Ferromagnetic nanoparticular films studied by optical and magneto-optical ellipsometry

T. Fördös,¹ O. Životský,¹ K. Postava,¹ D. Hrabovský,¹ J. Pištora,¹ J. Lančok,² M. Miglierini,³ M. Klementová³

¹Department of Physics, Technical University of Ostrava, 708 33 Ostrava, Czech Republic
²Institute of Physics, AS CR, Na Slovance 2, 182 21 Prague, Czech Republic
³Institute of Inorganic Chemistry of the AS CR, 250 68 Husinec-Řez 1001, Czech Republic

E-mail: kamil.postava@vsb.cz, lancok@fzu.cz

Abstract. Fe and Co nanoparticles embedded in SiO₂ matrix were prepared using dual radiofrequency magnetron sputtering. Films were characterized by high resolution transmission electron microscopy (HRTEM). Magnetic properties were measured using longitudinal magneto-optical Kerr effect. Magneto-optical hysteresis loop measurements show differences between Fe particular film with strong in-plane uniaxial anisotropy and Co one, which is almost isotropic in the plane of the film. Nanoparticular film thicknesses and volume fractions of particles were obtained using spectroscopic ellipsometer UVISEL. Ellipsometric data were fitted to the model based on the Bruggeman effective medium approximation.

1. Introduction
Ferromagnetic metal particles in dielectric matrix possess interesting magnetic and optical properties [1, 2]. Self-organized single crystal metal particles can be produced using three-dimensional (3D) epitaxial growth [3], which is promising technique for preparation of single-domain particular nanostructure, magnetic metamaterials, and magnetophotonic crystals. On the other hand optical and magneto-optical ellipsometry are very sensitive nondestructive methods suitable for study of magnetic anisotropy and magnetization reversal properties of nanostructured systems [4, 5]. Magneto-optic vector magnetometry enables measurement of the magnetization components in the structure during magnetic reversal.

In this paper, we describe preparation and characterization of layers consisting of the ferromagnetic Co and Fe single crystal particles embedded in SiO₂ matrix. Magnetic properties of the particular layers are studied using magneto-optical ellipsometry and optical properties of the layers are measured using spectroscopic ellipsometry. Section 2 deals with brief description of the sample structure, preparation technique, and structure of obtained particles. Magneto-optic setup and ellipsometer are described. Main results are summarized in Sec. 3.

2. Experimental

2.1. Samples preparation
Nanoparticular Co and Fe films were prepared by using dual radiofrequency (RF) magnetron sputtering with alternating targets. The vacuum chamber was evacuated down to 6×10⁻⁴ Pa and filled with Ar of pressure 1 Pa. Figure 1 shows structure of the samples. First, the SiO₂ film of
nominal thickness of 10 nm was deposited on the single-crystal Si substrate at room temperature. Particles of ferromagnetic metal was formed on the top of the SiO$_2$ film. The system of particles were covered by SiO$_2$, which filled interparticle volume, capped the nanoparticular layer, and smoothed the top surface. We have prepared systems with one ($N = 1$) and ten ($N = 10$) nanoparticular layers (see Fig. 1). Nominal effective thickness of the ferromagnetic nanoparticular layers are 3, 5, and 10 nm. The growth rates were 2.22 nm/min, 3.76 nm/min, and 1.48 nm/min for Fe, Co, and SiO$_2$, respectively. The HRTEM observations were made by using JEOL JEM 3010 (300 kV, LaB$_6$ cathode and 1.7 Å point resolution) with an energy dispersive X-ray detector. As one can see in Figure 2, Fe particles are joined to each other, while Co particles are mostly completely surrounded by host SiO$_2$. Average size of the Co particles and Fe clusters determined from HRTEM images is 8.4 nm and 12.6 nm, respectively, for the 3 nm thick layers.

**Figure 1.** Left subplot shows schematically the layer structure of the samples. Observations of Fe multilayer structure – 10 nm of SiO$_2$ (light layers) sandwiched with 10 nm (left TEM image) and 3 nm (right TEM image) layers of Fe nanoparticles embedded in SiO$_2$ matrix.

**Figure 2.** Observations of 3 nm layers of Fe nanoparticles (in the above pictures) and Co nanoparticles (in the below pictures) embedded in SiO$_2$ matrix.
2.2. Magneto-optical ellipsometry

Magneto-optical ellipsometry is a very sensitive measurement technique based on a polarization change of light reflecting by magnetic sample [4]. We can describe magneto-optical response of a sample by using the Jones reflection matrix

\[ R = \begin{bmatrix} r_{ss} & r_{ps} \\ r_{sp} & r_{pp} \end{bmatrix} \]  

where \( r_{ij} \) is the amplitude reflection coefficient. Now, two magneto-optical angles can be defined for incident \( s \)- and \( p \)-polarized light using approximation of small angles

\[ \frac{r_{sp}}{r_{ss}} = \frac{\tan \theta_s + i \tan \epsilon_s}{1 - i \tan \theta_s \tan \epsilon_s} \approx \theta_s + i \epsilon_s \]  

(2)

\[ \frac{r_{ps}}{r_{pp}} = \frac{\tan \theta_p + i \tan \epsilon_p}{1 - i \tan \theta_p \tan \epsilon_p} \approx \theta_p + i \epsilon_p \]  

(3)

In this paper we use the \( s \)-polarized incident light, thus the reflected light is polarized elliptically with the ellipticity \( \epsilon_s \) and the azimuth rotation \( \theta_s \).

\[ \text{Figure 3. Basic MO configurations} \]  

– polar, longitudinal and transverse component of magnetization vector.

Figure 3 shows three components of the magnetization vector. However, their contributions (frequently polar and longitudinal) can be mixed together. Therefore it is very important to make good separation of different contributions. One of the ideas is separating by using 180° sample and magnetic field rotation. This idea is based on fact, that the polar contribution is symmetric, while longitudinal is antisymmetric [6]. Thus, separation can be done by addition and substraction of measured data.

Our experimental set-up consists of semiconductor laser working at wavelength of \( \lambda = 670 \) nm, polarizer, quarter-wave plate as a retarder, Wollaston prism, and two photodiodes. The method is so called differential intensity method. We have measured longitudinal and transversal hysteresis loops for incident \( s \)-polarized light as a function of the external in-plane magnetic field \( H \). The angle of incidence was set to \( \varphi = 60^\circ \) and the maximum external magnetic field of \( H_{\text{max}} = 1 \) kOe was applied.

2.3. Optical ellipsometry

Ellipsometric characterization is based on the change of the incident light polarization by a sample. Classical ellipsometry enables to measure the amplitude and the phase of the complex reflection coefficients ratio

\[ \frac{r_{pp}}{r_{ss}} = \tan \psi \ e^{i \Delta} \]  

(4)

where \( r_{pp} \) and \( r_{ss} \) are the Fresnel reflection coefficients for \( p \)- and \( s \)-polarized light and \( \psi, \Delta \) are the ellipsometric angles.
We use the ellipsometer UVISEL produced by Jobin-Yvon Horiba company, which is based on phase modulation technology and covers the spectral range from 190 nm to 2100 nm. Our ellipsometric data were measured at the angle of incidence of 70° and were fitted to the model based on the Maxwell-Garnett and Bruggeman effective medium approximation describing nanoparticulate films.

3. Results
Longitudinal and transverse hysteresis loops of single layer \((N=1)\) of Co and Fe particles were measured for different sample azimuth angles. Figure 4 shows the hysteresis loops for easy and hard axis, and the polar plot of remanence effect. Clear two-fold in-plane uniaxial anisotropy confirms that coherent rotation of the magnetization dominates.

In contrast to the Fe sample, the Co nanoparticulate film is almost magnetically isotropic, which is evident from the polar remanence plot shown in Fig. 5. Also the shape of the longitudinal hysteresis loop is almost independent on the in-plane magnetic field direction. The origin of this magnetic isotropy is associated with the random distribution of the magnetic properties of nanoparticles, which is also confirmed by rounded shape of the longitudinal hysteresis loop (see Fig. 5).

We have studied multilayer samples \((N = 10)\) using spectroscopic ellipsometry to increase sensitivity to optical properties of ferromagnetic particular layers. Complex refractive indexes of the Co and Fe particles embedded in SiO\(_2\) matrix were modeled using the Maxwell-Garnett and Bruggeman effective medium approximations. The Maxwell-Garnett approach describes a system of very diluted collections of small particles completely surrounded by host material [7]. Let us consider the mixture where spherical inclusions, much smaller than the optical wavelength, with permittivity \(\varepsilon_i\) occupy random positions in the environment of permittivity \(\varepsilon_e\). Then the Maxwell-Garnett mixing formula can be written as

\[
\frac{\varepsilon_{\text{eff}} - \varepsilon_e}{\varepsilon_{\text{eff}} + 2\varepsilon_e} = f \frac{\varepsilon_i - \varepsilon_e}{\varepsilon_i + 2\varepsilon_e}
\]

where \(\varepsilon_{\text{eff}}\) is the effective permittivity and \(f\) denotes the volume fraction of the inclusions in the mixtures. The Bruggeman approach describes a composite of aggregated phases or random-
mixture structures. The essence is the absolute equality between the phases in the mixture. The Bruggeman formula can be written as

\[(1 - f) \frac{\epsilon_e - \epsilon_{eff}}{\epsilon_e + 2\epsilon_{eff}} + f \frac{\epsilon_i - \epsilon_{eff}}{\epsilon_i + 2\epsilon_{eff}} = 0 \] (6)

We have applied a simple model for multilayer sample \(N = 10\) consisting of the 3 nm thick Fe films by using basic optical constants of Fe and SiO\(_2\) [8, 9]. As can be seen in Figure 6, the Bruggeman model is more appropriate for Fe particles. The reason is associated with the fact, that most of the nanoparticles are joined to each other. The obtained thicknesses are: \(t_{SiO_2} = 11.5\) nm, \(t_{Fe} = 7.3\) nm and the volume fraction of Fe is \(f_{Fe} = 0.36\). On the other hand, ellipsometric data of Co particular film can be sufficiently well fitted also using the Maxwell-Garnett model, which indicates that the particles are more separated.

**Figure 6.** Ellipsometric angles \(\psi\) and \(\Delta\) of 3 nm thick Fe multilayers. Comparison between the Maxwell-Garnett model and the Bruggeman model shows that the later fits the data better.

### 4. Conclusions

Ferromagnetic nanoparticles of Co and Fe embedded in SiO\(_2\) matrix were prepared using the RF magnetron sputtering with alternating targets. Magnetic nanoproperties were studied using the magneto-optic vector magnetometry. In the case of Co nanoparticles smooth hysteresis curve was observed almost independent on magnetic field direction, which indicate randomly oriented magnetic moments of the particles. Strong in-plane uniaxial magnetic anisotropy was observed in the case of the Fe nanoparticles. Origin of the anisotropy is going to be a subject
of further study. Also applicability of the Bruggenmam effective medium approximation to fit the ellipsometric data indicates particle coalescence.

Acknowledgements
Partial support from the projects KAN 400100653 (Grant Agency of Academy of Sciences of the Czech Republic), MSM6198910016 (Ministry of Education of the Czech Republic), and CZ.1.05/2.1.00/01.0040 (RMTVC), and SP/2010150 is acknowledged.

References
[1] Terris B D and Thomson T 2005 J. Phys. D: Appl. Phys. 38 R199–R222
[2] Pawlak D A 2008 Scientia Plena 4 014801
[3] Zhou H, Kumar D, Kvit A, Tiwari A and Narayan J 2003 J. Appl. Phys. 94 4841–4846
[4] Postava K, Hrabovský D, Hamrlková J, Pištora J, Wawro A, Baczewski L T, Svěklo I and Maziewski A 2011 Thin Solid Films (to be published)
[5] Postava K, Hrabovský D, Životský O, Pištora J, Dix N, Muralidharan R, Caicedo J M, Sánchez F and Fontcuberta J 2009 J. Appl. Phys. 105 07C124 ISSN 0021-8979
[6] Ding H F, Pütter S, Oepen H P and Kirschner J 2001 Phys. Rev. B 63 134425–1–134425–7
[7] Sihvola A 2000 Electromagnetic Mixing Formulas and Applications IEE Publication Series (Institution of Engineering and Technology (IET))
[8] Palik E D (ed) 1991 Handbook of Optical Constants of Solids I, II, III (Academic Press)
[9] Johnson P B and Christy R W 1974 Phys. Rev. B 9 5056–5070