Fabrication of single crystalline EuO thin film with SrO buffer layer on SrTiO$_3$ substrate

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Abstract. We report the new fabrication method of single crystalline EuO thin film along the (100) plane and its crystal structure and magnetic property. EuO thin film was fabricated on SrO buffer layer created on SrTiO$_3$ substrate. The obtained EuO thin film was formed a single phase of the rock-salt structure with the lattice constant of 0.516 nm and showed ferromagnetism below the Curie temperature of 71 K. The examined physical properties are consistent with the bulk materials.

1. Introduction

Europium monoxide (EuO), a ferromagnetic semiconductor, with a Curie temperature ($T_C$) of 69 K, has been attracting attention due to its anomalous magneto-optical and transport properties [1-6]. In the case of electron doping with excess Eu or the substitution of Eu$^{2+}$ ions with Gd$^{3+}$ or La$^{3+}$ ions, $T_C$ increases to as high as 200 K and a metal-insulator transition (MIT) appears in which the electrical resistivity drops by more than 12 orders of magnitude with decreasing temperature [3,7-10]. When an external magnetic field is applied, the MIT temperature is shifted to the higher temperature side and the electrical resistivity decreases due to the colossal magnetoresistive (CMR) effect [9]. Since the CMR effect originates from localized Eu$^{2+}$ 4f electrons, the spin polarization of carriers is predicted to be fully 100% below $T_C$ [11-14]. Therefore EuO is one of candidate compounds for next-generation spintronics applications such as spin valves and spin switches.

To apply EuO for spintronics devices, it is important to grow a few atomic layers single crystalline EuO on the substrate. So far we report the fabrication and physical properties of high quality single crystal thin films of EuO with the thickness of 50 nm [15]. Well defined EuO thin films cannot be grown just on the SrTiO$_3$ substrate, because the other phase of Eu$_2$O$_3$ inevitably appears at the interface between the SrTiO$_3$ and EuO film due to oxygen diffusion. To solve this problem, we adopted BaO as a buffer layer between EuO thin films and SrTiO$_3$ substrate [16]. In this case, due to the large lattice mismatch between the BaO buffer layer and EuO (6.3%), EuO thin films with a few
atomic layers cannot be grown epitaxially on the BaO thin film [15-16]. To satisfy small lattice mismatch between EuO and buffer layer, we propose that SrO is used for buffer layer in between EuO thin film and SrTiO$_3$ substrate, as shown in Fig. 1. The relationship of the epitaxial growth is EuO[110] // SrO[110] // SrTiO$_3$[110] and EuO(001) // SrO(001) // SrTiO$_3$(001). SrO forms a rock-salt crystal structure with lattice constant of 0.5144 nm, which is the same crystal structure and similar lattice constant as EuO (The lattice mismatch between SrO buffer layer and EuO thin film is slightly -0.04 %) [17]. Thanks to this quite small lattice mismatch, EuO thin films with a few atomic layers are expected to be grown epitaxially on the SrTiO$_3$ substrate.

In this paper, we report the growth of single crystalline EuO thin film with SrO buffer layer on SrTiO$_3$, its crystal structure and magnetization property. The obtained EuO film with the thickness of 50 nm forms a single phase rock-salt structure with a lattice constant of 0.516 nm and shows a ferromagnetic transition at $T_C = 71$ K.

![Figure 1. (Color online) Atomic structure of EuO (001) thin film with SrO buffer layer onto SrTiO$_3$ substrate. Lattice constant of SrTiO$_3$, SrO and EuO and lattice mismatch between SrTiO$_3$–SrO and SrO–EuO are shown in the figure.](image)

### 2. Experimental Procedure

Single-crystalline EuO thin film was grown using a molecular beam epitaxy (MBE) method [16]. The thin films were grown on 0.05 wt% Nb-doped SrTiO$_3$ (001) single-crystalline substrates. First, the substrates were heated at 600°C for 1 hour under an ultra-high vacuum ($2.0 \times 10^{-7}$ Pa) to obtain clean surface. Then, strontium metal with a purity of 99.9 % was evaporated using an effusion cell under an oxygen pressure of $1.0 \times 10^{-4}$ Pa to create a buffer layer of SrO with a thickness of 5.0 nm. Finally, the EuO thin film was grown by the evaporation of Eu metal under an oxygen pressure of $8.0 \times 10^{-6}$ Pa on the SrO buffer layer at a substrate temperature of 400 °C. The crystal structure was observed using an X-ray diffractometer (XRD; RINT-Ultima III, Rigaku Ltd.) with a Cu Kα radiation. The magnetization was measured with a superconducting quantum interference device magnetometer (SQUID; MPMS-7, Quantum Design Ltd.).

### 3. Results and Discussion

Figure 2 shows the XRD patterns of 50 nm EuO thin films with and without SrO buffer layer on a SrTiO$_3$ substrate at room temperature, respectively. The Eu$_2$O$_3$ (310) peak at $2\theta = 31.5$ deg. was observed in the XRD pattern of the case without SrO buffer layer, which is due to an oxygen diffusion from SrTiO$_3$ to EuO. From the photoemission spectroscopy, only Eu$^{3+}$ 4f and O$^{2-}$ 2p state exist in EuO / SrO / SrTiO$_3$ and no other peak appear [18]. These results suggest that the SrO buffer layer protects the oxygen diffusion. The obtained lattice constant of EuO thin films with SrO buffer layer along the parallel to the surface normal is $0.516 \pm 0.001$ nm that is in good agreement with a previous report.
obtained EuO thin films with the case of BaO buffer layer (0.515 ± 0.001 nm) [15]. Therefore the obtained EuO thin film is confirmed to be rock-salt crystal structure as same as bulk materials [16]. The lattice mismatch between SrO buffer layer and EuO thin films is -0.3 %. Therefore due to the quite small lattice mismatch, the atomic flatness of EuO ultra-thin films is expected when SrO buffer layer is used. Note that peaks marked by open circles are unknown peak but are not originated from europium compounds.

Figure 3 shows the temperature-dependent magnetization of EuO thin film with SrO buffer layer, which is measured by applying a magnetic field of 500 Oe along the EuO [010] direction. Since the magnetization curve can be reproduced by the Brillouin function with spin moment $S = 7/2$ and $T_C = 71$ K, the EuO thin film is explained by simple Heisenberg ferromagnetism. $T_C$ is slightly higher than that of the bulk material (69 K). $T_C$ can be controlled by the stoichiometry, the lattice constant and the lattice distortion. In the present case, the obtained EuO thin film is confirmed to be formed a single-crystalline rock-salt structure and the same lattice constant as the bulk materials. Therefore, the inconsistency in the value of $T_C$ is considered to be due to the slight excess of Eu atoms. Our previous magneto-optical infrared imaging experiment on the EuO thin film with BaO buffer layer also suggested the excess of Eu ion leading to a magnetic polaron states [16, 19].

4. Conclusion
In conclusion, we successfully fabricated single-crystalline EuO thin film with SrO buffer layer on SrTiO$_3$ substrate using a molecular beam epitaxy method. The obtained EuO thin film was found to be a single crystalline ferromagnetic with $T_C = 71$ K. Due to the quite small lattice mismatch at the interface, we expect that single crystalline EuO thin films with a few atomic layers can be fabricated. The technique is useful for next-generation spintronics devices such as a spin filter.

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