論文の内容の要旨

論文題目

Systematic Study on Microscopic Ferromagnetism and High T_c in Anatase Co-doped TiO_2 Epitaxial Thin Films

（アナターゼ型CoドープTiO$_2$エピタキシャル薄膜の微視的強磁性と高T$_c$に関する系統的研究）

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Ferromagnetic oxide semiconductor Co-doped TiO_2 ($Ti_{1-x}Co_xO_{2-\delta}$; $(Ti,Co)O_2$) is a promising candidate of semiconductor spintronic materials. This compound has high ferromagnetic Curie temperature, T_C (> 600 K). Its ferromagnetism at room temperature can be controlled via electrostatic and chemical carrier doping. Although there are many reports on $(Ti,Co)O_2$especially for high quality film fabrication, bulk magnetization and controlling of magnetic property, there are still several issues on this compound related to microscopic ferromagnetism, magnetically dead layer in the surface and mechanism of the high T_C.

I conducted microscopic investigation of ferromagnetism in anatase $(Ti,Co)O_2$ thin film by means of the observation of magnetic domain structure. The observation of microscopic magnetic domain structure is an evidence of microscopic ferromagnetism and also a key to realize spintronic applications such as current-induced magnetic domain wall motion. In this dissertation, I clearly observed magnetic domain structure in $(Ti,Co)O_2$ at room temperature for the first time. Magnetic force microscope operated under vacuum condition enabled to resolve the individual magnetic domains, from which the micromagnetic parameters were also evaluated. In addition, I was able to relate the evolution of magnetic domain structure with the carrier density as well as the magnetization from bulk measurement. This result is consistent with the carrier mediated ferromagnetism reported previously.

To protect the film from magnetically dead layer at the surface, non-magnetic capping layer was developed in this experiment. With only a 2 nm-thick capping layer, the improved magnetization as well as magnetic anisotropy of anatase $(Ti,Co)O_2$ was observed at 300 K. By using the capping layer, the film thickness was able to be decreased to very thin thickness with maintaining the magnetization. As a result, the magnetic domain structure in ultrathin film was successfully observed at room temperature because of improved surface magnetization. In addition, the stability of film was substantially improved by the capping layer.

In order to investigate the origin of high T_C, the T_C vs. carrier density relationship was systematically studied. The T_C of anatase $(Ti,Co)O_2$ thin films as function of carrier density was investigated by using electrical transport and magnetization measurements at lower temperature than T_C in order to avoid unintentional change of carrier density at high temperature. A non-monotonic relationship of T_C vs. carrier density was obtained implying a formation of bound magnetic polarons in addition to carrier-mediated interaction. Some possible substructures which could response for the nucleation of magnetic polarons, were proposed by using density functional theory (DFT) calculation.

In conclusion, I succeeded in observing magnetic domain structure in $(Ti,Co)O_2$ at room temperature and microscopic ferromagnetism was confirmed by homogeneous emergence of magnetic domains. I also developed non-magnetic capping layer to protect surface oxidation resulting in the fabrication of ultrathin magnetically active film. The first report of systematic study of T_C vs. carrier density relationship in $(Ti,Co)O_2$ was presented suggesting a non-monotonic relationship. This suggests a possibility of bound magnetic polarons in concert with carrier-mediated interaction which electrons play an important role. It is expected that these results will contribute to magnetic domain manipulation and future development of spintronics devices using this compound. Furthermore, the presently proposed ferromagnetic mechanism could also provide a guideline to explore new high T_C ferromagnetic semiconductors.