Electronic Structure of Ferromagnetic Perovskite Manganite Thin Films Investigated by Hard X-ray Photoemission Spectroscopy

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Transition metal oxides exhibits a rich variety of electric and magnetic properties including colossal magnetoresistance, ferromagnetism with metallic conduction, perfect spin polarization, charge/orbital ordering due to the strong coupling between spin, and charge, and are also important in relation to applications pertaining to spintronic heterostructure devices working at room temperature. We report the bulk sensitive Hard X-ray core level photoemission spectroscopy to investigate the intrinsic electronic structure of perovskite manganites, typically the strained (La,Ba)MnO₃ thin films [1] showing enhanced ferromagnetism and electron doped (Nd,Ce)MnO₃ thin film [2].

In case of the strained (La,Ba)MnO₃ thin films, a new sharp satellite peak appeared at the low energy site of the Mn 2p₃/₂ main peak in a 20nm thick well-strained film with strongly enhanced ferromagnetism, whereas a broader signal was observed for the unstrained film with 300nm thickness as shown in Fig.1. Cluster calculations revealed that the intensity corresponded to the density of the state at the Fermi level relating to the magnitude of the ferromagnetic order. The satellite intensity also agreed quantitatively with the square of the magnetization.

In case of (Nd, Ce)MnO₃ thin film, it can be seen that there is a clear difference between NCeMO and LBMO thin films on the spectra, indicating the existence of Mn²⁺, i.e. the mixed valence state of Mn³⁺ and Mn⁴⁺ within NCeMO film as shown in Fig.2. It should be noted that these experimental results, especially the stoichiometry of NCeMO film on the microstructure and the mixed valence state of Mn²⁺ and Mn³⁺, are demonstrated for the first time.

For ferromagnetic manganite, Hard X-ray core level photoemission spectroscopy makes it possible to estimate the electronic structure directly related to the magnetization at deep depths even without any surface treatment, and we believe that it can play an important role in the development of novel functional magnetic, electrical and optical devices constructed using transition metal oxides.