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ScienceDirect
Applied Surface Science 253 (2006) 2108–2112
applied surface science
www.elsevier.com/locate/apsusc

Effect of post-annealing temperature on the microstructure and magnetic properties of Ce:YIG thin films deposited on Si substrates

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Received 9 January 2006; received in revised form 29 March 2006; accepted 3 April 2006
Available online 15 May 2006

Abstract

Amorphous $Ce_1Y_2Fe_5O_{12}$ (Ce:YIG) thin films deposited on single crystal Si(1 0 0) and thermally oxidized Si(1 0 0) substrates by pulsed laser deposition were annealed in the temperature range of 700–1000 °C in air. The annealing temperature dependence of microstructure and magnetic properties of Ce:YIG films was studied using X-ray diffraction combined with vibrating sample magnetometer. The results show that single phase of polycrystalline Ce:YIG thin films can be obtained by the post-annealing of as-deposited films at the temperature of 700 °C. However, two steps of phase segregation of Ce:YIG occur as the post-annealing temperature increases: at first, Ce:YIG is decomposed into YIG and non-magnetic CeO_2 when annealed at 800 °C; then YIG continues to be decomposed forming Fe_2O_3 when the temperature is increased up to 900 °C. Consequently, the saturation magnetization of Ce:YIG films decreases first and then increases with the post-annealing temperature going up, which indicates that the saturation magnetization of Ce:YIG films is mainly related to the phase composition of the films. Meanwhile, the presence of SiO_2 buffer layer can significantly enhance the saturation magnetization of Ce:YIG films.

PACS: 81.40.Ef; 81.40.Rs; 75.70.Ak

Keywords: Ce:YIG thin film; Post-annealing; Microstructure; Magnetic property

1. Introduction

Yttrium iron garnet ($Y_3Fe_5O_{12}$, YIG) belongs to a group of magnetic oxides with large magneto-optic (MO) effect. Ce-substituted yttrium iron garnet ($Ce_xY_{3-x}Fe_5O_{12}$, Ce:YIG) thin films were considered to be of great potential for applications in high performance non-reciprocal waveguide devices, integrated optical devices and magneto-optic memory media due to their high Faraday rotation coefficient and low propagation loss [1–4]. Ce:YIG thin films grown by radio-frequency (RF) sputtering [5], liquid phase epitaxial (LPE) growth [6] and pulsed laser deposition (PLD) [7,8] have been extensively investigated in recent years. Crystallized Ce:YIG films have been epitaxially grown on garnet substrates such as gadolinium gallium garnet ($Gd_3Ga_5O_{12}$ GGG). Since silicon (Si) is now the most important material in integrated circuits, its use as a substrate may realize the combination of Si electronic devices and optical waveguide on the same Si substrate [9,10]. However, the Si substrates have a different crystal structure compared with YIG, which leads to a considerably high lattice mismatch, so post-annealing is pre-requisite to obtain polycrystalline Ce:YIG films on Si substrate. To the best of our knowledge, growth of Ce:YIG films on Si substrate has rarely been investigated up to now. Especially, the phase segregation phenomenon of Ce:YIG induced by high temperature annealing has not been reported so far. In this work, we first deposited an amorphous film on Si substrate by PLD; then studied the influence of post-annealing temperature on the microstructure and magnetic properties of Ce:YIG thin films. In addition, the effect of a SiO_2 buffer layer was studied. It is found that after post-annealing at 700 °C, single phase polycrystalline Ce:YIG thin films were obtained. However, phase segregation phenomenon of Ce:YIG can be observed when the post-annealing temperature continues to increase; the buffer layer of

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doi:10.1016/j.apsusc.2006.04.002

SiO_2 makes the saturation magnetization of Ce:YIG significantly enhance. Here we present the results of this work.

2. Experimental procedure

Target with the composition of $Ce_1Y_2Fe_5O_{12}$ has been fabricated from a uniform mixture of CeO_2, Y_2O_3 and Fe_2O_3 powders (99.9% purity for all) by standard procedure of pressing and solid state sintering at 1300 °C in air. The substrates included two sets: one was single crystal Si(1 0 0) and the other was SiO_2/Si achieved by thermally oxidizing the surface of single crystal Si(1 0 0) to form a buffer layer of SiO_2. The thickness of SiO_2 layers was about 550 nm, measured from the cross-sectional SEM image of SiO_2/Si bilayer structure. Both the two sets of Si wafers were simultaneously placed on the substrate holder which was electrically heated to 300 °C and rotated during deposition. Films were prepared by PLD using a 248 nm KrF excimer laser operated with a 25 ns pulse width and at 10 Hz repetition rate in a vacuum chamber starting with a base pressure under 6.0×10^{-4} Pa. The deposition rate was about 4.0 nm min^{-1}. The as-deposited films were cut into small pieces and were annealed in the temperature range of 700–1000 °C in air for 3 h.

Microstructure of both as-deposited and post-annealed Ce:YIG thin films was determined by a D/max 2550V X-ray diffractometer (XRD) using a Cu Kα source of 1.542 Å. Magnetization curves were obtained using an HH-15 vibrating sample magnetometer (VSM) with the magnetic field applied parallel to the film plane and operated at room temperature. The oxidation states of Ce ions and Fe ions in the films were investigated by X-ray photoelectron spectroscopy (XPS), which was carried out on a RBD upgraded PHI-5000C ESCA system (Perkin-Elmer) with Mg Kα radiation ($h\nu$ = 1253.6 eV). The data was analyzed by using the RBD AugerScan 3.21 software.

3. Results and discussion

3.1. Variation of microstructure with the annealing temperature

As revealed from the XRD θ–2θ scan, the as-deposited $Ce_1Y_2Fe_5O_{12}$ films show amorphous behavior which, however, become well crystallized after post-deposition heat treatment. Fig. 1 shows the XRD patterns of annealed films deposited on single crystal Si(1 0 0), where Si-700, Si-800, Si-900 and Si-1000 denote the films after annealing at temperatures of 700, 800, 900 and 1000 °C, respectively. It can be seen from Fig. 1 that the film of Si-700 exhibits the best crystallinity of polycrystalline $Ce_1Y_2Fe_5O_{12}$ in our experiment. The diffraction lines at 2θ of 28.9°, 32.4° and 35.6° indicate the presence of cubic garnet YIG phase corresponding to (4 0 0), (4 2 0) and (4 2 2) crystalline planes, respectively. Compared to that of Si-700, the YIG(4 0 0) peak of Si-800 broadens towards low 2θ values, indicating the presence of CeO_2. After annealing the film at a higher temperature of 900 °C, the intensity of CeO_2(1 1 1) peak is considerably enhanced. Furthermore, a

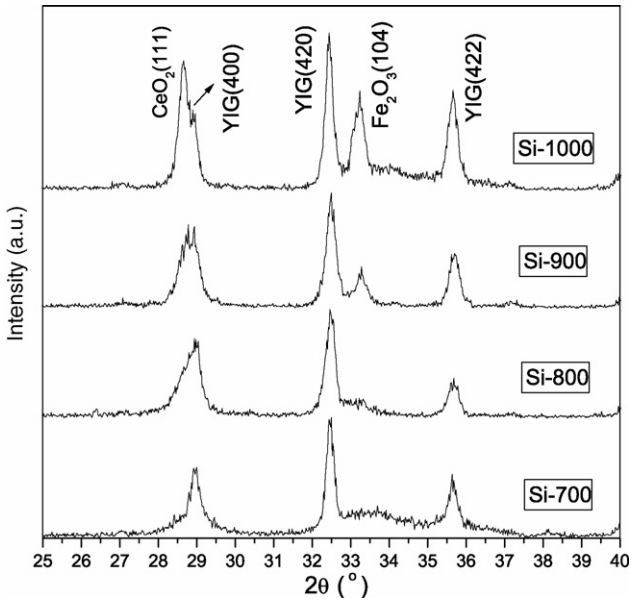

Fig. 1. XRD patterns of Ce:YIG films on Si(1 0 0) annealed at different temperatures.

new peak presenting Fe_2O_3(1 0 4) phase comes up at the 2θ angle of 33.3°. As the annealing temperature is raised to 1000 °C, both the peaks of CeO_2(1 1 1) and Fe_2O_3(1 0 4) boost up as can be seen from XRD pattern of Si-1000 in Fig. 1.

To improve the magnetic properties of the films, a buffer layer of SiO_2 was introduced. Fig. 2 illustrates the XRD patterns of films grown on SiO_2/Si. Similarly, the films after annealing at temperatures of 700, 800, 900 and 1000 °C are denoted as SiO_2-700, SiO_2-800, SiO_2-900 and SiO_2-1000, respectively. As determined from Fig. 2, it is also demonstrated that the phase segregation phenomenon of Ce:YIG occurs with the increase of annealing temperature, that is, the Ce:YIG decomposes into YIG and CeO_2 firstly, and then continues to

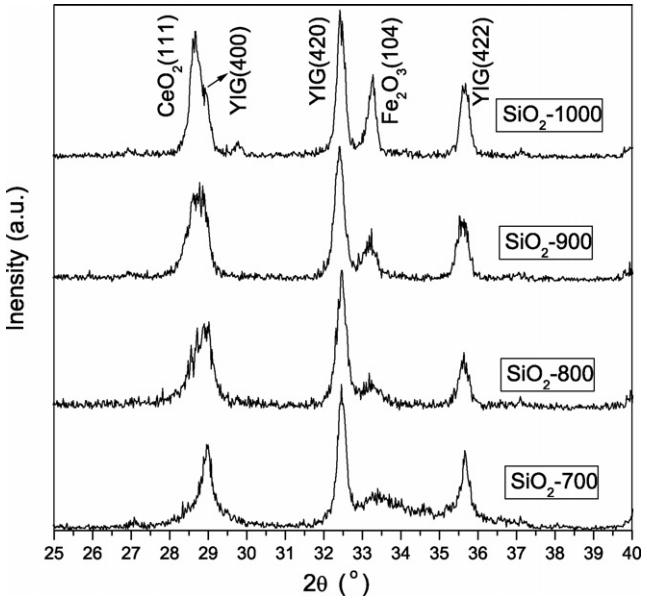

Fig. 2. XRD patterns of Ce:YIG films on SiO_2/Si annealed at different temperatures.