Magneto-Optical Properties of Interfaces in $[\text{NiO}/\alpha\text{-Fe}_2\text{O}_3]_N$ and $[\alpha\text{-Fe}_2\text{O}_3/\text{NiO}]_N$ Multilayers

R. Lopusník*, S. Visnovsky**, N. Keller**, M. Guyot**, R. Krishnan**, and M. Tessier**

*Institute of Physics, Charles University, Ke Karlovu 5, CZ-121 16 Prague 2, Czech Republic

**Laboratoire de Magnétisme et d'Optique, UVSQ-CNRS, 45, Ave des Etats Unis, F-78035 Versailles Cédex, France

Magneto-optic polar Kerr spectroscopic and magnetization studies on the oxide multilayers $\text{NiO}/\alpha\text{-Fe}_2\text{O}_3$ prepared by pulsed laser deposition on quartz substrates are reported. The spectra are consistent with the hypothesis that a layer of spinel ferrite NiFe_2O_4 is formed at the interfaces. The line width in the MO Kerr spectra are typical for the ferrimagnetic spinels containing exclusively ferric ions. The formulae which explain the role of the interface and propagation effects on the observed MO response in reflection and transmission are provided.

Key words: magneto-optic recording, magneto-optic spectroscopy, magnetic multilayers, oxide films, pulsed laser deposition

1. Introduction

Magnetic oxide films such as spinel ferrites, garnets and hexaferrites represent an alternative to metallic materials currently employed in magneto-optic (MO) recording. A variety of them including orthoferrites can be prepared by pulsed laser deposition (PLD) ¹⁻⁴⁾. The interdiffusion is important in multilayer preparation. To investigate this we studied $[\alpha\text{-Fe}_2\text{O}_3/\text{NiO}]_N$ multilayers. The choice of this system is because both oxides are antiferromagnetic and if interdiffusion occurs then NiFe_2O_4 will be formed which can be studied conveniently by magnetic and magneto-optic methods. The sequential ordering of the layers during the deposition is relevant and an asymmetry exists in the interfaces as has been demonstrated by us ^{2,3)}. Therefore we studied both $[\alpha\text{-Fe}_2\text{O}_3/\text{NiO}]_N$ and $[\text{NiO}/\alpha\text{-Fe}_2\text{O}_3]_N$ multilayers.

2. Experimental details

The oxide $[\text{NiO}/\alpha\text{-Fe}_2\text{O}_3]_N$ and $[\alpha\text{-Fe}_2\text{O}_3/\text{NiO}]_N$ multilayers were prepared by pulsed laser deposition (PLD) at a wavelength 355 nm using pulse of 6 ns with a repeat rate of 10 Hz and typical fluences of 10 J/cm². Fused quartz discs of 10 mm diameter and 0.3 mm thick served as substrates, which were held at 430 - 450 °C. The average deposition time for each layer was up to 30 min. The first layer on the substrate was either $\alpha\text{-Fe}_2\text{O}_3$ in $[\alpha\text{-Fe}_2\text{O}_3/\text{NiO}]_N$ or NiO in $[\text{NiO}/\alpha\text{-Fe}_2\text{O}_3]_N$ systems (Fig. 1). $N=1$ denotes the couple $\alpha\text{-Fe}_2\text{O}_3/\text{NiO}$ or $\text{NiO}/\alpha\text{-Fe}_2\text{O}_3$ and $N=1.5$ indicates that another $\alpha\text{-Fe}_2\text{O}_3$ or NiO layer was added, respectively. More details can be seen in our earlier work ^{2,3)}. In order to keep the

conditions similar for each layer, as regards thermal diffusion, we kept the total deposition and annealing times constant at 30 min, the temperature being the same for both (Table 1).

Table I Samples

System	N	Deposition (min)	Annealing (min)
$\text{NiO}/\alpha\text{Fe}_2\text{O}_3/\text{SiO}_2$	1	30	0
$\text{NiO}/\alpha\text{Fe}_2\text{O}_3/ \dots / \text{NiO}/\text{SiO}_2$	2.5	15	15
$\text{NiO}/\alpha\text{Fe}_2\text{O}_3/ \dots / \text{NiO}/\text{SiO}_2$	2.5	20	10

In addition to our previous studies in transmission ^{2,3)} we have carried out complex angle MO Kerr spectroscopy at nearly normal light incidence between 1.2 and 4.5 eV on the samples. This technique is sensitive to the presence of the thinnest magnetically ordered interface layers. Unless otherwise stated the spectra pertain to the multilayer side of the sample. The topmost layer in all our MO Kerr experiments was always NiO. Prior the spectroscopic experiment the polar Kerr rotation hysteresis loops were taken in an applied field up to 1.5 Tesla to ascertain complete saturation.

3. Results and discussion

Magnetization and Faraday rotation display oscillations with N. This indicates a magnetic and structural non equivalence of the $\text{NiO}/\alpha\text{-Fe}_2\text{O}_3$ and $\alpha\text{-Fe}_2\text{O}_3/\text{NiO}$ interfaces. The MO Kerr rotation loops with H_c of the order of 80 kA/m confirmed that the saturation has been attained in all the samples. Figure 2 shows the Kerr rotation in $[\alpha\text{-Fe}_2\text{O}_3/\text{NiO}]_1$, i.e., in the system with a single interface. The Kerr spectra of the system $[\text{NiO}/\alpha\text{-Fe}_2\text{O}_3]_{2.5}$ with two different deposition/annealing times are displayed in Fig. 3. An ellipticity spectrum is included for one of them to illustrate the compatibility required by the dispersion relations. The spectra taken on $[\text{NiO}/\alpha\text{-Fe}_2\text{O}_3]_{2.5}$ from the substrate side are shown in Fig. 4. The spectroscopic structure of the multilayer is superimposed on a monotonously increasing phase averaged MO rotation in the substrate. It is recalled that the layer closest to the substrate is again NiO.

Both the multilayer composition profile and the optical and MO constants of the individual layers must be accounted for to understand the observed MO effects. Antiferromagnetic NiO shows absorption which increases up to 4 eV. On the main absorption wing there

Fig. 1 Diagram of $[\text{NiO}/\alpha\text{-Fe}_2\text{O}_3]_N$ and $[\alpha\text{-Fe}_2\text{O}_3/\text{NiO}]_N$ multilayers.

is structure assigned to the crystal field transitions ^{5,6}. In hematite, $\alpha\text{-Fe}_2\text{O}_3$, the Fe^{3+} crystal field transitions are superimposed on the absorption edge situated in the same spectral range ⁷.

The MO Kerr spectrum of nickel ferrite, NiFe_2O_4 , is determined by four strong electronic transitions assigned either to the charge transfer from Ni^{2+} to octahedral Fe^{3+} , or to the intersublattice exchange between the tetrahedral and octahedral Fe^{3+} ⁸. The spectral lines of NiFe_2O_4 are broader due to a higher degree of covalency in bonding related to the presence of Fe^{3+} in non-centrosymmetric tetrahedral sites ⁹. The upper limit of the photon energy range for the spectra measured on the multilayer side is due to a scattering at diffused interfaces and to an increased

Fig. 2 MO polar Kerr rotation in $[\alpha\text{-Fe}_2\text{O}_3/\text{NiO}]_1$ multilayer.

absorption and reflectivity in the topmost NiO layer above 3.8 eV ^{5,6}.

All the MO Kerr spectra show a similar structure, although the peak amplitudes change with N and preparation conditions. The main features of the spectra can be explained from a model of the optical interaction in magnetic multilayers. The complex ellipsometric angles characterizing Faraday (χ_i) and normal incidence polar Kerr effect (χ_r) can be expressed analytically which allows us to appreciate the role of propagation and interface effects as well as non-linear thickness dependence of MO effects. For an incident monochromatic wave of zero azimuth and ellipticity we have ¹⁰

$$\chi_i^{(0,n+1)} = \frac{1}{M_0} \sum_{j=1}^n \frac{\Delta N_j}{2N_j} \left[i(1 - e^{-2i\beta_j}) I_j + 2\beta_j J_j \right] \quad (1)$$

$$\chi_r^{(0,n+1)} = \frac{1}{M_0 M_1} \sum_{j=1}^n \frac{\Delta N_j}{2N_j} \left[-i(1 - e^{-2i\beta_j}) K_j + 4\beta_j e^{-2i\beta_j} H_j \right] \quad (2)$$

where

$$\beta_j = N_j \frac{\omega}{c} t_j \quad (3)$$

characterizes the radiation penetration depth, (ω/c) is the radiation wavevector in a vacuum, $2\Delta N_j$ is the difference in the refractive indices for the circular polarizations, N_j and t_j are the complex index of refraction and the thickness of the j -th layer. The constants I_j , J_j , K_j and H_j depends on the Fresnel coefficients and exponential factors (3), n denotes the number of layers in the stack. The matrix elements M_0 and M_1 are reduced by a common factor $M_0 \propto M_{11}$, $M_1 \propto M_{21}$ where M_{ij} stand for the elements of the $\mathbf{M}$ -matrix characterizing the isotropic multilayer in the absence of magnetic

ordering ¹¹. The first term in the brackets is mostly due to the interface effects while the second one describes the propagation.

For a single magnetic interface layer sandwiched between the non-magnetic layers, $\text{air}(0)/\text{NiO}(1)/\text{NiFe}_2\text{O}_4(2)/\alpha\text{-Fe}_2\text{O}_3(3)/\text{SiO}_2(4)$ or $\text{air}(0)/\alpha\text{-Fe}_2\text{O}_3(1)/\text{NiFe}_2\text{O}_4(2)/\text{NiO}(3)/\text{SiO}_2(4)$ simplified closed form expressions can be written down for the Faraday and normal incidence polar Kerr effect, respectively, which provide some insight to the trends in MO effects when either the layer thickness or the radiation frequency change. We obtain

$$\chi_i = \frac{\Delta N_2}{2N_2} \left(1 + r_{01}r_{34}e^{-2i(\beta_1+\beta_2+\beta_3)}\right)^{-1} \quad (4)$$

$$\times [i(1 - e^{-2i\beta_2})(r_{01}e^{-2i\beta_1} - r_{34}e^{-2i\beta_3}) + 2\beta_2(1 - r_{01}r_{34}e^{-2i(\beta_1+\beta_2+\beta_3)})],$$

$$\chi_r = \frac{\Delta N_2}{2N_2} \left(1 + r_{01}r_{34}e^{-2i(\beta_1+\beta_2+\beta_3)}\right)^{-1} (r_{01} + r_{34}e^{-2i(\beta_1+\beta_2+\beta_3)})^{-1} \quad (5)$$

$$\times (1 - r_{01}^2)e^{-2i\beta_1} [-i(1 - e^{-2i\beta_2})(1 + r_{34}^2e^{-2i\beta_2}e^{-4i\beta_3}) + 4\beta_2e^{-2i\beta_2}r_{34}e^{-2i\beta_3}],$$

where r_{01} and r_{34} are the Fresnel reflection coefficients for the air - layer 1 and layer 3 - substrate interfaces, respectively. In our case the multilayers display diffuse interfaces and we can expect an amplitude reduction of the reflection coefficients at the boundaries between the magnetic layers and the sandwiching media (NiO and α -Fe₂O₃). We have therefore neglected the reflection at the interfaces of NiO/NiFe₂O₄ and α -Fe₂O₃/NiFe₂O₄ ($r_{12}=r_{23}=0$) and a weak effect of the reflections on the back side of the quartz substrate. The propagation effects disappear and Faraday rotation cannot be measured when the penetration length is much smaller than layer thickness, corresponding to $\exp(-2i\beta) \ll 1$. Then, as Eq. (5) shows, only the conventional MO Kerr effect for a single interface can be measured.

The above formulae allow us to understand why in the two multilayers with $N=2.5$ which differ in the deposition and annealing time, we observe such a large differences in their spectra (Fig. 3). For the layer thickness involved of the order of 10 nm the contributions from the propagation and interface effects to the MO response are of the same order of magnitude and small differences in optical constants may introduce appreciable changes in the spectra. Similar behavior has been observed on hexaferrite films¹²⁾.

The computer simulations¹³⁾ carried out for the multilayers $[\text{NiO}/\alpha\text{-Fe}_2\text{O}_3]_1$ and $[\alpha\text{-Fe}_2\text{O}_3/\text{NiO}]_1$ allowed us to appreciate the effect of a varying thickness of individual layers on the MO Kerr spectra. In agreement with our experiment and as predicted by Eq. (5), the most remarkable effect is produced by the weakly absorbing upper NiO layer which introduces a complex phase shift strongly dependent on its thickness. As a result, the MO Kerr rotation and ellipticity spectra become sensitive to the NiO layer thickness in the range from zero to 90 nm and therefore considerably differ from those observed in the ferrimagnetic NiFe₂O₄ surface or thin film.

In conclusion, we have reported MO spectra of the multilayers prepared by pulsed laser deposition with magnetic layers formed at the NiO/ α -Fe₂O₃ interfaces and provided the formulae which explain the effect of the interface and propagation effects on the observed MO response. The line width in the spectra are typical for the ferrimagnetic spinels containing exclusively ferric ions.

(a)

(b)

Fig. 3 MO polar Kerr effect in two multilayers $[\text{NiO}/\alpha\text{-Fe}_2\text{O}_3]_{2.5}$ which differ in layer deposition/annealing times; (a) Sample (20 min/10 min): rotation and ellipticity, (b) Sample (15 min/15 min): rotation. Measured from the multilayer side.

Fig. 4 MO polar Kerr rotation in two multilayers $[\text{NiO}/\alpha\text{-Fe}_2\text{O}_3]_{2.5}$ which differ in layer deposition/annealing times; (a) Sample (20 min/10 min), (b) Sample (15 min/15 min). Measured from the substrate side.

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