Thin Solid Films 520 (2012) 6446–6449

Contents lists available at SciVerse ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Dielectric tunability and magnetoelectric coupling in $LuFe_2O_4$ epitaxial thin film deposited by pulsed-laser deposition

M. Zeng [a,b], J. Liu [a], Y.B. Qin [c], H.X. Yang [c], J.Q. Li [c], J.Y. Dai [a,*]

[a] Department of Applied Physics, The Hong Kong Polytechnic University, Hong Kong, PR China
[b] Institute of Advanced Materials, School of Physics and Telecommunication Engineering, South China Normal University, Guangzhou 510006, PR China
[c] Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, PR China

ARTICLE INFO

Article history:
Received 12 October 2011
Received in revised form 12 June 2012
Accepted 13 June 2012
Available online 23 June 2012

Keywords:
Magnetoelectric coupling
Dielectric tunability
Charge-Ordering
Multiferroics
Pulsed-laser deposition
X-ray diffraction
Lutetium ferrite

ABSTRACT

C-axis orientated $LuFe_2O_4$ thin films on (001) sapphire substrates are epitaxially deposited by pulsed-laser deposition. Temperature-dependent resistance characterization reveals the ferrimagnetic transition at 237 K and charge-ordering transition at 340 K in the film. Importantly, the dielectric constant of the film can be significantly changed by both electric and magnetic fields. The dielectric tunability reaches 35% when an electric field of 5 V is applied, while this value reduces to 20% and 15%, respectively, when a magnetic field of 0.83 T is applied perpendicular and parallel to the film normal direction. This suggests a magnetically controlled dielectric tunability and strong magnetoelectric coupling, and is therefore promising for tunable device applications in film form.

1. Introduction

Multiferroics have attracted extensive interest of study because they exhibit simultaneously ferromagnetic and ferroelectric polarizations and a coupling between them, which offer rich physics and device concepts [1,2]. Recently, $LuFe_2O_4$, an emerging candidate for room-temperature multiferroics, has been extensively investigated for its anomalous dielectric and spin- and charge-ordering behaviors [3–14]. In particular, its giant dielectric tunability under magnetic and electric fields suggests that this material has great potential in applications on various electronic devices such as multiferroic memory and tunable microwave devices [4,6].

$LuFe_2O_4$ is a member of the RFe_2O_4 (R = rare earth element) family [14]. It has a rhombohedral crystal structure (space group $R\bar{3}m$, a = 3.44 Å and c = 25.28 Å) consisting of three double W-like hexagonal $Fe_2O_{2.5}$ and two double U-like $LuO_{1.5}$ layers [13]. Within each W layer, Fe atoms are arranged in triangles, leading to a frustrated spin and charge ordering of Fe^{2+} and Fe^{3+} ions [8,11–15]. Many physical properties of $LuFe_2O_4$ depend greatly on the spin- and charge-ordering states, such as ferroelectric, magnetoelectric and dielectric performances as well as phase transitions. The ferroelectricity in bulk $LuFe_2O_4$ was first reported by Ikeda et al. who attributed the ferroelectricity as charge ordering of Fe ions [3] because the centers of Fe^{2+} and Fe^{3+} ions do not coincide. The two-dimensional ferrimagnetic order appearing below 240 K has been suggested by neutron scattering studies and Monte-Carlo simulation [12,16]. The large magnetoelectric coupling has been attributed to the intricate interplay between charge and spin degrees of freedom with the crystal lattice and external electric and magnetic fields to some extent on a short-range order [8,9,17,18]. Also, a large response of the dielectric constant under small electric and/or magnetic fields has been attributed to the charge fluctuation on the Fe sites [4,6,18]. These suggest potential application of $LuFe_2O_4$ in which the charge and spin degrees of freedom of an electron can be controlled.

Physical properties of $LuFe_2O_4$ have been intensely studied in single crystal and ceramic forms [2–18], but are very rarely reported in thin film form. It is well-known that single phase oxide films have been proved to introduce properties due to interference lattice modulation and dimension effect [2]. In particular, the realization of thin film form $LuFe_2O_4$ is essential for this material to be used in integrated electronic devices. In this paper, we report the study of the $LuFe_2O_4$ film, in particular its dielectric tunability under both electric and magnetic fields, i.e. magnetoelectric coupling.

2. Experimental details

$LuFe_2O_4$ film was fabricated by pulsed-laser deposition on (001) sapphire substrates at 800 °C. The laser used in the deposition was a KrF (λ = 248 nm) excimer laser with a laser fluence of 3 J/cm^2. Ceramic target was fabricated through using high purity submicron powders of

---

[*] Corresponding author.
E-mail address: apdaijy@inet.polyu.edu.hk (JY. Dai).

0040-6090/$ – see front matter © 2012 Elsevier B.V. All rights reserved.
doi:10.1016/j.tsf.2012.06.058

Lu_2O_3 and Fe_2O_3 with Lu:Fe molar ratio of 1:4 (standard ceramic processing techniques). The substrate was placed parallel to the target at a distance of 7 cm. During the deposition, the chamber was kept at high vacuum of 1×10^{-5} Pa. The deposited films were then in-situ annealed at the growth temperature in an oxygen ambient of pressure up to 20 Pa for 1 h before being cooled down to room temperature. The crystallographic structure of the $LuFe_2O_4$ film was analyzed using a Bruker AXS D8 discover X-ray diffractometer with Cu Kα radiation ($\lambda=0.154$ nm). And the power of 40 kV and 40 mA and a high-resolution ω-2θ goniometer were used during the operation.

For electrical characterization, gold interdigital electrodes, dimensions of finger line spacing and width of 10 μm, and finger line length of 1 mm, were coated on the $LuFe_2O_4$ film surface. Current–voltage (I–V) measurement was carried out by using Keithley 2410 source meter with a protecting current of 0.02 A. The resistance dependence on temperature was measured by using Keithley 6517A high resistance electrometer. The sample was first cooled by liquid nitrogen to 77 K, held for 15 min, and then measurement was performed during heating up to 370 K with a rate at 1 °C/min. The temperature-dependent dielectric constant and loss were measured using a 4291B RF impedance analyzer under different frequencies. The dielectric tunability, n_τ, defined as $n_\tau=(\varepsilon(V)-\varepsilon(0))/\varepsilon(0)$, was characterized at room temperature when the magnetic field of 0.83 T was applied along different directions ($H//c$ axis and $H\perp c$ axis). In which ε (V) and ε (0) are the dielectric constants with and without electric field, respectively.

3. Results and discussion

The characteristic XRD pattern of $LuFe_2O_4$ film is shown in Fig. 1. The XRD pattern shows strong characteristic peaks of (003), (006) and (009) of $LuFe_2O_4$, indicating a good *c*-axis epitaxial growth. Also, some weak impure phase peaks are exhibited in the film. It is noted that the usage of a Fe-enriched target favors the formation of $LuFe_2O_4$ phase, and details of growing conditions and film structure were reported in our previous paper [19].

Fig. 2 plots I–V curves of the $LuFe_2O_4$ film under the applied voltage sweeping from −20 to 20 V for four cycles. It is clear that the I–V curves show nonlinear characteristic with gradually changed small hysteresis loops. Similar results have been reported in $LuFe_2O_4$ single crystal [7]. The mechanism of nonlinear I–V characteristic has been proposed by Zeng et al. [7] as the charge-ordering state breakdown in $LuFe_2O_4$. Furthermore, the shift of the hysteresis loops in our deposited $LuFe_2O_4$ film may be caused by the slow charge ordering melting in the film due to the pinning effect of substrate to the domain movement [20].

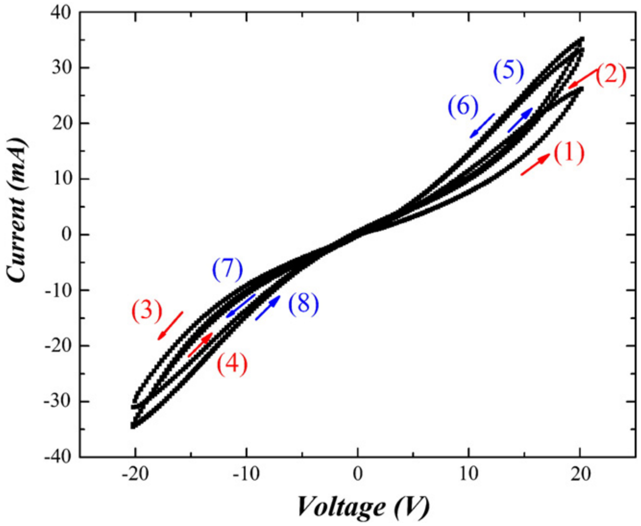

Fig. 2. Current versus voltage curves of $LuFe_2O_4$ film under the applied voltage sweeping from −20 to 20 V for four cycles. The voltage is applied along the number ordering.

Temperature-dependent resistance of the $LuFe_2O_4$ film is presented in Fig. 3. The significant resistance change at 237 K (Neel temperature, denoted as T_N) can be attributed to magnetic transition. This result suggests the existence of strong coupling in charge and spin orderings in the $LuFe_2O_4$ film. A similar result has been reported in other RFe_2O_4 families, such as YFe_2O_4 [21] and $ErFe_2O_4$ [22]. The most interesting result is that a resistance discontinuity is observed at 340 K (Curie temperature, denoted as T_{CO}). This abrupt resistance change can be attributed to the charge ordering transition, roughly coinciding with the onset of ferroelectricity in conventional ferroelectric materials. That is to say, when the system is above T_{CO}, the charge ordering induced polarization disappears.

This charge ordering transition in the $LuFe_2O_4$ film near 340 K was also observed in the temperature-dependent dielectric constant measurement as shown in Fig. 4. From Fig. 4(a), it can be seen that when temperature increases approaching to 340 K the dielectric constant increases sharply to an extremely large value and then followed by a sudden drop to negative value (not shown in this figure), i.e. there is a discrete point. Correspondingly, a sharp peak in the dielectric loss is presented as shown in Fig. 4(b). This feature can be explained by the charge-ordering transition at this temperature, which is similar to the first-order ferroelectric phase transition, where the dielectric constant reaches infinity and becomes discrete at Curie temperature. It is worth noting that this abnormally large dielectric constant and loss value is presented only in the low frequency region. The mechanism may be attributed to electron oscillation jumping between the Fe^{2+} and Fe^{3+}

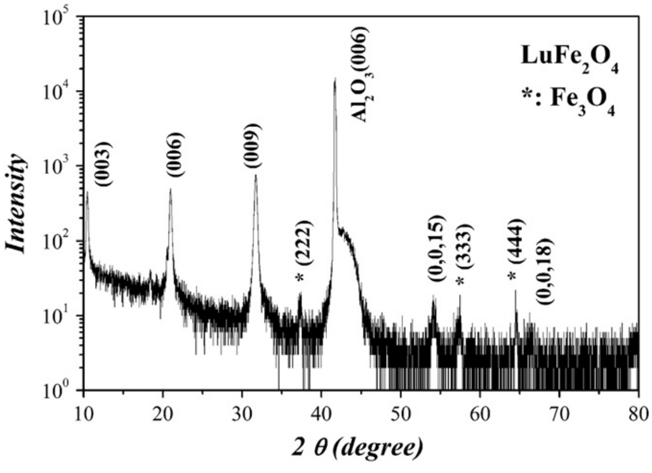

Fig. 1. XRD pattern of $LuFe_2O_4$ thin film on the (001) sapphire substrate.

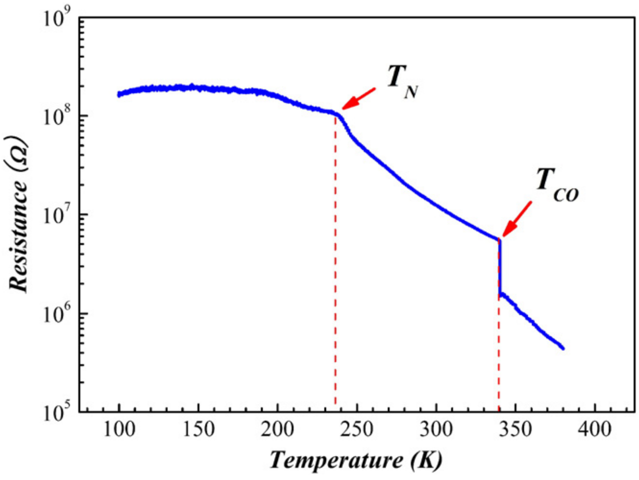

Fig. 3. Resistance dependence on temperature of $LuFe_2O_4$ film during heating.