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Influence of Oxygen Partial Pressure on Electrical and Magnetic Characteristics of Spinel $CoFe_2O_4$ Thin Films

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Abstract

Recently, semiconducting spinel ferrites have attracted increasing attention for the potential as room temperature spintronics devices. In this study, we investigated the effect of O_2 atmosphere during depositions on $CoFe_2O_4$ film for controlling the electrical transport property. The electrical and magnetic properties were evaluated for the $CoFe_2O_4$ films fabricated in various O_2 partial pressures.

1. Introduction

Spinel ferrite is one of the most promising materials in the spintronics field owing to half-metallicity, spin filter effect and high Curie temperature. Since such ferrites, $CoFe_2O_4$, $MnFe_2O_4$ and $ZnFe_2O_4$ etc., are the electric insulators, the uses in the spinctronics are restricted. If we succeed in controlling the electric conductivity, it extends potential as room temperature spintronics devices. In recent research, semiconducting characteristics of the spinel ferrites were realized by varying composition of cations[1][2][3]. On the other hands, $NiFe_2O_4$ grown by magnetron sputtering in Ar atmosphere (without oxygen gas) showed low resistivity [4]. Such low resistivity could be attributed to the coexisting of Fe^{2+} and Fe^{3+} in B sublattices, which caused by composition modulation or oxygen defects. Therefore, sophisticated controls of the O_2 partial pressure during the deposition is considered to realize fine-tuning of the resistivity in the spinel ferrite. However, few researches about controlling electrical properties by O_2 pressure have been reported so far. Furthermore, in previous researches, spinel ferrites were grown by PLD or sputtering from “oxide sources”. It is considered to be difficult to control the oxidation states of the films by such fabrication technique.

In this research, we fabricated $CoFe_2O_4$ film, which is ferrimagnetic insulator originally, by reactive molecular beam epitaxy method in various O_2 partial pressure using metal Fe and Co sources. This method is expected to realize the control of the oxidation state accurately, leading fine-tuning of the transport property. We also investigated the magnetic characteristics of the films because the oxygen defects may affects magnetic property.

2. Experiments

$CoFe_2O_4$ films were grown on MgO(100) substrate by reactive molecular beam epitaxy method (Base Pressure : ~10^{-8} Pa). The film structures were MgO(100)/$CoFe_2O_4$(30 or 50 nm)/Al_2O_3(3 nm). $CoFe_2O_4$ thin films were grown by co-evaporation of Fe and Co at 300℃ in O_2 atmosphere(0.1~ 4×10^{-4} Pa). Growth rate of $CoFe_2O_4$ was 0.4 Å/s. Al_2O_3 was deposited at room temperature for preventing excess oxidation of $CoFe_2O_4$ in the air. The epitaxial growth and surface morphology was observed by RHEED and AFM. We measured electrical resistivity by Van der Pauw method and four terminal method. We confirmed magnetization process by MOKE measurement. The oxidation states and composition ratio of the $CoFe_2O_4$ were estimated by XPS. We determined charge carrier from Seebeck coefficient.

3. Results and discussion

Fig. 1 shows the RHEED images for (a) 4×10^{-4} Pa, and (b) 3×10^{-5} Pa in the [001] azimuth. The $CoFe_2O_4$ film showed typical diffraction patterns from the (100) plane of the spinel structure. The sharp streaks suggest high crystallinity and flat surface in both films. The diffraction patterns of the films grown under 2×10^{-5} Pa were spotty and were not like the spinel structure (not shown). From AFM measurements, the surface roughness was less than 0.3 nm.

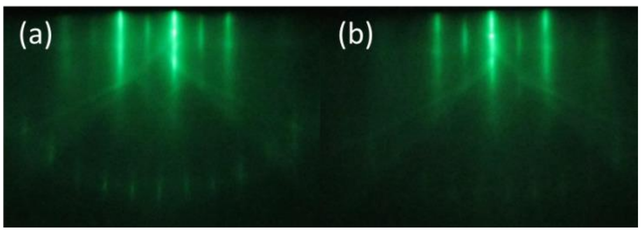

Fig.1. RHEED patterns for heteroepitaxial growth of $CoFe_2O_4$ thin films on MgO (100) in O_2 atmosphere (a) 4×10^{-4} Pa (b) 3×10^{-5} Pa

Fig.2 shows XPS results for the films grown in various O_2 pressure. Peak shift in Fig.2.(a) suggests that the deposition under low oxygen pressure yielded increase of Fe^{2+} /Fe^{3+} ratio. Fig.2.(b) exhibited clear peak shifts, which suggests the presence of the un-oxidized metallic Co. These results mean that the oxidation states are controllable by O_2 atmosphere during the deposition. Composition ratio between Fe and Co was also evaluated from the spectra. The result is Fe:Co = 1:1.92~1.96. Since composition ratio was constant, change of electrical resistivity came from the O_2 pressure, not from the composition fluctuation.

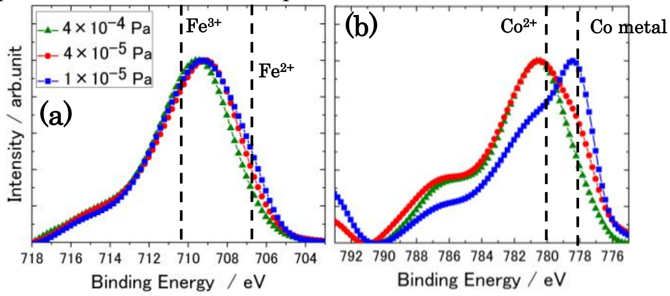

Fig.2. XPS spectra of (a) Fe $2p_{3/2}$ (b) Co $2p_{3/2}$ core level for the $CoFe_2O_4$ films.

Fig.3 shows O_2 partial pressure dependence of electrical resistivity of the films. $CoFe_2O_4$ films grown in low O_2 pressure exhibited low resistivity. The tendency indicates that the electrical transport is enhances by oxygen defects, namely coexisting of Fe^{2+} and Fe^{3+} in B sublattices or insufficient oxidation of Co.

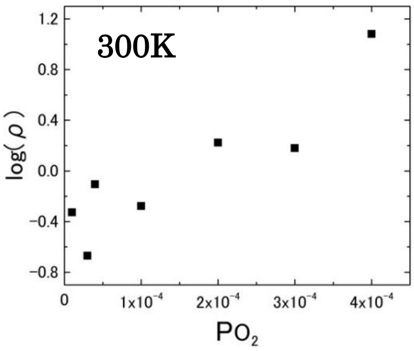

Fig.3. O_2 partial pressure dependence of Resistivity of the $CoFe_2O_4$ films grown in various O_2 pressures

Fig.4 shows (a) temperature dependence of resistivity ρ of the $CoFe_2O_4$ films grown in 4×10^{-4} Pa and (b) the Arrhenius plot. Semiconducting behavior was observed. The activation energy was estimated at 55 meV from the Arrhenius plot. That value is rather small compared to the previous study in which the films were fabricated by PLD[1]

Seebeck measurements were also carried out to determine the carrier type. The Seebeck coefficient for films grown in 4×10^{-5} Pa and 3×10^{-5} Pa were -140.6 μV/ K and -164.4 μ V / K, respectively. Negative value means that charge carrier was electron. Increment of Seebeck coefficients implied increasing of electron density.

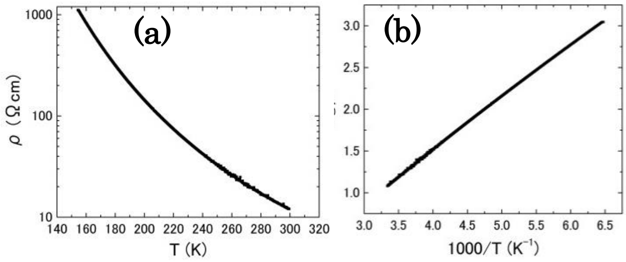

Fig.4. (a) Temperature dependence of resistivity ρ of the $CoFe_2O_4$ films grown in 4×10^{-4} Pa. (b) The ρ vs 1000/T plots are fitted to the Arrhenius law

Fig.5 shows magnetic hysteresis loop measured by MOKE in out-of-plane magnetic field (1.6T). The in-plane MOKE measurements in magnetic field of 0.2T showed no hysteresis loop (not shown). Although in-plane magnetic field was not sufficient, these results suggest that $CoFe_2O_4$ films have perpendicular magnetic anisotropy. All the loops in Fig. 5 have similar shapes, so that the magnetic property was not affected by O_2 partial pressure during deposition.

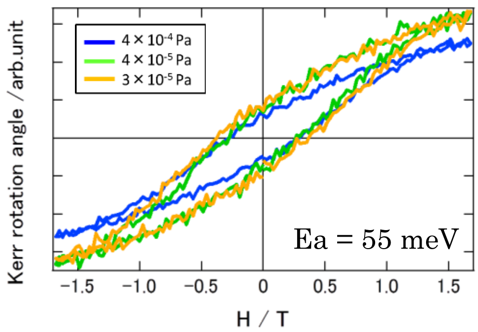

Fig.5. O_2 partial pressure dependence of Resistivity of the $CoFe_2O_4$ films grown in various O_2 pressures

4. Summary

We fabricated $CoFe_2O_4$ films in various O_2 pressure. From RHEED and AFM measurements, $CoFe_2O_4$ films have high crystallinity and vary flat surface. The films grown in low O_2 pressure showed low electrical resistivity, which is attributed to the presence of Fe^{2+} and metal Co. The temperature dependence of the resistivity obeyed the Arrhenius law. Such semiconductive transport properties extend the potential of spinel ferrites as new spintronics materials.

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