

論文の内容の要旨

論文題目

X-ray magnetic circular dichroism study of multiferroic and dilute magnetic materials

(X線磁気円二色性によるマルチフェロイックおよび希薄磁性物質の研究)

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Spintronics materials like multiferroics and dilute magnetic semiconductors (DMSs) have attracted a great deal of attention in the scientific community from the viewpoints of both academic research and practical applications. In order to elucidate the origin of ferromagnetism of these spintronics materials, it is necessary to investigate the electronic structure. Advances in experimental techniques such as synchrotron radiation and electron spectroscopies provide us with great opportunities to unravel the underlying physics producing the magnetic properties of the materials. In this thesis, I have investigated the electronic structure of spintronics materials using x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD).

Recently, thin films consisting of alternating ferrimagnetic spinel-type NiFe_2O_4 (NFO) and ferroelectric perovskite-type BaTiO_3 (BTO) layers $\text{BTO}/(\text{NFO}/\text{BTO})_n$ were prepared and exhibited the coexistence of ferroelectric and ferromagnetic ordering with strong magnetoelectric (ME) coupling between them [1, 2], but the origin of ME coupling is still remains unclear because in previous studies, the techniques used to characterize the ME coupling of NFO/BTO heterostructures were mostly measure macroscopic quantities. For that purpose, XAS and XMCD at the both Ni and Fe $2p \rightarrow 3d$ absorption edges are an ideal

technique to clarify this issue because they are element-specific microscopic probes. Here, I have studied the local electronic and magnetic states of Ni and Fe ions in the NFO/BTO multilayers grown on (001)-SrTiO₃ substrates using pulsed laser deposition with various NFO and BTO thicknesses by XAS and XMCD in the bulk-sensitive total-fluorescence yield (TFY) mode at room temperature. The measured Ni 2*p* and Fe 2*p* spectra indicate that the Ni ions are octahedrally coordinated by oxygen and are divalent (Ni²⁺) and that the Fe ions are trivalent (Fe³⁺) and are tetrahedrally or octahedrally coordinated by oxygen with opposite spin directions, consistent with the inverse spinel structure of NiFe₂O₄. With increasing number of layers, both Ni and Fe magnetic moments decrease. With decreasing NFO layer thickness, the average magnetic moment of the Ni ions decreases while the average magnetic moment of the Fe ions remain unaffected, meaning that Ni ions replaced by Fe ions at the interface lose ferromagnetic ordering. I found clear correlation between the ME coupling strength and the ferromagnetic moment of Fe. This suggests that the enhancement of ME coupling occurs at interfacial Fe-TiO₂ bonding. The Fe-TiO₂ bonding creates oxygen vacancies and the oxygen vacancies at the interface may play an important role to enhance the ME coupling.

Further, I performed XAS and the XMCD studies of Cd_{1-x-y}Mn_xCr_yTe thin films with Cr content *y* varied from 0 to 0.04 while keeping the Mn content *x* fixed around 0.20 grown on GaAs (001) substrates by the molecular beam epitaxy (MBE) technique. The prototypical DMS Cd_{1-x}Mn_xTe is a spin-glass (*x*<0.6) or antiferromagnetic (*x*>0.6). Shen *et al.* [3] found that Cr doping into Cd_{1-x}Mn_xTe turns the system from the antiferromagnetic spin glass to a ferromagnet. In order to elucidate the origin of ferromagnetism, interaction between Mn and Cr has to be clarified. I found that both Cr and Mn ions are divalent and that the spins alignment of Cr and Mn are parallel. The ferromagnetic moment of Mn increases with Cr concentration. I found the equal concentration of ferromagnetic Cr and Mn ions in Cd_{0.76}Mn_{0.2}Cr_{0.04}Te sample. These results suggest that in the presence of Cr ions in Cd_{1-x-y}Mn_xCr_yTe, the interaction between Mn spins changes from antiferromagnetic to ferromagnetic mediated by the Cr ions possible mechanism of ferromagnetic ordering between Mn and Cr ions is due to the double exchange interaction.

Next, I have studied the possible existence of orbital magnetic moment in multiferroic MnWO₄ using the Mn *L*_{2,3}-edge XMCD. Shanavas *et al.* [4] reported a

finite but very small orbital magnetic moment from ab initio electronic-structure calculations. By applying the orbital sum rule to the XMCD spectrum, despite the d^5 configuration of Mn^{2+} ion, a significantly large orbital magnetic moment was deduced. The distorted $(\text{MnO}_6)^{10-}$ octahedra play an important role in giving rise to the apparently large orbital magnetic and electric polarization in MnWO_4 . The orbital magnetic moment is in the same direction of the spin magnetic moment, indicating that Mn $3d$ states are more than half-filled. Using the CI cluster-model analysis, I found that the average $3d$ occupancy, n_{3d} , is equal to 5.09.

References

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