Magneto-optical Polar and Longitudinal Kerr Spectra of NiFe_2O_4 Single-crystals

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Abstract- Measurements and analysis of spectroscopic reflection ellipsometry and magneto-optical polar and longitudinal complex Kerr effect are reported on a natural (111) face of a high quality NiFe_2O_4 single-crystal. The experimental reflection data is expressed in terms of the permittivity tensor. This information is employed in modeling experimental polar Kerr spectra of a NiFe_2O_4 film and a satisfactory agreement has been achieved. These results demonstrate the importance of the spectroscopic magneto-optic reflection ellipsometry in the diagnostics of magnetic oxide films with thicknesses in the range 10^1 to 10^2 nm.

Key words: magneto-optical recording, thin films, ferrimagnetic oxides, pulsed laser deposition, nickel ferrite

1. Introduction

The magnetic oxide films are candidates for the next generation magnetic and magneto-optic (MO) recording media. The present work is motivated by our studies of nickel ferrite (NiFe_2O_4) films prepared by pulsed laser deposition¹⁾⁻³⁾. At the layer thicknesses in the range of 10^1 - 10^2 nm, a unique identification of the formation of NiFe_2O_4 layers is rather demanding. For this purpose, the MO spectroscopic technique represents a useful complementary tool sensitive to the chemical composition, crystallographic structure, layer thickness and the presence of spontaneous magnetization. The approach is analogous to that employed in the classical spectroscopic ellipsometry: one compares the experimentally observed MO spectra with the model system. However, the explanation of the MO response in these samples requires reliable optical and MO data for the NiFe_2O_4 layer in a broad spectral range, e.g. expressed in terms of the complex permittivity tensor. This information has been obtained by Krinchik *et al.* on polycrystalline samples using multiple angle transverse Kerr effect spectroscopy⁴⁾. The investigations by Krinchik *et al.*⁴⁾ on polycrystals and those by Kahn *et al.*⁵⁾ on single-crystals were limited to polar Kerr rotation (PKR) spectra. Note that the polar Kerr ellipticity (PKE) spectra have not yet been reported. The purpose of the present work is to provide a complete set of the MO polar Kerr rotation and ellipticity spectra and ellipsometry spectroscopic data necessary for a complete determination of the permittivity tensor in

NiFe_2O_4 single crystals. In order to check the consistency of the data in the spectral range of concern, complex longitudinal Kerr effect studies were also carried out (*i.e.*, rotation (LKR) and ellipticity (LKE)). The results are then applied to model the spectroscopic MO Kerr ellipsometry measurements of a NiFe_2O_4 layer which illustrates the pertinence and consistency of the determined permittivity tensor spectra.

2. Experimental details

The single-crystal of NiFe_2O_4 has been prepared by a flux technique from high purity raw materials. The crystals of this batch displayed very low line width of ferromagnetic resonance indicating a high degree of perfection of the sample. A natural (111) face of the crystal was used for the present measurements. Also a series of NiFe_2O_4 single layers were prepared by pulsed laser deposition at a wavelength of 355 nm using pulses of 6 ns with a repetition rate of 10 Hz and a typical fluency of 10 J/cm^2 . The samples were grown in an atmosphere of partial oxygen pressure of 60 mTorr. The temperature of the fused quartz substrates was maintained at 450°C during the deposition. The deposition time was varied from 36 min to 1440 min. The values of the layer thickness were estimated by means of stepmeter and magnetization measurements.

The optical and MO experiments were carried out in the photon energy range from 1.4 to 4.3 eV. The real index of refraction and extinction coefficient spectra were obtained by multiple angle spectroscopic ellipsometry using a Sentech ellipsometer SE 850. As expected, the best consistency of the ellipsometric data has been achieved at an angle of incidence of 75° .

The polar and longitudinal MO Kerr experiments were performed using a MO spectrometer employing azimuth modulation and compensation technique. Special care has been taken to minimize the effect of fringing magnetic fields on the optical elements. The small contribution originating from the fringing fields was determined from the effect of magnetic field on the reflection from an aluminium surface and subsequently eliminated.

The polar Kerr azimuth rotation and ellipticity spectra were measured at nearly normal light incidence.

Fig. 1 MO hysteresis loop measured on (111) natural face of NiFe_2O_4 single-crystal. The photon energy of the incident light was 2.8 eV.

The applied magnetic field of 1.3 Tesla was sufficient to saturate the magnetization of the sample, as illustrated by the MO hysteresis loop shown in Fig. 1. In the longitudinal geometry the corresponding azimuth rotation and ellipticity were determined at an angle of incidence of 45° with the incident wave polarized perpendicular to the plane of incidence. In this geometry the degree of magnetic saturation of the bulk NiFe_2O_4 single-crystal reached about 80%.

4. Results and discussion

The optical and magneto-optical spectra of NiFe_2O_4 display a structure typical for ferrimagnetic oxide compounds containing transition metal ions with unfilled 3d-subshell. The MO spectra are sensitive to the presence of a particular transition metal ion, its coordination and its valence. In multilayers, these properties may be employed for the detection of a ferrimagnetic layer with the expected composition. The inverse spinel ferrite NiFe_2O_4 contains ferric ions in both tetrahedral [Fe^{3+}] and octahedral (Fe^{3+}) sites along with nickel ions [Ni^{2+}] in octahedral sites. The ideal nickel ferrite with formula $[\text{Ni}^{2+}\text{Fe}^{3+}](\text{Fe}^{3+})\text{O}_4$, space group O_h , and has a cubic crystal structure with eight formula groups per unit cell. Strong exchange interactions between magnetic ions in tetrahedral and octahedral sites results in a ferrimagnetic ordering with a rather high Néel temperature of 858 K and saturation magnetization of 270 kA/m⁶.

The spectra of the optical constants for a NiFe_2O_4 single-crystal as determined from the ellipsometry are given in Fig. 2. In the studied spectral range we observe a satisfactory agreement of the real index of refraction, n , and the extinction coefficient, k , with experimental data observed on polycrystalline NiFe_2O_4 samples⁴⁾. The reduced amplitudes in the UV region obtained on the polycrystalline samples [4] might be due to

Fig. 2 Measured spectra of refraction index and extinction coefficient on (111) natural face of NiFe_2O_4 single-crystal.

depolarization on surface imperfection and grains of the polycrystalline samples the role of which is enhanced at the photon energy end of the spectral range.

The complex polar Kerr effect is shown in Fig. 3. The structure in the polar Kerr rotation spectra displays all the spectroscopic features observed in the detailed spectrum by Kahn *et al.*⁵⁾. The agreement with the spectrum by Krinchik *et al.*⁴⁾ is also reasonable. The amplitudes in our spectrum are somewhat smaller. At the lower photon energy end of the spectrum the Kerr rotation amplitudes tends to zero in agreement with ref. 4, which is expected in the spectral region situated below the absorption edge. The behavior is not reproduced in the spectrum by

Fig. 3 Polar Kerr rotation and ellipticity spectra measured on (111) natural face of magnetically saturated NiFe_2O_4 single-crystal.

Fig. 4 The real and imaginary part of the off diagonal element of NiFe_2O_4 single-crystal permittivity tensor.

Kahn *et al.*⁵⁾ who reported a significant rotation. Our polar Kerr rotation spectrum differs from that by Kahn *et al.*⁵⁾ by a component that is monotonous in photon energy. We believe that this component is due to the magnetic field dependent baseline originating from the fringing fields generated with the sample electromagnet employed in Ref. 5 and operated at high currents close to the saturation of the magnetic circuit. Note that our polar Kerr ellipticity spectrum is qualitatively consistent with the dispersion relations.

The real and imaginary parts, ϵ'_1 and ϵ''_1 , of the off diagonal tensor element ϵ_1 ($\epsilon_1 = i\epsilon_{xy} = \epsilon'_1 - i\epsilon''_1$) are related to the experimentally observed quantities, the polar Kerr rotation, θ_{KP} , the polar Kerr ellipticity, ϵ_{KP} , the real index of refraction, n , and the extinction coefficient, k , through^{7),8)}

$$\epsilon'_1 = -(C \epsilon_{KP} + D \theta_{KP}), \quad (1)$$

$$\epsilon''_1 = C \theta_{KP} - D \epsilon_{KP}, \quad (2)$$

where

$$D = -k^3 + 3n^2k - k, \quad (3)$$

$$C = n^3 - 3nk^2 - n. \quad (4)$$

The spectra of the real and the imaginary part of the off diagonal tensor component ϵ_1 are given in Fig 4. They agree with those obtained on polycrystalline samples by Krinchik *et al.*⁴⁾. A qualitative microscopic explanation of magneto-optical spectra of NiFe_2O_4 was proposed by Kahn⁵⁾ and Krinchik^{4),9)}. Recently Fontijn *et al.*¹⁰⁾ carried out a detailed analysis of the spectral variation of the off-diagonal tensor element and was able to explain them as a sum of four allowed electronic transitions. The lines of weaker oscillator strength observed below 2.5 eV may be assigned to the electronic transitions within Fe^{3+} and Ni^{2+}

polyhedra of mostly crystal field origin.

To check the compatibility between the obtained optical and magneto-optical constants of the NiFe_2O_4 single-crystal, we carried out complex longitudinal Kerr measurements that were subsequently compared with calculations based on the tensor elements deduced from our polar measurements. We employ classical Voigt formula¹¹⁾

$$\theta_{LK} - i\epsilon_{LK} = \frac{-i\epsilon_1 N_y N \cos \varphi (N_z)^{-1}}{(NN_z + \epsilon_0 \cos \varphi)(N \cos \varphi - N_z)}, \quad (5)$$

expressing longitudinal Kerr rotation θ_{KL} and ellipticity ϵ_{KL} for s-polarized incident light as a function of the angle of incidence φ , the refractive index N and the diagonal element of permittivity tensor ϵ_0 ($\epsilon_0 = \epsilon_{xx} = \epsilon'_{xx} - i\epsilon''_{xx}$). We denote $N_y = N \sin \varphi$, $N_z = (\epsilon_0 - N_y^2)^{1/2}$.

An excellent agreement between calculated and experimental spectra was obtained below a photon energy of 3 eV (Fig 5.). Above this energy the spectroscopic features are still well reproduced. The differences between the experiment and calculation at higher photon energies are most probably due to the wavelength dependent scattering on imperfections of the natural surface.

As mentioned above, our main focus was to apply the MO polar Kerr spectroscopy to the detection of NiFe_2O_4 layers formed during the pulsed laser deposition onto fused quartz substrate. We have carried out spectroscopic MO Kerr experiments on the film-substrate system and compared the result with the model. The experiment showed that for the model it is sufficient to consider a simplified system consisting of a magnetic Ni-ferrite film on an infinite fused quartz substrate. In the film thickness range of the order of $10^1 - 10^2$ nm both the propagation and interface effects in multiple reflections must be accounted for. The MO response of the system in

Fig. 5 Experimental longitudinal Kerr rotation and ellipticity spectra measured on (111) natural face of NiFe_2O_4 single-crystal are compared with modeling.

The approximation of small ellipsometric angles is restricted to the MO effects linear in the off-diagonal

$$\theta_{PK} - i\epsilon_{PK} = \frac{-i(1-r_{01}^2)\delta N_1}{2N_1(1+r_{01}r_{12}e^{-2i\beta_1})(r_{01}+r_{12}e^{-2i\beta_1})} [(1+r_{12}^2e^{-2i\beta_1})(1-e^{-2i\beta_1}) + 4i\beta_1r_{12}e^{-2i\beta_1}], \quad (6)$$

The single interface Fresnel coefficients are given by

$$r_{n-1,n} = \frac{N_{n-1} - N_n}{N_{n-1} + N_n}. \quad (7)$$

We have further denoted

$$\delta N_1 = \frac{1}{2}(N_+ - N_-), \quad (8)$$

with the complex refractive indices for right (+) and left (-) circular polarized light given by $(N_{\pm})^2 = \epsilon_{xx} \pm i\epsilon_{xy}$, $\beta_1 = N_1(\omega/c)t_1$ and t_1 the film thickness. Note that for a thick absorbing film it reduces to the case of an interface between isotropic ambient and bulk magnetic medium.

In Fig. 6, we show the measured PKR spectra of a NiFe_2O_4 film deposited onto a fused quartz substrate. The thickness of the film was 93 ± 7 nm as evaluated from the stepmeter and VSM magnetization measurements. Making use of formula (6) and the permittivity tensor data from Figs. 2 and 4, we have computed the spectra for the model system assuming the NiFe_2O_4 film thickness of 120 nm. The result is shown in Fig. 6 and we observe a satisfactory agreement with the experiment. The spectroscopic data obtained from the reflection ellipsometry and MO Kerr experiments reproduce well the behavior of thin films at a thickness where both the interface reflection and transmission as well as and propagation effects are important.

Fig. 6 Experimental (circles) and calculated (solid line) PKR spectra for a NiFe_2O_4 single layer deposited by pulsed laser ablation onto a fused quartz substrate.

tensor element and is given governed by the relation ¹²⁾

4. Conclusion

A complete set of the optical and magneto-optical data measured on a (111) natural face of a NiFe_2O_4 single crystal has been obtained in the photon energy range between 1.2 and 4.3 eV. The results extend the spectroscopic information available so far. The consistency of the experimentally determined optical constants and polar Kerr spectra was demonstrated by comparison of the measured and computed MO Kerr spectra or the longitudinal configuration. The spectra of the tensor elements obtained from classical reflection ellipsometry and from MO reflection spectroscopy at polar magnetization have been employed for the analysis of the NiFe_2O_4 layers. Spectroscopic reflection MO ellipsometry is a powerful and highly sensitive diagnostics tools for the study of the magnetic oxide thin films with a thickness in the range 10^1 - 10^2 nm.

Acknowledgments

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