Observation of a Transversal Nonlinear Magneto-Optical Effect in Thin Magnetic Garnet Films

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A simultaneous breaking of space and time-reversal symmetry leads to a coexistence of crystallographic $\chi^{(2)}_{ijk}(2\omega)$ and magnetization-induced $\chi^{(3)}_{ijkl}(2\omega)$ electric-dipole type contributions to the nonlinear optical susceptibility in the same medium. As a consequence, a new transversal nonlinear magneto-optical effect arises that is linear in **M**. This effect is experimentally demonstrated in thin films of magnetic garnets, as well as large effects quadratic in **M**. We suggest a novel method to distinguish the two susceptibility contributions based on their different transformation properties. [S0031-9007(97)02712-9]

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Space-inversion or time-reversal symmetry breaking, for example, at phase transitions or due to external forces, often has important consequences for physical properties. In linear optics, the breaking of *time-reversal* symmetry leads to a number of well known magneto-optical effects like Faraday rotation in transmission and Kerr rotation in reflection. For the nonlinear case, optical second harmonic generation (SHG) in the electric dipole approximation is only allowed in media with a broken *space-inversion* symmetry [1]. As a consequence, *nonlinear magneto-optical* effects can only be observed in materials in which both space-inversion and time-reversal symmetry are broken simultaneously. It appears that the overwhelming majority of magnetically ordered materials, metallic and dielectric, ferromagnetic and antiferromagnetic, are centrosymmetric in their bulk form. However, space inversion is broken at the surface and very recently SHG in reflection has been proven to be a versatile tool for studying magnetized surfaces and interfaces of metallic materials with a centrosymmetric bulk crystal structure [2–6].

In this Letter we present results on the experimental observation of a novel nonlinear magneto-optical effect in magnetic *bulk* materials in which the space-inversion and time-reversal operations are broken simultaneously. We show that due to the coexistence and the interference of these two contributions to the SHG a new *transversal* nonlinear magneto-optical effect arises, which is *linear in the magnetization.* This transversal nonlinear effect is in contrast to the well known transversal linear optical effect of magnetic birefringence (called the Voigt or Cotton-Mouton effect), which is quadratic in the magnetization [7]. We also show that for certain high symmetry directions a SH signal is generated *only in the presence of a magnetization.* We propose a method that allows an unambiguous separation between the crystallographic and the magnetization-induced contributions to the SHG signals. We have found that the corresponding tensor components are of comparable magnitude, with the magnetic ones vanishing above the Curie point.

The total nonlinear optical polarization of a medium (in the electric dipole approximation) can be written as

$$\begin{aligned} P_i(2\omega) &= P_i^{\mathrm{cr}}(2\omega) + P_i^{\mathrm{magn}}(2\omega) \\ &= \chi^{(2)}_{ijk}(-2\omega,\omega,\omega)E_j(\omega)E_k(\omega) \\ &\quad + \chi^{(3)}_{ijkl}(-2\omega,\omega,\omega,0)E_j(\omega)E_k(\omega)M_l(0), \quad (1) \end{aligned}$$

where $\mathbf{P}^{\mathrm{cr}}$ and $\mathbf{P}^{\mathrm{magn}}$ are the crystallographic and magnetic contributions, respectively, $E_j(\omega)$ and $E_k(\omega)$ are the fundamental optical fields, and $\mathbf{M}(0)$ is a spontaneous or magnetic-field induced static magnetization. Both the $\mathbf{P}^{\mathrm{cr}}$ and $\mathbf{P}^{\mathrm{magn}}$ contributions are of the *electric-dipole* character, because they are proportional only to $\mathbf{E}(\omega)$. They are simultaneously allowed in noncentrosymmetric media, but their properties are different.

(i) $\mathbf{P}^{\mathrm{cr}}$ is described by a *polar* tensor $\chi^{(2)}_{ijk}$ of rank 3, whereas $\mathbf{P}^{\mathrm{magn}}$ is described by an *axial* tensor $\chi^{(3)}_{ijkl}$ of rank 4. They exhibit characteristically different rotational anisotropy that may strongly depend on the magnetization orientation in the crystal, similarly as was predicted for magnetized surfaces [2].

(ii) In nonabsorbing materials $\chi^{(2)}_{ijk}$ is a real but $\chi^{(3)}_{ijkl}$ is an imaginary tensor [2,8]. The corresponding nonlinear waves have a 90° phase shift and thus cannot interfere. However, interference becomes allowed when one or both of them are complex. *This interference leads to the effects which are linear in the magnetization.*

(iii) The two contributions to $\mathbf{P}(2\omega)$ should vary differently as a function of temperature. $\mathbf{P}^{\mathrm{cr}}$ probes the degree of a crystal lattice noncentrosymmetry. It may depict anomalies at structural phase transitions. $\mathbf{P}^{\mathrm{magn}}$ should reflect a temperature variation of the magnetization and thus vanishes at the transition from a magnetically ordered to a paramagnetic state.

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To obtain a $\mathbf{P}^{cr} \neq 0$ for normal incidence the in-plane symmetry must be rather low [9], while for the interference, a simultaneous existence of a nonzero $\mathbf{P}^{magn}$ is necessary. Magnetic garnet films, epitaxially grown on substrates with a controllable lattice mismatch are excellent systems to test these ideas. Though bulk crystals of magnetic garnets like yttrium iron garnet $Y_3Fe_5O_{12}$ are centrosymmetric, in thin films space-inversion symmetry is broken due to a distortion of the crystal structure. This is evidenced by the observation of SHG [9–11] and linear magnetoelectric effects [12] in such films. In Ref. [9], a symmetry analysis of the crystallographic SHG contributions is given. Here, we are interested in the magnetization-induced SHG effects. To avoid any influence of the linear magneto-optical effects, like Faraday rotation or magnetic circular dichroism, we consider the transversal situation, with the magnetization in the film plane.

Low symmetry *(210) films* are characterized by the monoclinic point group m (C_{1h}), $x \| [001]$. Using Eq. (1) we get for the magnetization-induced nonlinear polarization

$$P_{XX}^{magn}(2\omega, \varphi) = E^2 M[B\cos^4\varphi - A\sin^4\varphi + (3/4)(A - B)\sin^2 2\varphi], \quad (2)$$
$$P_{YY}^{magn}(2\omega, \varphi) = 2E^2 M \sin 2\varphi (A\cos^2\varphi + B\sin^2\varphi),$$

where φ is the angle between the magnetization direction and the [001] axis. Capital indices XX and YY denote input-output polarizations of the light in the laboratory frame. The definition of the crystallographic axes x, y, and z in films of different symmetries was taken as in Ref. [9]. A, B, and C are combinations of the real and imaginary parts of $\chi^{(2)}_{ijk}$ and $\chi^{(3)}_{ijkl}$. Equation (2) implies that applying a magnetic field in the Y direction will lead to a change in the *transmitted* SHG intensity $I_{ij}(2\omega) = |P_{ij}^{cr} + P_{ij}^{magn}|^2$ for light propagating in the Z direction. Thus the coherent contributions of $\mathbf{P}^{cr}$ and $\mathbf{P}^{magn}$ to $I(2\omega)$ lead to a *transversal* nonlinear magneto-optical effect that contains both *linear* and *quadratic* terms in the magnetization.

(111) films have point group symmetry $3m$ (C_{3v}), $x \perp [\bar{1}10]$ [9]. Taking into account the relevant crystallographic [9] and magnetization-induced susceptibilities, we get the following equations for the SHG rotational anisotropy $I_{ij}(2\omega, \varphi)$:

$$I_{XX}(2\omega, \varphi) = E^4(A\cos^2 3\varphi + BM^2 + 2CM\cos 3\varphi), \quad (3)$$
$$I_{YY}(2\omega, \varphi) = E^4 A \sin^2 3\varphi .$$

A linear magnetic response is therefore expected for the XX (and YX) polarization combinations, while for YY (and XY) the $\mathbf{P}^{magn} = 0$.

For *(001)-oriented films,* the situation is different. Here, the point group symmetry is $4mm$ (C_{4v}), $x \| [100]$, leading to $\mathbf{P}^{cr} = 0$ for normal incidence. This means that here we expect a purely magnetization-induced SHG. From Eq. (1) we get for the rotational anisotropy in a (001) film

$$I_{XX}(2\omega, \varphi) = E^4 M^2(A - B\sin^2 2\varphi + C\sin^4 2\varphi),$$
$$I_{YY}(2\omega, \varphi) = (1/4)E^4 M^2 C \sin^2 4\varphi . \quad (4)$$

Because of the purely quadratic $\mathbf{M}$ dependence, the SHG intensity should not be sensitive to the magnetization reversal in (001) films.

Magnetic garnet films of three different types with substrate orientations (001), (111), and (210) were grown by liquid phase epitaxy. The samples differed in film and substrate compositions (see Table I). Thin wafers of cubic centrosymmetric $Gd_3Ga_5O_{12}$ (GGG) garnet and substituted GGG with a larger lattice parameter were used as substrates. The substrates are transparent at the fundamental and SHG frequencies and did not show any SHG signals.

The experiments were done in transmission at normal incidence with the light propagating along the Z axis (see Fig. 1) and with a magnetic field up to $H \simeq 2.3$ kOe applied along Y. Rotating the sample in the range $0 \leq \varphi \leq 360°$ around the Z axis we could register the SHG signal with the magnetization being kept along Y and the incoming and outcoming linear light polarization along X or Y. As we show below, such an approach allows an unambiguous separation between the crystallographic and magnetization-induced SHG signals.

The SHG signal was generated by the output at 0.841 μm (1.474 eV) of a mode-locked Ti-sapphire laser working at a repetition frequency of 82 MHz, a pulse width of about 100 fs at an average power on the sample between 100 and 250 mW. At this wavelength, the linear absorption of magnetic garnet films and bulk crystals is 10–20 cm^{-1} [13] and the fundamental beam propagates through thin films without any noticeable attenuation, while the polarization remains unchanged because the linear magneto-optical effects (Faraday rotation and ellipticity) are zero in the transversal geometry. Second order magnetic birefringence effects are small, $\Delta n \approx 10^{-5}$ [7] and do not change the polarization of the fundamental beam. The linear absorption is much higher at the second harmonic frequency (2.948 eV), $\alpha \simeq (5–6) \times 10^3$ cm^{-1} [13]. Therefore, in transmission experiments the detected SHG signal originates only from a back-side layer with a thickness of about 1 μm.

TABLE I. Basic parameters of the three samples.

Film	Thickness (μm)	Film composition	Lattice misfit (%)
(001)	5	$(YbPr)_3(FeGa)_5O_{12}$	0.28%
(111)	1	$(YLuBi)_3(FeGa)_5O_{12}$	−0.06%
(210)	10	$(YPrLuBi)_3(FeGa)_5O_{12}$	0.39%

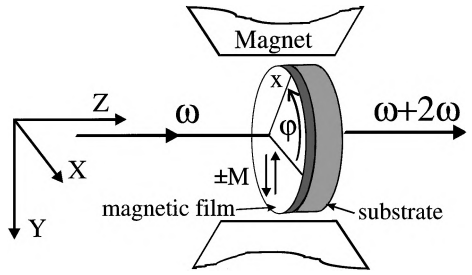

FIG. 1. Experimental geometry: sample rotates in a transverse magnetic field.

Figure 2 shows the rotational anisotropy of SHG in films of three types at $T = 295$ K. The upper and lower parts of each figure correspond to the I_{XX} and I_{YY} case, respectively, where subscripts XX and YY denote input-output polarizations of the light in the laboratory frame (see Fig. 1). The horizontal line passing through 0° and 180° on each plot corresponds to the mirror plane m, and only the top half (I_{XX} case) and the bottom half (I_{YY} case) are shown. The vertical line cutting each plot in the middle corresponds to m', the mirror reflection followed by the time-reversal operation. The symmetry element m' leads to the sign change of the magnetic contribution to the SHG intensity. The data were fitted to Eqs. (2)–(4), with A, B, and C being the only adjustable parameters. Apart from small deviations, the agreement with experiment is very good.

In (210) films strong SHG signals were observed in nonmagnetized and magnetized films for all polarization combinations. The SHG intensity in these films was 1 to 2 orders of magnitude larger as compared to other film orientations. The change in the SHG intensity due to the switching of the sample *in-plane* magnetization demonstrates the transversal nonlinear optical effect, linear in **M**, in perfect agreement with our prediction. In a nonmagnetized (111) film, an SHG signal was observed with a 60° periodicity in the rotational anisotropy. The magnetized (111) film showed a 120° periodicity for XX (and for YX) polarizations, with a 60° rotation between $+\mathbf{M}$ and $-\mathbf{M}$ states. No magnetic effect was observed for YY (and XY) polarizations. In the (001) film *no SHG signal was detected in the absence of a magnetic field.* However, as predicted, in a magnetized sample an SHG signal was observed, which was quadratic in **M** and thus insensitive to the sign of the applied magnetic field. Thus, an SHG response can be "turned on" with the help of a magnetic field. We have to mention that in films of this symmetry the SHG intensity was substantially lower than in the other films, which explains the larger scatter in the experimental points.

Although not shown here, magnetization-induced polarization rotation of the SHG light up to 90° was also observed in addition to the intensity changes. Similar to the latter, this rotation depends on the mutual directions of crystallographic axes, magnetization, and incoming light polarization.

In order to independently prove the different origins of the two contributions to the nonlinear polarization, the

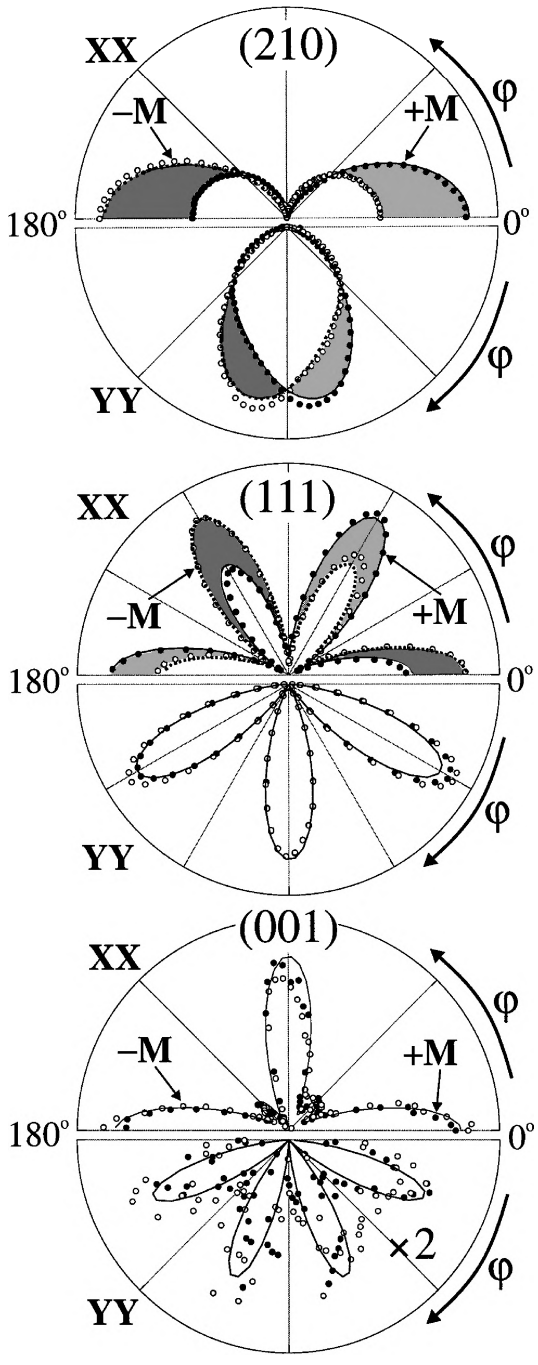

FIG. 2. Rotational anisotropy of the SHG intensity in garnet films of different symmetry: solid circles denote $+\mathbf{M}$ state, while open circles are for the $-\mathbf{M}$ state. XX and YY denote the input-output polarization combinations. Solid and dotted lines are the theoretical fits for $+\mathbf{M}$ and $-\mathbf{M}$ states, respectively, from Eqs. (2)–(4). Magnetic contrast (difference between the $+\mathbf{M}$ and $-\mathbf{M}$ theoretical fits) is indicated by dark (positive) and light (negative) shadowed areas.

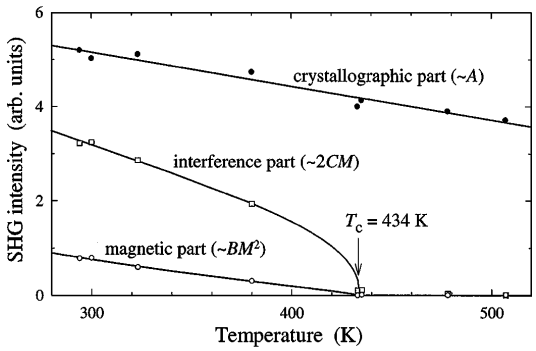

FIG. 3. Temperature variations of crystallographic, magnetization-induced, and interference terms in the SHG intensity for the (111) film.

SHG rotational anisotropy has been studied as a function of temperature for several films. The temperature variations of the crystallographic, interference, and pure magnetic contributions to the SHG intensity for the (111) film are shown in Fig. 3. The different terms are separated by fitting the experimental angular dependence to Eq. (3). The crystallographic contribution decreases linearly with temperature, whereas the magnetization-related contributions vanish at T_c. The interference term $\propto 2CM$ shows a $(1 - T/T_c)^{0.61(6)}$ dependence, whereas the pure magnetic part $\propto BM^2$ vanishes with a $(1 - T/T_c)^{1.05(8)}$ dependence, as expected from the proportionality to M. This makes an additional strong support for the derived Eqs. (2)–(4). These results allow a direct evaluation of a ratio between crystallographic and magnetization-induced susceptibilities. At room temperature we get $I^{\mathrm{magn}}/I^{\mathrm{cr}} \approx 0.16$ or $|\chi^{\mathrm{magn}}|/|\chi^{\mathrm{cr}}| \approx 0.4$. The fact that these two contributions are of the same order of magnitude is unexpected because the magnetization-induced part should normally be regarded as a perturbation of the crystallographic part due to magnetic ordering.

The origin of the crystallographic contribution (the loss of bulk inversion symmetry due to a growth induced lattice distortion) could independently be proven by SHG studies of thin samples cut from the *bulk* crystals of yttrium-gallium garnet $Y_3Fe_{5-x}Ga_xO_{12}(x \simeq 0.7)$. In contrast to that of thin films, their crystal structure is centrosymmetric. The SHG signals in these samples were several orders of magnitude lower than the SHG signals in thin films and, in fact, could be related only to a surface contribution. We therefore conclude that the crystallographic and magnetization-induced SHG in films of magnetic garnets are due to *bulk* electric-dipole mechanisms.

In conclusion, we have shown that a simultaneous breaking of space and time-reversal symmetry leads to a coexistence of two electric-dipole contributions to the nonlinear optical susceptibility: a crystallographic and a magnetic one. Such a coexistence can occur only in noncentrosymmetric media in an applied magnetic field or possessing magnetic ordering [14]. We have observed a new nonlinear transversal magneto-optical effect arising from their interference that is linear in the magnetization and is a bulk effect that is strongest for the lowest film symmetry. An unambiguous separation between the two contributions to the SHG was obtained from rotational anisotropy experiments in an applied field, and at room temperature they were found to be of the same order of magnitude.

Though we are able to explain the experimental data applying a simple symmetry analysis, a more profound microscopic theory is required to explain the absolute and relative values of the crystallographic and magnetization-induced nonlinear susceptibilities $\chi^{(2)}_{ijk}$ and $\chi^{(3)}_{ijkl}$. In fact, these two types of nonlinear susceptibility should also coexist in noncentrosymmetric molecules. However, to study time-noninvariant contributions to the nonlinear magneto-optical response, magnetically ordered materials are most suitable because of their large values of the exchange splitting of the electronic states.

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