Lei Li: Writing – review & editing, Formal analysis. Song Zhang: Writing – review & editing, Formal analysis. Jinlong Zhu: Writing – review & editing, Formal analysis. Rong Chen: Writing – review & editing, Funding acquisition, Conceptualization. Hao Jiang: Supervision, Project administration, Writing – review & editing, Funding acquisition, Conceptualization. Shiyan Liu: Supervision, Project administration, Writing – review & editing, Funding acquisition, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.
Data availability

Data will be made available on request.

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