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Magneto-optic iron-garnet thin films for integrated optical applications

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The presence of oxygen during deposition and annealing affects both the structure and composition of yttrium-iron-garnet thin films.

Optical isolators allow polarized light to transmit in only one direction. These devices are used to prevent unwanted feedback into photonics components such as amplifiers and lasers. The realization of optical isolators in an integrated waveguide configuration, however, while still a challenging proposition, will potentially result in reduced costs and dimensions, as well as in increased integrability and improved fabrication yield for integrated optics. Yttrium iron garnet ($Y_3Fe_5O_{12}$, or YIG) and its substitutes are among the materials of choice for integrated optical isolators due to their high Verdet constant, i.e., the nonreciprocal Faraday rotation of the polarization experienced by light in the presence of a magnetic field.

Pulsed laser deposition (PLD) is potentially the technique of choice for fabricating ferrite thin films, primarily because of its proven simplicity and efficiency.[1] However, PLD-fabricated garnet films on non-garnet substrates are usually amorphous and require post-deposition annealing. Here we demonstrate that the effect of oxygen inclusion is actually a major component in these processes. Interestingly, very few studies have been conducted to understand why and how oxygen affects the microstructure of magneto-optic thin films, which is closely linked to their physical characteristics. We investigate the effect of oxygen in both the deposition and the subsequent thermal annealing steps.

To study the role of oxygen during the fabrication process, we performed a series of six depositions at a fixed temperature of 800°C, while setting the oxygen pressure at a value of either 0 or 50mTorr. (More precisely, what we call 0mTorr is actually the background pressure in the deposition chamber, which was ~0.7mTorr). The detailed list of the deposition parameters is reported elsewhere.[2]

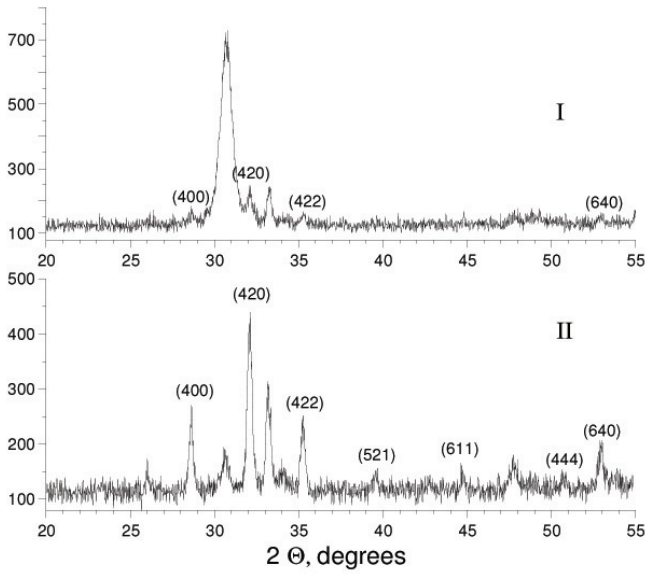

Figure 1. *X-ray diffraction spectra for sample 5 of an yttrium-iron-garnet (YIG) thin film after post-deposition heat treatment. (I) After first anneal in Ar; (II) after second anneal in air. The YIG peaks are denoted with corresponding (h k l) values. On the y axis: counts per second.*

We studied the structure of the samples using x-ray diffraction, and used energy-dispersive x-ray spectroscopy together with Rutherford backscattering spectroscopy for compositional analysis.

We performed the post-deposition heat treatment (anneal) for all six samples. To study the effect of oxygen during anneal, different Ar/O_2 gas ratios were used. See Table 1 for the list of

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Table 1. *Annealing parameters*

Sample (series)	Temperature, °C	Atmosphere	Time, h	Comments
1 (A)	800	Ar, air, O_2	2+2+2	no YIG, similar results after three different anneals
2 (A)	800	O_2	1.5	no YIG found
3 (A)	800	air	1	no YIG found
4 (A)	1000	O_2	1.5	no YIG found
5 (B)	800	Ar, air, O_2	2+2+2	YIG + concurrent phases
5 (B)	1000	air	1.5	4^{th} anneal, no change
6 (B)	800	air	1	regular YIG structure

parameters. We denote the four films deposited in vacuum (i.e., at 0mTorr of oxygen pressure) as the A series, and the two samples deposited at oxygen pressure of 50mTorr as the B series.

The as-deposited films from the A series were characterized by three crystalline phases: Y_2O_3; Fe as a ferrite phase; and Fe_2O_3. The films from the B series were mostly amorphous after deposition.

After annealing, A-series films demonstrated similar diffraction patterns, regardless of the four different deposition conditions, which came mostly from orthorhombic $FeYO_3$. In the annealed samples from the B series, a regular YIG polycrystalline structure with minor traces of other phases was detected only in sample 6. Whereas some YIG peaks were identified after the first annealing in argon, film 5 was characterized by the presence of $FeYO_3$ and Fe_2O_3. These YIG peaks were not as high and narrow as those in sample 6. Afterward, sample 5 was annealed again in air, then annealed a third time in pure oxygen at 800°C, and finally a fourth time in air, but at 1000°C. The most prominent evolution that we observed was the further growth of the YIG phase after the second annealing (conducted in air), as shown in Figure 1.

Prior to annealing, we found that the amount of oxygen in the A-series films was less than that in the B series, as would be expected: 58 and 73% at., respectively. Yttrium and iron concentrations were similar to one another for both series, with the iron content being 2–3% higher than that of yttrium. After annealing, both series demonstrated similar compositions, i.e., an oxygen content ranging from 68 to 73% at., and a higher concentration of iron with respect to yttrium (still in the 2–3% range).

In conclusion, we demonstrated that oxygen is important for forming YIG during pulsed laser deposition as well as during the subsequent post-deposition annealing. Without significant oxygen in the annealing atmosphere, what forms is generally not high-quality polycrystalline YIG, but a highly distorted lattice structure that includes other phases (as shown in Figure 1). Conversely, we showed that temperature has much less impact on this process.

Oxygen concentration in the as-deposited films fabricated in vacuum (the A series) is close to that of stoichiometric YIG, although none of the anneals that we performed led to the formation of this specific phase. Instead, $FeYO_3$ formed, in agreement with the typical triple phase diagram for Fe_2O_3, Y_2O_3, and Fe when iron is deficient.[3] We believe, therefore, that formation of regular YIG is due to an excess of oxygen. Indeed, a similar deficiency in iron content was also observed for the films belonging to the B series, yet perfect polycrystalline YIG was detected there.

We also note that the concentration of oxygen should be maintained above the stoichiometric value during the entire annealing process. Otherwise, the content of this gas in the films tends to reduce continuously because of high diffusion mobility, and the final result is formation of a degraded YIG structure.

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Michael Zaezjev received his PhD in physics for the slow-neutron study of oxygen in liquid potassium performed at Dubna, Russia. Since moving to Canada he has worked in the field of magneto-optic materials, developing an integrated optical isolator device.

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