Optical and magnetic properties of Al/NiFe and Al/Ge/NiFe nanosized films

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Abstract. Nanosized films with ferromagnetic layers are widely used in nanoelectronics, sensor systems and telecommunication. The physical and magnetic properties of nanolayers may significantly differ from those known for bulk materials due to fine crystalline structure, influence of interfaces, roughness, and diffusion. In this work, we are employing a spectral ellipsometry method, magneto-optical Kerr magnetometry and VSM to investigate the impact of layer thickness on the optical constants and magnetization processes for two and three layer films of the type Al/NiFe/sitall, Al/Ge/NiFe/sitall on sitall substrate for different thickness of the upper Al layers. The refractive indexes of two layer films are well resolved by spectral ellipsometry demonstrating their good quality. Modelling data for three-layer films show considerable discrepancy with the experiment, which can be related to a stronger influence of interfaces. The magnetization processes of two-layer films weakly depend on the type and thickness of the upper non-ferromagnetic layers. However, the coercivity of three layer films may significantly change with the thickness of the upper layer: more than twice when the thickness of Al layer increases from 4 to 20 nm.

1 Introduction

Thin films with magnetic layers are an important class of nanostructured materials for applications in nanoelectronics and spintronics [1-2]. Considerable interest in such systems is also related to a possibility of strengthening and modifying the magneto-optical effects, for example, by excitation of long-living modes of surface plasmon polaritons [3], or by spatial diffusion of spins in the ferromagnetic layers [4-6].

The technological production methods of thin films allow controlling the layer thickness with a precision of a nanometer. However, the properties of thin films may differ significantly from those of bulk materials [7-9]. This is due to the structural parameters such as the size of crystallites, the quality of the interfaces of intermediate layers, and diffusion. The structure of multilayer films can be complicated by the need to protect the functional layers, with the physical properties of interest. Therefore, the task is to control the physical properties of the individual layers along with geometry and overall magnetic response.

Ellipsometric methods are promising for such monitoring, since they permit the measurement of optical parameters of individual nanolayers (and growing layers) with high accuracy and selectivity in the process of production [10-15]. The determination of optical and geometrical parameters is based on the minimization of a functional involving a quadratic form of the difference between the experimental and modelling data. In the case of magnetic films it is of interest to combine the two approaches: traditional ellipsometry and magneto-optical (MO) magnetometry [16]. In particular case of the equatorial MO Kerr effect, a general approach within the concept of matrix optics can be used for the model calculations.

In this work, the methods of magneto-ellipsometry are used to study the optical parameters and magnetization processes of two-layer and three-layer films of Al/NiFe, Al/Ge/NiFe on sitall substrate for varying thicknesses of upper Al layer. The behaviour of the refractive index of individual layers depending on the film structure may disclose the role of interfaces on magnetic properties. We have found that the hysteresis curves of three-layer films undergo significant changes depending on the thickness of the upper nonmagnetic layer.

1.1 Magneto-ellipsometry analysis of multilayered films

The ellipsometric parameters are determined from the measurement of the ratio of complex reflection coefficients for the two polarizations of the light wave: (p) and (s), with the electric field parallel and perpendicular to the plane of incidence, respectively. This complex ratio is usually expressed in the polar form introducing the ellipsometric parameters ψ and Δ, which characterize the relative change in amplitudes and phase shift for p- and s-polarizations, respectively:

$$\frac{r_p}{r_s} = \tan\psi \exp i\Delta \qquad (1)$$

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In (1), r_p, r_s are the complex reflection coefficients of *p*- and *s*-polarized waves, respectively. Ellipsometric measurements are not direct. In general it is necessary to solve the inverse problem, which may pose considerable difficulties, then, it is very important to build initially an adequate model, which is already using the regression techniques to fit the experimental results.

The magneto-ellipsometry analysis of multilayered films is based on the determination of the characteristic matrices of individual layers, which can have a magnetization in the film plane and perpendicular to the plane of incidence (along the *x*-axis). This corresponds to the geometry of the equatorial Kerr effect, and allows simultaneous determination of magneto-optical and ellipsometric parameters, since the electromagnetic field components with the polarization *s* and *p* remain the eigenfunctions of the system. The influence of the change in magnetization of a layer within multilayered system with a given geometry is the change in the intensity and phase shift of the *p*-wave, whereas the scattering parameters of *s* –waves do not depend on the magnetization. This approach is more convenient for numerical calculations than using the Mueller matrix or Jones matrix.

In the model, the equatorial MO effect can be taken into account by the introduction of the antisymmetric tensor of the second rank for the matrix of the dielectric constant [17,18]:

$$\hat{\varepsilon}_j = \varepsilon_j \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & iQ_j m_{xj} \\ 0 & -iQ_j m_{xj} & 1 \end{pmatrix} \qquad (2)$$

In equation (2) the dielectric constant is written for a ferromagnetic layer (distinguished by index *j* in the multilayer system). It is assumed that the plane of incidence is the *y*-*z* plane, and the surface of the sample is *y*-*x* plane. $\varepsilon_j = (\tilde{n}_j + i\kappa_j)^2$, m_{xj} and Q_j are the complex permittivity, the projection of the normalized magnetization on the *x*-axis (perpendicular to the plane of incidence), and the magneto-optical constant of the j-th layer, respectively. For waves with *p*-polarization the reflection r_p and transmission t_p coefficients for a multilayer system are found from the matrix equation

$$\begin{pmatrix} 1 + r_p \\ (-1 + r_p)p_{p1} \end{pmatrix} = \hat{M}_p t_p \begin{pmatrix} 1 \\ -p_{pN} \end{pmatrix} \qquad (3)$$

$$p_{p1} = \frac{\cos\theta}{n_1}, \qquad p_{pN} = \frac{\cos\theta_N}{n_N} \qquad (4)$$

In (4), $\hat{M}_p$ is the characteristic matrix of the system, θ is the angle of incidence from the outside medium with the refractive index n_1, θ_N is the angle of refraction in the last medium (substrate) with a refractive index n_N. The matrix $\hat{M}_p$ is determined by the product of the corresponding matrices of the individual layers with the layer parameters: $n_j = \sqrt{\varepsilon_j}$ is the complex refractive index and h_j is the layer thickness:

$$\hat{M}_p = \prod_{j=2}^{N-1} \hat{M}_{pj} \qquad (5)$$

$$\hat{M}_{pj} = \begin{pmatrix} \cos(\beta_j h_j) - i\,\delta_j \sin(\beta_j z) & \frac{i}{p_{pj}} \sin(\beta_j h_j) \\ i p_{pj} \sin(\beta_j h_j) & \cos(\beta_j h_j) + i\,\delta_j \sin(\beta_j z) \end{pmatrix} \qquad (6)$$

$$\beta_j = k_0 n_j \cos\theta_j\,, p_{jp} = \frac{\cos\theta_j}{n_j}, \delta_j = iQ_j m_{xj} \tan\theta_j \qquad (7)$$

Here λ is the wavelength, $k_0 = 2\pi/\lambda$. The angle θ_j is defined as follows:

$$n_j \cos\theta_j = \sqrt{n_j^2 - n_1^2 \sin^2\theta}$$

Thus, in the developing approach the magnetic layers are treated within the same characteristic matrix formalism using the off-diagonal magneto-optical parameter Q_j.

For *s*-polarization the intensity of the reflected light does not depend on the magnetization. The basic equations remain the same with the replacement of p_{pj} to $p_{sj} = n_j \cos\theta_j$ and putting $\delta_j = 0$.

The characteristic matrices depend on the refractive indexes which have to be determined by comparing the experimental and modelling results. Since the ellipsometric method is not direct, certain assumptions on the permittivity spectra should be made. For example, in a certain range of wavelengths for layers with attenuation simple approximations (Cauchy-Urbach) are possible for complex refractive index [10]:

$$\tilde{n}_j(\lambda) = A_{nj} + \frac{B_{nj}}{\lambda^2} + \frac{C_{nj}}{\lambda^4} \qquad (8)$$

$$k_j(\lambda) = A_{kj} \exp\left(\frac{B_{kj}}{\lambda} - C_{kj}\right) \qquad (9)$$

With the help of this model it is possible to achieve a good match with the experimental data on the spectral characteristics of ψ and Δ to determine the optical parameters of the layers.

2 Materials and methods measurements

The investigated multilayered systems of Al/NiFe/ and Al/Ge/NiFe/ on a sitall substrate were fabricated by vacuum thermal evaporation method utilizing VUP-5M (vacuum universal post) using layer-by-layer condensation. The sample holder was rotating to insure good uniformity. In NiFe permalloy alloy the concentration of Ni was approximately 50%. After deposition of the bottom layer of permalloy with constant thickness of 20 nm on the substrate the samples were annealed in vacuum at T=550° K to refine the structural properties. The film was polycrystalline and isotropic in the plane. The thickness of the upper nonmagnetic layer of Al was varied whilst the thickness of the Ge layer kept fix at 2 nm. The quartz resonator method was used to measure the film thickness.

The ellipsometric parameters were measured using spectral ellipsometry (Ellipse 1891) based on the static measuring system with fixed incidence angle of 70º in the wavelength range of 350-1050 nm with a step of 2 nm. The fitting model with available literature data was applied in the range of 400-800 nm. At this stage, the magneto-optical (MO) measurements in the equatorial configuration were carried out separately by measuring the change in the intensity of *p*-polarized light at the wavelength of 632 nm. The magnetic hysteresis curves were also measured with a vibrating magnetometer BM-07 (VSM) at the magnetic field strength up to 200 mT. The sample position during the magnetic measurements varied to investigate the existence of in-plane magnetic anisotropy.

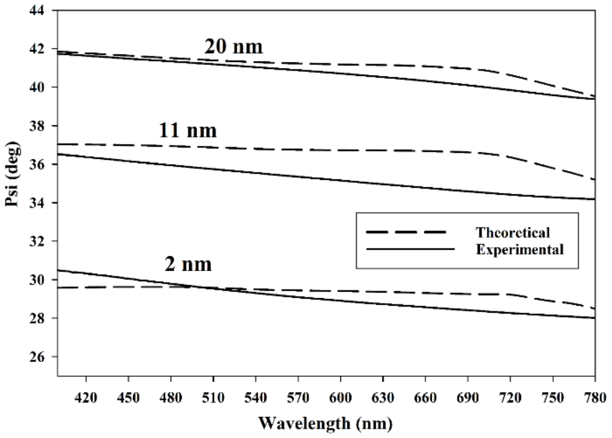

Fig. 1. Spectra of ellipsometric parameter ψ for Al/NiFe/sitall film system for different values of the thickness of Al layer. Solid lines – experiment, dotted lines- two-layer optical model.

3 Results and discussion

Fig. 1 shows the experimental and model spectra of the ellipsometric parameter ψ for the films Al/NiFe/sitall for different values of Al-layer thickness. The refractive indices n_j of the layers were approximated by formulas (7)-(8). The value of n (and thickness h) of individual layers were determined by achieving a good matching between the experimental and modelling data by minimizing a quadratic form which defines the difference between them. Thus, the coefficients in expansion (7)-(8) were determined. The process was initially completed for the Al layer of 20 nm thick. If the same optical parameters (n_j) are used to describe the ellipsometric data for thinner Al- layers, some discrepancies with the experiment could be seen, which indicates the dependence of n on the layer thickness (in the range of 10-15%) for $h < 20$ nm. The values of the refractive indexes of Al and NiFe layers determined from the ellipsometric spectra of Fig. 1 for Al-layer thickness h =20 nm are shown in Fig. 2. The obtained results quite well (less than 5%) coincide with the published data for bulk materials [19].

The magnetic hysteresis loops obtained by MO and VSM methods for the bi-layer system are shown in Fig. 3 for different values of h_{Al}. The hysteresis loops did not depend on the sample alignment proving the absence of in-plane anisotropy. The coersivity and the hysteresis shape were slightly dependent on the Al layer thickness. VSM measurements indicate the existence of rather large anisotropy since the loops does not go to saturation even in fields of 2 kOe.

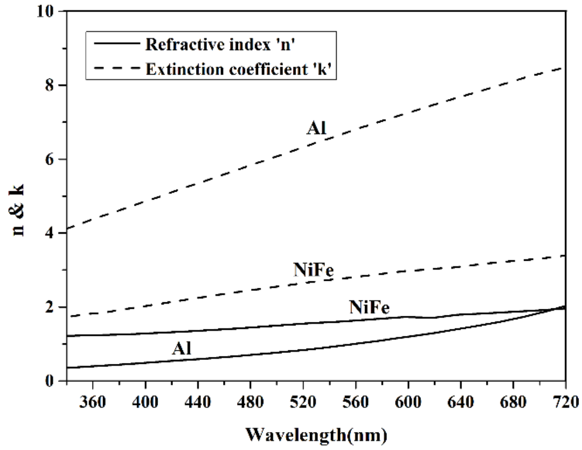

Fig. 2 .Complex refractive index $n = \tilde{n}_j + i\kappa_j$ of Al and permalloy layers determined from the experimental ellipsometric spectra of Fig. 1 for $h_{Al} = h_{NiFe} = 20$ nm. Solid lines represent the real part $\tilde{n}$, the dotted lines show the imaginary part *k*.

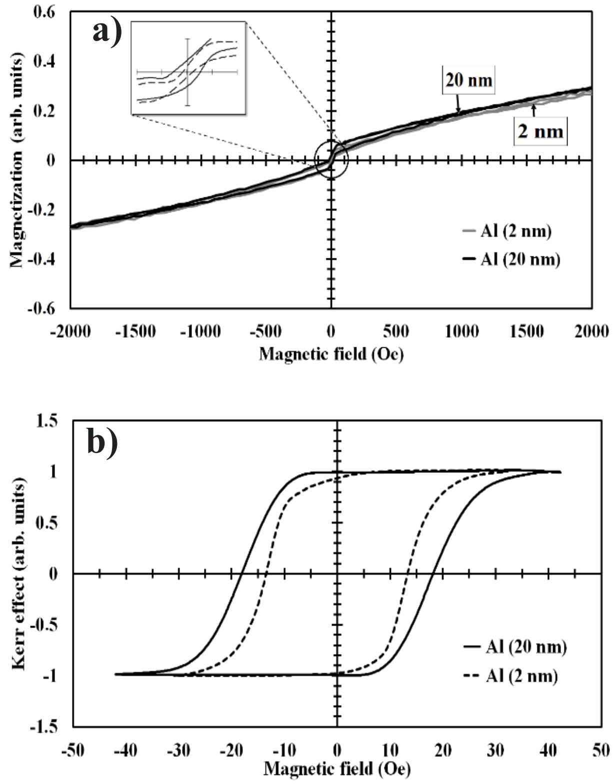

Fig. 3. Hysteresis curves for the films Al/NiFe/sitall for different thickness of Al layer. a) VSM method. b) MO method.

The greatest differences between the experimental and model data are obtained for three-layer films with an intermediate Ge layer when using the optical parameters determined for bilayer films, as can be seen from Fig. 4. The variation in n_j within a reasonable range did not produce matching either. This means that a simple three-layer model does not correspond to the real samples. Very probably, there is a mutual diffusion or roughness, and it is necessary to introduce additional intermediate layers or optical parameter gradients.

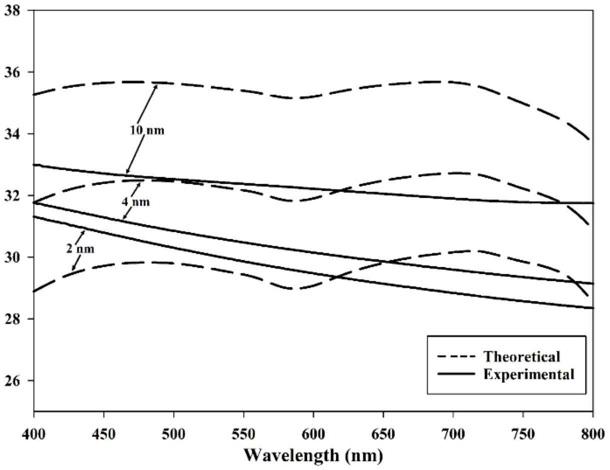

Fig. 4. Spectra of ellipsometric parameter ψ for the film Al/Ge/NiFe/sitall for different values of the thickness of the layer Al. Solid line – experiment, dashed lines– three-layer optical model.

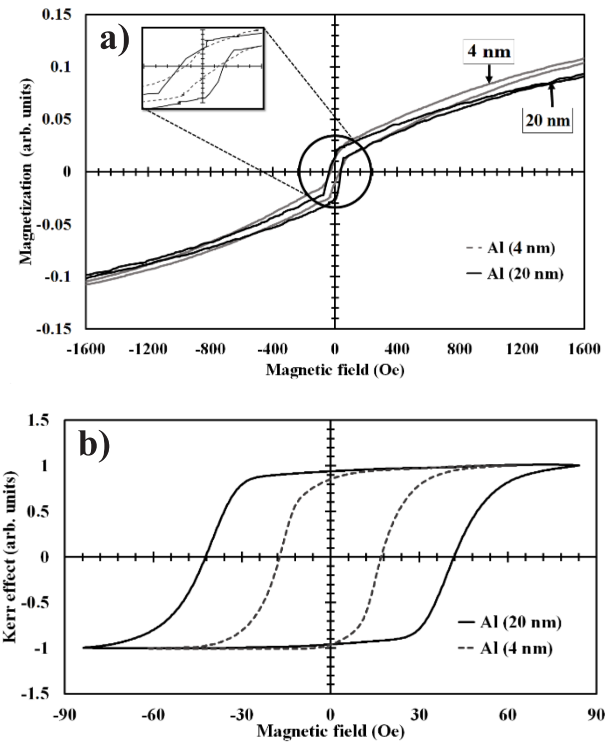

Fig. 5. Hysteresis curves for the films Al/Ge/NiFe/Sitall for different thickness of Al layer. a) VSM method. b) MO method.

In addition, the three-layer films demonstrated unusual strong dependence of coercivity on the thickness of the upper Al layer as shown in Fig.5. MO loops show greater coercivity dependence on h_{Al}, indicating a larger contribution of NiFe surface region in the coercivity increase. It is known that the coercivity of permalloy films is very sensitive to internal stress/strains. The deposition of Al layers did not result in noticeable changes in hysteresis loops (compare with Fig. 3). Therefore we attribute the coercivity increase with increasing h_{Al} to the structural changes produced by Ge layer at NiFe and Al interfaces.

4 Conclusion

Optical and structural parameters of the multilayer films of the type Al/NiFe/, Al/Ge/NiFe/sitall were investigated by ellipsometric and magneto-optical methods. For double-layer systems without the inner Ge layer there is a good agreement between the experimental and two-layer optical model data. Three-layer optical model for Al/Ge/NiFe/sitall does not fit the ellipsometric spectra, which indicates a more complex structure with possible intermediate layers. The measurement of the magnetic hysteresis loops for the three-layer films by MO and VSM techniques shows a strong dependence of coercivity on the thickness of the top layer, which also indicates the structural modification.

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