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In situ magneto-optical ellipsometry data analysis for films growth control

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ABSTRACT

In this work we present the way of ferromagnetic films study by means of magneto-ellipsometry. The method of interpretation of in situ magneto-optical ellipsometry spectra for real time growth control is described. The method has been successfully tested on Si/SiO_2/Fe films within the model of a homogeneous semi-infinite medium. As a result, the dielectric tensor components for Fe layer were calculated using a developed approach.

1. Introduction

In recent years, the magnetic materials for data storage and spintronic devices have deserved significant attention. There is a problem of an in situ real time control of nanomaterials synthesis [1] and their properties investigation because the in-air investigation of these structures is often impossible due to the high chemical activity of many materials used in this field. One of the best solutions of this problem is to use the optical and magneto-optical techniques. They are powerful, do not affect the sample and have some flexibility when being used in situ, directly in an ultrahigh vacuum chamber. We suggest that magneto-ellipsometry [2] meets all these requirements. This experimental technique combines ellipsometry [3] and the magneto-optical Kerr effect measurement within one setup with an ultra-high vacuum chamber and the electromagnet for magnetization reversal of the sample [4].

Although several attempts have been made to design a single setup for determining magneto-optical and conventional optical constants in it, few studies have focused on data processing that can be conducted right in the process of materials synthesis. Mok in his study [5] faced the necessity of carrying out an additional experiment. He had to determine magnetization in order to extract magnetic-field-dependent and independent parts in non-symmetric terms of a dielectric permittivity tensor. This indicates a need to write a sufficient data processing algorithm so that it is applicable to studying samples with magneto-ellipsometry measurement without using any other setups despite that one that is used for synthesis. The use of this software would reduce the time and increase efficiency of experiment data analysis.

In this paper, we suggest a new approach to real time control of obtaining material parameters of magnetic thin films that can be applied right in the process of their growth by means of the in situ magneto-optical ellipsometry. In the end, to address the validity of the proposed method, we carried out an experiment on Si(substrate)/SiO_2(layer)/Fe(layer) film study and compared the obtained values of material parameters with those in [6].

The results we present here indicate that the offered method is truly sufficient, simple, and reliable for in situ spectral magneto-ellipsometric measurements data interpretation.

2. Magneto-ellipsometry data analysis

Here we describe the method of interpretation of the magneto-ellipsometric measurements data. We consider the case of electromagnetic wave incidence from non-magnetic dielectric medium (characterized by the refraction index $N_0 = n_0 - ik_0$) onto ferromagnetic metal (the refraction index $N = n - ik$) in the visible light range.

In the setup, a Cartesian coordinate system is defined with the x-axis normal to the interfaces and pointing into the substrate from the sample surface. The y- and x-axis lie in the plane of incidence. There can be three configurations: longitudinal (L), transverse (T), and polar (P) defined according to the direction of the magnetization vector. According to the design of the setup [4], we consider T-configuration in which magnetization is z-axis directed, i.e perpendicular to the plane of incidence and parallel to the surface.

The key idea of the proposed approach is reported in [7], where it was applied toward the particular case of low magnetic field and

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consequently the use of small parameters. Here we consider a general case of experimental data processing for the model of a homogeneous semi-infinite medium without any constraints.

We suggest that magneto-ellipsometry technique gives an opportunity to determine all elements of the dielectric permittivity tensor ε of the magnetized ferromagnetic metal [8]

$$\hat{\varepsilon} = \begin{bmatrix} \varepsilon_{11} & \varepsilon_{12} & 0 \\ \varepsilon_{21} & \varepsilon_{22} & 0 \\ 0 & 0 & \varepsilon_{33} \end{bmatrix} = \begin{bmatrix} \varepsilon'_{11} - i\varepsilon''_{11} & -i(\varepsilon'_{12} - i\varepsilon''_{12})Q & 0 \\ i(\varepsilon'_{12} - i\varepsilon''_{12})Q & \varepsilon'_{11} - i\varepsilon''_{11} & 0 \\ 0 & 0 & \varepsilon'_{11} - i\varepsilon''_{11} \end{bmatrix} \tag{1}$$

where ε' and ε'' are real and imaginary parts of medium permittivity, respectively, $Q = Q_1 - iQ_2$ is a proportional to magnetization magneto-optical parameter. Diagonal tensor elements are responsible for refractive index and extinction coefficient, off-diagonal tensor elements are related to magnetooptical effects. So we get quite a lot of information on the sample if we know all the elements of dielectric permittivity tensor. The current work presents how to obtain the values of these elements from magneto-ellipsometric spectra without necessity of any additional ex situ measurements.

Let us denote the ellipsometric parameters in the non-magnetic condition as ψ_0 and Δ_0. In the case of magneto-ellipsomertic characterization of the sample the surface transverse magneto-optical Kerr effect results in the ellipsometric angles corrections $\delta\psi$ and $\delta\Delta$. Thus, the ellipsometric parameters become $\psi_0 + \delta\psi$, $\Delta_0 + \delta\Delta$. It means that we have four measured independent real-valued quantities (ψ_0, $\delta\psi$, Δ_0, $\delta\Delta$), as a result, we can derive four real-valued quantities (ε'_{11}, ε''_{11}, ε'_{12}, ε''_{12}).

There are four steps of data analysis:

1. Carrying out spectral ellipsometry (ψ_0, Δ_0) and magneto-optical Kerr effect measurements ($\psi_0 + \delta\psi$, $\Delta_0 + \delta\Delta$).

2. Calculation of spectral dependences of refractive index (n) and extinction coefficient (k)

$$N = n - ik = N_0 \sin\varphi_0 \sqrt{\frac{1 + \tan\varphi_0^2 (1 - \tan\psi_0 e^{i\Delta_0})^2}{(1 + \tan\psi_0 e^{i\Delta_0})^2}} \tag{2}$$

3. Theoretical calculation of the ellipsometric parameters ψ_0, $\delta\psi$, Δ_0, $\delta\Delta$. Here we rewrite the basic equation of ellipsometry in the following way:

$$\tan(\psi_0 + \delta\psi)\exp(i(\Delta_0 + \delta\Delta)) \quad = R_p R_S^{-1} = (R'_p - iR''_p)(R'_S - iR''_S)^{-1} \tag{3}$$

where R_p and R_s are complex reflection coefficients corresponding to in-plane p-polarization and out-of-plane s-polarization, respectively, real parts are marked by ′, imaginary by ″. According to mode conversion from the p- to the s-polarized channel we can write that

$$R_p = R_{pp} + R_{pS} = R'_{p0} + R'_{p1} - i(R''_{p0} + R''_{p1}) \tag{4}$$

$$R_S = R_{SS} + R_{Sp} = R_{S0} = R'_{S0} - iR''_{S0} \tag{5}$$

$$R'_{p0} = \alpha_1(AA_1 + 2BB_1) \tag{6}$$

$$R''_{p0} = \alpha_1(AB_1 - 2BA_1) \tag{7}$$

$$R'_{p1} = \alpha_1(A^2C - 4B^2C + 4ABD) \tag{8}$$

$$R''_{p1} = \alpha_1(A^2D - 4B^2D - 4ABC) \tag{9}$$

$$R'_{S0} = \alpha_2(A_2C_2 + 2B_2D_2) \tag{10}$$

$$R''_{S0} = \alpha_2(B_2C_2 - 2A_2D_2) \tag{11}$$

where we have distinguished the magnetic field contribution and marked it by subscript 1, non-magnetic summands – by subscript 0. By substituting the determined values of n and k into Eqs. (6)–(11) and using the following notations

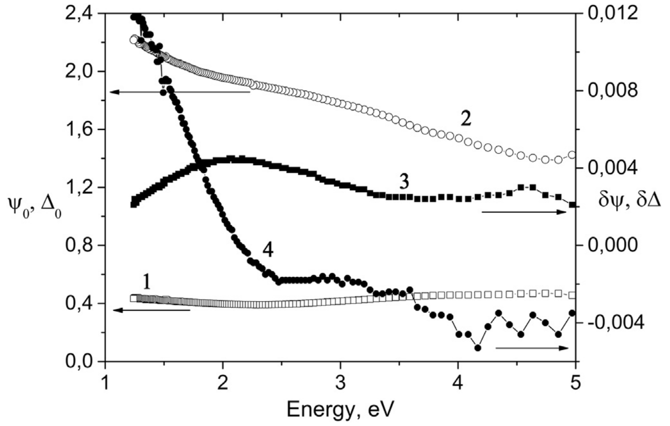

Fig. 1. The experimental and calculated values of ellipsometric parameters: 1 – ψ_0, 2 – Δ_0, 3 – $\delta\psi$, 4 – $\delta\Delta$.

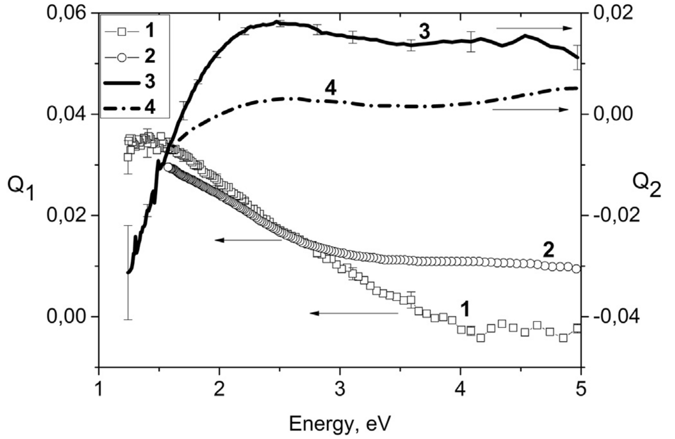

Fig. 2. Calculated values of real and imaginary parts of Fe magneto-optical coupling parameter $Q = Q_1 - iQ_2$ in comparison with those obtained for Fe in [6]: 1 – calculated values of Q_1, 2 – Q_1 in [6], 3 – calculated values of Q_2, 4 – Q_2 in [6].

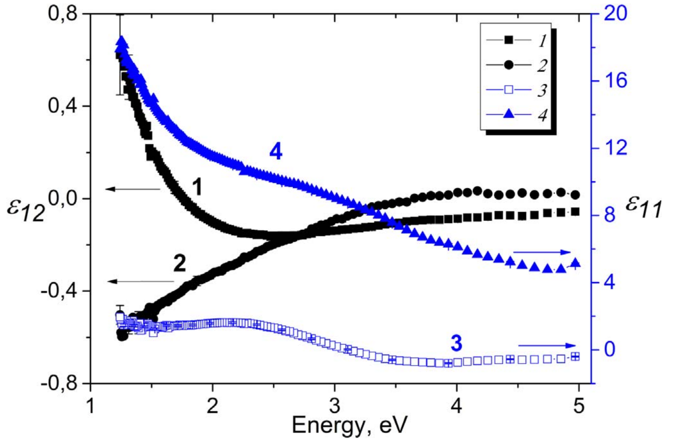

Fig. 3. The calculated values of real and imaginary parts of the Fe diagonal ε_{11} and off-diagonal ε_{12} dielectric permittivity tensor elements: 1 – ε''_{12}, 2 – ε'_{12}, 3 – ε'_{11}, 4 – ε''_{11}.

$\alpha_1 = (A^2 + 4B^2)^{-1} \quad \alpha_2 = (C_2^2 + 4D_2^2)^{-1}$

$A = \xi_1^2 + \xi_2^2 + \xi_3^2 + \xi_4^2 \quad B = -\xi_1\xi_2 - \xi_3\xi_4$

$C = 2(NS - PT) \quad D = 2(NT + PS)$

$A_1 = \xi_5\xi_6 - \xi_7\xi_0 + 2\xi_8\xi_9$

$B_1 = -2\xi_5 ac + 2\xi_7 bd + 2\xi_8\gamma_1$

$A_2 = \xi_6\xi_7 - \xi_5\xi_0 - 2\xi_8\xi_9$

$B_2 = -2\xi_7 ac + 2\xi_5 bd - 2\xi_8\gamma_1$

$C_2 = \gamma_3^2 - \gamma_4^2 + \gamma_5^2 - \gamma_6^2$

$D_2 = -\gamma_3\gamma_4 - \gamma_5\gamma_6$

$T = K_1(2W + V) + K_2(2U - 2X)$

$S = K_1(2U - 2X) - K_2(2W + V)$

$K_1 = Q_1(n_0^2 - k_0^2) - 2n_0k_0Q_2$

$K_2 = Q_2(n_0^2 - k_0^2) + 2n_0k_0Q_1$

$N = \Re(\sin\varphi_0)a - \Im(\sin\varphi_0)c$

$P = -\Re(\sin\varphi_0)c - \Im(\sin\varphi_0)a$

$U = nk\xi_6 + n_0k_0\xi_0 + \gamma_1\gamma_2$

$W = -2nkac - 2n_0k_0bd - \xi_9\gamma_2$

$V = \xi_1^2 - \xi_2^2 - \xi_3^2 + \xi_4^2$

$X = -\xi_1\xi_2 + \xi_3\xi_4$

$\xi_0 = b^2 - d^2 \quad \xi_1 = na + n_0b$

$\xi_2 = nc + n_0d \quad \xi_3 = ka + k_0b$

$\xi_4 = kc + k_0d \quad \xi_5 = n^2 + k^2$

$\xi_6 = a^2 - c^2 \quad \xi_7 = n_0^2 + k_0^2$

$\xi_8 = n_0k - nk_0 \quad \xi_9 = bc + ad$

$\gamma_1 = (ab - cd) \quad \gamma_2 = nk_0 + n_0k$

$\gamma_3 = n_0a + nb \quad \gamma_4 = n_0c + nd$

$\gamma_5 = k_0a + kb \quad \gamma_6 = k_0c + kd$

$\cos\varphi_0 = a + ic \quad \cos\varphi_1 = b + id$

where φ_0 and φ_1 are the angles of incidence and refraction, respectively, while a, b are real and c, d are imaginary parts of $\cos\varphi_0$ and $\cos\varphi_1$, respectively, we obtain all necessary expressions for ellipsometric angles calculation.

4. Fitting to the experimental ellipsometric angles by the Nelder–Mead method [9]. As a result it yields the spectral dependences of real (Q_1) and imaginary (Q_2) parts of magneto-optical parameter Q. Thus, we have information about all elements of the dielectric permittivity tensor.

3. Results and discussion

In order to demonstrate the method of interpretation of in situ magneto-ellipsometry measurements data, the sample in the form of polycrystalline Fe layer on the surface of Si(100)/SiO_2 was studied. The process of SiO_2/Si(100) substrate primary chemistry is specified in [10]. Fe film was made by ultrahigh vacuum thermal evaporation with deposition on the cool substrate, nevertheless the proposed data analysis approach can be also applied to data from the molecular beam epitaxy setups. Final thicknesses of SiO_2 and Fe layers were 3.84 nm and 160.5 nm, respectively. During all the measurements the angle of incidence was fixed at 56°. While carrying out magneto-optical ellipsometry measurements the magnetization reversal of the sample was in the ± 2 kOe field.

Then, the proposed algorithm for the interpretation of magneto-ellipsometric measurement data for the model of a homogeneous semi-infinite medium was used. The values of refractive index, extinction coefficient and magneto-optical parameter Q of Fe layer in Si/SiO_2/Fe structure were obtained using the presented above approach. They were used for calculating ψ_0, $\delta\psi$, Δ_0, $\delta\Delta$. In Fig. 1 the experimental and calculated values of these parameters are presented. Experimental data are shown by lines and calculated – by symbols. One can see that they fit each other. It is not surprising as the error of approximation by Nelder–Mead method was put to be 0.0001.

Finally, we obtained magneto-optical coupling parameter Q values from these curves (Fig. 2), consequently we have completely determined all elements of the dielectric permittivity tensor (Fig. 3). The comparison of the Fe magneto-optical parameter Q with [6] shows a good qualitative agreement. Quantitatively, the curves are not similar as the thicknesses of Fe layer in the works differ: our sample (Si/SiO_2/Fe) was 160.5 nm and the sample in [6] (Si(100)/Fe) was 60 nm thick. In both cases the Fe thickness is greater than the optical skin depth so the results can be compared.

Thus, a new opportunity of in situ simultaneous characterization of optical and magneto-optical properties of films without carrying out any additional ex situ measurements has been demonstrated by means of magneto-ellipsometry. This approach can be used for thin films synthesis control.

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