A spectroscopic study of the nonlinear magneto-optical response of garnets

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In magnetic garnet films crystallographic $\chi^{(2)}_{ijk}(2\omega)$ and magnetization-induced $\chi^{(3)}_{ijkl}(2\omega)$ electric-dipole type contributions to the nonlinear optical susceptibility may coexist due to a simultaneous breaking of space and time-reversal symmetry. A spectroscopic study shows different behavior of these two contributions as a function of the photon energy. © *1997 American Institute of Physics.* [S0021-8979(97)28308-8]

Though different theoretical issues of nonlinear optics of magnetic materials have been discussed since the early sixties (see Ref. 1 and references therein), nonlinear optical phenomena in magnetically ordered materials have been observed only recently. In the electric dipole approximation, bulk second harmonic generation (SHG) requires the breaking of space-inversion symmetry. 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.[2–8] Monolayer sensitivity, quantum well oscillations, and huge nonlinear magneto-optical effects were observed. Nonlinear Kerr rotation close to 90° was indeed observed for thin Fe films which should be compared to their linear rotation of about 0.04°.[9]

Bulk crystals of magnetic garnets like yttrium iron garnet $Y_3Fe_5O_{12}$, possess a crystallographic as well as a magnetic centrosymmetric structure.[10] Consequently, SHG is forbidden in the electric-dipole approximation. Thin films of magnetic garnets possess magnetic and magneto-optical properties different from those in bulk crystals. It was shown that in thin films of magnetic garnets the inversion symmetry may be broken due to a distortion of the crystal structure (see, e.g., Ref. 11). Therefore, such garnet films are very suitable materials to elucidate different mechanisms of nonlinear magneto-optical effects, not only for this group of materials, but for other magnetic materials as well. Recently we demonstrated the possibility to separate the crystallographic and magnetic contributions to the nonlinear optical response in these films.[12] The magnetic part is found to emerge below the transition temperature T_C, and at room temperature the two contributions are of the same order of magnitude. This article is devoted to a spectroscopical study of these different contributions. A clear difference in their wavelength dependence is observed.

In the electric-dipole approximation the nonlinear optical polarization $\mathbf{P}(2\omega)$ of a magnetic medium possessing a spontaneous magnetization $\mathbf{M}(0)$ can be written in the form

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

where $E_j(\omega)$ and $E_k(\omega)$ are the fundamental optical fields. A polar tensor $\chi^{(2)}_{ijk}$ of rank 3 describes the crystallographic contribution. It is allowed in crystals lacking inversion symmetry. An axial tensor $\chi^{(3)}_{ijkl}$ of rank 4 describes the magnetization-induced contribution. It is also allowed in noncentrosymmetric crystals. These two contributions to the nonlinear polarization $\mathbf{P}(2\omega)$ may coexist in the same medium. They are both spontaneous and no external field is required to observe them in a single-domain state. However, there are important differences between them. First, they possess different transformation properties under the symmetry operations of the medium, and as a consequence they vary differently when the incident polarization $\mathbf{E}_i(\omega)$ varies with respect to the crystal axes.[12,13] Second, these two contributions should vary differently as a function of temperature and magnetization.[12] It appears that they also have different behavior as a function of the photon energy. Indeed, specific tensor elements can be connected with specific parts of the energy band structure. Therefore it might be possible to look for the origin of the nonlinear (magneto-) optical response in this way.

The magnetic films were grown by a liquid phase epitaxial method. The (111) films of the composition $(YLuBi)_3(FeGa)_5O_{12}$ were grown using thin wafers of gadolinium gallium garnet $Gd_3Ga_5O_{12}$ (GGG) as substrates. The (210) films with the composition $(YPrLuBi)_3(FeGa)_5O_{12}$ were grown on GGG substituted wafers with a larger lattice parameter.

The SHG signal was generated by the output 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 in the wavelength range of 720–860 nm. The use of this type of laser gave a significant increase of the SHG output as compared to previously reported experiments performed with longer pulses (10–12 ns) and longer wavelength (1.06 μm) from Nd-YAG lasers.[14–16] Using the same experimental setup we could compare the relative SHG intensities from magnetic garnet

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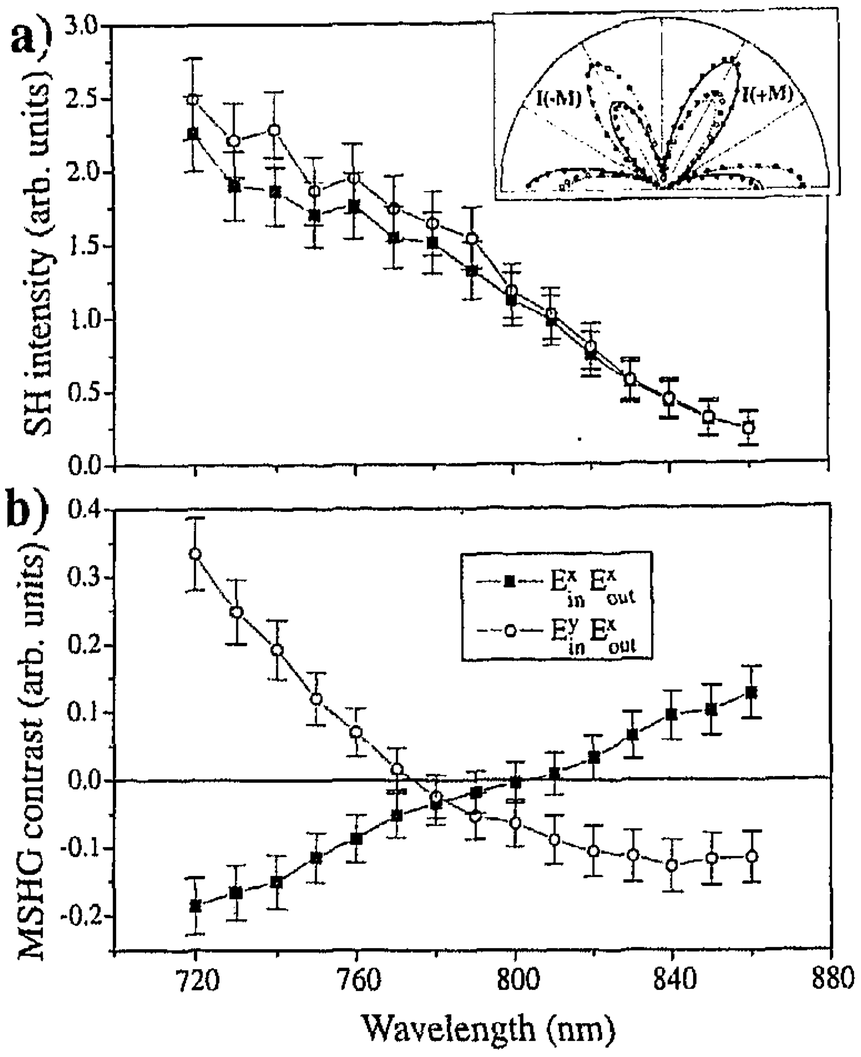

FIG. 1. Second harmonic intensity (a) and MSHG contrast (b) for the (111)-oriented garnet film as a function of the incident wavelength and for different polarization combinations (indicated in the figure). Inset shows the rotational anisotropy pattern (*XX* polarizations) for opposite directions of the applied magnetic field.

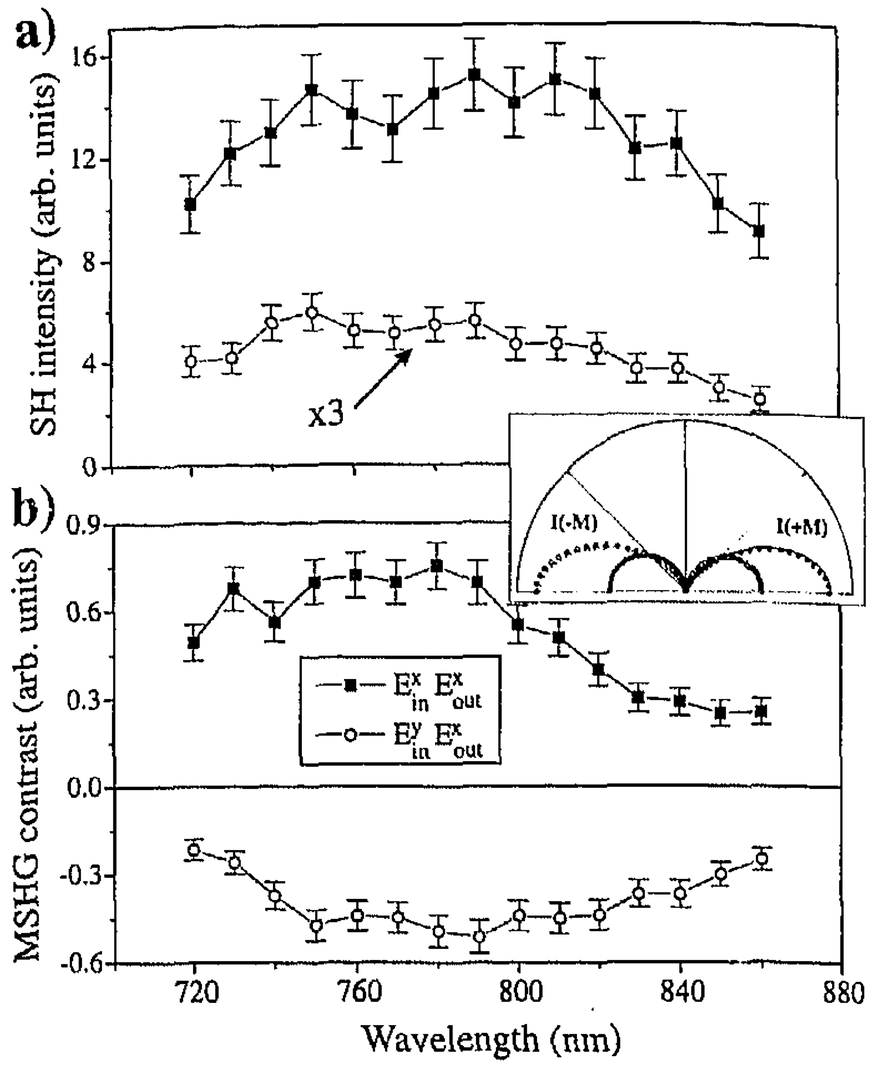

FIG. 2. The same as Fig. 1 but for the (210)-oriented sample.

films and from ferromagnetic metal surfaces. The SHG signals were several orders of magnitude higher in the garnet films using the same excitation power. This is directly related to the fact that for the garnet films we deal with a *bulk* response. At λ=720–860 nm (1.44–1.72 eV) the linear absorption coefficient of magnetic garnet films and bulk crystals is of the order of $\alpha \approx 10$–$20\ \mathrm{cm}^{-1}$,[10] but at the frequency of the second harmonic $\alpha \approx 10^4\ \mathrm{cm}^{-1}$. Therefore in transmission experiments the detected SHG signal can only escape from a backside layer with a thickness of about 1 μm. Under such circumstances the phase-matching conditions are unimportant.[1]

Most of the experiments were done in transmission geometry with a laser beam propagating along the z axis and with a magnetic field up to $H=2.3$ kOe applied along the Y axis in the film plane. Rotating the sample by 360° around the Z axis we could register the rotational anisotropy of the SHG signal with the magnetization being kept along the Y axis and the incoming and outcoming linear light polarization being fixed along the X or Y axis. Insets in Figs. 1 and 2 show the anisotropic MSHG response for the (111) and (210) films, respectively, for $\mathbf{M} \| Y$ and $\mathbf{M} \| -Y$. The plots are symmetric with respect to the horizontal axis. Clearly, these dependencies reflect the symmetries of the corresponding surfaces. Such an approach allows unambiguous separation between the crystallographic and magnetization induced SHG contributions due to their different transformation properties.[12] For every wavelength, we measured both second harmonic intensity (normalized by a reference signal from a quartz crystal) and MSHG contrast.

Figure 1 shows the generated second harmonic intensity and MSHG contrast as a function of the incident light wavelength for the (111)-oriented sample. Here, the magnetization-induced contrast is defined as

$$\rho = \frac{I(+\mathbf{M}) - I(-\mathbf{M})}{I(+\mathbf{M}) + I(-\mathbf{M})} \tag{2}$$

measured at the azimuthal angle where the signal has the maximum [60° for the (111) sample and 0° for the (210) one]. The same data for the (210)-oriented sample are plotted in Fig. 2. In spite of the similar linear optical properties of these samples, there is a considerable difference between their second harmonic behavior. What is however more important for our present consideration, one can see [at least, for the (111) sample] that the SHG intensity and the MSHG contrast vary differently as a function of wavelength for the same sample. Although the spectral dependence of the SHG intensity for both polarization combinations is very similar, the MSHG contrast behaves very differently for the two cases. Whereas for the *XX* polarization combination ρ monotonously decreases with increasing energy, changing sign at $\lambda \approx 800$ nm, the opposite happens for the *YX* polarizations. From the rotational anisotropy equations (see Ref. 12) and formula (2) we can obtain the following equations for ρ:

$$\begin{aligned} \rho_{XX}(2\omega) &= 4CM/(2A + BM^2), \\ \rho_{YX}(2\omega) &= -\tfrac{4}{3}CM/(2A + \tfrac{2}{9}BM^2), \end{aligned} \tag{3}$$

where A, B, and C are combinations of the real and imaginary parts of the nonlinear susceptibility tensor elements. The opposite signs of the MSHG contrast for the two contributions in these equations are nicely confirmed by the experiments for both (111) and (210) samples [Figs. 1(b) and 2(b)]. On the other hand, Eq. (3) predicts the values for ρ to be approximately three times different ($2A \gg BM^2$, see Ref. 12), while experimentally this ratio is considerably smaller. This may be partly due to the effects of the local fields. More quantitative study would therefore be very desirable.

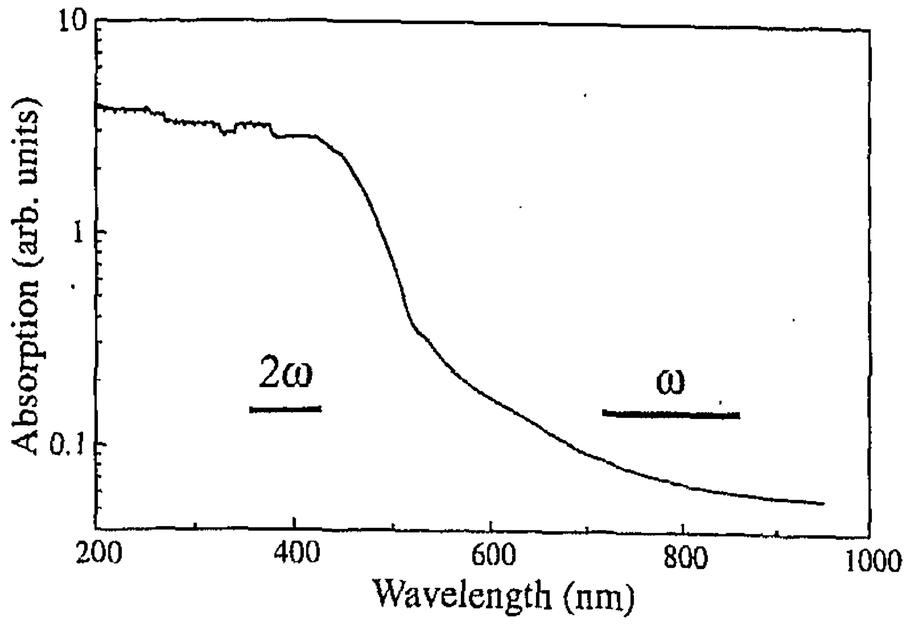

FIG. 3. Absorption spectrum measured on (111)-oriented garnet film.

It is interesting to compare the observed second harmonic behavior with the absorption spectrum for the same sample (see Fig. 3). While the latter one is rather flat for the second harmonic wavelength region (marked 2ω), MSHG demonstrates a strongly dispersive behavior. However, the dependence of the SHG intensity as a function of wavelength is similar (but stronger) to the absorption curve in the region of ω and is therefore probably related to one-photon absorption. As this also plays a role in the local field effects (that end up to the 4th power in the output intensity) this may explain the stronger wavelength dependence of MSHG relative to that of absorption. One should also mention that the SHG process is governed by selection rules which are different from those in linear optics. This fact may also lead to the different behavior of one-photon and two-photon optical properties.

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