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Development of *in situ* magneto-ellipsometry for studying correlation between the optical and magneto-optical properties of ferromagnetic thin films

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Abstract. In this work we present the way of nanostructured films study by means of magneto-ellipsometry. The method of interpretation of *in situ* magneto-optical ellipsometry spectra from the *in situ* molecular beam epitaxy setup with an integrated magneto-ellipsometric real time synthesis control is described. The method has been successfully tested on Fe/SiO₂/Si nanostructures 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 influence on the sample and have some flexibility when using *in situ*, directly in 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].

In this paper, we suggest a new approach of real time control of obtaining material parameters of magnetic thin films right in the process of molecular beam epitaxy by means of the *in situ* magneto-optical ellipsometry. In the end, we demonstrate how our method works by presenting the results of Fe(layer)/SiO₂(layer)/Si(substrate) film study.

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) onto ferromagnetic metal (the refraction index N) in the visible light range. The magnetization vector is z-axis directed. YX is a plane of incidence, YZ is a



boundary plane The key idea of the proposed approach is reported in [5], where it was applied toward the particular case of 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 [6]

$$[\varepsilon] = \begin{bmatrix} \varepsilon_{11} & \varepsilon_{12} & 0 \\ \varepsilon_{21} & \varepsilon_{22} & 0 \\ 0 & 0 & \varepsilon_{33} \end{bmatrix} = \begin{bmatrix} \varepsilon'_{11} - i\varepsilon''_{11} & -i(\varepsilon'_{12} - i\varepsilon''_{12})(Q_1 - iQ_2) & 0 \\ i(\varepsilon'_{12} - i\varepsilon''_{12})(Q_1 - iQ_2) & \varepsilon'_{11} - i\varepsilon''_{11} & 0 \\ 0 & 0 & \varepsilon'_{11} - i\varepsilon''_{11} \end{bmatrix}, \quad (1)$$

where ε' and ε'' are a 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 magneto-optical effects. Let us denote the ellipsometric parameters in the non-magnetic condition as ψ_0 and Δ_0 . In the case of magneto-ellipsometric 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}).

Table 1. The notations used in the developed approach.

1	2	3	4
$\alpha_1 = (A^2 + 4B^2)^{-1}$	$C = 2(NS - PT)$	$\xi_0 = b^2 - d^2$	$T = K_1(2W + V) + K_2(2U - 2X)$
$\alpha_2 = (C_2^2 + 4D_2^2)^{-1}$	$D = 2(NT + PS)$	$\xi_1 = na + n_0b$	$S = K_1(2U - 2X) - K_2(2W + V)$
$A = \xi_1^2 + \xi_2^2 + \xi_3^2 + \xi_4^2$	$\gamma_1 = (ab - cd)$	$\xi_2 = nc + n_0d$	$K_1 = Q_1(n_0^2 - k_0^2) - 2n_0k_0Q_2$
$A_1 = \xi_5\xi_6 - \xi_7\xi_0 + 2\xi_8\xi_9$	$\gamma_2 = nk_0 + n_0k$	$\xi_3 = ka + k_0b$	$K_2 = Q_2(n_0^2 - k_0^2) + 2n_0k_0Q_1$
$A_2 = \xi_6\xi_7 - \xi_5\xi_0 - 2\xi_8\xi_9$	$\gamma_3 = n_0a + nb$	$\xi_4 = kc + k_0d$	$N = Re(\sin \varphi_0)a - Im(\sin \varphi_0)c$
$B = -\xi_1\xi_2 - \xi_3\xi_4$	$\gamma_4 = n_0c + nd$	$\xi_5 = n^2 + k^2$	$P = -Re(\sin \varphi_0)c - Im(\sin \varphi_0)a$
$B_1 = -2\xi_5ac + 2\xi_7bd + 2\xi_8\gamma_1$	$\gamma_5 = k_0a + kb$	$\xi_6 = a^2 - c^2$	$U = nk\xi_6 + n_0k_0\xi_0 + \gamma_1\gamma_2$
$B_2 = -2\xi_7ac + 2\xi_5bd - 2\gamma_1\xi_8$	$\gamma_6 = k_0c + kd$	$\xi_7 = n_0^2 + k_0^2$	$V = \xi_1^2 - \xi_2^2 - \xi_3^2 + \xi_4^2$
$C_2 = \gamma_3^2 - \gamma_4^2 + \gamma_5^2 - \gamma_6^2$	$\cos \varphi_0 = a + ic$	$\xi_8 = n_0k - nk_0$	$W = -2nkac - 2n_0k_0bd - \gamma_2\xi_9$
$D_2 = -\gamma_3\gamma_4 - \gamma_5\gamma_6$	$\cos \varphi_1 = b + id$	$\xi_9 = bc + ad$	$X = -\xi_1\xi_2 + \xi_3\xi_4$

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)

$$N = n - ik = N_0 \sin \varphi_0 (1 + tg^2 \varphi_0 (1 - (tg \psi_0 \exp(i\Delta_0)))^2 (1 + (tg \psi_0 \exp(i\Delta_0)))^{-2})^{1/2} \quad (2)$$

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:

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

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

$$R_p = R_{pp} + R_{ps} = R'_{p0} + R'_{p1} - i(R''_{p0} + R''_{p1}); \quad R_S = R_{SS} + R_{sp} = R_{S0} = R'_{S0} - iR''_{S0} \quad (4)$$

$$R'_{p0} = \alpha_1(A_1A + 2B_1B); \quad R'_{p1} = \alpha_1(A^2C - 4B^2C + 4ABD); \quad R'_{S0} = \alpha_2(A_2C_2 + 2B_2D_2) \quad (5)$$

$$R''_{p0} = \alpha_1(AB_1 - 2BA_1); \quad R''_{p1} = \alpha_1(A^2D - 4B^2D - 4ABC); \quad R''_{S0} = \alpha_2(B_2C_2 - 2A_2D_2) \quad (6)$$

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 equations (5, 6) and using the notations presented in Table 1, where φ_0 and φ_1 are the angles of incidence and refraction respectively, we obtain all necessary expressions for ellipsometric angles calculation.

4 Fitting to the experimental ellipsometric angles by the Nelder–Mead method. 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 SiO₂/Si(100) was studied. Fe film was made by ultrahigh vacuum thermal evaporation with deposition on the cool substrate.

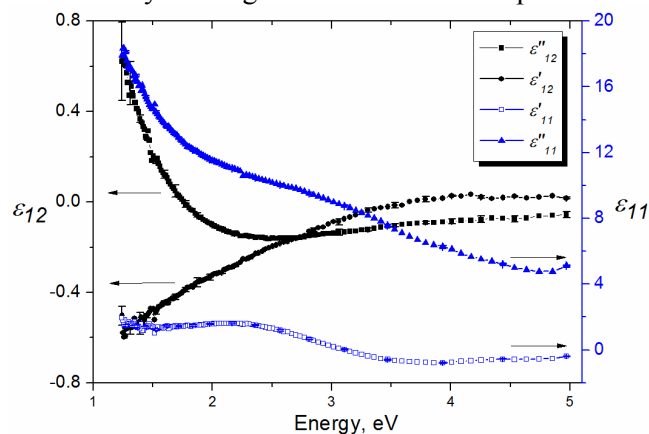


Figure 1. The calculated values of real and imaginary parts of the Fe diagonal tensor element ε_{11} and off-diagonal tensor element ε_{12} (during measurements the angle of incidence was fixed at 56° , the magnetization reversal of the sample in the ± 2 kOe field, the thicknesses of SiO₂ and Fe layers were 3.84 nm and 160.5 nm, respectively).

In conclusion, the algorithm for the interpretation of magneto-ellipsometric measurement data has been proposed for the model of a homogeneous semi-infinite medium. The values of refractive index, extinction coefficient and magneto-optical parameter Q of Fe layer in Fe/SiO₂/Si structure were obtained using the presented approach. Comparison of the Fe magneto-optical parameter Q with [7] shows a good agreement. We have completely determined all elements of the dielectric permittivity tensor (Figure 1). Thus, the opportunity of simultaneous characterization of optical and magneto-optical properties of nanostructures has been demonstrated by means of magneto-ellipsometry.

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