Domain and domain wall contributions to optical second harmonic generation in thin magnetic films

I. L. Lyubchanskii  
Donetsk Physical-Technical Institute, National Academy of Sciences of Ukraine, 340114 Donetsk, Ukraine  

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

A symmetry analysis of nonlinear-optical imaging of domains and domain walls in thin magnetic films is presented. Different contributions to the second harmonic (SH) response depending on the local magnitude of the film magnetization $M$ as well as on its spatial derivatives are calculated. The polarization of the SH light is shown to be a sensitive function of the relative weight of different magnetization-induced contributions, the film symmetry, and the type of the domain wall. © 1997 American Institute of Physics. [S0021-8979(97)26508-4]

Recently, a new technique of nonlinear-optical domain imaging has been reported.\(^{1,2}\) This technique uses the magnetization-induced second harmonic generation (MSHG) and has several advantages in respect to the linear-optical tools. First of all, the nonlinear interactions giving rise to second-harmonic generation (SHG) have symmetry properties which differ essentially from those describing the linear-optical effects. In particular, SHG is known to be extremely sensitive to the presence of inversion symmetry which forbids the normally strongest electric dipole contribution to the SHG. For centrosymmetric media this symmetry is lifted at surfaces and interfaces, providing a high surface and interface sensitivity of MSHG.\(^{3,4}\) In the second place, the magneto-optical effects are typically much stronger in the nonlinear MSHG response relative to those in linear optics.\(^{3,5}\) Third, MSHG may be used to study ferromagnetic as well as antiferromagnetic domain structures.\(^{1}\)

In the present work we report results of a symmetry analysis of MSHG imaging of domain and domain walls in thin ferromagnetic films. We show that the inhomogeneous magnetization of the domain walls may provide an additional important source of MSHG via gradient terms which depend on the spatial derivatives of the magnetization.\(^{6,7}\) The analysis of the polarization of different MSHG sources is performed for both Bloch- and Néel-type domain walls in films having different symmetry.

Within the dipole approximation the light at the double frequency $2\omega$ is generated by the nonlinear polarization

$$P_i(2\omega) = \chi_{ijk} E_j(\omega) E_k(\omega),$$

(1)

where $E(\omega)$ is the electric field of incident fundamental wave at frequency $\omega$. For notation reasons, here we omit the frequency arguments and skip the usual superscript $(2)$ for the nonlinear susceptibility tensor $\chi_{ijk}^{(2)}(2\omega,\omega,\omega)$. For magnetic media the nonlinear optical susceptibility tensor $\chi_{ijk}$ may be presented as a sum of different terms

$$\chi_{ijk} = \chi_{ij}^{(0)} + \chi_{ijk}^{(1)} M_L + \chi_{ijklm}^{(2)} M_L M_M + \chi_{ijklm}^{(3)} \nabla_i M_M + \chi_{ijklm}^{(4)} M_L \nabla_j M_M \nabla_k M_N + \cdots,$$

(2)

where $\chi^{(0)}$ is the nonmagnetic part of $\chi$ while $\chi^{(1)}$ and $\chi^{(2)}$ describe the effect of the local magnetic order. Capital letters are used to denote the indices of the axial magnetization vector $M$. In Eq. (2) we also introduce gradient terms $\chi^{(3)}$ and $\chi^{(4)}$ which are nonvanishing in the presence of a nonuniform magnetization. Similar gradient terms were introduced into the theory of linear-optical domain imaging.\(^{8}\) We note that all tensors with an odd number of polar (small) indices vanish for centrosymmetric media. In that case only the gradient terms ($\chi^{(3)}$ and $\chi^{(4)}$) contribute to the nonlinear source $P(2\omega)$.

Below we particular focus on the nonlinear-optical properties of different thin magnetic garnets. The theoretical consideration is however more general since it is based only on symmetry arguments and therefore can be applied to other magnetic systems with the same symmetry. The bulk of a perfect garnet crystal is cubic and centrosymmetric. In thin films grown on a not perfectly matching substrate, however, the inversion symmetry is lifted via a distortion of the lattice during the film growth\(^{9}\) so that all terms on the right-hand side of Eq. (2) are symmetry-allowed. On the other hand, the lattice distortion is assumed to be weak so that the lattice is close to the centrosymmetric arrangement in the perfect crystal. This assumption is essential for an experimental detection of the gradient effects on MSHG. One obviously expects that in most cases the gradient terms in Eq. (2) are relatively small corrections to the leading nonmagnetic $\chi^{(0)}$ and local $\chi^{(1)}, \chi^{(2)}$ magnetic terms. In a thin garnet film with a nearly-centrosymmetric lattice, however, the importance of these terms may be reduced so that the relative weight of the gradient terms ($\chi^{(3)}$ and $\chi^{(4)}$) is enhanced. We also note that the garnets possess a complicated magnetic structure with multiple sublattices. Some symmetry operations should therefore be combined with lattice translations by a vector smaller than the size of the unit cell. Here we do not consider the effect of these translations assuming that the unit cell size is much smaller than the wavelength and the wall thickness.

A coordinate system is introduced with the $z$ axis being normal to the film, and the $x$ and $y$ axes lying in the film plane. We assume that there is at least one symmetry reflection plane normal to the (nonmagnetic) film which coincides with the $y=0$ plane. The consideration is performed for films with an even-fold rotation symmetry as well as with an odd-
As can be seen from Table I, there is no SHG light generated in the domain wall where a parallel magnetization is present. In the Bloch wall A, e.g., the nonlinear polarization can be written as

$$P_y^{(1)}(2\omega) = \alpha_{xyzy}^{(1)} E_y^{(2)}(\omega) M_x,$$

(3)

For the Néel wall A, on the other hand, the MSHG response arising from the second term of Eq. (2) is polarized along the y axis.

In a similar way, one has no MSHG from the third term (\(\alpha_{xyz}^{(2)}\)) in the domains. The only nonvanishing contribution of, e.g., the Bloch domain wall A and the Néel wall B is given by

$$P_y^{(2)}(2\omega) = \alpha_{xyz}^{(2)} E_y^{(2)}(\omega) M_x M_z.$$

(4)

We note, however, that \(M_z M_z\) is an odd function of the coordinate along the wall normal. Since the "brightness" of the wall image is proportional to the polarization \(P_y^{(2)}(2\omega)_{dy \times dy}\), that is averaged over the resolution length \(d_y\) of the optical objective, the nonlinear source (4) generated in a thin wall (much thinner than \(d_y\)) does not contribute to the image. Taking the next term of Eq. (2) into account, one has a contribution to MSHG in transmission at normal incidence

$$P_y^{(3)}(2\omega) = \alpha_{xyz}^{(3)} E_y^{(2)}(\omega) \nabla_x M_z$$

(5)

for both Bloch and Néel domain walls along y (A walls). In B walls this contribution is along x. The MSHG source (5) is therefore always polarized along the wall and is independent of the type of the wall. The last term of Eq. (2) can produce a contribution via terms containing \(M_z \nabla_x M_x\) and therefore vanishes after integration over the domain wall. The results of the present analysis are summarized in Table I. For the fundamental light polarized along the y axis we find exactly the same polarizations for the different contributions to the MSHG image.

As can be seen from Table I, there is no SHG light generated within the domains in the even rotation symmetry film. Also, only y-polarized MSHG can be generated in the Néel wall if the fundamental beam is polarized along x or y axes. Therefore, if the MSHG light is not purely polarized along y, the wall is of the Bloch type. Moreover, the relative weight of the \(\chi^{(1)}\) and \(\chi^{(3)}\) contributions can then be found from the polarization properties of the MSHG light generated by the Bloch domain wall.

Now we analyze the odd-fold rotation symmetry films. The (nonmagnetic) film has a symmetry plane normal to the y axis while the \(x \rightarrow -x\) is not a symmetry operation. The magnetization within the domain is again assumed to be

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**TABLE I. Polarization of the MSHG wave generated in an even rotation symmetry film via different terms of Eq. (2).** The fundamental beam is polarized along either x or y directions. We also explicitly show how these contributions are related to different components of the film magnetization.

<table>
<thead>
<tr>
<th>Term</th>
<th>Domain 1</th>
<th>Domain 2</th>
<th>Wall A</th>
<th>Wall B</th>
<th>Wall A</th>
<th>Wall B</th>
</tr>
</thead>
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<tr>
<td>n</td>
<td>(M_{xx})</td>
<td>(M_{yz})</td>
<td>(M_{xx})</td>
<td>(M_{yz})</td>
<td>(M_{xx})</td>
<td>(M_{yz})</td>
</tr>
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<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>1</td>
<td>(P_y \times M_z)</td>
<td>(P_y \times M_z)</td>
<td>(P_y \times M_z)</td>
<td>(P_y \times M_z)</td>
<td>(P_y \times M_z)</td>
<td>(P_y \times M_z)</td>
</tr>
<tr>
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<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>3</td>
<td>(P_y \times \nabla_x M_z)</td>
<td>(P_y \times \nabla_x M_z)</td>
<td>(P_y \times \nabla_x M_z)</td>
<td>(P_y \times \nabla_x M_z)</td>
<td>(P_y \times \nabla_x M_z)</td>
<td>(P_y \times \nabla_x M_z)</td>
</tr>
<tr>
<td>4</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
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</table>
TABLE II. Polarization of MSHG generated in an odd rotation symmetry garnet film via different terms \( \chi^{(n)} \) of Eq. (2).

<table>
<thead>
<tr>
<th>( n )</th>
<th>Domain</th>
<th>Bloch wall A</th>
<th>Bloch wall B</th>
<th>Néel wall A</th>
<th>Néel wall B</th>
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</thead>
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<tr>
<td>0</td>
<td>( P_x )</td>
<td>( P_x )</td>
<td>( P_x )</td>
<td>( P_x )</td>
<td>( P_x )</td>
</tr>
<tr>
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<td>( P_x \alpha M_z )</td>
<td>( P_x \alpha M_y )</td>
<td>( P_x \alpha M_x )</td>
<td>( P_x \alpha M_y )</td>
<td>( P_x \alpha M_y )</td>
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<tr>
<td>2</td>
<td>( P_x \alpha M_z )</td>
<td>( P_x \alpha M_y )</td>
<td>( P_x \alpha M_x )</td>
<td>( P_x \alpha M_y )</td>
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<td>( P_x \alpha M_z )</td>
<td>( P_x \alpha M_y )</td>
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<td>( P_x \alpha M_y )</td>
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<td>( P_x \alpha M_y )</td>
<td>( P_x \alpha M_y )</td>
</tr>
</tbody>
</table>

along the film normal \( z \). Following the same arguments as were used above, one can find the symmetry-allowed contributions to the SHG wave generated in transmission by a fundamental wave at normal incidence. The results are collected in Table II.

The lowering of the film symmetry is seen to result in a much larger number of nonvanishing contributions. The MSHG image of the magnetic domain structure is therefore expected to be more complex. One can nevertheless find some simple rules which may be used to analyze the image. For example, the polarization of the SHG response from the domains possesses information about the relative importance of the local terms that are even \( \chi^{(0)} \) or odd \( \chi^{(1)} \) in the magnetization. Then, the Néel wall B (parallel to the symmetry plane \( y=0 \)) should look like a "dark" line if only the \( y \)-polarized MSHG is recorded. The "brightness" of the Néel wall A (relative to the "brightness" of the domains) then brings information about the relative importance of the terms linear in the magnetization \( \chi^{(1)} \) and \( \chi^{(3)} \). Similarly, if the domain walls are of the Bloch type, the relative "brightness" of the wall A in \( y \)-polarized MSHG light tells us the relative weight of the two gradient terms \( \chi^{(3)} \) and \( \chi^{(4)} \), etc.

In conclusion, a symmetry analysis of MSHG images of the domain structures of thin magnetic films is performed. Four different magnetization-induced contributions are taken into account. Two of them (terms \( \chi^{(1)} \) and \( \chi^{(2)} \)) depend on the local value of the magnetization and contribute to the MSHG response from domains and domain walls. We also introduce gradient terms \( \chi^{(3)} \) and \( \chi^{(4)} \) which are proportional to the spatial derivatives of the magnetization. The latter contributions are the only dipole-allowed source of MSHG in centrosymmetric media and may produce an important contribution to the MSHG image of domain walls in crystals where the inversion symmetry is lifted. We study the polarization rules for the MSHG images of odd- and even-fold rotation symmetry thin films measured in transmission at normal incidence. It is shown that MSHG imaging of the domain structure with the use of different polarizations may bring information about the type of the domain wall and the relative weight of different contributions to the MSHG response.

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