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Composition dependence of electronic states and valencies in epitaxial LaNi_{1.x}Mn_xO₃ thin films studied by X-ray absorption spectroscopy

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Introduction

Spintronics has attracted much attention as a new technology that takes advantage of not only electric charge but also spin. La, NiMnO₆ is a ferromagnetic semiconductor that has a potential of applying to spintronics devices because of its ferromagnetic transition temperature ($T_c \sim 280$ K). The ferromagnetism of LaNi_{0.5}Mn_{0.5}O₃ is explained by the mechanism due to a superexchange interaction in Ni-O-Mn, which is predicted theoretically by Kanamori-Goodenough (G-K) rules. However, electronic states of transition metals responsible for the ferromagnetic interaction have not been elucidated yet. G-K rules predict both $Ni^{2+}(d^8)$ -O- $Mn^{4+}(d^3)$ [1] and $Ni^{3+}(d^3)$ -O- $Mn^{3+}(d^4)$ [2] interactions to exhibit a ferromagnetic behavior. In this study, X-ray absorption spectroscopy (XAS) of LaNiO₂-LaMnO₃ composition spread films was carried out to evaluate the valence states of transition metal ions for studying the origin of ferromagnetism in LaNi_{0.5}Mn_{0.5}O₃.

Experimental

Epitaxial LaNiO₃-LaMnO₃ composition spread films were fabricated on LaAlO₃(100) substrates using a pulsed laser deposition method. During deposition, the substrate temperature was kept at 600 °C at an oxygen pressure of 100 mTorr. Composition and structures of the films were characterized by X-ray fluorescence (XRF) and X-ray diffraction (XRD), respectively. XAS spectra were obtained by measuring the sample drain current at an undulator BL-2C beamline.

Results and discussion

Figure 1 shows Ni $2p \rightarrow 3d$ absorption spectra (a) and Mn $2p \rightarrow 3d$ absorption spectra (b) at room temperature. With increasing the incorporated Mn into LaNi³⁺O₃, Ni $2p_{3/2}$ single peak is becoming split into double peaks where the lower energy peak is emerging, as shown in Fig. 1 (a). Mederde *et al.* reported that Ni $2p_{3/2}$ spectra of NdNi³⁺O₃ and PrNi³⁺O₃ are quite different from that of Ni²⁺O [3]. By comparing our Ni $2p_{3/2}$ XAS spectra with the reported spectra, it is indicated that the valency of Ni varies from Ni³⁺ to Ni²⁺ as a result of Mn substitution in

LaNi³⁺O₃. Mn $2p_{3/2}$ spectra also show changes in shape with the substitution of Ni by Mn, as shown in Fig. 1 (b). This suggests that the replacement of Ni by Mn increases the valency of Mn from Mn³⁺ to Mn⁴⁺, which is consistent with La_{1-x}Sr_xMnO₃ XAS spectra obtained by Abbate *et al*. [4]. Thus, the valency changes from Ni³⁺ to Ni²⁺, and from Mn³⁺ to Mn⁴⁺ due to the charge transfer from Mn to Ni were clearly observed. This charge transfer results in a ferromagnetic semiconductor LaNi_{0.5}Mn_{0.5}O₃ consisting of mainly Ni²⁺ and Mn⁴⁺. Therefore, it is suggested that a superexchange interaction between Ni²⁺ and Mn⁴⁺ via an oxygen ion rather than Ni³⁺ and Mn³⁺ is the origin of ferromagnetism in LaNi_{0.5}Mn_{0.5}O₃.

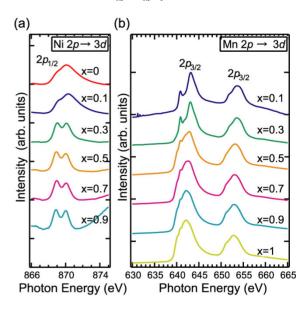


Fig. 1: XAS spectra of LaNi_{1-x}Mn_xO₃: (a) Ni $2p \rightarrow 3d$, (b) Mn $2p \rightarrow 3d$.

References

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