Optical properties of LuFeO$_3$ thin films prepared by pulsed laser deposition

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Abstract. The LuFeO$_3$ thin films were successfully prepared by pulsed laser deposition on silicon and LaNiO$_3$-coated silicon substrates, respectively. The crystalline structure of the films has been studied by x-ray diffraction and indicated that all the films were well-crystallized. Atomic force microscope was used to characterize the surface morphology of the films and the surface was uniform and dense. Raman spectra measurements were carried out to study the lattice vibration modes of the films in the 200-1000 cm$^{-1}$ wave number range. Polarization-electric field hysteresis loops of the LuFeO$_3$ thin films were measured at applied electric field.

1. Introduction

Ferroelectric materials are used in modern electric devices, such as memory elements, filtering devices and high-performance insulators [1-4]. RFeO$_3$ compounds, belonging to the rare-earth orthoferrite, are a big family, where R is a trivalent rare earth cation. They have orthorhombic distorted perovskite structures (space group Pnma) [5]. They have been found a host of fascinating magnetic properties including canted anti-ferromagnetism and spin reorientation transitions [6]. LuFeO$_3$ (LFO) is one of the RFeO$_3$ families.

Recently, the charge–frustrated system LuFe$_2$O$_4$ and its dielectric and multiferroic characteristics have attracted much attention of the scientists [7]. LuFeO$_3$ can be seen the giant dielectric constant step [8]. Kumar [9] has prepared hexagonal metastable phase from undercooled LuFeO$_3$ melting with low oxygen partial pressure.

In this work, the LuFeO$_3$ thin films were prepared on silicon and LaNiO$_3$/Si substrates by pulse laser deposition. The structural, electrical and optical properties of the films were investigated.

2. Experimental

The LuFeO$_3$ thin films were prepared by pulsed laser deposition (PLD) using the KrF ($\lambda_{ex} = 248$ nm) pulsed excimer laser ablating a LuFeO$_3$ target. The films were deposited on Si substrate and LaNiO$_3$-coated Si (LNO) substrate, respectively. The base pressure of the deposition chamber was $7 \times 10^{-4}$ Pa. The films were deposited in oxygen or argon atmosphere for 30 minutes. The substrate temperature is 750-800 °C. After deposition, the films were in situ heat preservation in oxygen or argon atmosphere.

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The structure of the films was investigated by x-ray diffraction (XRD) using D/max2200. The surface morphology of the films was examined by tapping mode atomic force microscopy (AFM) (Veeco Dimension 3100). For the investigations of the electrical properties of the films, top Pt electrodes with area of $9 \times 10^{-4} \text{ cm}^2$ were sputtered on the films. Ferroelectric property was measured by Precision Materials Analyzer (Precision Premier II) at 1 KHz with different applied electric field. The optical properties of the films were studied by Raman spectra using Jobin-Yvon LabRAM HR 800UV micro-Raman spectrometer equipped with He–Cd 325 nm (3.82eV) laser as an exciting source.

3. Results and discussion

Figure 1 shows the XRD patterns of the LFO thin films deposited on LNO substrate. The crystal structure of the LFO film at room temperature was determined to be perovskite structures. All the samples have been well-crystallized and give x-ray peaks corresponding to LFO except for the impure phase, which turns out to be Si. The peaks at (110) and (220) are strong and sharp, which indicate that the PLD method has grown the films with preferred well orientation at 750 °C.

![Figure 1. XRD patterns for the LuFeO₃/LNO thin films deposited at 750 °C and 800 °C, respectively](image)

Microstructure and surface morphology of the LFO thin films deposited at 750 °C were characterized by AFM. The surfaces of the films were uniform and dense, as shown in Figure 2. The grain size and root-mean-square roughness were obtained from the analysis of the AFM images. The grain size was estimated to be 50 nm. The root-mean-square roughness was calculated to be 1.231 nm.
Figure 2. AFM images (1×1 μm²) of the films. (a) two dimensional and (b) three dimensional

Figure 3 shows the room-temperature polarization-electric field (P-E) hysteresis loops of the LFO thin films measured at applied electric field. The remnant polarization (P_r) was detected is 1.38 μC/cm², and the coercive field (E_c) was 74.8 kV/cm. The loop was not saturated, indicating that the leakage current of the films is high.

Figure 3. Ferroelectric P-E hysteresis loops of LuFeO₃ films

Figure 4 shows the Raman spectra of the LuFeO₃ films grown on Si substrate by PLD. For single crystal, Venugopalan [6] proved that there were sixty phonon modes expected for RFeO₃ crystals. At the center of the Brillouin zone, the irreducible representation is given as 7A_g+5B_1g+7B_2g+5B_3g+8A_u+10B_1u+8B_2u+10B_3u, where A_g, B_1g, B_2g and B_3g were Raman-active modes of species, three infrared modes are species B_1u, B_2u and B_3u, and A_u as the inactive mode.
Figure 4. Raman spectra of the LuFeO$_3$ films deposited on Si substrate by PLD

As shown in Figure 4, the Raman peaks are almost the same whatever the films deposited in oxygen and argon atmosphere at 800 °C, respectively. But the intensity of the Raman peaks on the films deposited in argon is stronger than that in oxygen. The peaks appear at 315, 393, 465, 516, 650 cm$^{-1}$, respectively. There are assignments and symmetries of $A_g$ and $B_{1g}$ spectra, and comparison from present results and the literature [6] is in Table 1. The positions of Raman peaks at present results are slightly different from the literature, which can be attributed to be crystalline state. The films of the present study are polycrystal, and that of the literature was single crystal. In other words, the preparation process changes the oxygen bonding and disorder that is reflected in the vibration frequencies of mode involving oxygen [10].

| Present results (cm$^{-1}$) | $A_g$ [6] (cm$^{-1}$) | $B_{1g}$ [6] (cm$^{-1}$) |
|---------------------------|----------------------|------------------------|
| 315                       | 278                  | 279                    |
| 393                       | 350                  | 350                    |
| /                         | 427                  | 425                    |
| 465                       | 450                  | /                      |
| 516                       | 516                  | 518                    |
| 650                       | 654                  | /                      |

4. Conclusions

The lutetium ferrite thin films were prepared using pulsed laser deposition. The x-ray diffraction results indicate that the LuFeO$_3$ thin films on LNO at 750 °C show a highly orientation. The polarization–electric field hysteresis loops of the film can prove that the LFO film has ferroelectric property. Uniform and dense surface morphology is observed in the AFM micrograph. The first-order Raman scattering gave the peak positions at 315, 393, 465, 516 and 650 cm$^{-1}$, respectively.

Acknowledgments

The author thanks to Mr. Wenfei Xu for AFM investigation, Mr. Xiangui Chen for optical investigation, Dr. Lin Sun and Mr. Nuofan Ding for experimental work. This work was supported by the National Natural Science Foundation of China (No. 60990312), the State Key Basic Research
Program of China (2007CB924902), Shanghai Leading Academic Discipline Project (B411) and Science and Technology Commission of Shanghai Municipality (10JC1404600).

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