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Stress-induced magnetic anisotropy of CoFe₂O₄ thin films using pulsed laser deposition

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Abstract

Cobalt ferrite (CoFe₂O₄) thin films (\approx 70 nm) were epitaxially grown on TiO₂-terminated (001) SrTiO₃ substrates by pulsed laser deposition (PLD). Films with very smooth surface, which follow the terrace of the substrate, were obtained at temperatures below 600 °C. The magnetic properties of CoFe₂O₄ can be controlled by changing the deposition parameters. The in-plane magnetic anisotropy can be explained as induced by the compressive stress in films growing at low temperature and low oxygen pressure. Tuneable magnetic properties by PLD make CoFe₂O₄ more attractive for practical application, especially to create multi-functional devices in combination with perovskite ferroelectric films.

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Ferrite films are of particular attractive for microwave devices because of their low conductivity and large permeability at high frequency [1]. On the other hand, with large magnetocrystalline anisotropy and magnetostriction, high chemical and mechanical stabilities, they offer many possibilities for future applications such as magnetic and magneto-optic recording [2], tunnel magnetoresistance devices [3], ferrofluids [4] and medical applications [5]. For these applications, highly oriented growth of ferrite films is necessary. In order to promote the epitaxial growth of ferrite films, low mismatch MgO substrate [6] or $CoCr_2O_4$ buffered SrTiO₃ and MgAl₂O₄ substrates [7] were used.

In this article, we investigate the properties of epitaxial $CoFe_2O_4$ films directly grown on (001) $SrTiO_3$ substrates using pulsed laser deposition (PLD). The magnetic

anisotropy can be tuned by changing the deposition condition such as temperature and oxygen pressure. The large in-plane magnetic anisotropy is obtained without using buffered layers. The role of deposition parameters on the microstructure and magnetic properties of the films will be discussed.

The films of 70 nm were grown by PLD using a KrF excimer laser ($\lambda = 248 \text{ nm}$) with pulse duration of 25 ns. Polycrystalline CoFe₂O₄ target, obtained by complexometric synthesis [8], and one-side polished single-crystal (001) SrTiO₃ substrates were used in experiments. The substrate was positioned 6 cm from the target and the substrate temperature (T_s) was held constant during deposition, ranged from 500 to 700 °C. The PLD system was operated at an energy density of 2.5 J/cm² and a laser frequency of 5 Hz. Films were deposited in oxygen environment with the ambient pressure (pO₂) varying from 0.02 to 0.1 mbar. After deposition, films were cooled down to room temperature in 1 bar of oxygen.

The crystallographic structure analyses using an X-ray diffractometer reveal that CoFe₂O₄ films are epitaxial by

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the presence of a strong (004) peak around 43°. Atomic force microscopy images of films grown at different T_s , as presented in Fig. 1, show that up to 600 °C films have very smooth surface (RMS = 0.4 nm) and follow the terrace of SrTiO₃ substrate. At 700 °C, the surface becomes rougher due to the presence of particles with the size of 60 nm.

The electrical resistivities of these films, obtained by a four-point probe, are in the range of $10^5 - 10^6 \Omega$ cm. The inplane and perpendicular magnetic hysteresis loops, measured by a vibrating sample magnetometer, are plotted in Fig. 2. For low-temperature-grown films, the in-plane loops exhibit a large hysteresis with coercivity in the range of 100-120 kA/m. This in-plane magnetic anisotropy, however, is less pronounced at higher temperature. That observation can be explained in terms of the strain in the film, originated from the lattice mismatch between the film and the substrate. Since the lattice parameter of CoFe₂O₄ is 8.392 A, films grown on SrTiO₃ substrate (lattice parameter of 3.905 Å) are under compression in the film plane while are under tension perpendicular to the film plane. Due to its negative magnetostriction, a strong in-plane stress anisotropy can be induced and dominates magnetocrystalline anisotropy. With an increase in the substrate temperature, the stress is released and the later becomes more competitive as shown by the comparable perpendicular hysteresis loops.

We now estimate magnitude of the stress anisotropy and compare to the magnetocrystalline anisotropy. The stressinduced anisotropy field is given by $3\lambda_{100}\sigma/M_s$ [9] in which $\lambda_{100} = -590 \times 10^{-6}$ is the magnetostriction coefficient, $\sigma = Y \varepsilon$ is a uniaxial stress, M_s is the saturation magnetisation (Young's modulus $Y = 1.5 \times 10^{12} \text{ dyn/cm}^2$ and the strain ε estimated from the difference between bulk and film lattice constants). Regarding the magnetocrystalline anisotropy field of $2K_1/M_s$ with the magnetocrystalline anisotropy constant $K_1 \sim 3 \times 10^6 \text{ erg/cm}^3$, the anisotropy constant associated with the magnetoelastic coupling can be expressed as $K = 3\lambda_{100}\sigma/2$. From the measured d spacing for the (004) reflection, the lattice parameter c of 8.412 \AA is derived for the film grown at 600 °C. Assuming that the compression and tension are comparable, K is estimated of $3.2 \times 10^6 \text{ erg/cm}^3$, which is on the same order as K₁.

 $CoFe_2O_4$ films are also grown at different oxygen pressures. The results show that with $pO_2 \leq 0.05$ mbar films exhibit a strong in-plane magnetic anisotropy. At low pressure, films are oxygen deficiency because oxygen diffuses out of the as-deposited layer. This causes the compression of the unit cell and enhances the in-plane magnetic anisotropy.

Epitaxial $CoFe_2O_4$ films have been directly grown on $SrTiO_3$ substrate by PLD. At low substrate temperature and low oxygen pressure, the films have smooth surface and in-plane magnetic anisotropy. It allows the deposition



Fig. 1. AFM images of films grown at different Ts: (left) 500 °C, (center) 600 °C, (right) 700 °C.



Fig. 2. In-plane (solid points) and perpendicular (open points) loops of films grown at different T_s.

of further epitaxial layers, e.g. perovskite ferroelectrics, for practical application as multi-functional devices.

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