Optical and magneto optical activity on partially sintered $\text{Y}_3\text{Fe}_5\text{O}_{12}$ materials

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Abstract

The optical and magneto-optical activities of yttrium iron garnet are shown to depend on the sintering stage. Low density yttrium iron garnet presents a lower optical activity than the high density garnet. The magneto-optical activity decreases at energies in which the optical transitions are associated to crystal field, indicating a relationship between the optical activity and the distortion of the structure. Finally, a calculation routine has been given to obtain the different elements of the dielectric tensor in bulk materials.

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Introduction

Yttrium iron garnet (YIG) shows a narrow ferromagnetic resonance in microwaves which renders it useful to design microwave devices such as resonators, oscillators, phase shifters and even metamaterials [1-3]. In the visible range, it shows an important magneto-optical activity combined with a low absorption. For this reason, it has been used as the ferromagnetic element in the novel field of magneto-plasmonics in which non ferromagnetic noble metal nanostructures embedded in a magneto-optically active matrix enhance the magneto-optical response of the system [4,5]. In this case, the coupling of magnetism with the collective oscillations of free electrons of noble metals leads to an increase of the magneto optical properties of the matrix. In particular, the combination of YIG with noble metals or doped with different elements has led to an enhancement of its magneto-optical response or has allowed tailoring its optical properties [6-10].

However, the optical and magneto-optical characterization are usually performed in different set ups, which may lead to errors in case the sample presents surface inhomogeneities. Since the advent of generalized ellipsometry and magneto-optical ellipsometry [11-16], it is possible to obtain both the values of the complex refractive index, N , and the complex magneto optical constant, Q , in a single set up without changing the sample position or taking reference measurements as in the reflectometry set ups, which is of particular importance when the system presents inhomogeneities as in the case of wedged samples [17]. In this paper, the calculation of the magneto optical constant, Q , from two measurements under an in plane saturating magnetic field in the transverse Kerr configuration is shown. It is also shown how the magneto optical activity is decreased in not fully dense YIG materials.

Experimental

YIG ferrite powders having stoichiometric compositions of $\text{Y}_3\text{Fe}_5\text{O}_{12}$ were prepared by the solid state reaction method. The raw materials, Fe_2O_3 (Aldrich, 99.9% purity) with the particle size of around 5.0 μm and Y_2O_3 (Cerac, 99.99% purity) with the particle size of around 9.0 μm , were mixed in propanol and ball milled for 1 h. The powders were calcined in air at 1200 °C for 2 h to obtain pure yttrium iron garnet. These powders were uniaxially pressed at 30 MPa and sintered at a heating rate of 50 °C min^{-1} in a spark plasma sintering apparatus (FCT-HP D25/1) under an applied pressure of 80 MPa and in vacuum (10^{-1} mbar). The final sintering temperatures were 900 and 925 °C, with holding times of 1 and 15 min, respectively.

The density of the specimens was measured by powder picnometry (model GeoPyc 1360, MICROMERITICS[®]) using a theoretical density of 5.172 $\text{g}\cdot\text{cm}^{-3}$ [18]. Powder X-ray diffraction analysis (D8 Advance, BRUKER) was used to determine the crystalline phases of the milled powder. The average size grain of the sintered samples was characterized by scanning electron microscopy (SEM, DSM950 Zeiss). The hysteresis loops were measured with an alternative gradient magnetometer (AGM, MicroMagTM, Princeton Measurements Corporation). Ellipsometry (SOPRA GES E5) was used to measure the dielectric constant of the sintered samples.

Results

Powders sintered at 900 and 925 °C present densities of 84 and 93 % as measured by powder picnometry. X-ray diffraction data (Fig. 1) show that in both cases, the diffraction peaks corresponding to the garnet phase are the only peaks observed. Just minor differences related to the structural quality are observed. In particular, the peaks at 33 and 34° are more intense and much better defined in the sample sintered at 925 °C,

which indicates a slightly better crystalline quality. The hysteresis loops (inset in Fig. 1) show that both samples are magnetically soft, as expected, and are already saturated around 1 kOe.

Fig. 1. X-ray diffractograms for two samples; one sintered at 900 °C for 1 minute and the other sintered at 925 °C for 15 minutes. Inset shows hysteresis loops for the same samples.

It is the aim of this work to determine how the processing conditions determine the optical and magneto-optical activity of the YIG materials. Fig. 2a and b show the ellipsometry data, $\tan\psi$ and $\cos(\Delta)$ as a function of the saturating magnetic field applied (2.5 kOe). As can be observed, just minor differences are due to the application of the magnetic field. However, large differences are observed between the samples, despite no relevant differences were shown in the X-ray diffraction data. The sample sintered at 925 °C shows better defined features, in particular, the $\cos(\Delta)$ value varies from 1 down to 0.4, whereas in the sample sintered at 900 °C, $\cos(\Delta)$ decreases from 1 down to 0.75, which is indicative of a high difference in the absorption. Provided that the transverse Kerr effect (TKE) is related to the absorption, important differences are expected between both samples.

By defining the TKE, taken from ellipsometry measurements as $TKE = 100 \cdot (\tan^2\Psi_+ - \tan^2\Psi_-) / (\tan^2\Psi_+ + \tan^2\Psi_-)$, no effect is observed below 2.5 eV (Fig. 2c), as

expected from a non absorbing sample. Also, a change in the sign of the activity is observed around 3.5 eV, as in the ellipticity in the polar configuration [19]. Whereas no relevant differences are observed in the magneto-optical activity between both samples over 3.5 eV, the samples sintered at 900 °C shows a lower TKE in the 2.5-3.5 eV range than the one sintered at 925 °C, which correlates well with the lower absorption of this sample in the same range. Obviously, this effect is not density related, as it is energy-dependent.

Fig 2. Ellipsometry parameters, $\tan(\Psi)$ (2a) and $\cos(\Delta)$ (2b), as a function of the sign of a saturating magnetic field and transverse Kerr effect (2c) for the sample sintered at 925 °C.

As the ellipsometric ratio is being measured, from these two measurements, with no need of repositioning the sample, varying the angle of incidence or taking a reference measurement, all the information concerning the complex refractive index, N , and the complex magneto-optical constants, Q , can be obtained. Experimentally, in an ellipsometry experiment, the following relationship is obtained

$$\rho = \frac{R_{pp} + R_{sp}\chi^{-1}}{1 + R_{pp}R_{ps}\chi} \quad (1)$$

where χ is the ratio of the amplitudes of the waves p and s (A_p/A_s) and, by definition,

$$R_{pp} \equiv \frac{r_{pp}}{r_{ss}} = \tan \Psi_{pp} \exp(i\Delta_{pp}) \quad (2)$$

$$R_{ps} \equiv \frac{r_{ps}}{r_{pp}} = \tan \Psi_{ps} \exp(i\Delta_{ps}) \quad (3)$$

$$R_{sp} \equiv \frac{r_{sp}}{r_{ss}} = \tan \Psi_{sp} \exp(i\Delta_{sp}) \quad (4)$$

where r_{ij} are the elements of the Jones matrix. According to the transfer matrix formalism developed by Zak et al. [20,21], $R_{ps} = 0$, which reduces eq (1) to:

$$\rho = R_{pp} + R_{sp}\chi^{-1} = \frac{r_{pp}}{r_{ss}} + \frac{r_{sp}\chi^{-1}}{r_{ss}} \quad (5)$$

Also, from references [20] and [21], for a substrate, the following expressions for r_{pp} , r_{sp} and r_{ss} can be deduced:

$$r_{pp} = \frac{N \cos \theta - \alpha_z - i\alpha_y Q}{N \cos \theta + \alpha_z + i\alpha_y Q} \quad (6)$$

$$r_{ss} = \frac{\cos \theta - N\alpha_z}{\cos \theta + N\alpha_z} \quad (7)$$

$$r_{sp} = \frac{i\alpha_y Q}{\alpha_z(N\alpha_z + \cos \theta)(N \cos \theta + \alpha_z + i\alpha_y Q)} \quad (8)$$

where, Q is the magneto optical constant, that relates to the diagonal and off diagonal components of the dielectric tensor through $\varepsilon_{xy} = iQ\varepsilon_{xx} = iQN^2$, $N = n + ik$, the complex refractive index, θ is the angle of incidence, $\alpha_y = \frac{\sin \theta}{N}$ and $\alpha_z = \sqrt{1 - \alpha_y^2}$.

Considering that r_{ss} has no dependence with Q and usually $|Q| \ll 1$, one gets that, to a very good approximation, r_{sp} is odd in Q and r_{pp} has a term that does not depend on Q

plus a term linear in Q . Therefore, by averaging ρ obtained at a positive applied field (ρ_+) and ρ obtained at a negative applied field (ρ_-), $\rho_0 = (\rho_+ + \rho_-)/2$ (valid up to terms in $|Q|^2$), the refractive indices, and therefore the diagonal elements of the dielectric tensor, are obtained

$$N = \sin \theta \sqrt{1 + \left(\frac{1 - \rho_0}{1 + \rho_0} \right)^2 \tan^2 \theta} \quad (9)$$

As expected from Fig. 3a and b, the optical absorption is significantly lower in the sample sintered at 900 °C than in the sample sintered at 925 °C. This implies that differences in the magneto-optic behaviour, the origin of which arises from a difference in absorption between right and left circularly polarized light, between the samples are expected.

Figure 3.- Real (a) and imaginary (b) part of the refractive index and off-diagonal components of the dielectric tensor (c).

On the other hand, substituting the values in equations (6-8), one finds that, discarding terms on $|Q|^2$ and higher:

$$\rho_+ - \rho_- = \frac{2i\alpha_y Q \cdot (\cos\theta + N\alpha_z)}{(\cos\theta - N\alpha_z) \cdot (N\cos\theta + \alpha_z)^2} \left[\alpha_z - N\cos\theta + \chi^{-1} \frac{N\cos\theta + \alpha_z}{\alpha_z \cdot (N\alpha_z + \cos\theta)} \right] \quad (10)$$

from which the magneto-optical constant Q , and the off diagonal elements by applying $\varepsilon_{xy} = iQN^2$, can easily be obtained (Fig. 3c). Again, the most relevant differences are found in the 2.5-3.5 eV energy range, which is associated to crystal field transitions [22]. This result suggests that when a material is quenched during sintering, before reaching high densities, due to the operating mass transport mechanisms, the crystal structure is partially distorted to a point that is not easily identifiable by X-ray diffraction methods, but enough to induce important changes in the optical and magneto-optical behaviour of the material. Once the samples are sintered to high densities and there is basically no mass transport, the optical activity goes to bulk.

Conclusions

In conclusion, it has been shown that the magneto-optical activity is modified during the process of sintering, although X-ray diffraction shows no relevant differences. However, ellipsometry and magneto-optical ellipsometry have been shown to be valuable tools to detect those changes. In particular, yttrium iron garnet poorly sintered samples, quenched in the initial sintering stage, show a lower optical activity than high density samples, quenched in the final sintering stage. In the same sense, the magneto-optical activity is severely affected at energies in which the optical transitions are associated to crystal field transitions, which indicates a relationship exists between the optical activity and the distortion of the structure at an atomic level. Finally, a

formalism that allows calculating the magneto-optical constants from ellipsometry experiments in bulk materials based on the transfer matrix formalism has been established.

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