Nonlinear Magneto-Optical Microscopy

V. Kirilyuk, A. Kirilyuk, and Th. Rasing
Research Institute for Materials, University of Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands

(Received May 11, 1998; Accepted August 26, 1998)

We report the development of a new magnetic imaging technique: nonlinear magneto-optical microscopy. Magnetization induced second harmonic generation (MSHG) is used to image the domain structure in ultrathin CoNi/Pt films. Due to both very high magneto-optical contrast and enhanced interface sensitivity, fine details of magnetization reversal on CoNi/Pt interfaces become visible with the MSHG imaging that are not detectable by usual magneto-optics. The problem of separating the images from different multilayer interfaces is discussed.

Keywords: magneto-optical imaging, optical second harmonic generation, magnetic domains, ultra thin films

1. Introduction

Technologically, thin magnetic films have found a lot of important applications, e.g. as recording layers of computer hard disks or in giant magnetoresistive sensors. The key process in all these devices is the switching of the film magnetization direction. This process of magnetization reversal plays therefore a primary role here and has been thoroughly studied for many systems. This is also why numerous magnetic imaging techniques were developed and used for domain structure studies in ultra thin films.

When reducing the film thickness, film interfaces become more and more important, to a large extend determining the magnetic properties of a thin film. Curie temperature, magnetic anisotropy, coercive field, etc. are in a direct correlation with the interface structure. This is especially valid when the hysteresis properties of such an ultra thin film are considered. In turn, the magnetic hysteresis is a consequence of the magnetic domain structure behavior, both static and dynamic.

This paper reports the development of a new magnetic imaging technique: magnetization-sensitive optical second harmonic generation. The technique is selectively sensitive to the interfaces of a multilayer structure and displays huge magneto-optical effects. The potential magneto-optical recording material - CoNi/Pt multilayers was taken as an example for the reported study. The magneto-optical contrast appeared to be extremely high in the nonlinear case eliminating the need for the image treatment and thus facilitating the measurements.

2. Method and experimental setup

SHG arises from the nonlinear polarization \( P(2\omega) \) induced by an incident laser field \( E(\omega) \). In the electric dipole approximation this polarization can be written for a magnetic material as:

\[
P(2\omega) = \left| \chi_{ijkl}^{\pm} (M) \right| E_j(\omega) E_k(\omega)
\]

where \( \chi_{ijkl}^{\pm} \) are respectively the even and odd elements of the nonlinear susceptibility tensor \( \chi^{(2)} \) which fulfill \( \chi_{ijkl}^{\pm} (-M) = \pm \chi_{ijkl}^{\pm} (M) \), as suggested by Ru-Pin Pan et al. The (001) (or isotropic) surface and in the transversal magneto-optical configuration, with \( M \) perpendicular to the sample surface, this tensor is given by:

\[
\chi^{(2)} = \begin{pmatrix}
xxx & xyy & xzz & 0 & xzt & 0 \\
0 & 0 & 0 & yzz & 0 & yyy \\
xzt & yzz & zzz & 0 & zxx & 0
\end{pmatrix}
\]

The elements shown in italic are even in the magnetization, while those in boldface are odd. Due to this form of the \( \chi^{(2)} \) tensor, in magnetic materials the SHG intensity depends on the magnetization direction. For example for the \( S_{in} P_{out} \) polarization combination (i.e. \( E(\omega) \parallel y \)) we obtain:

\[
I^{2\omega}(\pm M) = \left| X_{xyy} \pm X_{xyz} \right|^2 = \left| X_{xyy} \right|^2 + \left| X_{xyz} \right|^2 \pm 2\left| X_{xyy} X_{xyz} \right|
\]

The change of the SHG intensity is thus due to the interference between even and odd ("crystallographic" and "magnetic") tensor components. It is straightforward to define a magnetic signal as the MSHG asymmetry:

\[
\rho = \frac{I^{2\omega}(+M) - I^{2\omega}(-M)}{I^{2\omega}(+M) + I^{2\omega}(-M)}
\]

For our experiments we used a Ti:sapphire laser operating at a repetition rate of 76 MHz and a pulse width of about 100 fs. The incoming laser light was filtered and focused onto the sample (see Fig. 1). Polarization control of both incident fundamental and reflected SH light was achieved by means of polarizers. The image of the sample was created with a help of a microscope objective (x20, N.A. 0.35). Appropriate optical filtering was used before the signal was detected by a liquid nitrogen cooled CCD camera.

The object of the present study was the magnetization reversal in thin CoNi films sandwiched by Pt that were recently investigated by MSHG. The CoNi/Pt multilayers have been found to be a promising magneto-optical
recording material because of their low Curie temperature and strong magneto-optical effects in the short wavelength region\textsuperscript{12}. Prepared by sputtering, the samples showed very strong dependence of their magnetic interface properties on the sputtering Ar pressure\textsuperscript{14,15}.

In the transversal magneto-optical geometry, the magnetic contrast \( p \) (Eq. (4)) for the most effective \( P_{in}, P_{par} \) polarization combination was around 80\%, eliminating the usual (for the linear imaging) procedure of background subtraction as unnecessary. For the sake of comparison, it was possible to measure in the same geometry also the linear optical image of the domain structure, using the polarized light from a white light source (halogen lamp) for the sample illumination.

3. Results and discussion

Fig. 2 shows the linear (a) and nonlinear (b) images of the same domain structure measured in a 9 nm thick CoNi film. The size of the MSHG image is restricted to the diameter of the focused laser beam. We should underline here that the MSHG image is shown without any contrast improvement nor background subtraction that is used to obtain the linear image.

Roughly speaking, the domain pattern is very similar on these two images; however all fine details of the domain pattern are visible in the MSHG image only. For example, the change of the contrast along the cross section between the white lines cannot be inferred from the linear picture.

The magnetization reversal process was studied next starting from a fully saturated sample (Fig. 3(a)). The magnetic field of the same value (approximately 60 Oe) was applied for given time intervals (usually 20 seconds) followed each time by an image accumulation (10 min. per image). The change of the domain pattern after each subsequent field application indicates the long time scale, thermally activated, domain wall dynamics that was extensively studied in ultrathin films with perpendicular magnetic anisotropy\textsuperscript{7-14}.

Again, as in the MSHG image of Fig. 2, there are some details of the domain structure with a contrast of some 10-20\% with respect to the contrast of the opposite domains. For example, images (c-g) clearly show several
faint stripes extending along the diagonal of the images. All of them disappear later, in a completely saturated sample (b). For the moment, we cannot give them an unambiguous interpretation as interface structures; this might be also some subdomains that are homogeneous across the film but have their magnetization direction slightly different from that of the main domain.

In order to answer this question, a precise comparison of the linear and MSHG images is necessary. However, the low quality of the linear images (due to the very small magneto-optical response in the transversal geometry) did not allow us to do so.

4. Future developments

In Ref. 14, a method is developed that allows one to separate the contributions to MSHG coming from the different interfaces of the multilayer structure. Multiple scattering calculations are used, in order to compute the SHG in a system with different interfaces. Then, using experimental data (images) as an input to the fitting program, it is possible to obtain the values of the nonlinear optical tensor separately for different interfaces. In standard MSHG measurements, angle-of-incidence dependence of the SHG intensity and/or magnetic signal is used as the required set of data. Actually, in the process of imaging with a high-aperture objective, the signals are integrated over a large range of the angles of incidence. The most straightforward way to get the required data set would be to image the same area at a variable aperture, and to treat the obtained set of images with the multiple scattering approach. This is, however, a subject for future studies; no such apertures were used at present.

Another problem that has to be solved is the limited size of the MSHG image. Because of the quadratic dependence of the nonlinear signal on the incoming light intensity, simple defocusing of the beam decreases the signal very quickly below the detection limit. To avoid the infinite image accumulation time, scanning of the fundamental beam should be employed.

In spite of several questions to be answered and problems to be solved, the advantages of the technique are clear and can be successfully used for the studies of the magnetic domain structures in ultra thin films and at their interfaces.

Acknowledgements We thank M.A.M. Haast and J.C. Lodder for kindly supplying us with samples. Part of this work was supported by an HCM institutional fellowships ERBCHBGCT930444 and ERBCHRXTCT940563 and the European TMR network ERBFMRXCT960015 (NO-MOKE).

References