Impurity band conduction and AHE in anatase Co-doped TiO₂ ferromagnetic semiconductor

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Oxide ferromagnetic semiconductors are considered to be viable alternatives to III-V based ferromagnetic semiconductors for practical spintronic devices primarily due to their room temperature ferromagnetism. While there is significant debate about the origin of ferromagnetism in transition metal doped oxide semiconductors, several reports point to an intrinsic origin and suggest the ferromagnetism is possibly mediated by free carriers, originates from magnetic polaron formation, or exchange in an impurity band. Nevertheless, a conclusive proof has not yet been presented. This also applies to Cobalt doped titanium dioxide (CTO), which we have studied here. In addition to magnetic and structural properties, we have used the Hall and Anomalous Hall Effect (AHE) to characterize the electrical transport in Co-doped TiO₂ to study the role of carriers in ferromagnetism. We show that Co-doped TiO₂ in anatase phase exhibits a hitherto not observed hysteretic AHE effect consistent with the magnetic response, as well as a transition to impurity band conduction.

Cobalt doped TiO₂ thin films were grown by pulsed laser deposition on TiO₂ terminated SrTiO₃ substrates at relatively low temperatures and low pressures to stabilise the anatase phase. Growth parameters such as oxygen pressure, target-substrate distance and laser repetition rate were varied. Cobalt doping was kept at 1.4 at % in order to stay below the solubility limit. Structural analysis by X-Ray and Transmission Electron Microscopy reveals only the anatase phase up to a film thickness of 150nm, and no evidence of phase segregation or Co-cluster formation. The films are ferromagnetic at room temperature with clear hysteresis and a slight perpendicular anisotropy. The magnetic moment of CTO was determined to be around 1.9 $\mu_{\rm B}$ / Co atom. Electronic transport measurements were carried out in the range of 10-300K and a comparison with un-doped TiO₂ films grown under similar conditions was made. From the Hall transport measurements, the Co-doped films were found to be n-type semiconductors with a relatively low carrier density of the order of 10^{18} /cm⁻³. The films have a resistivity of about 0.1 Ω cm and rather high mobility (10-50 cm²/Vs) at room temperature. Thus, they have reasonably good semiconductor properties. Studies as a function of temperature reveal that cobalt doping induces a maximum in the Hall coefficient at 100K indicating that the conduction mechanism changes from conductance by carriers thermally activated into the conduction band, to impurity band conduction at lower temperatures. In contrast, the un-doped films grown under similar condition show the usual exponential variation of the Hall coefficient, indicating carrier freeze out at lower temperatures typical for a semiconductor. Significantly, Co-doped TiO₂ in anatase phase also exhibits a hitherto not observed AHE effect over the full temperature range including room temperature, with hysteresis consistent with the magnetic response.

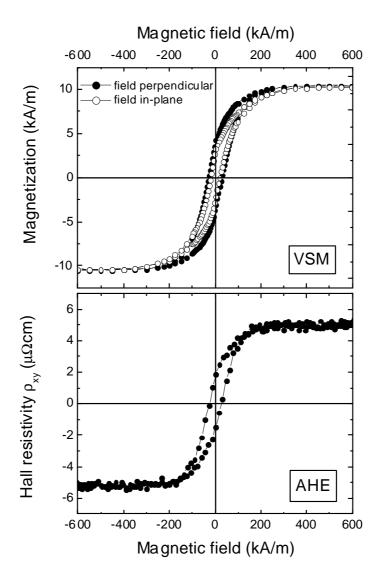


Fig 1. Magnetic hysteresis loops (top panel) and Anomalous Hall Effect (bottom panel) observed on Cobalt doped TiO_2 films at room temperature. For the AHE curve the linear background slope due to the ordinary Hall effect has been substrated.