Linear and nonlinear magneto-optical diffraction from one-dimensional periodic structures

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Nonlinear magneto-optical diffraction from periodically structured samples has been studied using magnetization-induced second harmonic generation (MSHG). Several orders of diffraction were clearly observable in MSHG, along with a strong dependence on the magnetization. Sizeable asymmetry between the MSHG signal measured in positive and negative diffraction peaks, especially visible in second order, are explained by Fresnel factors. It was found that first-order diffraction hysteresis loops differ from all others by showing an “overshoot” at magnetization reversal, both in MSHG and in the linear magneto-optical Kerr effect. Tentatively this behavior is explained as due to inhomogeneous reversal of the magnetization in the stripes. © 2003 American Institute of Physics. [DOI: 10.1063/1.1557761]

Recent theoretical work has indicated the potential of nonlinear magneto-optical diffraction to study periodic magnetic arrays.¹ The nonlinear magneto-optical technique of magnetization-induced second harmonic generation (MSHG) has been shown to possess several advantages over linear magneto-optics. Most of all, due to its surface/interface sensitivity, the nonlinear magneto-optical response provides information about the magnetization of surfaces and buried interfaces separately from the bulk.² Due to different selection rules and higher-order optical tensors, other magneto-optical effects also arise in MSHG.³ These effects have allowed, e.g., magneto-optical observation of antiferromagnetic domains⁴ and of complicated domain patterns in strained films of yttrium–iron–garnet.⁵ Therefore, it might be expected that nonlinear magneto-optical diffraction would allow us to get more detailed information about periodic structures as well as about contributions by domain walls.

The aim of this work is to show the possibility of both observing and investigating in more detail the MSHG diffraction from one-dimensional periodic structures. For comparison, linear magneto-optical Kerr effect (MOKE) diffraction was studied as well.

Several orders of diffraction were clearly observable in the MSHG response from a patterned CoNi film, with a strong dependence on the magnetization. The peculiar shape of the hysteresis loops in the first-order diffraction of both MSHG and MOKE can tentatively be explained by inhomogeneous reversal of the magnetization of the stripes.

In the electric-dipole approximation, SHG is expressed through the second-order polarization $P(2\omega)$ induced in a medium by an incident electromagnetic wave $E(\omega)$:

$$P_i(2\omega) = \chi_{ijk}^{(2)} E_j(\omega) E_k(\omega). \quad (1)$$

The third-rank polar tensor $\chi^{(2)}$ vanishes in any cen-

trosymmetric medium. Hence, a symmetry breaking surface or interface is a source of SHG, and gives rise to the extreme interface sensitivity of the technique. The presence of magnetization does not influence the bulk inversion symmetry but does change the symmetry of the interface, making magnetic probing also interface sensitive.⁶ For an isotropic surface in a transverse magneto-optical configuration ($\mathbf{M} \parallel \mathbf{y}$, xz is the plane of incidence) the nonlinear magneto-optical tensor $\chi^{(2)}$ can be written as

$$\chi^{(2)} = \begin{pmatrix} \mathbf{xxx} & \mathbf{xyy} & \mathbf{xzz} & 0 & xzx & 0 \\ 0 & 0 & 0 & yzy & 0 & \mathbf{yxy} \\ zxx & zyy & zzz & 0 & \mathbf{zzx} & 0 \end{pmatrix}. \quad (2)$$

The elements shown in bold face are *odd* in the magnetization (roughly proportional to it).⁷

For the MSHG measurements, a pulsed laser beam from a Ti-sapphire laser (76 MHz $\times$ 100 fs pulses) with a wavelength of 760 or 810 nm was focused onto the sample. The polarization of the incoming fundamental laser beam was chosen using a Babinet Soleil compensator and a polarizer was used afterward to ensure a high degree of light polarization. An analyzer was used to choose the polarization of outgoing SHG that was detected after proper filtering with a photomultiplier. To test different components of the nonlinear optical tensor, different input–output polarization combinations ($P_{\text{in}}-P_{\text{out}}, S_{\text{in}}-P_{\text{out}}, S_{\text{in}}-S_{\text{out}}, P_{\text{in}}-S_{\text{out}}$) were used, where P and S polarizations are defined as parallel and perpendicular to the plane of incidence, respectively.

For the MOKE hysteresis measurements, a Wollaston prism in combination with a differential two-diode detection scheme was employed. The angle of incidence was always 45°, and the detection arm could be rotated in order to measure the magneto-optical signal as a function of the angle of diffraction. Transverse magneto-optical geometry was used for all experiments.

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FIG. 1. MOKE and MSHG hysteresis loops measured for different orders of diffraction for the sample with $d=5\ \mu\text{m}$. Note that the second order for MSHG corresponds to the same beam direction as the first-order MOKE.

The samples were prepared in a computer-controlled sputtering system, base pressure of 5×10^{-8} mbar, with argon as the sputtering gas. The deposition rates were kept low (1.7–2.0 Å/s for Pt and 0.4–0.6 Å/s for CoNi) to assure smooth layer growth and good control of the layer thickness. A 40 nm thick Pt buffer layer was deposited on a Si(001) substrate followed by a magnetic CoNi layer (thickness of around 10 nm) and covered with a 5 nm thick Pt cap layer. After deposition, the samples were lithographically patterned to produce one-dimensional structures that consisted of magnetic stripes. Two different periods, $5\ \mu\text{m}$ (the width of the stripes is equal to $4\ \mu\text{m}$) and $0.3\ \mu\text{m}$ (the width of the stripes is equal to $0.2\ \mu\text{m}$), were used.

Figure 1 shows hysteresis loops measured in different diffraction orders for the sample with $d=5\ \mu\text{m}$. In this configuration (a combination of angle of incidence, wavelength

FIG. 2. MOKE and MSHG hysteresis loops measured for different orders of diffraction for the sample with $d=0.3\ \mu\text{m}$.

and structure period) only first-order linear diffraction could be seen. The same direction corresponds to second-order MSHG diffraction. On the other hand, several diffracted orders were observed at negative angles. For the sample with period $d=0.3\ \mu\text{m}$ (see Fig. 2) only minus first-order MSHG diffraction could be observed at the fundamental wavelength of 810 nm. To measure the MOKE hysteresis loops in the same direction, a blue GaN laser with $\lambda=405\ \text{nm}$ was used.

An interesting feature of diffracted MSHG is the reversal of magnetic contrast with respect to that measured in the specular direction. This inversion, valid for both samples, did not appear in the linear MOKE. Note that contrast reversal was observed in an early study of MOKE diffraction,⁸ although no explanation was given.

The shape of the first-order diffraction hysteresis loops differs from all others by showing “overshoot” at magnetization reversal (see Fig. 1). This overshoot has already been observed before in the linear MOKE⁹ and can tentatively be explained by inhomogeneous reversal of the magnetization in the stripes. It is reasonable to suppose that, due to the lithography process, the edges of stripes have a higher density of defects and therefore larger coercive field H_c , due to the pinning. The total diffracted response can be described by a complex tensor reflectivity function $\hat{f}=(x;\alpha,\beta)$, where α and β are angles of incidence and diffraction, respectively. $\hat{f}$ is periodic in x with the structure period D . The net diffracted field can be expressed as

$$\mathbf{E}^{\text{diff}} \propto \int_0^D \hat{f}(x;\alpha,\beta) \mathbf{E}^{\text{in}} e^{i2\pi \cdot (\sin \alpha - \sin \beta)x/\lambda} dx. \quad (3)$$

If $\hat{f}=\text{const}$, $\mathbf{E}^{\text{diff}} \equiv 0$ (i.e., when there is no structure at all). This form suggests that features placed approximately half a period from each other contribute opposite signs to first-order diffraction. This can therefore explain why the magnetic response of the edges of our wide (80% of the period) lines contribute opposite to the response of the center part. Subtraction of the two loops with different coercive field would result in the observed shape. This increased coercive field has tentatively been confirmed for the $5\ \mu\text{m}$ sample by magneto-optical microscopy measurements.

FIG. 3. Dependence of the MSHG intensity for two opposite magnetic directions as a function of the angle of detection.

An interesting observation is that although the same overshoot is present in the MSHG loops as well, it is not symmetric. It can be shown that this asymmetry is due to the fact that the measured MSHG response contains both terms that are linear as well as terms that are quadratic in magnetization $\mathbf{M}$. The MSHG intensity as a function of magnetization $\mathbf{M}$ in terms of *odd* and *even* effective tensor components can be written as

$$\begin{aligned} I(2\omega, \mathbf{M}) &\propto |\chi^{\text{even}} + \chi^{\text{odd}}(\mathbf{M})|^2 \\ &= |\chi^{\text{even}}|^2 + |\chi^{\text{odd}}(\mathbf{M})|^2 \\ &\quad + |\chi^{\text{even}}| \cdot |\chi^{\text{odd}}(\mathbf{M})| \cos(\varphi), \end{aligned} \quad (4)$$

where φ is the relative difference in phase between χ^{even} and $\chi^{\text{odd}}(\mathbf{M})$. That is, when the odd contribution is not small with respect to the even contribution, the response is not linear in the magnetization. As can be seen, the MSHG loop becomes asymmetric when either the relative phase between the odd and even components is large or when their ratio is large. In fact the contribution quadratic in $\mathbf{M}$ becomes comparable to the linear term when the ratio $\chi^{\text{odd}}/[\chi^{\text{even}} \cos(\varphi)]$ is large. Thus both the relative phase between the odd and even components as well as their ratio may influence the hysteresis shape significantly.¹⁰

Figure 3 shows the dependence of the MSHG intensity for two opposite magnetic directions as a function of the angle of detection. Obviously, the MSHG signal is limited to narrow diffraction peaks. The background is practically equal to the dark current of the photomultiplier. This means the absence of any large-scale roughness that may cause strongly diffusive SHG.^{11,12}

It is seen in Fig. 3 (as well as in Fig. 1) that there is sizeable asymmetry between positive and negative diffraction peaks, especially visible in second order. While the average intensity of the second-order diffraction signal is five times larger than that of the minus second one, the magnetic

asymmetry is an order of magnitude smaller. This is explained by a change in the relative contribution of even and odd tensor elements. The MSHG intensity is given by

$$I(\pm \mathbf{M}) \propto |a\chi_{zzz} \pm b\chi_{xzz}|^2, \quad (5)$$

with a and b denoting the effective Fresnel factors. This leads to

$$I(\pm \mathbf{M}) \propto |\sin^2(\alpha)\sin(\beta)\chi_{zzz} + \cos^2(\alpha)\cos(\beta)\chi_{xzz}|^2, \quad (6)$$

where angles α and β are denoted in Fig. 1. Obviously, the increase of diffracting angle β increases the contribution from χ_{zzz} and decreases that from χ_{xzz} , resulting in a decrease of magnetic asymmetry. Given the fact that usually χ_{zzz} is the largest component, it also explains the increase in average intensity.

In conclusion, nonlinear magneto-optical diffraction was studied in one-dimensional arrays of magnetic stripes. Some interesting features of MSHG diffraction were found. First, there is sizeable asymmetry between the MSHG signal measured in positive and negative diffraction peaks, visible especially in second order. This can be simply explained by the influence of corresponding Fresnel factors. Next, the asymmetric shape of the hysteresis loops themselves originates from the very large amplitude of the magnetic signals. Finally, it was found that first-order diffraction hysteresis loops differ from all others by showing an overshoot at magnetization reversal, both in MOKE and MSHG. Tentatively we explain such behavior as due to inhomogeneous reversal of the magnetization in the stripes. It will be necessary to check these results in more detail by domain imaging these structures.

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