

Magneto-Ellipsometry Investigations of Multilayer Nanofilms of Fe and Co

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Spectral ellipsometry technique is demonstrated to be a useful tool for the investigation of optical and magneto-optical parameters of magnetic heterostructures. Ellipsometry parameters ψ , Δ were measured in the range of 350-1000 nm. Solving the inverse problem by a regression method the refractive index and thickness of the layers were deduced. Magneto-optical spectra were detected in the configuration of equatorial Kerr effect which is fully compatible with the ellipsometry measurements. Spectroscopic ellipsometry method of magnetic structure characterisation can be used for *in situ* monitoring of magnetic film growth in various processes such as magnetron sputtering, electron beam evaporation and ion beam sputtering.

Key words: Magneto-ellipsometry, Magnetic nanofilms, Kerr effect, *In situ* ellipsometry

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1. INTRODUCTION

Thin films and multilayers have become an important class of nanostructured materials with immense possibility of tailoring their properties in order to achieve the desired functionality. Recent technological advances in thin film deposition techniques have made it possible to deposit layers with a thickness in the range of a few nanometers to produce multilayer structures for various applications [1, 2].

For the potential applications in electronics metallic magnetic multilayers are of prime interest although they have a lower spin polarisation at room temperature than other materials (for example, Fe₃O₄) because of their high conductivity and stability. The principle structure for spintronics is composed of ferromagnetic metal films separated by dielectric or semiconducting layers [3, 4]. Based on such structures, a record value of magnetoresistance is obtained at tunnelling through MgO [5] or PrO₁₁ [6] barrier. In many systems, Si is also used as interlayer between the magnetic layers. A large number of works are devoted to the creation of new elements of spintronics where several methods of spin current control are proposed [7-9]. Magnetic multilayers are also used for the development of magnetic sensors and recording heads [10, 11].

In all these applications, a precise control of layer thickness, and good interface quality in terms of sharpness and smoothness of the interface, play a crucial role in determining the quality of the final device. The preparation of thin films is therefore of importance in both fundamental research and practical applications. A large number of techniques have been developed for deposition of thin films, such as electron beam evaporation and sputtering which are most commonly used techniques for thin film deposition. They can be made compatible with *in situ* characterisation using optical methods. In the case of magnetic multilayers, spectroscopic ellipsometry and magneto-optical investigations could be used simultaneously for *in situ* characteriza-

tion during the film growth or after film deposition which is equally important [12-14].

Spectroscopic ellipsometry is routinely used to measure thickness and refractive index of transparent thin films. Absorbing films are more difficult to characterize because the optical constants (n and k) are often correlated with the film thickness. This calls into question the uniqueness of the results and the use of additional characterisation techniques could be important. In the case of magnetic films, it is of interest to employ magnetic methods to get information on magnetic parameters and as an additional tool for structure characterisation.

Magneto-optic Kerr effect (MOKE) investigations could be combined with ellipsometry methods for *in situ* monitoring. Care should be taken for applying MOKE during the film growth since the application of a magnetic field may influence the film structure [15]. On the other hand, the magnetic field could be applied during the film growth to create a desired anisotropy. In the present paper we have demonstrated successful application of ellipsometry and equatorial MOKE for characterisation of Fe and Co multilayers.

2. FORMALISM

The phenomenological description of optical and magneto-optical parameters is based on the matrix form of the permittivity [16, 17]. Considering that the plane of incidence is the y, z plane and the magnetisation is along the x -axis (equatorial Kerr effect), the permittivity of isotropic magnetic layer has diagonal components ε and off-diagonal components $\varepsilon_{yz} = -\varepsilon_{zy} = -iQm_x$. Here Q is the magneto-optical constant and m_x is the x -directional cosine of the magnetisation. In this case, the p - (H_x, H_y, H_z) and s - (E_x, E_y, E_z) polarised waves remain the eigenfunctions and the consideration is convenient to make in terms of Abeles matrices (see for details [18]).

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The presence of the off-diagonal term in the permittivity does not change the propagation conditions for *s*-polarisation. The characteristic matrix for layer *j* and *s*-polarisation is of the form

$$\widehat{M}_{sj} = \begin{pmatrix} \cos(\beta_j h_j) & \frac{i}{q_{pj}} \sin(\beta_j h_j) \\ iq_{pj} \sin(\beta_j h_j) & \cos(\beta_j h_j) \end{pmatrix} \quad (1)$$

$$q_{sj} = n_j \cos \theta_j, \beta_j = k_0 n_j \cos \theta_j, k_0 = \frac{2\pi}{\lambda} \quad (2)$$

Here n_j , θ_j , h_j are the refractive index, angle of refraction and thickness of layer *j*, λ is the wavelength. The characteristic matrix of the multilayered system is found by multiplication of the matrices \widehat{M}_{sj} :

$$\widehat{M}_s = \prod_{j=2}^{N-1} \widehat{M}_{sj} \quad (3)$$

Index $j = 2$ is related to the first layer and $j = N - 1$ to the last layer. Index $j = 1$ corresponds to the medium from which light strikes the film and $j = N$ is for the substrate. The reflection parameter r_s from the total system is found from

$$\begin{pmatrix} 1 + r_s \\ (-1 + r_s) \cos \theta / n_1 \end{pmatrix} = \widehat{M}_s \begin{pmatrix} t_s \\ -t \cos \theta_N / n_N \end{pmatrix} \quad (4)$$

Here t_s is transmission parameter which is not of interest here but for convenience we keep it in the matrix form of the equation.

In the case of *p*-polarisation, the form of the wave equation in the magnetic layer is the same but the wave number changes to k_m

$$k_{mj} = k_0 \sqrt{\varepsilon \left(1 + \left(\frac{\varepsilon_{\perp}}{\varepsilon} \right)^2 \right)}, \quad \varepsilon_{\perp} = i\varepsilon Q m_x \quad (5)$$

There is also the following relationship between magnetic and electric fields:

$$ik_{mj} \sin \theta_j H_x = ik_0 (\varepsilon_{\perp} E_y + \varepsilon E_z) \quad (6)$$

$$\frac{\partial H_x}{\partial z} = -ik_0 (\varepsilon E_y - \varepsilon_{\perp} E_z) \quad (7)$$

In (6) the dependence on *y* in the form of $\exp(ik_0 \tilde{n} \sin \theta y)$ was used. As a result the characteristic matrix given by equation (1) of the magnetic layer changes. The diagonal components becomes

$$M_{pj}^{11} = \cos \beta h_j - iQ m_x \tan \theta_j \sin(\beta h_j) \quad (8)$$

$$M_{pj}^{22} = \cos \beta h_j + iQ m_x \tan \theta_j \sin(\beta h_j) \quad (9)$$

The off-diagonal components change in a usual way with replacement of q_{sj} by

$$q_{pj} = \frac{\cos \theta_j}{n_j} \quad (10)$$

With these changes, the equation for r_p has the form of (4).

This approach gives a compact form for the ellipsometric parameter ρ of multilayered magnetic system

$$\rho = \tan \psi \exp(i\Delta) = \frac{r_p}{r_s}$$

The reflection coefficients r_s , r_p are determined from matrix equation (4) where the corresponding parameters are given by equations (1), (2), (8)-(10). This form is convenient for computer simulation.

3. SAMPLE PREPARATION

Multilayers of Fe and Co with Si, SiO₂ interlayers were obtained on Si and glass substrates by electron beam evaporation. Advantage of the evaporation process is that, it is carried out in clean ultra high vacuum conditions ($\sim 10^{-9}$ mbar) thereby depositing a film with minimum contamination. Since the mean free path of the evaporated atom is very large compared to the chamber dimension under ultra high vacuum conditions, the distance between the evaporation source and the substrate can be kept large enough to have uniform deposition over a large substrate area. The deposition can be done on elongated stripe substrates so there will be a uniform gradient in film thickness and it will be possible to investigate the change of the properties with the thickness. Preparation of individual samples varying in thickness would be time consuming. The substrate temperature is 250 °C. The deposition rate was kept very low, at the level of 0.02 nm/min. The stripes with total length of 600 mm consisted of separate 15×10 mm substrates which were used for optical and magnetic measurements.

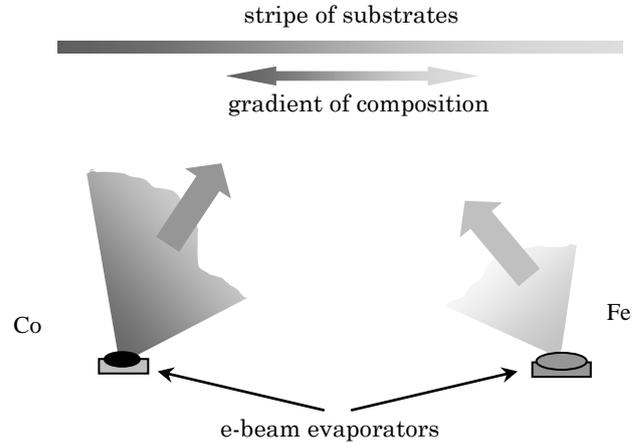


Fig. 1 – Scheme of $\text{Co}_x\text{Fe}_{1-x}$ films deposition in a wide range of Co concentration by a method of two electron beam sources

Ferromagnetic $\text{Co}_x\text{Fe}_{1-x}$ alloy films in a wide range of Co concentration were deposited by co-precipitation Co and Fe onto stripes of glass substrates by using two electron beam sources (see Fig. 1). In this way a compositional gradient was obtained along the strips of substrates ranging from $x = 0.16$ to $x = 0.81$. A series of samples with different magnetic properties were obtained.

4. EXPERIMENTAL RESULTS AND DISCUSSION

Spectroscopic static ellipsometer was used for measurements. The magnetic field was applied along the sample plane and perpendicular to the plane of incidence so equatorial Kerr configuration was realised. The field magnitude was 120 Oe. In ac regime, the frequency was 100 Hz. The ellipsometric parameters are proportional to the sample magnetisation so detecting the modulated signal the magnetisation curve can be deduced for different thicknesses.

Firstly, to test the system sensitivity we investigated Co layers on glass substrate for various thicknesses. A detectable modulated signal corresponds to a thickness of about 1 nm. It could be that the Co films at such thicknesses are in a superparamagnetic state. On the other hand, detecting a magnetic signal at such thickness is at the limit of system resolution. The observed loops at ~ 1 nm film thickness show that the sensitivity of the magneto-ellipsometry is quite good. Fig. 2 shows the change in coercivity with the film thickness. The coercivity firstly increases with the thickness reaching maximum at about 3 nm and then decreases staying almost constant at thickness larger than 10 nm. This demonstrates the effect of substrate on domain pinning. This result is in a good agreement with that obtained in [14] for *in situ* Co film deposition.

Similar results can be obtained for the coercivity of Fe and $\text{Co}_{50}\text{Fe}_{50}$ films on glass substrates. In the case of Fe on Si substrate, it appears that a thin FeSi layer of about 1.5 nm is spontaneously formed.

It is known that antiferromagnetic coupling in multilayers Fe/Si is realised through non magnetic silicides FeSi with semimetallic properties. In general, Fe and Si can form a variety of stable bulk binary alloys. The nature of iron-silicides spontaneously formed during film growth was the subject of many studies (see, for example, [19]). It may arise from the interdiffusion during the growth. Spontaneous formation of FeSi could be proposed in our investigations. This conclusion is made from the comparison of modelling results and the experimental optical and magneto-optical studies. However we do not exclude that the situation could be more complicated due to the interlayer roughness.

Optical and magneto-optical spectra were measured for a series of samples Fe/Si with a thin layer of Fe (average thickness 1.5-5 nm with a step of 0.5 nm) with 20 nm Si on a glass substrate. Fig. 3 shows ellipsometric parameter ψ for a thickness of the upper layer of 3 nm. The results were compared with modelling for two systems Fe/Si/glass and Fe/FeSi/Si/glass. For modelling, the reference data for optical constants of $\text{Fe}_{50}\text{Si}_{50}$ were used [19]. It is seen that the model with additional layer of FeSi of about 1.5 nm gives a better fitting. It could be argued that the presence of interlayer roughness was not considered.

More convincing data are obtained from the analysis of magneto-optical spectra which are shown in Fig. 4. The relative ellipsometric spectra for two opposite directions of the magnetic field obtained after averaging 10 measurements demonstrate that the magnitude of the magneto-optical effect is not proportional to the upper layer thickness. If the existence of the FeSi layer of about 1.5 nm is assumed there is a very good correspondence with the experimental results.

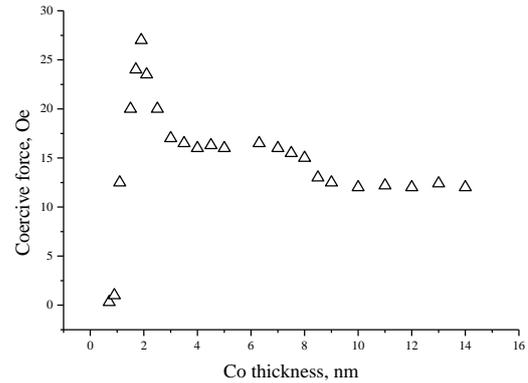


Fig. 2 – Coercive force vs thickness of Co film on glass substrate

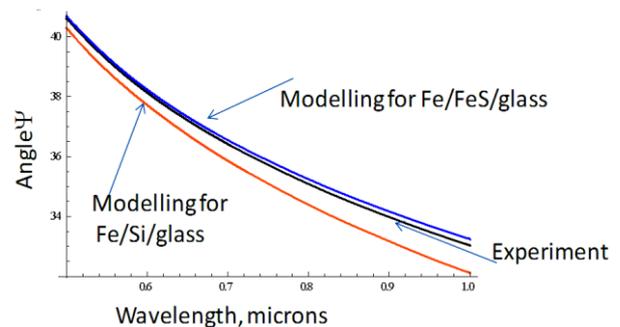


Fig. 3 – Experimental and simulation spectra of angle ψ for Fe/Si/glass samples with the top layer of 3 nm. The results of modelling compared to experimental ones suggest the formation of interlayer of FeSi of about 1.5 nm

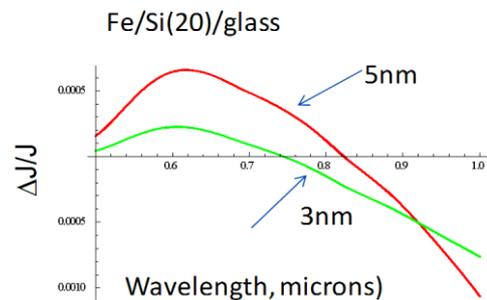


Fig. 4 – Experimental spectra of the relative intensity change at two opposite directions of the external magnetic field for a system of Fe/Si/glass for the upper layer thickness of 3 nm and 5 nm

5. CONCLUSIONS

Generalised magneto-ellipsometry was used to investigate thin multilayers of Co and Fe obtained by electron beam evaporation. The method is proved to have high sensitivity for measurement of nanosized films. It was demonstrated that for very thin films the coercivity increases with increasing thickness, but for thicker films its value becomes almost independent of thickness. In the case of Fe/Si multilayers, spontaneous formation of thin FeSi layer of thickness about 1.5 nm was demonstrated.

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