Preparation and Characterization of Ordered Double Perovskite SrLaVMoO₆ Thin Films

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Abstract

Epitaxial thin films of SrLaVMoO₆ with a double perovskite structure, were grown on (111) SrTiO₃ substrates by magnetron sputtering. The optimized (111) films exhibited a clear (111) diffraction peak, which is a superlattice reflection of the double perovskite unit cell, indicating clear B-site ordering. XPS results of the Mo 3d core level spectra depicted broadening or multiple features. A spin polarization *P* value of 0.83 was measured using superconducting planer junctions.

1. Introduction

Half-metallic antiferromagnets (HMAF) exhibit both fully spin-polarized charge transport and zero net magnetic moment. The prospect of getting a fully spin-polarized current from a metal that has no magnetic field provides not only fertile playground for fundamental research but also conceivable new spintronic devices that rely on the spin polarization of the carriers. Although there have been several theoretical studies based on the band structure calculations that predict candidates for HMAF, no experimental reports show existence of HMAF. Recently, we have observed the coexistence of antiferromagnetic nature and high spin polarization in polycrystalline bulk SrLaVMoO₆, suggesting that SrLaVMoO₆ is a strong candidate for a HMAF [1]. Fabrication of ordered SrLaVMoO₆ thin films is interesting from viewpoints of electronic applications and fundamental studies of HMAF. In this paper, we will present the extended study of structural characterization as well as the transport properties of thin films of SrLaVMoO₆. We have found that (111) epitaxial thin films of SrLaVMoO₆ grown under optimized growth conditions show clear signs of B-site ordering and pronounced electronic properties.

2. Experimental Procedure

SrLaVMoO₆ thin films were grown on SrTiO₃ (111) substrates using a magnetron sputtering technique from a target with stoichiometric composition. Thin films were deposited at temperatures of 800 °C in a mixed sputtering gas of Ar + 1 ~ 5% H₂ at 10 Pa. Phase purity, out-of-plane orientations and B-site (V/Mo) ordering of thin films were examined using x-ray diffraction (XRD) with a Cu K α radiation source. The electrical resistivity was measured by the standard four-probe method. Photoemission measure-

ments were performed using an Mg K α (1253.6 eV) x-ray source. Spin polarization was measured using superconducting planar junctions with a superconducting NbN electrode. The junction structure for conductance-voltage (G-V) measurements is as follows; SrLaVMoO₆ (50 nm) / MgO (0.7 nm) / NbN (100 nm).

3. Results and Discussion

XRD patterns for SrLaVMoO₆ films on SrTiO₃ (111) substrates grown at 800 °C under different oxygen partial pressures are shown in Fig. 1. Samples B was deposited in more optimum condition than samples A. Sample B exhibited a clear (111) diffraction peak, which is a superlattice reflection of the double perovskite unit cell, indicating clear B-site ordering. A is a disordered sample and B is an ordered sample.

The temperature dependence of resistivity ρ of samples A and B is shown in Fig. 2. Both samples show metallic behavior, but only sample B behaves like the SrLaVMoO₆ bulk. The residual resistivity ρ_0 for sample A is 0.01 mΩcm, which is much smaller than that (2.1 mΩcm) of sample B. The changes in the local structures caused by the B-site disordering influence the electrical conduction. Detailed discussion on this topic will be presented later.

In order to clarify the valence states and discuss the electronic structure of SrLaVMoO₆ films, x-ray photoemission spectroscopy (XPS) measurements were carried out. The Mo 3d core-level spectra of samples A and B are depicted in Fig. 3. The peak positions of the Mo 3d spectra for samples A and B indicate that the formal valence state of Mo ions is dominantly Mo^{4+} (4 d^2), which agrees well with the expectation from the simple ionic picture of antiferromagnetic SrLaVMoO₆. The overall line shape of the Mo 3d spectra for samples A and B shows significant broadening or multiple features, which is very similar to the case for Mo sites in half-metallic ferrimagnetic Sr₂FeMoO₆ [2], indicating an unusual electronic state of Mo. The spin polarization (P) values of samples A and B were determined to be 0.65 and 0.83 by fitting the curves which are calculated by the modified BTK theory [3] using three parameters: a spin polarization (P), a superconducting gap (Δ), and a dimensionless barrier parameter (Z) that describes the strength of interfacial scattering to the normalized conductance [G(V)/G_n] versus voltage curves obtained at 4.2 K

by using superconducting planer junctions. *P* value of sample B is higher than that of sample A.

The results of this study can be discussed in terms of B-site ordering. In sample A, a superlattice reflection was not observed, which indicates a disordered structure. Only disordered films showed the metallic Mo peak in the XPS spectra. This result is consistent with a general phenomenon in ordered type double perovskite compounds that deviations of valence states from ideal states cause B-site disordering. It is reported that in disordered SrLaVMoO₆, chemical fluctuation within very small spatial domains gives rise to local LaVO₃- and SrMoO₃-like regions [4]. Our disordered films may also include such a chemical fluctuation. In particular, perovskite SrMoO3 shows the lowest electrical resistivity among oxide compounds, so it is thought that the lower resistivity seen in Fig. 2 is due to the conductive path through the local SrMoO₃. On the other hand, sample B showed superlattice reflections, improved valence states, and higher spin polarization than these disordered sample A. The B-site ordering obviously influenced the electrical properties.



Fig. 1 XRD patterns for SrLaVMoO₆ films on SrTiO₃ (111) substrates grown at 800 °C under different oxygen partial pressures.



Fig. 2 Temperature dependance of resistivity ρ of disordered SrLaVMoO_6 films and ordered one.

4. Summary

(111)-oriented SrLaVMoO₆ films obtained with clear signs of B-site ordering exhibited significant broadening or multiple features in the Mo 3*d* spectrum as well as high spin polarization P value of 0.83 was obtained. Our present research has also demonstrated that epitaxial films grown along the (111) direction are promising for examining and elucidating the intrinsic electronic properties of HMAF materials with a double perovskite structure.

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Fig. 3 The Mo 3d core-level spectra of disordered SrLaVMoO₆ films and ordered one.