

Dissertation Abstract

論文の内容の要旨

Dissertation title: X-ray magnetic circular dichroism study of oxide-based magnetic materials and half-metallic alloys

(酸化物磁性体とハーフメタル合金の X 線磁気円二色性による研究)

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The study of spintronics materials such as diluted magnetic semiconductor (DMSs), multiferroic and half-metallic alloys is one of the most attractive fields in science from the viewpoints of both academic research and applications. In order to clarify the origin of ferromagnetism of these spintronics, it is necessary to investigate the electronic structure. In this thesis, we have investigated the electronic structure of spintronics materials using x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD).

The first discovery of room-temperature ferromagnetism in Co-doped TiO_2 by Matsumoto *et al.* [1] has aroused great interest in the search for such materials and a number of studies have been made to investigate whether the ferromagnetism is carrier-mediated or not [2-3], but the issue still remains controversial. XMCD at the Co $2p \rightarrow 3d$ absorption (Co $L_{2,3}$) edge is an ideal technique to clarify this issue because it is an element-specific magnetic probe. Our previous XMCD study has revealed that the ferromagnetism is not due to segregated Co metal clusters but is due to Co^{2+} ions in the TiO_2 matrix [4]. However, the XMCD signal intensities were an order of magnitude lower than that expected from the bulk magnetization [4]. We performed XAS and XMCD studies on rutile Co-doped TiO_2 by the surface-sensitive total electron yield (TEY) mode and the bulk-sensitive total fluorescence yield (TFY) mode and found that Co ions in the bulk indeed have a large moment of $0.8\text{-}2.2\mu_B/\text{Co}$ [5]. Then we extended the same approach to anatase Co-doped TiO_2 and studied the correlation between magnetism and transport properties.

We have performed the XAS and the XMCD studies of $(1-x)\text{BiFeO}_3\text{-}x\text{BiCoO}_3$ (BFCO) thin films (where $x = 0$ to 0.30) grown on $\text{LaAlO}_3(001)$ substrates using a chemical solution deposition technique. The XAS results indicated that the Fe ions were in the Fe^{3+} state and that the Co ions are in the Co^{3+} . XMCD results showed that the Fe ions were ferromagnetic at room-temperature and that the Co ions were in the paramagnetic. The XMCD measurements also revealed that antiferromagnetically are Fe^{3+} ions tetrahedrally co-ordinated by oxygen and Fe^{3+} ions octahedrally co-ordinated by oxygen. The magnetic moment of the Fe ions increases up to 20% Co content and after that it decreases. However, the Co magnetic moment was nearly independent of Co content unlike Fe, and the peak at 20% Co showed only a minor influence. The magnetization deduced from XMCD is larger than that obtained by SQUID measurements, indicating the enhancement of ferromagnetism within ~ 5 nm from the surface, probed by the total electron yield (TEY) method.

We studied the magnetic and electronic states of $\text{Co}_2\text{Mn}_\beta\text{Si}_{1.93}$ (CMS)/MgO and $\text{Co}_2\text{Mn}_\beta\text{Ge}_{0.38}$ (CMG) magnetic tunnel junctions by (Mn) composition (β) dependent by means of XMCD

measurements. In particular, the Mn composition (β) dependence of the Mn and Co magnetic moments was investigated. With an increase of β in the CMG films, the spin magnetic moment of Mn decreased which is consistent with Picozzi *et al.*'s [6] calculations which indicate that the anti-site (or excess) Mn is either paramagnetic or nonmagnetic. The Mn $L_{2,3}$ -edge XAS showed a Mn^{2+} -like multiplet structure in MnO, in contrast to lower value of β . The Co spin magnetic moment for all samples was obviously larger and/or equal to a theoretical value of $1\mu_B$. For Co-rich region, there is the possibility of the existence of CoMn because $m_{\text{spin}}(\text{Co})=1.5\mu_B$ is larger than $1.06\mu_B$ for Co at the regular Co site, Co_{Co} which is also consistent with Picozzi *et al.* [6]. These Co-rich films composition imply the presence of Co antisites that would lower the spin polarization at the Fermi level. For Mn-rich region, $m_{\text{spin}}(\text{Co})$ is almost the same as theoretically predicted value, i.e., there is no effect of excess Mn on $m_{\text{spin}}(\text{Co})$. A Co^{2+} -like multiplet structure in CoO was not observed in any films, indicating that, the Co atoms were not oxidized. For CMS the $m_{\text{spin}}(\text{Mn})$ behavior was same as CMG and in this case we did not observed oxidation of Mn. However, $m_{\text{spin}}(\text{Co})$ was almost independent of β . This is because the Co-rich CMS might have had more or less the same amount of CoMn as in Co-rich CMG.

References

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