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A combined nonlinear and linear magneto-optical microscopy

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New possibilities for magnetic domain studies are demonstrated using a combination of nonlinear magneto-optical microscopy and a conventional linear polarizing microscope. The use of an optical response that is governed by a higher rank tensor offers sensitivity to additional combinations of magnetization directions and optical wave vector and polarization, which is demonstrated in magnetic garnet films of different crystallographic orientations. We observed a nontrivial modulated domain structure in a (210) film and a clear domain contrast for a (111) film, where the linear image only indicated simple up-down domains and no domain contrast for these two situations, respectively. © 1997 American Institute of Physics. [S0003-6951(97)01717-8]

Recently, magnetization induced optical second harmonic generation (MSHG) has been shown to be a very sensitive tool to probe magnetic surfaces and interfaces.¹⁻³ This nonlinear optical technique has great capabilities in studying magnetic properties of antiferromagnets,^{4,5} stratified metal structures and buried interfaces.⁶ Because the symmetry properties of the nonlinear interactions differ essentially from those in linear optics it would be interesting to visualize magnetic domain patterns by nonlinear magneto-optical microscopy, particularly for situations where the surface/interface magnetization is (expected to be) different from the bulk.

In this letter, nonlinear magneto-optical microscopy and its application to the visualization of domain structures in epitaxially grown magnetic garnet films is demonstrated. The proposed method allows images to be obtained both from the SH and linear response. This combination appears to be very powerful because the nonlinear and linear images contain complementary magnetic information. As all our experiments were done in transmission at normal incidence, only the magnetization component perpendicular to the film surface could be directly visualized with the fundamental light (via the linear magneto-optical Faraday effect), whereas in-plane components were probed by the second harmonic. In the nonlinear microscope the MSHG response was imaged using the very same setup, after filtering out the fundamental light. The linear polarization of the incoming light was rotated between 0° and 180° with respect to the sample symmetry plane m . In this way, subdomains that have different in-plane components, were distinguished. The observed MSHG contrast was correlated with the in-plane magnetization component using recent studies of MSHG rotational anisotropy.^{7,8}

The experimental set-up of our nonlinear magneto-optical microscope is schematically presented in Fig. 1. As a light source we used a Ti:sapphire laser operating at a repetition rate of 82 MHz with a pulse width of about 100 fs and at the wavelength of 775 nm. A half-wavelength plate was used to rotate the linear polarization of the incoming light. The laser beam was focused on the sample onto a spot of about 70 μm diameter. The average power of the pump

beam on the sample was 100 mW, resulting in a peak power of nearly 4 GW/cm². We magnified the exposed area by a $\times 40$ (N.A.=0.65) objective in combination with an achromatic concave lens. After appropriate filtering the generated second harmonic intensity was imaged with a cooled charge coupled device (CCD) camera. The subtraction of the gaussian-like background was applied afterwards to remove the spot-profile inhomogeneity in the image intensity due to the pump beam.

As test structures for our nonlinear magneto-optical microscope, differently oriented magnetic garnet films with thicknesses around 10 μm were probed. Garnet films are interesting subjects because of the fact that their structural, magnetic and magneto-optical properties may be widely varied by changing the film composition and the composition and orientation of the substrate. Magnetic garnet films were grown by a liquid phase epitaxial method, on (210) and (111) gadolinium gallium garnet (GGG) substrates. These substrates are centrosymmetric, nonmagnetic and transparent at the fundamental and SHG wavelengths. Because of a growth anisotropy and a lattice mismatch between the substrate and the magnetic film, the inversion symmetry in the thin garnet films is broken.⁹ The point group symmetry of these magnetic films results from the substrate orientation and is $3m$ (C_{3v}) for (111) and m (C_{1h}) in the case of (210).¹⁰ In these noncentrosymmetric structures both crystallographic and magnetization-induced bulk contributions to the nonlinear polarization are allowed.⁷ Therefore they can interfere in a magnetized sample or in a sample with a spontaneous magnetic (domain) structure: $I_{2\omega} \propto |\chi^{cr} \pm \chi^{mag}|^2$, where \pm depends on the direction of \mathbf{M} and thus changes from domain to domain. It is this interference term that will be responsible for the magnetic contrast.

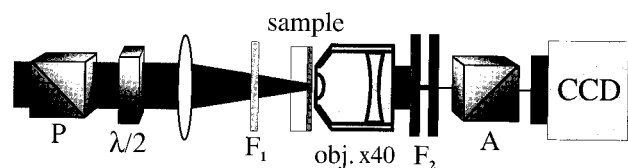


FIG. 1. Schematic microscopy setup.

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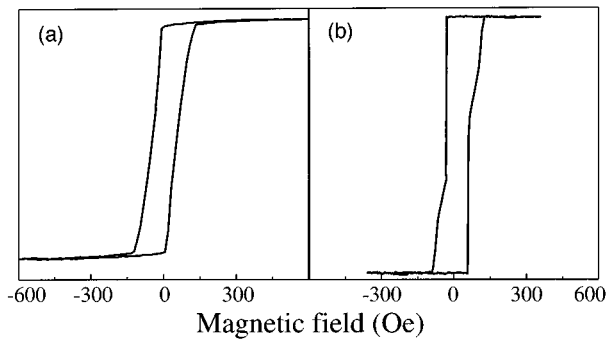


FIG. 2. Faraday hysteresis loops for (a) (210) oriented and (b) (111) oriented films.

Faraday hysteresis measurements (Fig. 2) show that both films have a remanence magnetization, but only in the (111)-film is the magnetization exactly perpendicular to the film surface. In the (210)-sample the magnetization is tilted at an angle of about 18° with respect to the film normal. This value was derived from the difference between the saturation and the remanence magnetization, taking into account that at remanence the sample is in a single-domain state, which is supported by direct imaging.

The domain pattern of the demagnetized films was initially tested using our setup as a linear Faraday microscope. Fig. 3(a) shows a typical labyrinth type domain structure for the (210) film where the dark/light areas indicate “up” and “down” domains.

Next, second harmonic images of this very same domain structure were taken, for various values of the incoming linear polarization with respect to the crystal symmetry plane m [Figs. 3(b)–(f)]. The SH images were recorded without analysing the outgoing light polarization. Remarkable changes in the magnetic contrast and in the SH intensity for the (210) garnet film were thus found. At 0° the SHG domain pattern appears to be exactly the same as in the linear light. To follow all subsequent changes appearing in the magnetic structure, the domain walls in this image are marked with dashed lines [Fig. 3(b)]. Rotating the incoming light polarization by 10° , a subdivision of the original domains was clearly observed [Fig. 3(c)]. This subdivision is even more sharp at larger angles and at 35° the SH intensity in the subdomains I and III becomes equal [Fig. 3(d)]. At 90° and at 145° the magnetic structure looks very similar to the cases 3(b) and 3(d) respectively, but the contrast appears to be shifted by half a domain width [Figs. 3(e), (f)]. To analyse the observed images, one should recall that in this configuration of normal incidence (and without polarization analysis), MSHG can only probe in-plane magnetization components, as follows from the MSHG selection rules.⁸ This means that different SH intensities correspond to different in-plane magnetizations. Therefore, from Fig. 3 we can conclude that four domain types appear to exist in the (210)-film ($\mathbf{M}_I \neq \mathbf{M}_{II} \neq \mathbf{M}_{III} \neq \mathbf{M}_{IV}$).

To analyse all recorded images (taken in steps of 5°) we started from the rotational anisotropy measurements (see Ref. 7), i.e. from the dependencies of the SH intensity on the azimuthal position of the sample with respect to the incoming light polarization and fixed (in-plane) magnetization. By

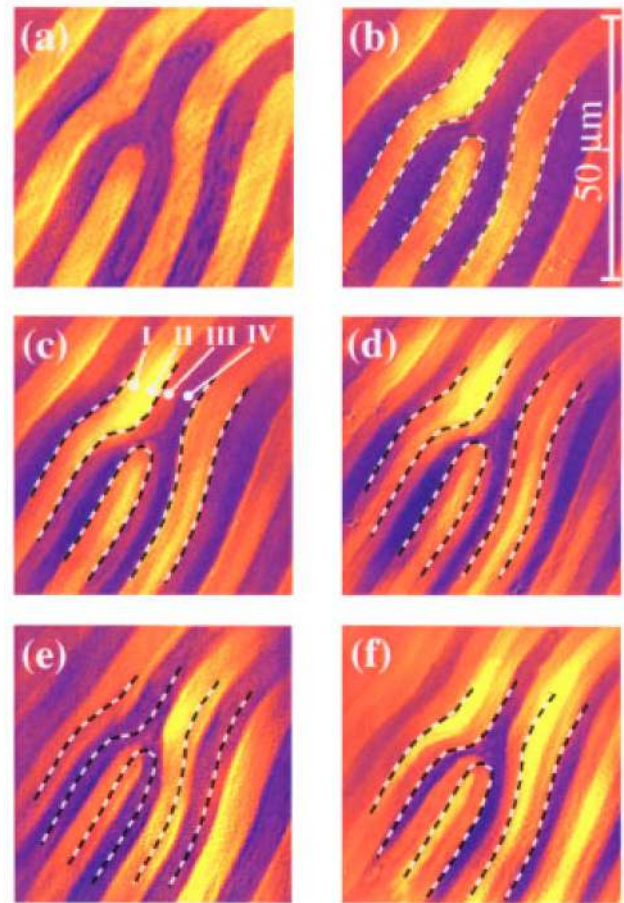


FIG. 3. (a) Linear and (b)–(f) second harmonic images of the magnetic domain structure in (210) oriented film. Input polarization was: (b) 0° ; (c) 10° ; (d) 35° ; (e) 90° ; (f) 145° with respect to the symmetry plane m .

a straightforward transformation, we obtain similar dependencies as a function of the incoming light polarization, with the in-plane \mathbf{M} fixed in the sample. It was then possible to estimate the in-plane magnetization in every subdomain. In total, an appropriate model of the (210) domain structure was derived (Fig. 4) with every “up” or “down” domain subdivided into two subdomains with different in-plane components. The in-plane magnetizations of the neighbouring domains are not collinear and the absolute value of the in-plane magnetization is the same in every subdomain. We should note that the observed image appeared to exist through the whole film, as was checked by changing the focusing depth, and can thus not be related to closure domains at the surface.

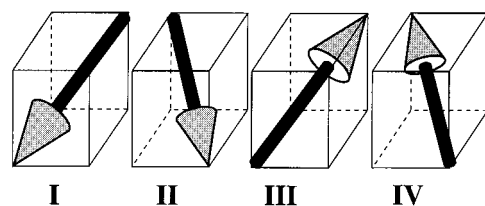


FIG. 4. Magnetization directions in four different domains in (210) film, as derived from the images of Fig. 3. Symmetry plane m coincides with the side-face plane.

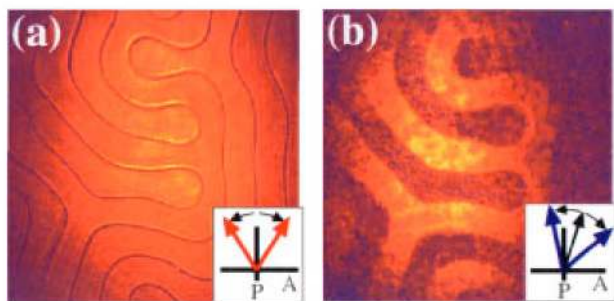


FIG. 5. Linear (a) and second harmonic (b) images of the magnetic domain structure in (111) oriented film obtained with crossed input and output polarizers.

Thus, a nontrivial modulated domain structure is shown to exist in the (210) oriented garnet film, which becomes only distinguishable using combined linear and nonlinear magneto-optical microscopy.

Similar measurements on the (111) film showed no magnetic contrast which means that no in-plane magnetization exists in the (111) sample. An analyser was then used to study the MSHG polarization rotation. To avoid an influence of the linear polarization rotation effect, a carefully crossed polarizer/analyzer configuration was used. Surprisingly, the outcoming SH signal showed a strong contrast in this case [Fig. 5(b)], though the linear picture showed no contrast between the domains at all [Fig. 5(a)]. Hence, a *different Faraday rotation value* for the SH light is found in the different domains. Indeed, the resulting SH polarization is a vector sum of crystallographic and magnetic contributions, the latter having different sign for the two opposite domains. As soon as the polarization of the crystallographic part is not equal to that of the fundamental light (except for certain high-symmetry directions), the final SH polarization states are not symmetric with respect to the incoming light polarization (see insets in Fig. 5). This configuration clearly demonstrates the difference in the mechanism responsible for the magneto-optical contrast in the two cases, and may be used to make a correlation between the domain structure and crystallographic axes. The contrast disappears when the incoming light polarization coincides with one of the sample symmetry planes.

In conclusion, we have demonstrated a new type of nonlinear magneto-optical microscopy that, in combination with standard linear microscopy, yields a wealth of additional and complementary information about magnetic domain structures. In particular, the higher rank tensor and a polarization analysis of the incoming fundamental and/or second harmonic light gives information about the magnetic structure that cannot be obtained in a single configuration with linear microscopy. It can also be shown that magnetization gradient near domain walls can give rise to additional contrast⁸. The sensitivity of MSHG to the breaking of crystallographic inversion symmetry also gives new possibilities for the observation of interface domain structures. This, together with the already demonstrated sensitivity for antiferromagnetic domains⁵, makes the further development of the nonlinear magneto-optical microscopy very promising.

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