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Optical second-harmonic generation in magnetic garnet thin films

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Abstract. Second-harmonic generation (SHG) at normal incidence in transmission geometry was studied in Bi-containing magnetic garnet thin films on gadolinium-gallium garnet (GGG) substrates of (111) and (210) types. A Q-switched Nd-YAG laser was used as a radiation source. The intensity of the SHG was lower than that in crystalline quartz but it was nevertheless reliably detectable. The study of the SHG intensity as a function of the rotation angle of the films around their normals showed that the signal is well described by phenomenological expressions derived under the assumption that the point symmetry of the films is C_{1h} ((210) substrate) or C_{3v} ((111) substrate). The SHG intensity was found to be independent of temperature in the range 290–405 K, thus excluding a 'magnetic' origin of the SHG. These experiments as well as those previously published on the linear magnetoelectric effect prove that there is no inversion centre in magnetic garnet thin films.

1. Introduction

Magnetic garnet thin films have been the subject of both basic and applied research during the last 10–15 years and some results of these studies have been analysed [1, 2]. The active interest in the structural, magnetic and magnetooptical properties of magnetic films is related to the fact that all these properties may be widely varied by changing the magnetic film composition, the composition and orientation of the substrate etc. Though the parent garnet crystal structure is characterized by the cubic centrosymmetric space group $Ia3d$ (O_h^{10}) [3], the study of film structure and magnetic properties showed that films have uniaxial or orthorhombic symmetry. The origin of this symmetry lowering is due to a growth anisotropy and to a mismatch between the lattice parameters of the non-magnetic substrate and the magnetic film. The crystallographic orientation of the substrate is also of primary importance.

Recently it was found that an electric field induces a linear variation of the magnetooptical Faraday rotation in magnetic garnet films (linear magnetoelectric effect) [4, 5]; this obseration is evidence for the absence of an inversion centre in the film structure. This observation is probably not easy to prove by x-ray analysis and it may not be of primary importance for the analysis of magnetic or linear magnetooptical properties. Nevertheless, the lack of an inversion centre is important for at least two reasons: (i) for the development of a more adequate model of film growth on a substrate; and (ii) for the investigation of those phenomena where the lack of an inversion centre is a necessary prerequisite.

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Besides the linear magnetoelectric effect mentioned above, the absence of an inversion centre is obligatory for the existence of the piezoeffect, the linear electrooptical effect, SHG etc, i.e. those phenomena that are described by polar tensors of odd rank.

In the present paper we report on the study of SHG in Bi-containing magnetic garnet thin films on substrates of gadolinium–gallium garnet $Gd_3Ga_5O_{12}$ (GGG) of two different orientations (111) and (210). As far as we know only one paper [6] has been published recently on the SHG in a magnetic film on a (111) type substrate of yttrium–iron garnet $Y_3Fe_5O_{12}$. Our data prove that the 'magnetic' mechanism of destroying the inversion centre as suggested in [6] is not valid in magnetic garnet thin films.

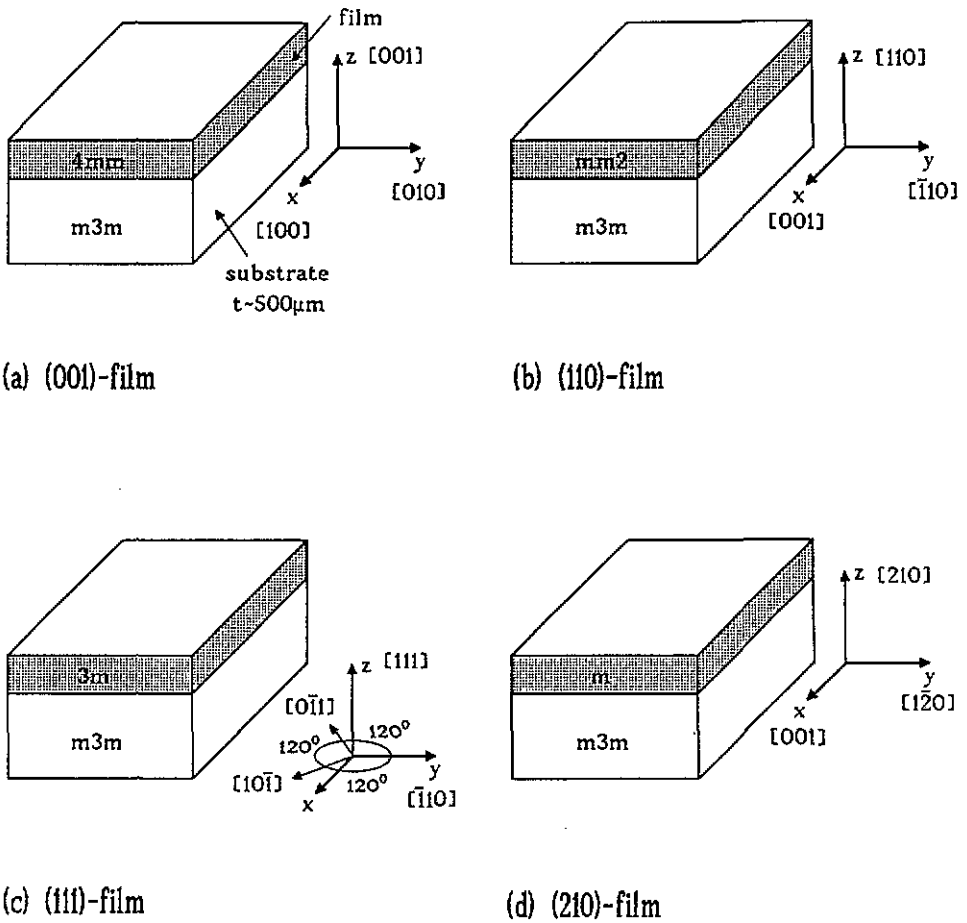

Figure 1. Symmetry of magnetic garnet thin films on GGG substrates for four different orientations.

2. Symmetry analysis

Magnetic garnet thin films can be grown on substrates of different orientations [1, 2], but we restrict our analysis to films of (001), (110), (111) and (210) types (see figure 1). The

lowering of the film symmetry stems from the elastic deformation caused by a mismatch of the lattice parameters between the substrate and the film, as well as from the growth-induced gradients of the lattice parameters of the films, their composition, etc. In order to derive the point group G of a film we have to retain only those symmetry elements of the cubic garnet point group $m3m$ that leave the film invariant. All symmetry elements of the new point group should leave the normal of the surface (the growth direction) invariant, and consequently the new point groups are polar and the operation of inversion is forbidden. Physical properties described by polar tensors of odd rank are forbidden in the cubic group $m3m$, but they become allowed in thin films. For those properties that are described by tensors of even rank, e.g. the magnetic anisotropy, the loss of the inversion centre $\bar{1}$ is not important and the group $G' = G \times \bar{1}$ may be used instead of the group G. In fact this simplification has always been used in the analysis of magnetic properties of thin films. In the following, we will discuss the symmetry properties and the SHG in thin films of four types.

(i) (001)-type films (figure 1(a)). The fourfold axis [001] is directed along the normal of the film; the mirror plane and the twofold axes that are perpendicular to [001] are lost and the point group of the film becomes 4 mm (C_{4v}). SHG is allowed in this point group and the non-linear polarization $P_i^{nl}(2\omega)$ has the following components:

$$\begin{aligned} P_x^{nl} &= 2\chi_{15}E_xE_z \\ P_y^{nl} &= 2\chi_{15}E_yE_z \\ P_z^{nl} &= \chi_{31}(E_x^2 + E_y^2) + \chi_{33}E_z^2 \end{aligned} \tag{1}$$

where χ_{ij} are components of the non-linear susceptibility tensor [7]. These equations show that the SHG for (001) films is forbidden at normal incidence but allowed at oblique incidence.

(ii) (110)-type films (figure 1(b)). The twofold axis [110] is along the normal of the film surface and the point group of the film is $mm2$ (C_{2v}). The non-linear polarization has the following components:

$$\begin{aligned} P_x^{nl} &= 2\chi_{15}E_xE_z \\ P_y^{nl} &= 2\chi_{24}E_yE_z \\ P_z^{nl} &= \chi_{31}E_x^2 + \chi_{32}E_y^2 + \chi_{33}E_z^2. \end{aligned} \tag{2}$$

Again as in the previous case SHG is forbidden at normal incidence and allowed at oblique incidence.

(iii) (111)-type films (figure 1(c)). The threefold axis is along the normal of the film and the relevant non-centrosymmetric point group is $3m$ (C_{3v}). The non-linear polarization has the following form:

$$\begin{aligned} P_x^{nl} &= \chi_{11}(E_x^2 - E_y^2) + 2\chi_{15}E_zE_x \\ P_y^{nl} &= -2\chi_{11}E_xE_y + 2\chi_{15}E_zE_y \\ P_z^{nl} &= \chi_{31}(E_x^2 + E_y^2) + \chi_{33}E_z^2 \end{aligned} \tag{3}$$

where the y axis is taken perpendicular to the mirror plane m. (3) shows that SHG is allowed at normal and oblique incidence.

(iv) (210)-type films (figure 1(d)). The only symmetry element for these films is the mirror plane m, which is perpendicular to the film surface and contains the axes [210] and [1$\bar{2}$0]. Thus the relevant point group is monoclinic m (C_{1h}) and the non-linear polarization has the following form:

$$\begin{aligned} P_x^{nl} &= \chi_{11}E_x^2 + \chi_{12}E_y^2 + \chi_{13}E_z^2 + 2\chi_{15}E_zE_x \\ P_y^{nl} &= 2\chi_{26}E_xE_y + 2\chi_{24}E_zE_y \\ P_z^{nl} &= \chi_{31}E_x^2 + \chi_{32}E_y^2 + \chi_{33}E_z^2 + 2\chi_{35}E_xE_z. \end{aligned} \tag{4}$$

Thus SHG for (210) films is allowed both at normal and oblique incidence.

We are interested in the angular dependence of the SHG intensity I at normal incidence of the pump radiation as function of rotation angle φ of the film around its normal. Another parameter is the angle α between the axes of the polarizer and the analyser placed in front of and behind the film, respectively. The following equation is easily derived for the intensity I:

$$I \simeq [\cos(3\varphi + \alpha) + T(3\cos\alpha\cos\varphi - \sin\alpha\sin\varphi)]^2 \tag{5}$$

where $T = (\chi_{11} + \chi_{12})/(\chi_{11} - 3\chi_{12})$ for (210) films and $T = 0$ for (111) films. For the derivation of (5) we assumed that the optical anisotropy of the film is small, i.e. we set $\epsilon_{ij} = \epsilon_0\delta_{ij}$. This anisotropy is usually of the order of 10^{-4} and thus may be neglected in our case. We also ignored a magnetooptical rotation of the polarization of the light since all experiments were made on demagnetized or multi-domain samples. The Kleinman relation $\chi_{12} = \chi_{26}$ was used when deriving (5).

3. Experimental details

The SHG was studied in transmission geometry at normal incidence (see figure 2). We used a Q-switched Nd-YAG laser ($\lambda = 1.064$ μm) with the pulse length $\tau_p = 8$ ns and the pulse energy $W_p = 5$ mJ. No attempt was made to measure the absolute values of the SHG intensity, nor to find the phase-matching conditions. Qualitative measurements showed that the SHG intensity in (210) films was one to two orders of magnitude lower than in a sample of crystalline quartz.

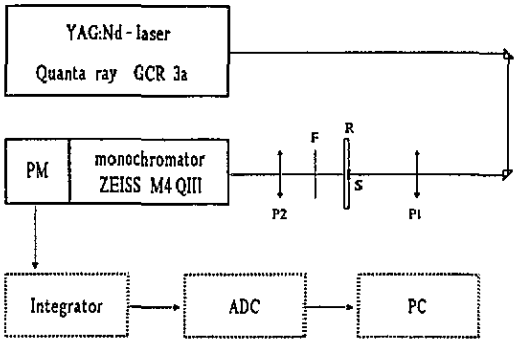

Figure 2. Experimental set-up: P1, linear polarizer for the fundamental wave; P2, polarizer for the SHG; S, sample; R, device for rotating the sample; F, optical filter for supressing the fundamental wave; PM, photomultiplier; ADC, analogue-to-digital converter; PC, personal computer.

Experiments were made on epitaxial magnetic garnet films grown on substrates of GGG with two orientations, (210) and (111). Some parameters of the sample are shown in table 1. Most of the experiments were performed at room temperature $T = 295$ K and some experiments on film No 2 were also carried out at $T = 400$–405 K. All experiments were carried out without any magnetic field, i.e. the samples were in a demagnetized state. The domain structure was observed with a polarizing microscope. In all samples except sample No 4 the domain structure was of the labyrinth type. No domain structure was seen in sample No 4, most probably as a consequence of its small thickness. The Curie temperature was measured in sample No 3 and was found to be $T_C = 415$ K.

Table 1. Characteristics of the films.

No	Film composition	Substrate orientation	Film colour	Film thickness (μm)	Domain structure	SHG intensity	SHG period (deg)
1	$(YBiPrLn)_3(FeGa)_5O_{12}$	(210)	Brown	10.5	Labyrinth	Strong	180
2	$(YBiPrLn)_3(FeGa)_5O_{12}$	(210)	Brown	~10	Labyrinth	Strong	180
3	$(YBi)_3(FeGa)_5O_{12}$	(111)	Brown	4.7	Labyrinth	Medium	60
4	$(YBi)_3(FeGa)_5O_{12}$	(111)	Light yellow	<1	No domain structure observed	Weak	60
5	$(YBiPrLn)_3(FeGa)_5O_{12}$	(111)	Brown	~5	Labyrinth	No signal detected	

4. Experimental results

Typical experimental results for (210)-type films are shown in figure 3. In two samples of this type SHG was rather easily detected and was reproducible after readjustment of the optical elements and samples. In figure 3 the angular dependence on φ of the SHG intensity is shown for three different values of the angle α between the polarization axes of the polarizer and the analyser. The dependence clearly shows a 180° periodicity as expected from (5). The results are well described quantitatively by (5), despite the approximations made in deriving this equation.

The SHG in film No 2 was also studied at high temperature ($T = 405$ K) for three different values of α ($\alpha = 0°$, 45°, 90°). No noticeable changes were found either in the intensity or in the shape of the SHG as a function of the angle φ, though this temperature is close to the magnetic transition temperature T_C =415–420 K to the paramagnetic state. No noticeable changes of the signal were observed after the subsequent cooling of the sample to room temperature. In our opinion these experiments prove a 'non-magnetic' mechanism of SHG in garnet films.

In two magnetic films of the (111) type the SHG signal showed a 60° periodicity as function of the angle φ, as expected from (5) (figure 4). In accordance with (5) the change of the angle α leads to a shift of the SHG curve by 30° along the φ axis, but in contrast to the (210)-type films, the shape of the angular variation does not change. The absolute intensity of the SHG in (111) films was lower than that in (210) films by at least a factor of ten. In one film of (111) type (No 5) no signal was detected.

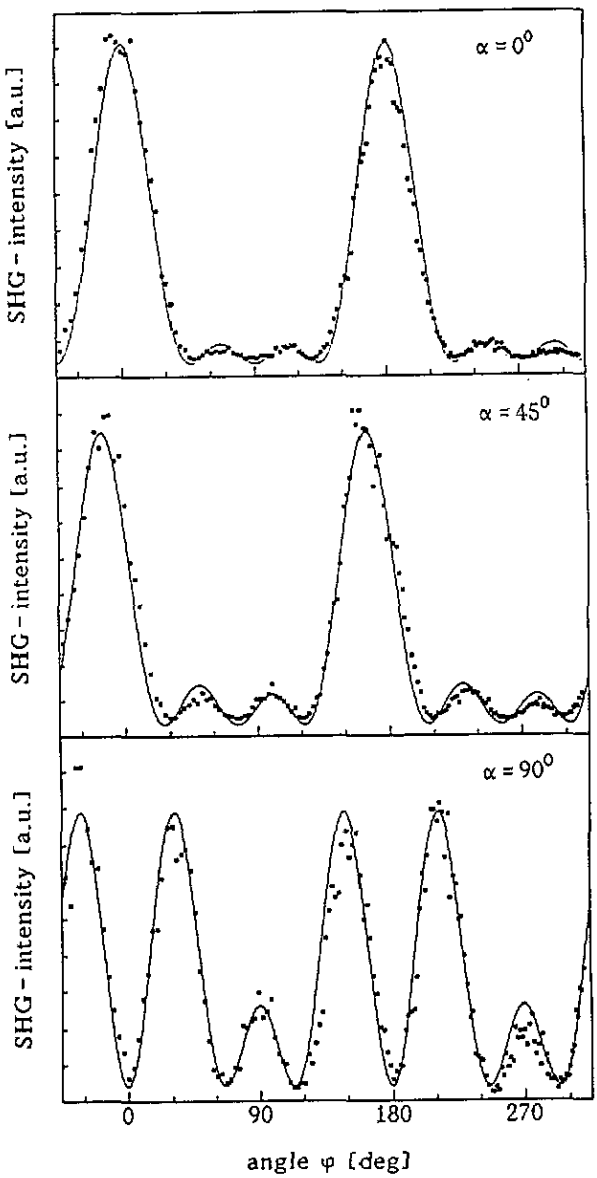

Figure 3. SHG intensity of a (210)-type film (No 2) as a function of the rotation angle φ around its normal. α is the angle between the polarization axes of the polarizer and the analyser. All theoretical curves were calculated by use of (5) ($T = 0.36$ K).

5. Discussion

A good agreement between the experimental data and the calculated angular variations of the SHG intensity (see figures 3 and 4) confirms that the origin of symmetry lowering in thin magnetic films is the substrate orientation and growth direction. The last point is probably of primary importance for destroying the inversion symmetry in the film structure. Although the SHG study cannot give a direct proof of the microscopic mechanism that destroys the

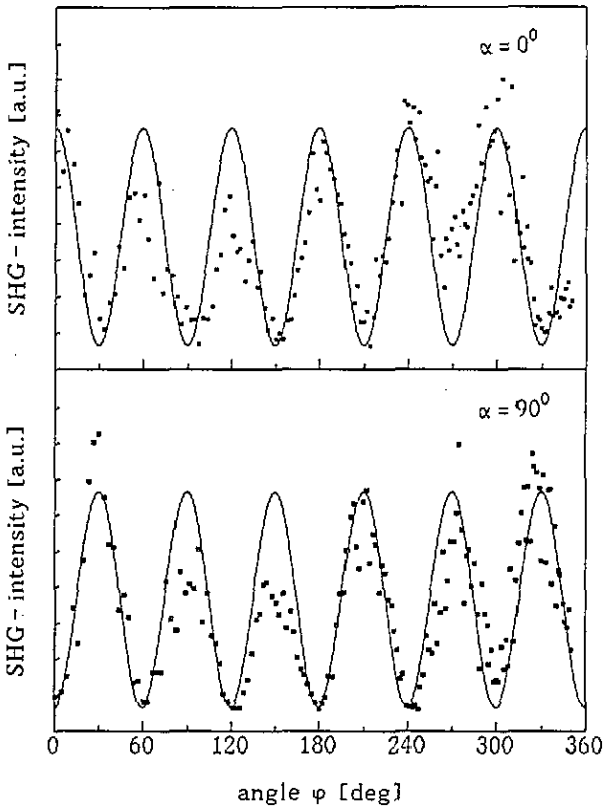

Figure 4. SHG intensity of a (111)-type film (No 3) as a function of the rotation angle φ around its normal. Theoretical curves were calculated by use of (5) ($T = 0$).

inversion symmetry, this study is of great help in solving this interesting problem. A more systematic study of SHG in thin films as a function of the substrate orientation, the lattice mismatch between substrate and film, the composition of the films, etc should be made.

Nevertheless it is possible to obtain SHG even in crystals with an inversion centre. As was pointed out by Terhune *et al* [8] higher-order effects due to electric-quadrupole or magnetic-dipole transitions can lead to frequency doubling. As higher-order effects they are expected to be much weaker than SHG due to electric-dipole transitions in a system without a centre of inversion. For our experiments we can rule out higher-order effects for at least two reasons. (i) If higher-order effects are detectable they should be seen in all films. We have seen no SHG in film No 5. (ii) The observation of a linear magnetoelectric effect [4, 5] has already proved the lack of an inversion centre. Although the microscopic mechanisms of the linear magnetoelectric effect and SHG should be completely different, the absence of an inversion centre is obligatory for both phenomena. It is also worth noting that in both cases the effects are substantially larger in (210) films than in (111) films. This is qualitative proof of larger non-centrosymmetric distortions in (210) films. A confirmation of this qualitative statement by other experiments sensitive to a loss of the inversion symmetry (e.g. piezoeffect or linear electrooptical effect) would be of great interest.

In a recent paper [6] a 'magnetic' mechanism was suggested as a possible source for the breaking of the inversion symmetry in magnetic garnets. This mechanism has also been discussed in theoretical papers [9–11]. In our opinion a magnetic mechanism may

be effective in antiferromagnetic crystals of a magnetoelectric type, e.g. in Cr_2O_3 in which below $T_N = 307$ K (among symmetry elements of the magnetic point group) the space–time inversion operation $\bar{1}'$ exists, but the space inversion $\bar{1}$ is forbidden. In ferrimagnetic materials, to which magnetic garnets belong, the breaking of the space inversion is possible only due to rare-earth sublattice ordering of a special kind [9] or to the lowering of the symmetry of the iron sublattices induced by a strong applied magnetic field (magnetic canted phase). Thus we believe that a magnetic mechanism should be excluded as a possible source for the breaking of the inversion symmetry in magnetic garnet films.

6. Conclusions

SHG is readily observed in transmission in magnetic garnet thin films on GGG substrates of (210) and (111) types at normal incidence of the pump radiation of an Nd–YAG laser. We have shown that symmetry properties of the SHG are well explained by taking into account the orientation of the substrate and assuming the existence of a polar axis along the growth direction, i.e. along the normal of the film. A non-magnetic mechanism of SHG in garnet films is also supported by the absence of any noticeable temperature dependence of the SHG intensity in the temperature range 295–405 K, in which the spontaneous magnetization strongly varies. We believe that SHG can be used as an effective tool in a more systematic study for developing adequate models of thin-film growth, structure and symmetry. Further studies with higher accuracy and in a magnetic field may also reveal magnetic contributions to the SHG in garnet films.

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