Ellipsometric selective sensitivity to magnetic nanostructures

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Abstract
Magneto-optic (MO) ellipsometry of ferromagnetic materials is extremely sensitive to ultra-thin films, multilayers, and nanostructures. It gives a possibility to measure all components of the magnetization vector in the frame of the magneto-optic vector magnetometry and enable us to separate magnetic contributions from different depths and materials in nanostructures, which is reviewed in this article. The method is based on ellipsometric separation using the selective MO Kerr effect. The figure of merit used to quantify the ellipsometric selectivity to magnetic nanostructures is defined on the basis of linear matrix algebra. We show that the method can be also used to separate MO contributions from areas of the same ferromagnetic materials deposited on different buffer layers. The method is demonstrated using both: (i) modeling of the MO ellipsometry response and (ii) MO measurement of ultra-thin Co islands epitaxially grown on self-organized gold islands on Mo/Al\textsubscript{2}O\textsubscript{3} buffer layer prepared using the molecular beam epitaxy at elevated temperatures. The system is studied using longitudinal (in-plane) and polar (perpendicular) MO Kerr effects.

Keywords: generalized ellipsometry, magneto-optical ellipsometry, material sensitivity, magnetic nanostructures

1. Introduction
Complexity of recent magnetic nanostructures requires improvement of characterization techniques and development of new measurement procedures. Among nondestructive and non-invasive techniques, the ellipsometric methods propose high sensitivity to films with thicknesses down to an atomic layer. Magneto-optical (MO) effects, i.e. change of optical properties originating from magnetized state of materials, enables applications of ellipsometric methods to magnetic measurement. The magneto-optical ellipsometry profits from all advantages of standard ellipsometry and moreover it is less affected by surface roughness and nonmagnetic films in a structure.

Basic application of MO ellipsometry is the hysteresis loop measurement based on detection of the MO signal proportional to magnetization as a function of external magnetic field. The method is used to monitor the magnetization reversal, magnetic anisotropy, hysteresis properties etc [1]. Its combination with microscopic observation, often called Kerr microscopy [2], is used to observe magnetic domains during magnetization reversal. Despite observed domain size is limited by the resolution of optical microscopy, the Kerr microscopy has significantly contributed to understanding of domain nucleation and domain wall motion. Important information related to electronic structure of a material is obtained from spectral dependence of MO effect. Magneto-optic spectroscopic ellipsometry usually based on modulation technique can be combined with standard ellipsometry to obtain spectral dependence of the complete permittivity tensor. Specific techniques based on MO ellipsometry are ultrafast magneto-optics, Brillouin light scattering [3], or second harmonic MO generation.

All the above mentioned applications of MO ellipsometry techniques profits from high near-surface sensitivity, nondestructive character, and possibility to measure all components of the magnetization vector in the frame of the magneto-optic vector magnetometry [4, 5]. Moreover, the magneto-optic Kerr effects enable us to separate magnetic contributions from different depth [6–10] and from different materials [11, 12] in multilayer systems and self-organized nanostructures [13].

In this paper, the MO ellipsometry is applied to characterize cobalt ultra-thin film deposited on self-assembled gold islands on Mo buffer layer. Selective sensitivity of both the longitudinal and polar MO effect to ferromagnetic Co grown on Au islands and Co grown directly on Mo buffer in the nanostructure is demonstrated experimentally and explained using model. In Sec. 2 we review some aspects of MO ellipsometry and MO vector magnetometry crucial for measured data understanding and processing for selectively sensitive MO ellipsometry. Section 3 deals with depth and material sensitivity of MO ellipsometry, procedure of separation of MO effect from different phases, and definition of the figure of merit for the separation. Section 4 presents the main original results of MO selective sensitivity affected by buffer layer. Separation of magnetic contribution is explained using model and processing of experimental MO data of Co film grown on nanostructured Au/Mo buffer.

2. Magneto-optical ellipsometry
In this section we define the magneto-optical angles, discuss basic MO configurations and possibility of magnetization com-
ponent separation in the frame of the MO vector magnetometry [4]. Careful separation of magnetization components and different MO effects is the first step, which precede to separation of contributions from different materials.

2.1. Basic definitions

Usual description of MO response of a sample is based on the Jones reflexion matrix

\[ R = \begin{pmatrix} r_{ss} & r_{sp}(M_p, M_L) & r_{pp}(M_T) \\ r_{sp}(M_p, M_L) & r_{pp}(M_T) \\ r_{pp}(M_T) & r_{pp}(M_T) & r_{pp}(M_T) \end{pmatrix}, \]  

(1)

where \( r_{ij}, \ i, j = s, p \) denote the amplitude reflection coefficients. The ratios of the off-diagonal and diagonal reflection coefficients are used for definition of two MO ellipsometric angles

\[
\frac{r_{sp}}{r_{ss}} = \tan \psi_{sp} \exp(i\Delta_{sp}) = \frac{\tan \theta_s + i \tan \epsilon_s}{1 - i \tan \theta_s \tan \epsilon_s} \approx \theta_s + i \epsilon_s, \\
\frac{r_{pp}}{r_{sp}} = \tan \psi_{pp} \exp(i\Delta_{pp}) = \frac{\tan \theta_p + i \tan \epsilon_p}{1 - i \tan \theta_s \tan \epsilon_p} \approx \theta_p + i \epsilon_p. \tag{2}
\]

The first forms define the generalized ellipsometric angles \( \psi_{sp,pp} \) (related to the amplitudes) and \( \Delta_{sp,pp} \) (represent the phases) defined in Refs. [14–16]. Similar definitions were also used in Refs. [17–19]. Note that the ratio \( r_{pp}/r_{ss} = \tan \psi \exp(i\Delta) \) defines the standard ellipsometric angles [20].

The second definition of the magneto-optic angles \( \theta_{sp,pp} \) and \( \epsilon_{sp,pp} \) has clear meaning: For the incident linear polarizations in the usual MO ellipsometry configurations use: (i) differential intensity measurement based on Wollaston prism and differential signal between two photodetectors, (ii) polarization modulation technique using the photoelastic modulator (PEM), and (iii) the azimuth modulation and nulling method based on Faraday modulator cells. Another aspect of the MO ellipsometry is the measurement of MO angles differences related to given magnetization state (usually saturated state by external magnetic field).

Small amplitude of MO effect has also important consequences in *additivity* of various MO contributions. MO effect can be usually understood as a small perturbation of optical properties (for instance small anisotropy or gyrotropy) originating from magnetization in materials. Therefore, the total MO effect can be obtained as addition of particular contributions. Here we consider MO contributions from different depth, films, materials, constituents, etc. Additivity of MO effects will be used in Sec. 3 and 4 for separation of different materials in magnetic nanostructure. This linear approximation is usually fulfilled (in all cases discussed in this paper) and it can be confirmed by a model.

2.2. Magneto-optic vector magnetometry

Contributions of different magnetization components (usually mixing of polar and longitudinal MO effects) appear in a similar way as the contributions from different materials. Therefore, important task of magneto-optical ellipsometry is careful separation of different magnetization components. We usually distinguish three basic components of the magnetization vector (see Fig. 1): the polar component \( M_p \) is perpendicular to the film (surface), the transverse component \( M_T \) is perpendicular to the plane of incidence, and the longitudinal one lies in the plane of film and in the plane of incidence. Dependencies of the amplitude reflection coefficients on different magnetization components is schematically shown in Eq. (1).

We note that discussion here is restricted to linear MO effects in systems with cubic or higher symmetry, the effects of quadratic MO terms were discussed in Refs. [5, 21, 22]. The transverse Kerr effect can be observed only using the \( r_{pp} \) reflection coefficient [23].

On the other hand, both the longitudinal and polar Kerr effect can be monitored using the conversion reflection coefficients \( r_{tp}, r_{ps} \), or using the MO angles defined by (2). For their separation, one can use property of in-plane longitudinal and transversal effect, which disappear for normal incidence (for films with higher symmetry). However, changing the angle of incidence can be inconvenient and exactly normal incidence is hard to be used experimentally. Another possibility is based on different symmetries of polar and longitudinal Kerr effects [24]. Interesting method was proposed by Ding et al. [25] based on two measurement for reversed incidence geometries. In this paper, we use modified approach based on rotation of the sample and magnetic field around the sample normal by the angle of 180°. Figures 2a,b show two configurations used for polar and longitudinal MO effect separation by addition and subtraction of both measured signals, respectively. Note that both the sample and also the magnetic field have to be rotated. Daboo et al. [26, 27] proposed to measure both in-plane magnetization components by rotation of the sample and magnetic field by 90°. The method has significant advantage of direct comparison of both components, because both are measured using the longitudinal MO effect. Therefore, measurements of four sample orientations shown on Fig. 2 result in separation of three magnetization components denoted \( M_p, M_L, M_T \). Table 1 summarizes the effects of the sample and magnetic field rotation on different off-diagonal permittivity tensor elements. The polar MO effect is symmetric by 180° rotation, while longitudinal and transverse effects are antisymmetric. Note that the quadratic (second-order) effect related to product \( M_L M_T \) appears in \( \epsilon_{xy} \), has the same symmetry as the polar MO effect (see Table 1), and can be separated from the longitudinal one [28].
Figure 1: Basic MO configurations – polar, longitudinal and transverse component of magnetization vector.

Figure 2: Separation of the magnetization components using sample rotation. Magnetic field has to rotate together with the sample.

Table 1: The off-diagonal permittivity tensor components as a function of sample rotation \( \varphi \) used for magneto-optic vector magnetometry. Transformation of permittivity tensor was obtained by rotation of the coordinate system around \( z \)-axis. Component notation corresponds to the coordinate system shown on Fig. 1.

<table>
<thead>
<tr>
<th>measured by</th>
<th>( \varphi = 0^\circ )</th>
<th>( \varphi = 90^\circ )</th>
<th>( \varphi = 180^\circ )</th>
<th>( \varphi = 270^\circ )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( e_{xy} )</td>
<td>( r_{px} )</td>
<td>( r_{px} )</td>
<td>( e_{yx}(M_P) )</td>
<td>( e_{yx}(M_P) )</td>
</tr>
<tr>
<td>( e_{xz} )</td>
<td>( r_{px} )</td>
<td>( r_{px} )</td>
<td>( -e_{xz}(M_L) )</td>
<td>( e_{xz}(M_L) )</td>
</tr>
<tr>
<td>( e_{yz} )</td>
<td>( r_{px} )</td>
<td>( r_{px} )</td>
<td>( e_{yz}(M_T) )</td>
<td>( -e_{yz}(M_T) )</td>
</tr>
</tbody>
</table>

Figure 3: Separation of the magnetization components using sample rotation. The diﬀerent behavior is essential for depth selective sensitivity and comes from phase change by propagation through Cobalt above the film. Even more illustrative is a plot in the complex magneto-optic plane, where the \( x \) and \( y \) axis correspond to \( \theta \) and \( \epsilon \), respectively (lower subplot of Fig. 3). Each point corresponds to the film in diﬀerent depth. The points far from the origin represent ﬁlms close to the surface, while the points close to the origin correspond to deeper films (amplitude of the eﬀect decrease due to absorption and limited penetration depth). However, also the phase of the complex MO eﬀect change, which is used for depth selectivity. For example two vectors show contributions from diﬀerent depths. We note that optimal separation is obtained for perpendicular vectors of the same length.

3. Selective sensitivity of MO ellipsometry

In this section, the basic principle of MO selective sensitivity is described using an example of depth sensitivity in reﬂection from a magneto-optic bulk material. We discuss extension of the method to distinguish MO contributions from constituents in nanostructures (periodic multilayers or self-organized nanostructures).

3.1. Basis of MO selectivity to magnetic nanostructures

Basic idea of MO selective sensitivity is demonstrated on the case of depth sensitivity of the MO eﬀect in a cobalt bulk. The model is based on additivity of MO eﬀects discussed in Sec. 2.1. Figure 3 shows a model of magneto-optical ultrathin (1 Å thick) Co ﬁlm buried in non-magneto-optic Co bulk in the depth \( t \). The similar modeling was originally proposed by Traeger [6] and Hubert [7]. Fig. 3 shows the complex polar Kerr eﬀect obtained for normal incidence. Upper subplot of Fig. 3 shows dependence of the Kerr rotation \( \theta \) and ellipticity \( \epsilon \) as a function of the buried MO ﬁlm depth. While the sensitivity of the rotation \( \theta \) monotonically decrease due to absorption in Co, the ellipticity \( \epsilon \) shows more complex behavior and even changes sign. The diﬀerent behavior is essential for depth selective sensitivity and comes from phase change by propagation through Cobalt above the ﬁlm. Even more illustrative is a plot in the complex magneto-optic plane, where the \( x \) and \( y \) axis correspond to \( \theta \) and \( \epsilon \), respectively (lower subplot of Fig. 3). Each point corresponds to the ﬁlm in diﬀerent depth. The points far from the origin represent ﬁlms close to the surface, while the points close to the origin correspond to deeper ﬁlms (amplitude of the eﬀect decrease due to absorption and limited penetration depth). However, also the phase of the complex MO eﬀect change, which is used for depth selectivity. For example two vectors show contributions from diﬀerent depths. We note that optimal separation is obtained for perpendicular vectors of the same length.

Based on above discussed dependence of the Kerr rotation and ellipticity one can separate contributions from diﬀerent depths using several methods. One possibility is to adjust some parameter (the angle of incidence, wavelength [8]) in such a way that the measured MO quantity becomes insensitive to the contribution from one depth and only sensitivity to desired depth remains. However, this method is not very convenient because of complicated alignment of the parameter. More eﬀective technique was proposed by Ferré et. al [8, 9] using adjustable compensating element into the MO ellipsometer. The authors used the Babinet-Soleil compensator, which enables continuous transformation of the Kerr rotation into ellipticity and vice verse. Such adjustment corresponds to rotation of the complex MO plane (lower subplot of Fig. 3) around the origin. Despite of general applicability, the method requires sev-
3.2. Methods for separation of different contributions

Separation of MO effects from different depths or materials is obtained in three steps [11, 13]: (i) measurement of different MO quantities (for example the MO Kerr rotation $\theta$ and ellipticity $\epsilon$) showing different sensitivity to the contributions, (ii) determination of weight coefficients from the model of the structure or from some knowledge of magnetic behavior, and (iii) numerical separation of contributions from different depth or materials in the nanostructure. This section describes this procedure in details.

Based on additivity of MO effects the obtained signals can be expressed as weighted sums of MO contributions from different magnetic phases. Here we describe dependence of the Kerr rotation $\theta$ and ellipticity $\epsilon$ on two magnetic phases, however generalization to more measured quantities (different incident polarizations, angles of incidence, or inspected light wavelengths) and more magneto-optic phases is straightforward. Based on linearity $\theta = a_1 m_1 + a_2 m_2$ and $\epsilon = b_1 m_1 + b_2 m_2$, where $m_1$ and $m_2$ are the normalized magnetizations of two phases. It is convenient to write the relations in the matrix form:

$$\Phi = \begin{bmatrix} \theta \\ \epsilon \end{bmatrix} = \begin{bmatrix} a_1 & a_2 \\ b_1 & b_2 \end{bmatrix} \begin{bmatrix} m_1 \\ m_2 \end{bmatrix} = A \mathbf{M},$$

where $\Phi$ represents the vector consisting of measured signals by different MO quantities, $\mathbf{M}$ is the vector of normalized magnetizations of different phases in the structure, and the matrix $A$ contains the weight coefficients. The length of the vector $\mathbf{M}$ has to be less than or equal to the length of the vector $\Phi$. In the following we expect the same length and squarness of the matrix $A$. If we determine the elements of the matrix $A$, which will be discussed later, the magnetization of both phases can be obtained using the matrix inversion:

$$\mathbf{M} = A^{-1} \Phi.$$  

There are two aspects of the inversion (4). Firstly, the procedure to determine elements of matrix $A$ is discussed. We have two possibility: (i) calculation of the weight constants $a_{1,2}$ and $b_{1,2}$ from a model, which is straightforward, but it is affected by precision of input optical, magneto-optical, and geometric parameters of the structure. In many cases of practical interest the model is approximate and gives only qualitative agreement with the experiment. Therefore we often use the second possibility (ii) estimation of weight coefficients in the $A$ matrix directly from experimental data using our knowledge of magnetic behavior of magnetic phases [11, 13]. We, for example know, that for maximal external magnetic field both magnetic phases are saturated and we may expect that for a certain field one phase is already saturated, while the other is not. Separation on this basis was applied in Refs. [11, 13].
Secondly, we have to care, whether the matrix inversion \( A^{-1} \) is mathematically possible and how the experimental errors in measurement of \( \Phi \) and imperfect determination of the matrix \( A \) influence errors in obtained \( M \). It is clear, that if two vectors in the complex MO plane (see Fig. 3) are collinear (both MO quantity is identically sensitive to two magnetic phases), the lines in the matrix \( A \) are linearly dependent. The matrix is singular and inversion is not possible. On the other hand, if the vectors in the complex MO plane are perpendicular, the matrix is well conditioned and the matrix inversion can be calculated. We can define the figure of merit describing possibility of magnetic phases separation using the matrix condition number \( \kappa(A) \).

When we measure the MO effect \( \Phi \) with the tolerance \( \Delta \Phi \), then relative error of magnetic contributions

\[
\frac{||\Delta M||}{||M||} \leq \kappa(A) \frac{||\Delta \Phi||}{||\Phi||} \tag{5}
\]

is proportional to the condition number \( \kappa(A) \) [37, 38]

\[
\kappa(A) = \frac{||A^{-1}||}{||A||} = \frac{\sigma_{\text{max}}(A)}{\sigma_{\text{min}}(A)} \tag{6}
\]

where \( ||A|| \) is the norm of the matrix \( A \). In our calculations we used the spectral norm [37, 38] and the condition number is expressed from the ratio of the maximal \( \sigma_{\text{max}}(A) \) and the minimal \( \sigma_{\text{min}}(A) \) singular numbers obtained from the singular value decomposition of the matrix \( A \). \( \kappa(A) \) is closed to 1 for well conditioned matrix and magnetic contributions are well separated. However, for large \( \kappa(A) \), the MO ellipsometry is not selectively sensitive to different magnetic phases [36].

4. Experimental demonstrations

This section deals with demonstration of the selective sensitivity in the case of ferromagnetic cobalt film deposited on the self-organized Au islands on the Mo buffer layer. Figure 4 shows schematically the sample structure. The sample was grown on a monocristalline sapphire (1120) substrate in molecular beam epitaxy (MBE) system. The Au islands are self-assembled on the 20 nm thick Mo(110) epitaxial layer at elevated temperature of 500 °C. Such patterned buffer was covered by the ferromagnetic Co (0001). The measured data presented in this paper were obtained for Co film thickness of approximately 2 nm. The system was capped by 5 nm thick Au film to protect Co from oxidation. Therefore, two ferromagnetic sub-systems coexist in the sample: (i) Au/Co/Au sandwiches – for the part where the Co layer is deposited on the Au islands and (ii) Au/Co/Mo sandwich – as the Co layer is grown between islands on the surrounding Mo surface. Direct growth of Co on Mo buffer results in uniaxial magnetic anisotropy originating from lattice mismatch between Co and Mo [39].

MO selective sensitivity of the sample was modeled using coherent summation of contribution from both subsystems. The total Jones matrix (1) was obtained by weighted summation of particular Jones matrices of \( R_{\text{Au/Co/Au}} \) and \( R_{\text{Au/Co/Mo}} \):

\[
R = p \cdot R_{\text{Au/Co/Au}} + (1-p) \cdot R_{\text{Au/Co/Mo}}, \tag{7}
\]

where \( p \) is the surface fraction of Au islands. MO angles were calculated from the total \( R \) using Eq. (2). Figure 5 shows the modeled MO contributions of both magnetic fractions in the complex MO plane as a function of the surface fraction \( p \). Left and right subplots represents the polar and longitudinal Kerr effects, respectively. Blue circles and red squares correspond to contributions of Co on gold islands and Mo buffer, respectively. Both the polar and longitudinal MO effects show clear possibility of selective sensitivity to two magnetic subsystems.

The sample was studied using MO Kerr ellipsometer based on differential intensity detection. Semiconductor laser beam of wavelength 670 nm modulated using Polarizer/PEM/Polarizer system at 100 kHz incident at the angle of incidence of 60° on the sample. Polarization of the reflected beam is measured using the achromatic quarter-wave compensator and the Wollaston prism with two detector systems. In-plane direction of external magnetic field is computer controlled together with motorized rotation of the sample. In this paper, we present data obtained for \( p \)-polarized incident light.

Figure 4: Schematic plot of the sample structure of Co grown on self-assembled Au islands

Figure 6 shows MO hysteresis loops after separation of all three components of the magnetization vector measured using the Kerr rotation \( \theta_p \) and ellipticity \( \epsilon_p \) (separation procedure is described in Sec. 2.2, Fig. 2). The loops obtained using the Kerr rotation \( \theta_p \) (upper line in Fig. 6) are significantly different from the loops of the Kerr ellipticity \( \epsilon_p \). Therefore, existence of two contributions from different magnetic systems is clearly evident. We suppose that the upper \( \theta_p \)-loops comes mainly from the Au/Co/Au islands. They contribute also to the lower \( \epsilon_p \)-
loops, but the effects of the surrounding Cobalt deposited on Mo buffer dominate in the lower $\varepsilon_p$-loops. In the following we discuss magnetic behavior: (i) both magnetic phases exhibit the polar magnetization components for in-plane magnetic field. However, the polar components are relatively small and the magnetizations are only slightly inclined out of the plane. (ii) Co grown on Mo buffer shows coherent magnetization rotation and strong in-plane magnetic anisotropy, which is explain by lattice mismatch [39], (iii) Longitudinal loop for Au/Co/Au islands is wider and smoother indicating the higher coercive field its distribution for individual islands.

5. Conclusions

Magneto-optical selective sensitivity is described on the basis of rigorous modeling and experimental observation in ultrathin Co films on nanostructure of self-assembled gold islands. Despite the fact that the lateral dimension of nanostructure is smaller than inspected light wavelength, we are able to separate magnetic response of Co deposited on Au island and on surrounding Mo buffer. Such possibility originates from different MO phase resulted from effect of buffer layer, which significantly affect also magnetic properties. The presented method is general and we believe that it is applicable for various systems of multilayers, periodic structures, and nanocomposites measured by different MO effects.

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