RBS and XRD Characterization of Yttrium Iron Garnet Thin Films

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Abstract. Magnetic materials such as yttrium iron garnet (YIG or Y3Fe5O12) present a great importance for their magneto-optic properties. They are potential materials used for applications in the domain of optical telecommunications for example. In this work, we have investigated YIG thin films deposited on substrates of quartz and GGG (gadolinium gallium garnet or Gd3Ga5O12). Using Rutherford backscattering spectrometry (RBS) we characterized the performed layers (thickness and stoichiometry) in order to correlate the films preparation conditions with the quality of the final material. We determined the optimal energy of the alpha particles beam used for RBS measurements and we fitted the experimental spectra using the SIMNRA simulated code. Our RBS results showed that the films have a stoichiometry close to that of the starting material. In addition, we found that the film thickness is proportional to deposition time but inversely proportional to the substrate temperature. Moreover, using x-ray diffraction (XRD) we determined the annealing effect on the structure of the profile of our thin films.

1. Introduction
Recent studies conducted on the preparation of YIG thin films have shown its importance especially in fields related to the field of magneto-optical applications in microwave frequency domains [1, 2]. The YIG is a ferrimagnetic garnet crystal with the composition of Y3Fe5O12, with a substantial Faraday rotation in large parts of the optical and microwave spectrum [3]. In our work, YIG thin films were grown using the technique of radio frequency (RF) magnetron sputtering [2-4]. This technique possesses several advantages as being a dry process during deposition, and having the possibility of high purity starting material, ability to sputter dielectric materials, commonly held industrial process and high compatibility with semiconductors technology. The radio frequency sputtering films were grown on quartz and GGG substrates. The Rutherford backscattering technique (RBS) has proven to be very effective for characterization of the thickness and stoichiometry of thin films [5, 6] having an advantage of being nondestructive. This technique allows us to correlate the quality of the films to the different growth conditions and choose the optimal conditions. The x-ray diffraction technique (XRD) serves as a powerful tool to investigate any variation in the structure of the prepared films due to the annealing process [7].

2. Experimental Techniques:
The samples were prepared by radio frequency (RF) magnetron sputtering. Table.1 shows the different sputtering conditions for different prepared samples.
Table 1 Sputtering conditions for different prepared samples

| Target          | Y₃Fe₅O₁₂                |
|-----------------|-------------------------|
| Substrate       | Quartz or GGG           |
| Argon flow      | 50sccm                  |
| Input power     | 100W                    |
| Sputtering time | between 1h30 and 3h00   |
| Annealing       | 740 °C                  |
| Annealing       | Air                     |
| Annealing       | 2h                      |
| Substrate position | 4.6 cm                |

The Rutherford back scattering measurements were performed using a 5SDH pelletron tandem accelerator of 1.7 MV located at the Lebanese Atomic Energy Commission under normal incident beam and in a random direction to avoid channeling. The measurements were done by using two different energies of alpha-particles beam: 2 MeV and 3.5 MeV.

In our experiments, X-ray measurements were done using a D8 Discover X-ray diffractometer from Bruker AXS systems at the Central Research Laboratory (CRSL) in the American University of Beirut (AUB). The X-ray tube emits radiation of wavelength $\lambda = 1.5418$ Å.

3. Results and discussions:

The different RBS spectra were processed with the SIMNRA simulation code [8]. Indeed, RBS measurements are insensitive to the chemical state of the atoms and the quantitative interpretation of the measurement results is absolute and in most cases unambiguous, as the physical processes underlying ion-matter interactions are well understood [9]. The use of the classical RBS at 2 MeV He⁺⁺ beam was not useful for some of the samples that have thicker YIG layer. However, the use of alpha particle beam at 3.5 MeV, with 5 µC fluence, was enough to determine the YIG layer thickness for all prepared samples. Usually, the sensitivity on the thickness determination for a thin film is better at 2 MeV than 3.5 MeV, as the electronic stopping power $(dE/dx)_e$ is higher. However, when comparing the thickness of some of the thinner YIG layers obtained by both energies, the difference in values was found to be less than 3%. Besides, especially for GGG substrate, the O yield in the 2 MeV RBS spectra was overlapped by those of Gd and Ga. Hence, the use of 3.5 MeV enhanced the oxygen yield, since at this energy the oxygen cross section is non-Rutherford, while those of Ga and Gd are still Rutherford. The main results obtained from the RBS measurements after comparing the two substrates are that, the two substrates, the film thickness showed a similar trend and almost the same values were obtained for samples taken in the same conditions (Table 2). One can see that the thickness is proportional to the deposition time when comparing $yq_1$ vs. $yq_2$ and $yg_1$ vs. $yg_2$ grown during 180 minutes and 90 minutes, respectively. However, the general tendency is to be confirmed in future work by considering other values of the deposition times.
Fig. 1 illustrates the variation of the film thickness versus substrate temperature for quartz and GGG. There is a decreasing trend with increasing temperature so the thickest layers were obtained at room temperature.

**Table 2** Thickness, determined by RBS, of the YIG film deposited on quartz (yq) and GGG (yg) as function of the deposition time and the substrate temperature

| Sample | Substrate | Deposition | Film |
|--------|-----------|------------|------|
| No     | temperature (°C) | time (min.) | thickness (μm) |
| yq₀    | 25        | 90         | 890  |
| yq₁    | 500       | 180        | 1137 |
| yq₂    | 500       | 90         | 830  |
| yq₃    | 800       | 90         | 727  |
| yq₄    | 25        | 90         | 900  |
| yq₅    | 500       | 180        | 1180 |
| yq₆    | 500       | 90         | 812  |
| yq₇    | 800       | 90         | 785  |
| yg₀    | 25        | 90         | 900  |
| yg₁    | 500       | 180        | 1180 |
| yg₂    | 500       | 90         | 812  |
| yg₃    | 800       | 90         | 785  |

Fig. 1 illustrates the variation of the film thickness versus substrate temperature for quartz and GGG. There is a decreasing trend with increasing temperature so the thickest layers were obtained at room temperature.

For quartz substrate, Fig. 2 shows the RBS spectrum of the yq₂ samples and the variation of the stoichiometric ratio of cation (Y+Fe)/anion (O) of the YIG films with the substrate temperature. The chemical composition of the films prepared at T = 25°C, 500°C and 800°C are Y₂.5₁Fe₅.4₂O₁₂, Y₂.5₁Fe₅.3₅O₁₂ and Y₃.₆₆Fe₅.₈₅O₁₂, respectively. At 500°C and 800°C, the sputter yield of the positive ions (Y+Fe) is higher than that of the negative ions (O), so the ratios cation/anion of the films (0.74 and 0.79 respectively) are higher than that of the target (0.67). In a general manner, the room
temperature sample is the closest to the stoichiometric YIG target and the ratios \((Y+Fe)/O\), \(Y/O\) and \(Fe/O\) are increasing when the substrate temperature increases.

Figure 2. Shows the experimental and simulated RBS spectra obtained for the film grown on quartz substrate sample \(yq\) showing the edge of \(Y, Fe, Si\) and \(O\) and the variation of the chemical stoichiometry (expressed as cation/anion ratio) of the film with quartz substrate temperature.

Fig.3 shows the RBS spectrum of the \(yg_0\) sample prepared at room temperature and the variation of the different cation/anion ratio. A similar behaviour of the quartz substrate is also observed for the GGG substrate. The chemical composition of the films prepared at \(T= 25^\circ C, 500^\circ C\) and \(800^\circ C\) are \(Y_{3.07}Fe_{5.24}O_{12}\), \(Y_{3.38}Fe_{5.32}O_{12}\) and \(Y_{3.50}Fe_{5.47}O_{12}\), respectively. At room temperature, the chemical composition of the YIG film is the nearest to the stoichiometric YIG target. In general, the YIG films elaborated on GGG substrate are closer to the stoichiometric YIG target than those elaborated on quartz substrate.

Figure 3. Shows the experimental and simulated RBS spectra obtained for the film grown on GGG substrate sample \(yg_0\) showing the edge of \(Y, Fe, Gd, Ga\) and \(O\) and the variation of the chemical stoichiometry (expressed as cation/anion ratio) of the film with GGG substrate temperature.
The GIXRD obtained spectra of YIG samples grown on quartz are shown in figure 4. The XRD results show the dependence of the crystallinity structure with the substrate temperature.

![Figure 4](image1.png)

**Figure 4.** Shows the annealing effect on the structure of the profile of our samples. This effect can be shown by comparing the results with and without annealing and the deposition temperature effect with annealing at 740°C.

4. Conclusion
The application of the RBS technique was very helpful for the study of stoichiometry and thickness of YIG thin films deposited, by RF sputtering, onto quartz and GGG substrates. This study was necessary to clarify some trends of YIG film growth depending on the sample preparation conditions. Indeed, it was found that the film thickness is proportional to the deposition time while it is inversely proportional to the substrate temperature. The films grown on GGG substrate are closer to the stoichiometric YIG than those grown on quartz and a better stoichiometry is obtained at room temperature for both substrates. The preliminary results of XRD show the dependance of the cristallinity structure with the substrate temperature.

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