

Synthesis of Ruddlesden-Popper type $\text{Sr}_3\text{Cr}_2\text{O}_7$ epitaxial thin films

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Ruddlesden-Popper type perovskite oxides $A_{n+1}B_n\text{O}_{3n+1}$ have been intensively studied because of their varieties of physical properties stemming from the layered structure and tunable composition. Among them, $\text{Sr}_3\text{Cr}_2\text{O}_7$ is an antiferromagnetic Mott insulator with a characteristic singlet orbital ordering based on the spin and orbital degrees of freedom [1]. In addition, its superconductivity was theoretically predicted for the $3d^2$ electronic state [2]. However, $\text{Sr}_3\text{Cr}_2\text{O}_7$ has been scarcely synthesized because high temperature and pressure are necessary, in general. In this study, we developed a synthetic route of c -axis oriented $\text{Sr}_3\text{Cr}_2\text{O}_7$ epitaxial thin film with high crystallinity by pulsed laser deposition (PLD).

A high-density $\text{Sr}_3\text{Cr}_2\text{O}_8$ pellet synthesized by spark plasma sintering was adopted as a PLD target and ablated by KrF excimer laser ($\lambda = 248$ nm). The $\text{Sr}_3\text{Cr}_2\text{O}_7$ films were synthesized on $(\text{LaAlO}_3)_{0.3}(\text{SrAl}_{0.5}\text{Ta}_{0.5}\text{O}_3)_{0.7}$ (LSAT; lattice mismatch: 1.0%) (100) and SrTiO_3 (STO; lattice mismatch: 1.9%) (100) single crystal substrates at substrate temperatures of 800 °C and 815 °C, respectively, in Ar atmosphere, which is beneficial for reducing the oxidation state of Cr ion from 5+ to 4+ [3].

In the X-ray diffraction (XRD) θ - 2θ patterns, only 00 l diffractions without any impurity peak were observed for the $\text{Sr}_3\text{Cr}_2\text{O}_7$ thin films on both LSAT and STO substrates (Fig. 1). Epitaxial growth of the $\text{Sr}_3\text{Cr}_2\text{O}_7$ thin films were confirmed by the 10 $\bar{1}7$ spot peaks in reciprocal space mapping. Sharp rocking curves around the 00 $\bar{1}0$ peak with full width at half maximum of 0.030° and 0.46° were observed for the thin films on LSAT and STO substrates, respectively, indicating good crystallinity. For the thin film on LSAT, particularly, an atomically flat surface with a step-and-terrace structure was observed (Fig. 2). These results indicated first successful synthesis of high-quality $\text{Sr}_3\text{Cr}_2\text{O}_7$ epitaxial thin films.

[1] J. Jeanneau *et al.*, Phys. Rev. Lett. **118**, 207207 (2017).

[2] D. Ogura *et al.*, Phys. Rev. B **96**, 184513 (2017).

[3] S. Fukuda *et al.*, Appl. Phys. Lett. **116**, 123101 (2020).

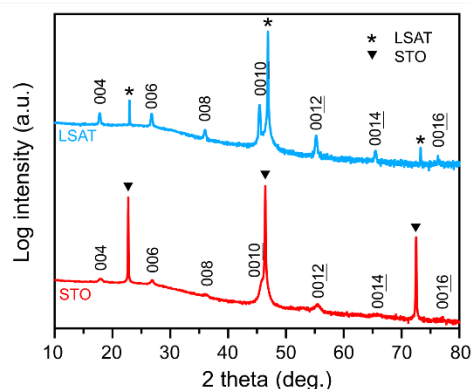


Fig. 1. X-ray diffraction θ - 2θ patterns for $\text{Sr}_3\text{Cr}_2\text{O}_7$ epitaxial thin films on LSAT (100) and STO (100) substrates.

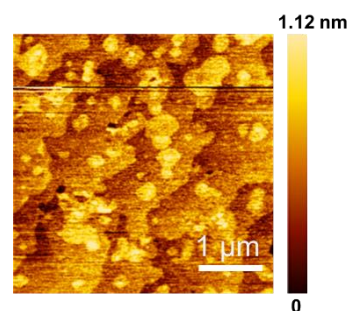


Fig. 2. Atomic force microscope image for $\text{Sr}_3\text{Cr}_2\text{O}_7$ epitaxial thin film on LSAT (100) substrate.