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Theory of nonlinear magneto-optical imaging of magnetic domains and domain walls

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A symmetry analysis of nonlinear magneto-optical imaging of magnetic domains and domain walls is presented. We introduce gradient terms giving rise to the magnetization-induced second-harmonic generation (MSHG) via spatial derivatives of the magnetization. The nonvanishing independent elements of the relevant tensors are derived for cubic media. Different contributions to the MSHG image from domains and domain walls are analyzed for thin magnetic films with different symmetry. It is shown that measurements of polarization properties of the MSHG response may yield information about the relative importance of different magnetization-induced contributions and also the type of domain walls. [S0163-1829(97)03529-7]

I. INTRODUCTION

The properties of magnetic films are strongly affected by their domain structure and the structure of the domain walls.[1,2] To visualize magnetic domains optical techniques are often used. Traditionally, one uses the linear magneto-optical effects: the Faraday and Kerr rotation of linearly polarized light. Recently, a technique of nonlinear-optical domain imaging has been reported,[3,4] that uses magnetization-induced second-harmonic generation (MSHG) and has several advantages with 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 SHG. For centrosymmetric media this symmetry is lifted at surfaces and interfaces, providing a high surface and interface sensitivity of MSHG.[5,6] In the second place, the nonlinear magneto-optical effects are typically much stronger than the linear ones[5,7] (rotations close to 90° have been reported and intensity changes of near 100%). Thirdly, MSHG may be used to study ferromagnetic as well as antiferromagnetic domain structures.[3,8]

Literature up to now has mainly discussed MSHG in uniformly magnetized (single domain) bulk media, thin films, and surfaces.[8–13] In this work we consider the more general case of MSHG in magnetic structures with domains and domain walls. The spatial derivatives of the magnetization near domain walls are shown to yield additional sources of the nonlinear-optical response.[9,14] For a centrosymmetric medium these gradient terms give the only nonvanishing dipole-allowed contribution to MSHG. We derive the nonvanishing independent elements of the relevant tensors for cubic centrosymmetric media. As an example, we study the MSHG response in transmission of a thin magnetic garnet film at normal incidence. Four different magnetization-induced contributions (local and gradient, both linear and bilinear in the magnetization) to the MSHG images are taken into account. The analysis is performed for both Bloch- and Néel-type domain walls in films having different symmetry. We demonstrate that the relative importance of different magnetization-induced contributions may be obtained from an analysis of the polarization properties of the MSHG response. The type of the domain wall can be also determined in such a study.

II. NONLINEAR SOURCES IN MAGNETIC DOMAINS AND DOMAIN WALLS

The dielectric response of magnetic media is known to be a function of their magnetization **M**. The dielectric constant can be written as

$$\epsilon_{ij}(\mathbf{M})=\epsilon_{ij}^{(0)}+f_{ijK}M_K+g_{ijKL}M_KM_L, \tag{1}$$

where $\epsilon_{ij}^{(0)}$ is the part of $\epsilon_{ij}(\mathbf{M})$ independent of the magnetic subsystem, and f_{ijK} and g_{ijKL} are linear and bilinear magneto-optical tensors, which describe the Faraday (Kerr) effect and magnetic birefringence, respectively.[15] In Eq. (1) we use small letters to denote the indices of polar vectors and capital letters to denote the indices of the axial vector **M**. The higher-order magnetization-induced terms are normally much less important. In the presence of magnetic inhomogeneities like domain walls, for example, one has to take into account the spatial derivatives of the magnetization and Eq. (1) should be rewritten as

$$\epsilon_{ij}(\mathbf{M})=\epsilon_{ij}^{(0)}+f_{ijK}M_K+g_{ijKL}M_KM_L+F_{ijkL}\nabla_kM_L + G_{ijKlM}M_K\nabla_lM_M+\cdots. \tag{2}$$

The F_{ijkL} and G_{ijKlM} terms give rise to so-called gradient effects[16] in the linear magneto-optical response.

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TABLE I. Independent nonzero elements of the $\chi^{(3)}_{ijklM}(-\omega_1-\omega_2;\omega_1,\omega_2)$ tensor describing the three-wave mixing in a cubic centrosymmetric medium. The elements are denoted as $ijklM$ for compactness.

$\chi_1^{(3)}$	$zyyyX=xzzzY=yxxxZ=-yzzzX=-zxxxY=-xyyyZ$
$\chi_2^{(3)}$	$yzyyX=zxzzY=xyxxZ=-zyzzX=-xzxxY=-yxyyZ$
$\chi_3^{(3)}$	$yyzyX=zzxzY=xxyxZ=-zzyzX=-xxzxY=-yyxyZ$
$\chi_4^{(3)}$	$yyyzX=zzzxY=xxxyZ=-zzzyX=-xxxzY=-yyyxZ$
$\chi_5^{(3)}$	$yzxxX=zxyyY=xyzzZ=-zyxxX=-xzyyY=-yxzzZ$
$\chi_6^{(3)}$	$yxzxX=zyxyY=xzyzZ=-zxyxX=-xyzyY=-yzxzZ$
$\chi_7^{(3)}$	$xyzxX=yzxyY=zxyzZ=-xzyxX=-yxzyY=-zyxzZ$
$\chi_8^{(3)}$	$yxxzX=zyyxY=xzzyZ=-zxxyX=-xyyzY=-yzzxZ$
$\chi_9^{(3)}$	$xyxzX=yzyxY=zxzyZ=-xzxyX=-yxyzY=-zyzxZ$
$\chi_{10}^{(3)}$	$xxyzX=yyzxY=zzxyZ=-xxzyX=-yyxzY=-zzyxZ$

The second-order optical response can be treated in a similar way. Within the dipole approximation the light at the double frequency 2ω is generated by the nonlinear polarization

$$P_i(2\omega)=\chi^{(2)}_{ijk}(-2\omega;\omega,\omega)E_j(\omega)E_k(\omega), \qquad (3)$$

where $\mathbf{E}(\omega)$ is the electric field of incident fundamental wave at frequency ω. For notation reasons, we will omit the superscript and frequency arguments for the nonlinear optical susceptibility tensor $\chi^{(2)}_{ijk}$ in the following. Similar to Eq. (2), for a magnetic system χ_{ijk} may be presented as a sum of different terms

$$\chi_{ijk}(\mathbf{M})=\chi^{(0)}_{ijk}+\chi^{(1)}_{ijkL}M_L+\chi^{(2)}_{ijkLM}M_LM_M+\chi^{(3)}_{ijklM}\nabla_lM_M + \chi^{(4)}_{ijkLmN}M_L\nabla_mM_N+\chi^{(5)}_{ijklMmK}\nabla_lM_M\nabla_nM_K + \cdots, \qquad (4)$$

where $\tilde{\chi}^{(0)}$ is the nonmagnetic part of $\tilde{\chi}$ while $\tilde{\chi}^{(1)}$ and $\tilde{\chi}^{(2)}$ describe the effect of the *local* magnetic order. $\tilde{\chi}^{(3)}$, $\tilde{\chi}^{(4)}$, and $\tilde{\chi}^{(5)}$ are gradient terms which are nonvanishing in the presence of a nonuniform magnetization. Equations (2) and (4) can be derived from the expression for the free energy (Ginzburg-Landau functional[17–20]) of a ferromagnet subjected to an external electromagnetic field if the spatial derivative of the ferromagnetic order parameter is taken into account.

We note that all tensors with an odd number of polar (small) indices vanish for centrosymmetric media. In that case only the linear-gradient terms ($\propto\tilde{\chi}^{(3)}$ and $\tilde{\chi}^{(4)}$) contribute to the nonlinear source $\mathbf{P}(2\omega)$. In contrast, the linear-gradient terms F_{ijkL} and G_{ijKlM} describing the linear-optical response vanish in the presence of inversion symmetry whereas those dependent on the local value of the magnetization [see Eq. (2)] are symmetry allowed.

A symmetry analysis of the different terms on the right-hand side of Eq. (4) is given below. As an example we will discuss the nonlinear-optical properties of magnetic garnets and garnet films of different symmetry. 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. In Sec. III we start with the nonvanishing terms in Eq. (4) ($\propto\tilde{\chi}^{(3)}$ and $\tilde{\chi}^{(4)}$) for cubic centrosymmetric media. In Secs. IV and V the analysis of MSHG for even- and odd-fold rotation symmetry garnet *films* is presented. For them we assume that the inversion symmetry of a bulk garnet crystal is lifted. This assumption is based on the experimentally observed *bulk* SHG response from the garnet films in transmission.[21] On the other hand, the lattice distortion is assumed to be weak so that the lattice is close to the centrosymmetric arrangement in the undistorted crystal. This assumption is essential for an experimental detection of the magnetization-gradient effects on MSHG. One obviously expects that the gradient terms in Eq. (4) are relatively small corrections to the leading nonmagnetic $\tilde{\chi}^{(0)}$ and local magnetic $\tilde{\chi}^{(1)}$ and $\tilde{\chi}^{(2)}$ terms. In a thin garnet film with a ''nearly-centrosymmetric'' lattice, however, the importance of these terms can be reduced so that the relative weight of the linear gradient terms ($\tilde{\chi}^{(3)}$ and $\tilde{\chi}^{(4)}$) is enhanced. We also note that terms involving second derivatives of the magnetization (like the $\tilde{\chi}^{(5)}$ term) vanish for centrosymmetric media so that their weight is presumably low in the MSHG response of garnet films. Therefore, such terms are not considered below.

III. SYMMETRY PROPERTIES OF NONLINEAR MAGNETO-OPTICAL TENSORS IN CUBIC CENTROSYMMETRIC MEDIA

As mentioned above, only the $\tilde{\chi}^{(3)}$ and $\tilde{\chi}^{(4)}$ tensors of Eq. (4) are nonvanishing in a centrosymmetric medium. We introduce the crystallographic coordinate system xyz with the axes along the (100), (010), and (001) directions. For the symmetry class O_h (describing the undistorted garnet crystal[22]) it is sufficient to consider the following symmetry operations: three mirror reflection with respect to the $x=0$, $y=0$, and $z=0$ planes and three rotations by 90° around the principal coordinate axes. Under rotations the polar and axial vectors are changed in the same manner whereas the reflection symmetries should be combined with the time inversion giving rise to an extra change of the sign of the axial vector $\mathbf{M}$. For example, under reflection with respect to one of the mirror planes the normal component of a polar vector changes its sign whereas that of an axial vector is not affected. In contrast, components of a polar vector parallel to the symmetry plane remain the same whereas those of an axial vector change their sign. Along this line, one finds that in general there are 60 nonvanishing elements (10 of them are independent) of a tensor with four polar and

TABLE II. Independent nonzero elements of the $\chi^{(4)}_{ijkLmN}(-2\omega;\omega,\omega)$ tensor. It is assumed that $i\neq j\neq k\neq i$.

$iiiIiI$, $iiiIjJ$, $iiiJiJ$, $iijIiJ=ijiIiJ$, $jiiIiJ$, $iiiJjI$,
$iijIjI=ijiIjI$, $jiiIjI$, $iijJiI=ijiJiI$, $jiiJiI$, $ijjIiI$, $jijIiI=jjiIiI$,
$iijJkK=ijiJkK$, $iijKjK=ijiKjK$, $iikJjK=ikiJjK$, $ijjIkK$,
$ijkIjK=ikjIjK$, $ijjKiK$, $ijkJiK=ikjJiK$, $ijjKkI$, $ijkJkI=ikjJkI$

one axial indices which are displayed in Table I. Such a tensor $\chi^{(3)}_{ijklM}(-\omega_1-\omega_2;\omega_1,\omega_2)$ is relevant, for example, for sum frequency generation[23] in the presence of two different fundamental waves. However, in the case of second-harmonic generation (degenerate three-wave mixing), there is an additional relation between different elements of $\chi^{(3)}(-2\omega;\omega,\omega)$ which is coming from the permutation symmetry of the incoming fields

$$\chi^{(3)}_{ijklM}=\chi^{(3)}_{ikjlM}, \tag{5}$$

which leads to

$$\chi^{(3)}_2=\chi^{(3)}_3,\quad \chi^{(3)}_5=\chi^{(3)}_6,$$

$$\chi^{(3)}_7=0,\quad \chi^{(3)}_9=\chi^{(3)}_{10}. \tag{6}$$

Therefore, the number of nonzero elements of $\tilde{\chi}^{(3)}$ is reduced to 54 and the number of independent elements is reduced to 6. We also note that the number of independent elements might be further reduced to 3 if the dispersion of $\tilde{\chi}^{(3)}$ is small and can be neglected (the so-called Kleinman's conjecture[24]). This condition is however not fulfilled for magnetic garnets and the Kleinman's conjecture is not used in the present work.

Along the same line, considering the symmetry operations together with the permutation symmetry of the incoming fields

$$\chi^{(4)}_{ijkLmN}=\chi^{(4)}_{ikjLmN}, \tag{7}$$

we find that there are 183 nonzero (21 independent) elements of the tensor $\chi^{(4)}_{ijkLmN}$, which have an even number of all three crystallographic indices as is illustrated in Table II. Any combination of x (X), y (Y), and z (Z) can be substituted instead of i (I), j (J), and k (K). For example, the second element of Table II gives

$$xxxXyY=xxxXzZ=yyyYxX=yyyYzZ=zzzZxX=zzzZyY,$$

etc.

IV. NONLINEAR MAGNETO-OPTICAL IMAGING OF GARNET FILMS WITH EVEN-FOLD ROTATION SYMMETRY

In the present section we consider even-fold rotation symmetry magnetic garnet films. We introduce a laboratory coordinate system with the z axis along the normal to the film, and the x and y axes being parallel to the film plane. Following Ref. 21, we assume that the inversion symmetry of the film is lifted due to a small lattice distortion as a result of the film growth. The $x\rightarrow -x$ and $y\rightarrow -y$ mirror reflections are nevertheless assumed to be symmetry elements of the film lattice (without taking the magnetic subsystem into account). The film therefore has also even-fold rotation symmetry. Such a symmetry is expected for magnetic garnet films grown on (001) and (110) nonmagnetic substrates[21] (C_{2v} and C_{4v}, respectively). For such a film the nonvanishing independent elements of the nonmagnetic $\tilde{\chi}^{(0)}$ and local magnetic $\tilde{\chi}^{(1)}$ tensor are derived before.[10,24] One can easily extend the symmetry analysis to find the nonzero elements of the $\tilde{\chi}^{(2)}$ tensor. Table III summarizes the results for films with C_{2v} and C_{4v} symmetry. For compactness, among the elements that are related via permutation symmetries

$$\chi^{(0)}_{ijk}=\chi^{(0)}_{ikj},$$

$$\chi^{(1)}_{ijkL}=\chi^{(1)}_{ikjL},$$

$$\chi^{(2)}_{ijkLM}=\chi^{(2)}_{ikjLM}=\chi^{(2)}_{ijkML}=\chi^{(2)}_{ikjML}, \tag{8}$$

only one of them is displayed.

The nonvanishing elements of the magnetic gradient tensors $\tilde{\chi}^{(3)}$ and $\tilde{\chi}^{(4)}$ are the same as those given in Tables I and II, while some of the relations between them are broken so that the number of independent elements is larger than that in

TABLE III. Nonvanishing elements of the $\chi^{(0)}_{ijk}$, $\chi^{(1)}_{ijkL}$, and $\chi^{(2)}_{ijkLM}$ tensors in films with C_{4v} and C_{2v} symmetry. The equality signs in brackets (=) do not hold under the C_{2v} symmetry. The element $\chi^{(1)}_{zxyZ}$ taken in square brackets vanishes under the C_{4v} symmetry. Only one of the elements related via permutation symmetry (8) is shown.

$\chi^{(0)}$	$zzz, zxx\ (=)\ zyy, xxz\ (=)\ yyz$
$\chi^{(1)}$	$xyzZ\ (=)\ -yxzZ, [\ zxyZ\ ], xxxY\ (=)\ -yyyX,$
$\chi^{(2)}$	$xyyY\ (=)\ -yxxX, yxyY\ (=)\ -xyxX, xzzY\ (=)\ -yzzX, zxzY\ (=)\ -zyzX$ $zzzZZ, zzzXX\ (=)\ zzzYY, zxxZZ\ (=)\ zyyZZ, zxxXX\ (=)\ zyyYY,$ $zxxYY\ (=)\ zyyXX, xxzZZ\ (=)\ yyzZZ, xxzXX\ (=)\ yyzYY, xxzYY\ (=)\ yyzXX$ $zxyXY, xyzXY\ (=)\ yxzXY, zzxXZ\ (=)\ zzyYZ, xzzXZ\ (=)\ yzzYZ,$ $yyxXZ\ (=)\ xxyYZ, xyyXZ\ (=)\ yxxYZ, xxxXZ\ (=)\ yyyYZ$

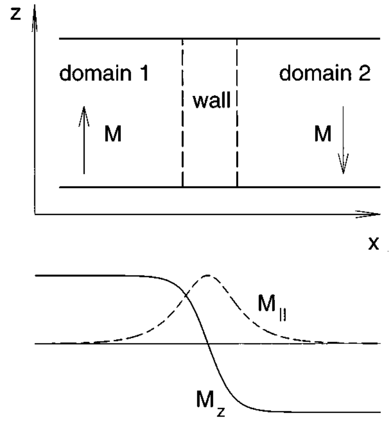

FIG. 1. Schematic side view of the thin magnetic film considered in Secs. IV and V. The lower panel schematically shows variations of the normal M_Z and parallel $M_{\|}$ components of the magnetization. In the Bloch-type wall $M_{\|}$ is along y (i.e., the magnetization **M** rotates out of the xz plane) whereas for the Néel wall it is along x (i.e., **M** rotates within the xz plane).

a perfect cubic crystal. As an example, we consider the nonvanishing elements of the $\overleftrightarrow{\chi}^{(3)}$ tensor that are given in the first line of Table I. Among them there are three independent elements $zyyyX=-zxxxY$, $xzzzY=-yzzzX$, and $yxxxZ=-xyyyZ$, for the films with the C_{4v} symmetry. For the C_{2v} symmetry films all six elements are independent.

We consider the particular case of two magnetic domains and the vertical (parallel to the film normal z) domain wall between them (Fig. 1). We assume that the domain wall is normal to the x axis (which is special because the $x=0$ and $y=0$ planes are the mirror planes). Furthermore, we assume that within the domains the magnetization vector **M** is along z (but antiparallel for two neighboring domains) because of the growth-induced magnetic anisotropy. In the domain wall, however, a parallel component of **M** is present. In the case of a Bloch wall this component is parallel to the wall (along the y axis) whereas for a Néel wall it is normal to the wall (along x). We also assume that the thickness of the domain wall is much smaller than the optical wavelength λ whereas the size of the domains is much larger than λ.

Now we analyze MSHG in transmission through such a thin garnet film. The fundamental beam is assumed to be incident along the normal to the film surface and purely polarized along either the x or y direction. The nonlinear polarization $\mathbf{P}(2\omega)$ along z does not radiate into the z direction, so only the components along x and y generate SHG light. The (linear-optical) Faraday rotation of the polarization of the fundamental and SHG light within the thin magnetic film is assumed to be small and will be neglected.

Because of the in-plane inversion symmetry, we find that the nonmagnetic part of the nonlinear optical susceptibility (described by the $\overleftrightarrow{\chi}^{(0)}$ term) does not contribute to the SHG response in either the domains or the domain wall. The contribution of the term linear in the magnetization ($\propto\overleftrightarrow{\chi}^{(1)}$) of Eq. (4) also vanishes in the domains but gives a finite contribution in the domain wall where a parallel magnetization is present (cf. Table III). For a Bloch wall the parallel components of the nonlinear polarization $\mathbf{P}^{(1)}(2\omega)$ arising via the $\overleftrightarrow{\chi}^{(1)}$ term can be written as

$$P_x^{(1)}(2\omega)=\chi^{(1)}_{xxxY}E_x^2(\omega)M_Y+\chi^{(1)}_{xyyY}E_y^2(\omega)M_Y,$$

$$P_y^{(1)}(2\omega)=2\chi^{(1)}_{yxyY}E_x(\omega)E_y(\omega)M_Y. \qquad (9)$$

For a purely x or y polarized fundamental beam the MSHG light generated via the $\overleftrightarrow{\chi}^{(1)}$ term is solely polarized along x. For a Néel wall, where the tangential magnetization is along x, the MSHG response arising from the $\overleftrightarrow{\chi}^{(1)}$ term is polarized along the y axis.

In a similar way, one has no MSHG from the third term ($\propto\overleftrightarrow{\chi}^{(2)}$) in the domains. For the x- or y-polarized fundamental beam the only nonvanishing contribution of the Bloch domain wall is given by

$$P_y^{(2)}(2\omega)=\chi^{(2)}_{yiiYZ}E_i^2(\omega)M_YM_Z, \quad i=x,y. \qquad (10)$$

We note, however, that $M_Y(x)M_Z(x)$ is an odd function of the coordinate x along the wall normal. Realizing that the thickness of domain walls is typically much smaller than the wavelength of the SHG light, one cannot resolve the nonlinear-optical response from the opposite sides of the domain wall. Therefore, the light generated by the polarization (10) in the left-hand side of the domain wall interferes destructively with that generated in the right-hand side. As a result, the nonlinear polarization (10) does not contribute to the MSHG image of the wall.

Inspecting Table I, one finds a contribution of the $\overleftrightarrow{\chi}^{(3)}$ term to MSHG in transmission at normal incidence

$$P_y^{(3)}(2\omega)=\chi^{(3)}_{yiixZ}E_i^2(\omega)\nabla_xM_Z, \quad i=x,y \qquad (11)$$

for both Bloch and Néel domain walls. The contribution arising via the $\overleftrightarrow{\chi}^{(3)}$ term is therefore not sensitive to the type of the domain wall. From Table II we find that the $\overleftrightarrow{\chi}^{(4)}$ term of Eq. (4) gives rise to the nonlinear polarization

$$P_x^{(4)}(2\omega)=\chi^{(4)}_{xiiJxJ}E_i^2(\omega)M_J\nabla_xM_J, \quad i=x,y; \quad J=X,Y,Z. \qquad (12)$$

However, $M_J\nabla_xM_J$ is an odd function of the coordinate x along the normal to the wall and therefore the contribution of $P^{(4)}(2\omega)$ vanishes after integration over the domain wall.

A similar analysis can be performed for a fragment of the wall which is normal to the y axis. For compactness, in the following we denote such a wall fragment by B whereas a part of the wall considered above (normal to x) is denoted as A (Fig. 2). In magnetic films with the labyrinth domain structure one can always find such fragments. Moreover, using a small external magnetic field one can also achieve a stripe domain structure where all the walls are parallel to a given direction. In this case one can prepare the domain structure where all the walls are either A or B walls. Because of the even-fold rotation symmetry of the film, for a wall B we find exactly the same nonvanishing contributions to the MSHG image. Since in a wall B the parallel component of the magnetization $\mathbf{M}_{\|}$ and the direction of the spatial derivative of the magnetization ∇M_Z are rotated by 90° with respect to those in a wall A, the nonlinear polarization $\mathbf{P}(2\omega)$ is also rotated by 90°. The results of the present analysis are summarized in Table IV. Using these results we also generated the expected images of Bloch and Néel walls A and B which are shown in Fig. 3. The MSHG light along the y (vertical)

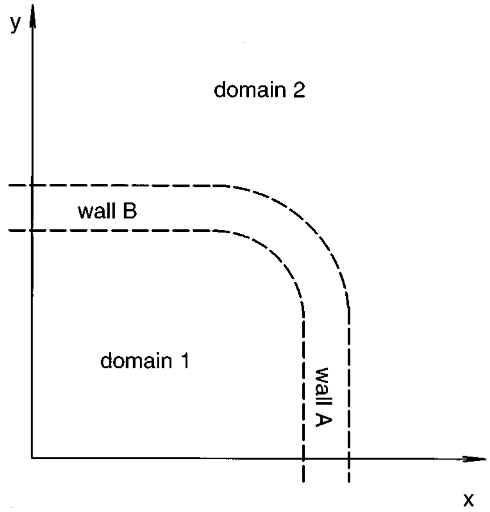

FIG. 2. Sketch of the system studied in Secs. IV and V. The $y=0$ plane coincides with the mirror symmetry plane of the film lattice. The $x=0$ plane is also the mirror symmetry plane for an even-fold rotation symmetry film (Sec. IV), while $x\rightarrow -x$ is not a symmetry operation for an odd-fold rotation symmetry film (Sec. V).

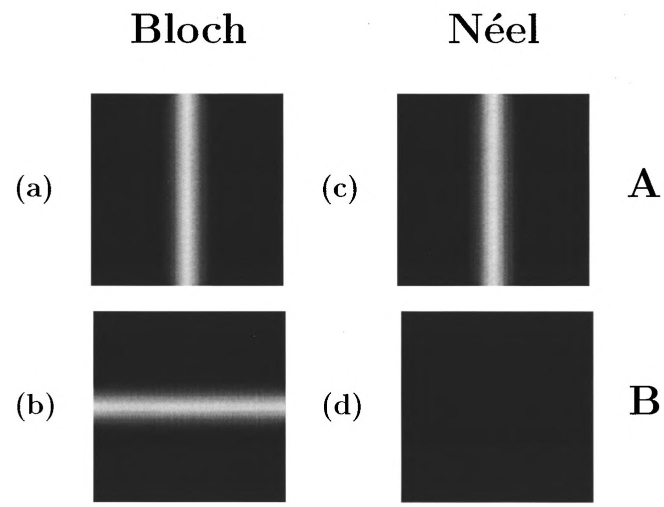

FIG. 3. Simulated images of fragments A (panels a, c) and B (b, d) of a Bloch (a, b) and Néel (c, d) wall in an even-fold rotation symmetry film. The MSHG response is polarized along the vertical y azimuth. The fundamental field is assumed to be polarized purely along the x (horizontal) or y azimuth. The thickness of the domain wall is much smaller than the resolution of the imaging objective. The brightness of the wall seen in panel (a) depends on the relative magnitude of the $\overleftrightarrow{\chi}^{(3)}$ term whereas that seen in panel (b) depends on the relative magnitude of the $\overleftrightarrow{\chi}^{(1)}$ term.

azimuth is recorded. The relevant elements of the $\overleftrightarrow{\chi}^{(1)}$ and $\overleftrightarrow{\chi}^{(3)}$ tensors are assumed to be of the same order of magnitude.

As can be seen from Table IV and Fig. 3, for the even rotation symmetry films there is no SHG light generated within the domains. Also, in a Néel wall the MSHG light is only polarized parallel to the wall if the fundamental beam is polarized either along the wall or the wall normal. Therefore, if the MSHG response of a wall is not purely polarized along the wall, the wall must be of the Bloch type. Note, however, that this rule is valid only for walls which are along the mirror symmetry planes of the film (along x and y). Therefore, if a domain wall in a thin garnet film is not oriented along one of the mirror symmetry planes [or the fundamental field $\mathbf{E}(\omega)$ has nonzero projections on both the wall normal and the wall, see Eq. (9)], nonparallel polarized MSHG light may be generated even in a Néel wall.

Further inspection of Table IV shows that for Bloch domain walls A or B the relative weight of the local $\overleftrightarrow{\chi}^{(1)}$ and gradient $\overleftrightarrow{\chi}^{(3)}$ contribution can be found from the polarization properties of the MSHG light. The higher the weight of the gradient $\overleftrightarrow{\chi}^{(3)}$ term, the stronger the MSHG component polarized along the Bloch domain wall. We also note that contributions of the local $\overleftrightarrow{\chi}^{(1)}$ and gradient $\overleftrightarrow{\chi}^{(3)}$ term to the MSHG image of domain walls depend differently on the wall thickness d_w. The thicker the domain wall, the wider the region contributing to the MSHG image of the wall. On the other hand, an increase of d_w leads also to a reduction of the magnitude of the spatial derivative of the magnetization $\nabla\mathbf{M}$. As a result, the weight of the gradient $\overleftrightarrow{\chi}^{(3)}$ term does not depend on the wall thickness d_w. E.g., for a wall A the weight of this term is proportional to

$$\int_{\text{wall}} dx\, \nabla_x M_z = \Delta M_z,$$

where ΔM_z is the change of M_z across the wall. In contrast, the weight of the local $\overleftrightarrow{\chi}^{(1)}$ term is proportional to d_w. The latter fact can be used to estimate the thickness of a Bloch domain wall from nonlinear-optical measurements even if d_w is much smaller than the wavelength. For example, consider a Bloch domain wall A. As mentioned above, the

TABLE IV. Polarization of the nonvanishing MSHG sources $\mathbf{P}_{\|}^{(n)}(2\omega)$ in an even-fold rotation symmetry film arising via different $\overleftrightarrow{\chi}^{(n)}$ terms of Eq. (4). The fundamental beam is polarized along x or y directions. We also explicitly show how these contributions are related to different components of the film magnetization.

Term n	Domain $\mathbf{M}\|z$	Bloch wall A $\mathbf{M}_{\|}\ \|y$	Bloch wall B $\mathbf{M}_{\|}\ \|x$	Néel wall A $\mathbf{M}_{\|}\ \|x$	Néel wall B $\mathbf{M}_{\|}\ \|y$
0					
1		$P_x \propto M_Y$	$P_y \propto M_X$	$P_y \propto M_X$	$P_x \propto M_Y$
2					
3		$P_y \propto \nabla_x M_Z$	$P_x \propto \nabla_y M_Z$	$P_y \propto \nabla_x M_Z$	$P_x \propto \nabla_y M_Z$
4					

TABLE V. Polarization of the MSHG wave generated in an odd-fold rotation symmetry garnet film via different $\tilde{\chi}^{(n)}$ terms of Eq. (4). The two terms in the last line of the table ($\propto\tilde{\chi}^{(4)}$) may have different weight.

n	Domain	Bloch wall A	Bloch wall B	Néel wall A	Néel wall B
0	P_x	P_x	P_x	P_x	P_x
1	$P_y \propto M_Z$	$P_x \propto M_Y$	$P_y \propto M_X$	$P_y \propto M_X$	$P_x \propto M_Y$
2	$P_x \propto M_Z^2$	$P_x \propto M_Y^2$	$P_x \propto M_X^2$	$P_x \propto M_X^2$	$P_x \propto M_Y^2$
3		$P_y \propto \nabla_x M_Z$	$P_x \propto \nabla_y M_Z$	$P_y \propto \nabla_x M_Z$	$P_x \propto \nabla_y M_Z$
4		$P_y \propto M_Y \nabla_x M_Z$ $+ M_Z \nabla_x M_Y$	$P_y \propto M_X \nabla_y M_Z$ $+ M_Z \nabla_y M_X$	$P_x \propto M_X \nabla_x M_Z$ $+ M_Z \nabla_x M_X$	$P_x \propto M_Y \nabla_y M_Z$ $+ M_Z \nabla_y M_Y$

''brightness'' of the wall with the use of x-polarized MSHG light is proportional to the product of the film nonlinear susceptibility $\chi_{xiiY}^{(1)}$ ($i=x,y$) and d_w. The magnitude of the $\chi_{xiiY}^{(1)}$ element can be independently determined if a strong enough external magnetic field along the y axis is applied. The film is then uniformly magnetized along y and the MSHG response solely arises via $\chi_{xiiY}^{(1)}$.[13]

V. NONLINEAR MAGNETO-OPTICAL IMAGING OF GARNET FILMS WITH ODD-FOLD ROTATION SYMMETRY

Now we assume that the garnet film has a mirror symmetry plane normal to the y axis, while the $x \to -x$ is not a symmetry operation for the lattice. The film therefore has one- or three-fold rotation symmetry. This symmetry is expected, for example, for magnetic films grown on a (111) nonmagnetic garnet substrate.[21] We analyze the MSHG image of two neighboring domains and two parts A and B of a domain wall (Fig. 2). The magnetization within the domains is again assumed to be 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 V.

For the odd-fold rotation symmetry films we find that there is a nonvanishing nonmagnetic contribution to the MSHG response arising via the $\chi_{xii}^{(0)}$ ($i=x$ or y) element of $\tilde{\chi}^{(0)}$. Obviously, this contribution does not depend on the magnetic subsystem and is polarized along x if the fundamental beam is purely polarized either along x or y. A quadratic magnetic-induced response with the same polarization is also generated via the $\tilde{\chi}^{(2)}$ term. Note, however, that the latter contribution may be different in the domain and domain wall because the magnitude of the $\chi_{xiiZZ}^{(2)}$ element may differ from the magnitude of the $\chi_{xiiXX}^{(2)}$ and $\chi_{xiiYY}^{(2)}$ elements. The local term $\tilde{\chi}^{(1)}$ that is linear in the magnetization gives rise to a y-polarized MSHG component in the domains generated by the nonlinear polarization

$$P_y^{(1)}(2\omega)=\chi_{yiiZ}^{(1)}E_i^2(\omega)M_Z, \quad i=x,y. \tag{13}$$

This contribution changes its sign across the wall because of the reversal of M_Z. Due to the destructive interference between the two sides of the domain wall, the polarization (13) should therefore give no contribution to the image of the wall. On the other hand, the $\tilde{\chi}^{(1)}$ term gives rise to an additional x-polarized contribution to the MSHG image of the Bloch wall A and Néel wall B via the polarization

$$P_x^{(1)}(2\omega)=\chi_{xiiY}^{(1)}E_i^2(\omega)M_Y, \quad i=x,y. \tag{14}$$

The Bloch wall B and Néel wall A ($M_\parallel \| x$) give a similar contribution polarized along y.

The gradient terms $\tilde{\chi}^{(3)}$ and $\tilde{\chi}^{(4)}$ obviously vanish in a domain with a uniform magnetization. On the other hand, as is discussed in detail above, they may produce an important contribution to the MSHG image of a domain wall, especially in thin films with a lattice structure that is slightly distorted from its centrosymmetric arrangement. Taking the $\tilde{\chi}^{(3)}$ term into account, we find that in addition to the polarization (11), symmetry allows also contributions of the type $\chi_{yiixX}^{(3)}\nabla_x M_X$ ($\neq 0$ for Bloch wall A) and $\chi_{xiixY}^{(3)}\nabla_x M_Y$ ($\neq 0$ for Néel wall A) which could give a difference between a Bloch and Néel type domain wall A. These terms and similar terms in Bloch and Néel walls B however vanish after integration across the wall. Therefore, the $\tilde{\chi}^{(3)}$ term contributes to the MSHG image of the wall solely via the nonlinear polarization (11), in the same way as for the even-fold rotation symmetry films. This contribution is not sensitive to the type (Bloch or Néel) of the wall. Along the same line, for a Bloch wall A we find that the only symmetry-allowed and nonvanishing (after integration across the wall) contribution of the $\tilde{\chi}^{(4)}$ term is given by the polarization

$$P_y^{(4)}(2\omega)=\chi_{yiiYxZ}^{(4)}E_i^2(\omega)M_Y\nabla_x M_Z + \chi_{yiiZxY}^{(4)}E_i^2(\omega)M_Z\nabla_x M_Y, \quad i=x,y \tag{15}$$

along y. The corresponding contribution of a Néel wall A

$$P_x^{(4)}(2\omega)=\chi_{xiiXxZ}^{(4)}E_i^2(\omega)M_X\nabla_x M_Z + \chi_{xiiZxX}^{(4)}E_i^2(\omega)M_Z\nabla_x M_X, \quad i=x,y \tag{16}$$

is polarized along x, i.e., the $\tilde{\chi}^{(4)}$ term is sensitive to the type of the wall. We also note that in general $\chi_{yiiYxZ}^{(4)} \neq \chi_{yiiZxY}^{(4)}$ in Eq. (15) and $\chi_{xiiXxZ}^{(4)} \neq \chi_{xiiZxX}^{(4)}$ in Eq. (16). Therefore, the short expression $\propto M_X\nabla_x M_Z + M_Z\nabla_x M_X$ and similar expressions used in the last line of Table V should be understood as ''a contribution that is $\propto M_X\nabla_x M_Z$ plus a contribution that is $\propto M_Z\nabla_x M_X$.'' The two terms contributing to the sum may have different weight.

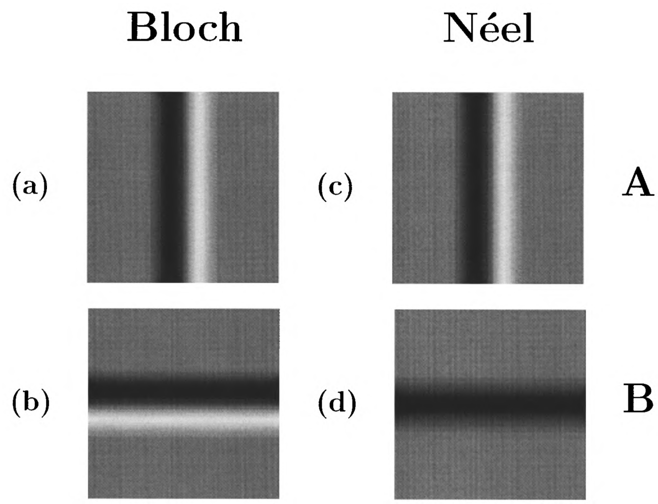

FIG. 4. Simulated images of fragments A (panels a, c) and B (b, d) of a Bloch (a, b) and Néel (c, d) wall in an odd-fold rotation symmetry film. The MSHG response is polarized along the vertical y azimuth. The fundamental field is assumed to be polarized purely along the x or y azimuth.

Using some guesses of the relative magnitude of the different terms, we also generated several possible images of Bloch and Néel domain walls which are shown in Fig. 4 (MSHG polarized along the y axis) and Fig. 5 (x-polarized MSHG images). The relative weight of the MSHG contributions originating from the walls are assumed to be of the same magnitude. For the MSHG contributions of the domains the sum of the relevant elements of the $\overleftrightarrow{\chi}^{(0)}$ and $\overleftrightarrow{\chi}^{(2)}$ tensors are taken to be three times larger than those of the $\overleftrightarrow{\chi}^{(1)}$ tensor. The thickness of the domain wall is assumed to be much smaller than the resolution of the imaging objective.

As is seen from Table V the lowering of the film symmetry results in a much larger number of nonvanishing contributions in comparison to that for the even-fold rotation symmetry films (cf. Table IV). The MSHG image of the magnetic domain structure is therefore expected to be more complex (Figs. 4, 5). One can nevertheless find some simple rules which may be used to analyze the image. We consider the Néel walls, as an example. The polarization of the MSHG response from the domains possesses the information about the relative importance of the local terms that are even ($\overleftrightarrow{\chi}^{(0)}$ and $\overleftrightarrow{\chi}^{(2)}$) or odd ($\overleftrightarrow{\chi}^{(1)}$) in the magnetization. Then, in the Néel wall of type B (parallel to the symmetry plane $y=0$) all terms give rise to the MSHG response polarized along x. Therefore, Néel walls B should appear like ''dark'' lines if only the y-polarized MSHG is recorded [see Fig. 4(d)]. The ''brightness'' of a Néel wall A [Fig. 4(c)] brings then information about the relative importance of the terms which are linear in the magnetization ($\overleftrightarrow{\chi}^{(1)}$ and $\overleftrightarrow{\chi}^{(3)}$). Note the black-white character of the images shown in Figs. 4(a)–4(c). It is related to the fact that the y-polarized component of the domain contribution changes its sign across the domain wall. The interference of this contribution with the MSHG response of the wall results in the observed black-white images. We also note that the magnitude of the element $\chi^{(1)}_{yiiX}$, $i=x,y$, contributing to the MSHG response of the Néel wall A [Fig. 4(c)], can be independently estimated in an additional measurement of MSHG generated in a uniformly magnetized magnetic film using an external magnetic field. Based on such an estimate of the $\overleftrightarrow{\chi}^{(1)}$ term, one can therefore evaluate the relative importance of the gradient $\overleftrightarrow{\chi}^{(3)}$ term.

Using Table V, similar rules can be formulated for the domain walls of the Bloch type. For example, the relative ''brightness'' of the wall A in y-polarized MSHG light gives us the relative weight of the two gradient terms $\overleftrightarrow{\chi}^{(3)}$ and $\overleftrightarrow{\chi}^{(4)}$, etc.

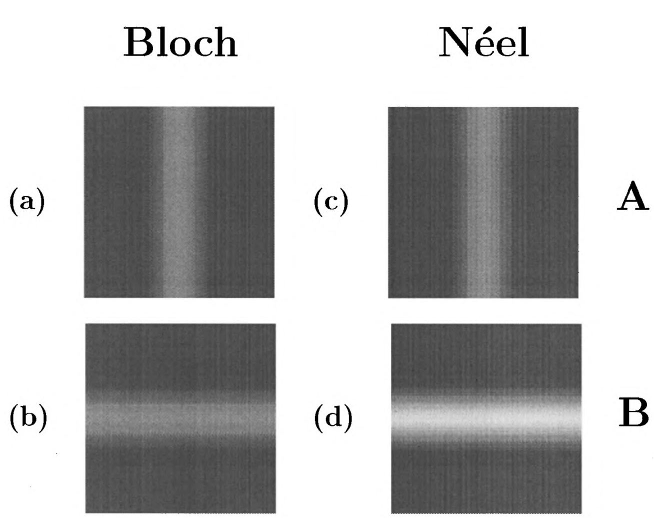

FIG. 5. The same as in Fig. 4 but for the MSHG response polarized along the horizontal x azimuth.

VI. CONCLUSION

A symmetry analysis of MSHG images of the domain structures of thin magnetic garnet films is performed. Four different magnetization-induced contributions are taken into account. Two of them ($\overleftrightarrow{\chi}^{(1)}$ and $\overleftrightarrow{\chi}^{(2)}$) depend on the local value of the magnetization and contribute to the MSHG response of both the domains and the domain walls. Nevertheless, the polarization of the wall contribution differs from the polarization of MSHG generated in the domains because of the rotation of the magnetization vector. We also introduce gradient terms ($\overleftrightarrow{\chi}^{(3)}$ and $\overleftrightarrow{\chi}^{(4)}$) 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 we therefore derive the nonvanishing independent elements of the relevant tensors.

We have studied the polarization rules for the MSHG images of thin garnet films measured in transmission at normal incidence. The MSHG images of odd- and even-fold rotation symmetry films are analyzed. It is shown that measurements of film images using different polarizations may bring information about (i) the type of the domain wall and (ii) the relative weight of different contributions to the MSHG response. The polarization rules are illustrated by simulations of possible images of domain walls using some assumptions about the relative magnitude of different magnetization-induced sources. It is shown that in some particular cases one can also estimate the thickness of the domain wall even if it is much smaller than the optical wavelength.

We point out that in the experiments reported in Ref. 4 the polarization of the MSHG response was not analyzed. In such a case the magnetic contrast is solely related to the tangential component of the magnetization **M**.[13] The present results indicate that polarization measurements of MSHG images of thin magnetic films can be used for a more detailed study.

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[1] *Physics and Engineering Application of Magnetism*, edited by Y. Ishikawa and N. Miura (Springer, Berlin, 1991).

[2] A. Hubert, *Theory of Domain Walls in Ordered Media* (Springer-Verlag, Berlin, 1974).

[3] M. Fiebig, D. Frohlich, G. Sluyterman, and R. V. Pisarev, Appl. Phys. Lett. **66**, 2906 (1995).

[4] A. Kirilyuk, V. Kirilyuk, Th. Rasing, V. V. Pavlov, and R. V. Pisarev, J. Magn. Soc. Jpn. **20**, 361 (1996); V. Kirilyuk, A. Kirilyuk, and Th. Rasing, Appl. Phys. Lett. **70**, 2306 (1997).

[5] Th. Rasing, M. G. Koerkamp, B. Koopmans, and H.v.d. Berg, J. Appl. Phys. **79**, 6181 (1996); Th. Rasing, J. Magn. Soc. Jpn. **20**, 13 (1996).

[6] J. Reif, J. C. Zink, C. M. Schneider, and J. Kirschner, Phys. Rev. Lett. **67**, 2878 (1991); H. Wierenga, W. de Jong, M. Prins, Th. Rasing, R. Vollmer, A. Kirilyuk, H. Schwabe, and J. Kirschner, *ibid.* **74**, 1462 (1995).

[7] B. Koopmans, M. G. Koerkamp, Th. Rasing, and H.v.d. Berg, Phys. Rev. Lett. **74**, 3692 (1995).

[8] For theory of the nonlinear optics in antiferromagnets see, e.g., V. N. Muthukumar, R. Valentí, and C. Gros, Phys. Rev. Lett. **75**, 2766 (1995); A. Dähn, W. Hübner, and K. H. Bennemann, *ibid.* **77**, 3929 (1996).

[9] N. N. Akhmediev, S. B. Borisov, A. K. Zvezdin, I. L. Lyubchanskii, and Yu. V. Melikhov, Sov. Phys. Solid State **27**, 650 (1985).

[10] R.-P. Pan, H. D. Wei, and Y. R. Shen, Phys. Rev. B **39**, 1229 (1989).

[11] W. Hübner and K. H. Bennemann, Phys. Rev. B **40**, 5973 (1989); **52**, 13 411 (1995).

[12] S. B. Borisov, N. N. Dadoenkova, I. L. Lyubchanskii, and V. L. Sobolev, Sov. Phys. Solid State **32**, 2127 (1990).

[13] V. V. Pavlov, R. V. Pisarev, A. Kirilyuk, and Th. Rasing, Phys. Rev. Lett. **78**, 2004 (1997).

[14] I. L. Lyubchanskii, Phys. Solid State **37**, 987 (1995); J. Magn. Magn. Mater. **157/158**, 307 (1996).

[15] R. V. Pisarev, in *Physics of Magnetic Dielectrics*, edited by G. A. Smolenskii (Nauka, Leningrad, 1974) (in Russian).

[16] R. Schäfer and A. Hubert, Phys. Status Solidi A **118**, 271 (1990); V. Kambersky, *ibid.* **125**, K117 (1991).

[17] L. D. Landau and V. L. Ginzburg, Zh. Eksp. Teor. Fiz. **20**, 1064 (1950).

[18] L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Pergamon, New York, 1959).

[19] C. Kittel, *Thermal Physics* (Wiley, New York, 1969).

[20] P. S. Pershan, Phys. Rev. **130**, 919 (1963).

[21] R. V. Pisarev, B. B. Krichevtsov, V. N. Gridnev, V. P. Klin, D. Frölich, and Ch. Pahlke-Lerch, J. Phys., Condens. Matter. **5**, 8621 (1993).

[22] A. Oleš, F. Kajzar, M. Kucab, and W. Sikora, *Magnetic Structures Determined by Neutron Diffraction* (PWN, Warszawa, 1976).

[23] A. V. Petukhov, Phys. Rev. B **52**, 16 901 (1995).

[24] Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).