Resistivity Control of VO₂ Thin Film Stabilized by Li⁺ Diffusion of LiCoO₂ Layer

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1 Introduction

The metal-insulator transition (MIT) phenomena of VO₂ thin film can use as the resistive switching device by electric double layer transistor using ionic liquid [1]. However, the use of ionic liquids is not practical. To realize a more practical device, an all-solid-state redox element consisting of a Fe₃O₄/Li₂O-SiO₂-ZrO₂ /LiCoO₂(LCO) multilayer structure film is realistic [2]. By using this device structure, it is possible to create neuromorphic devices and physical reservoir devices, and has the potential to become next-generation devices that can perform complex processing with low power consumption.

In this study, we prepared the VO₂/LCO multilayer films on Al_2O_3 (0001) substrate and examined their structural and electrical properties in order to explore the possibility of all-solid-state transistor using Li-ion conductor electrolyte and VO₂ thin films.

2 Experiment

Figure 1(a) show the device structure of LCO and VO₂ thin films. The LCO thin film was deposited on Al₂O₃ (0001) substrate by RF magnetron sputtering. After forming a LiCoO₂ film on the left half of the substrate, a metal mask was attached and a VO₂ thin film was deposited on the light half using a V metal target and O₂ gas. The thin film section with a width of 1.5 mm where VO₂ and LCO overlap is a VO₂/ LCO multilayer structure. The substrate temperatures of these thin films were maintained at 700°C during deposition processes.

The crystal structures were confirmed using X-ray diffraction (XRD). The temperature dependence of the electrical resistance and ionic conductivity were measured using the DC two-terminal method and AC impedance method, respectively. The electronic structures of valence band and conduction band were characterized by photoemission spectroscopy (PES) and X-ray absorption spectroscopy (XAS), respectively, which are installed at the undulator beamline BL-2A in the PF at KEK in Japan.

3 Results and Discussion

Figure 1(b) shows the XRD patterns of the LCO (~100 nm), VO₂/LCO and VO₂ (~120 nm) thin films as shown in Fig. 1(a). The LCO film exhibits a weak (003) peak at ~19.0 °. The VO₂ film exhibits a single peak of (020) at ~40.0 °. In the VO₂/LiCoO₂ multilayer film, the presence of a LiCoO₂ was not confirmed, and only the (020) peak of VO₂ was observed.

Figure 2 shows the valence bands and conduction bands obtained from the PES and XAS spectra, respectively, of LCO, VO₂ and VO₂/LCO thin films measured at 300K. The valence band and conduction band of LCO film is mainly composed of the Co 3d



Fig. 2: Valence and conduction bands of LCO, VO₂ and VO₂/LCO layers at ~300K.

hybridized with O 2p state. The VO₂ film at 300 K consists of the lower Hubbard band (LHB) and upper Hubbard band (UHB) of V 3*d* components. The energy separation between LHB and UHB reflects the electron correlation energy (U_{dd}). The U_{dd} increases significantly in the VO₂/LCO film. This suggests that the number of 3*d* electrons in the VO₂ film changes by the diffusion of Li ions, inducing strong electron correlation.

<u>References</u>

- [1] M. Nakano et al, Nature **487** (2012) 459.
- [2] W. Namiki et al., ACS. Nano 14 (2020) 16065.

Research Achievements

1. M. Takayanagi and T. Higuchi *et al.*, "Accelerated/ decelerated dynamics of the electric double layer at hydrogen-terminated diamond/Li⁺ solid electrolyte interface". Materials Today Physics **31** (2023) 101006.

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