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Journal of Magnetism and Magnetic Materials 299 (2006) 307–311
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

Large second-harmonic kerr rotation in $GaFeO_3$ thin films on YSZ buffered silicon

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Received 21 February 2005; received in revised form 27 April 2005
Available online 25 May 2005

Abstract

Epitaxial thin films of gallium iron oxide ($GaFeO_3$) are grown on (0 0 1) silicon by pulsed laser deposition (PLD) using yttrium-stabilized zirconia (YSZ) buffer layer. The crystalline template buffer layer is in situ PLD grown through the step of high-temperature stripping of the intrinsic silicon surface oxide. The X-ray diffraction pattern shows *c*-axis orientation of YSZ and *b*-axis orientation of $GaFeO_3$ on Si (1 0 0) substrate. The ferromagnetic transition temperature ($T_C \sim 215$ K) is in good agreement with the bulk data. The films show a large nonlinear second harmonic Kerr rotation of $\sim 15^\circ$ in the ferromagnetic state.

PACS: 75.60.Ej; 78.20.Ls; 42.65.Ky

Keywords: Thin films; $GaFeO_3$; MOKE; Pulsed laser deposition; Magnetization

1. Introduction

Multifunctional materials have attracted significant interest in recent years due to the coupling of their ferromagnetic and piezoelectric/ferroelectric properties, which is of great interest from the fundamental as well as applied points of view [1–5]. $GaFeO_3$ (GFO) is an interesting example in this context, which exhibits ferromagnetic and pyroelectric properties simultaneously [6]. This material was first discovered by Remeika et al. [6] and its crystallographic properties (orthorhombic structure) were described by Abrahams et al. [7] on the basis of unit cell volume and space group given by Wood et al. [8]. Observation and possible mechanisms of magnetoelectric effects in this ferromagnet were described by Rado [9] and

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0304-8853/$ - see front matter
doi:10.1016/j.jmmm.2005.04.017

Frankel et al. [10], and a sizable magnetic anisotropy was reported in $Ga_{2-x}Fe_xO_3$ single crystals by Levine et al. [11].

Recently, magnetization-induced second-harmonic generation (MSHG) and X-ray directional dichroism have been investigated in single-crystal GFO [12,13]. Remarkably, large effects have been found due to the intrinsically non-centro-symmetric nature of this crystallographic system. For building technologically viable device systems it is important to develop such multifunctional materials as crystalline thin films on different substrates, most desirably on Si. In this paper we describe the growth, observation of large SHG Kerr effect, and the magnetic properties of GFO thin films grown by pulsed-laser deposition on crystalline YSZ-template-buffered Si. We also present results for the films grown on crystalline YSZ substrates.

2. Experimental

High-purity powders of Ga_2O_3 (99.999%) and Fe_2O_3 (99.998%) were ground in the agate-mortar in the proper stoichiometric ratio. The resultant mixture were cold pressed in the pellet form and calcined at 1050 °C for 24 h. These pellets were reground thoroughly, pelletized and sintered at 1150 °C for 24 h to obtain stoichiometric GFO. High-purity commercially available YSZ target was used for the corresponding buffer layer growth on (0 0 1) Si. A KrF excimer laser ($\lambda = 248$ nm) was used for ablation. The laser energy was held at 1.8–2.0 J/cm^2. The base pressure was below 1×10^{-6} Torr before every deposition. High-quality epitaxial growths of GFO on YSZ substrate (1 0 0) and on YSZ buffered Si (1 0 0) were realized at an oxygen growth pressure of 400 mTorr. After each deposition, the sample was cooled to room temperature in the same oxygen pressure of 400 mTorr. The substrate temperature was kept at 650 °C for GFO deposition. Growth of crystalline template buffer layer of YSZ on (0 0 1) Si was realized without the need for any chemical process for removal of the surface oxygen as described in Ref. [14]. This method avoids the use of highly toxic HF solution to strip the oxide from the surface of Si substrate.

Film thickness and epitaxial quality were analyzed by Rutherford backscattering ion-channeling spectroscopy. Magnetization measurements were performed with a SQUID magnetometer (Quantum Design, MPMS). Second-harmonic generation studies were carried out with a Coherent Ti-sapphire femtosecond laser tuned to 750 nm. The incoming *s*-polarized light was nearly normal to the film surface and was focused to a spot size of about 100 μm. A photomultiplier tube was used to detect the second harmonic light from the film. SHG measurements were done at room temperature (above T_C) and at low temperature (100 K, i.e. below T_C) in an applied field of 3 kOe, which was applied in the film plane, normal to the polarization.

3. Results and discussion

GFO crystallizes into orthorhombic structure with lattice constants $a = 8.72$ Å, $b = 9.37$ Å and $c = 5.07$ Å. Lattice constants of YSZ and Si substrates are 5.12 and 5.41 Å, respectively. The lattice matching between the three materials is as follows: GFO/YSZ is 99% matched along *c*-axis and YSZ/Si is 94.7% matched. Figs. 1(a) and (b) show the X-ray diffraction (XRD) patterns of GFO grown on YSZ substrate (1 0 0) and on YSZ buffered Si (1 0 0), respectively, showing *b*-axis orientation of the GFO layer. The high-crystalline quality of the YSZ buffer layer is also clear. The *b*-axis growth of the GFO layer is significant since it yields the highest nonlinear Kerr effect for this crystal [12]. Fig. 1(c) shows the rocking curve full width at half-maximum (FWHM) of GFO/YSZ and GFO/YSZ/Si. Further, the FWHM of GFO/YSZ is ~0.44° (as compared to 0.24° of single-crystal YSZ substrate), establishing a high orientational quality of the film.

The ion backscattering channeling data for GFO on (0 0 1) single-crystal YSZ is shown in Fig. 2. The random spectrum matches well with the simulated one. The significant reduction in the backscattering yield for the Ga signal (minimum yield $\chi_{min} \cong 12\%$) upon channeling confirms the XRD observations of high-quality epitaxial growth of the films on single-crystal YSZ