Spin Canting and Orbital Order in Spinel Vanadate Thin Films

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Introduction

The spinel vanadates are geometrically frustrated spin systems, of interest due to the structural, magnetic, and electronic phase transitions that occur when exposed to perturbations. Of particular interest is CoV_2O_4, because of the localized-itinerant crossover physics that can be investigated by tuning the V-V distance. Furthermore, this material shows a collinear ferrimagnetic transition at T_N = 150 K, as well as a magnetic transition at T* = 90 K that has been suggested as a transition to a non-collinear state showing spin canting, accompanied by orbital ordering [1]. However, the 90 K transition has been difficult to observe reproducibly in powders and single crystals, thought to be caused by the materials' proximity to itinerancy. Experiments on powders have observed a weak first order structural phase transition that accompanies the magnetic transition at T* [2]. We have grown high quality thin films of CoV_2O_4 for the first time, and have stabilized an orthorhombic structure of the material, resulting in a stabilization of the T* transition, as well as dramatic reorientation of the low temperature magnetic structure [3].

Experimental

Thin films of CoV_2O_4 were grown using pulsed laser deposition on (001) $SrTiO_3$ substrates (lattice mismatch of ~7.5%), from a homemade pressed pellet of CoV_2O_6. X-ray diffraction measurements were performed to verify the high quality of the films, and to determine the structure, epitaxy, and crystallinity of the samples. Magnetic measurements were performed on films using a Quantum Design Magnetic Properties Measurement System at the National High Magnetic Field Lab to determine the macroscopic magnetic properties of the material. Neutron scattering measurements were performed at Oak Ridge National Lab. Epitaxial CoV_2O_4 thin films exhibit an orthorhombic structure with lattice parameters *a* = 8.36(2) Å, *b* = 8.24(5) Å), and *c* = 8.457(3) Å, i.e., different from the cubic bulk structure (*a* = 8.4 Å). Films with thicknesses varying between 50 -300 nm all have the same lattice parameters, and show dramatic differences in the magnetic properties in comparison to bulk [3].

Results and Discussion

Figure 1 shows a representative magnetization measurement measured as a function of temperature with the magnetic field applied along different crystallographic directions. There are two clear magnetic transitions in the material at T_N = 150 K, and T* = 90 K. The T_N transition is a ferrimagnetic transition similar to that of bulk. However, the structural changes in our material strongly stabilize the T* transition, which is much weaker in bulk. Furthermore, unlike the minor spin-canting the T* transition is attributed to in bulk, our films undergo dramatic reorientation of the spins, from an out-of-plane easy axis in the ferrimagnetic state, to an in-plane easy axis below T*, implying the single ion anisotropy of the Co ions must be changing. Neutron scattering measurements in zero field confirm a low-temperature single-domain state with an easy axis along the [110] direction, and spin canting of the V ions of at least ~20°. Our findings imply longer-ranged orbital order and increased localized character of V moments relative to bulk, indicating epitaxial strain as a viable tuning method of orbital order and itinerancy in spinel vanadates, and other frustrated antiferromangets [3].

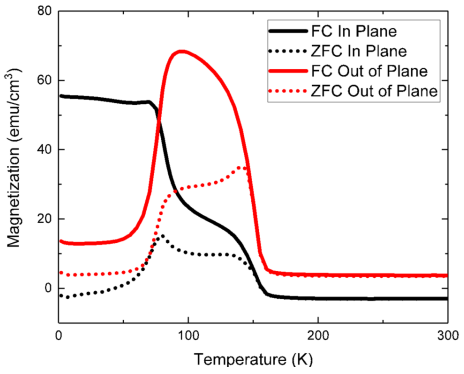

Fig. 1: Representative magnetization vs. temperature curve (t = 100 nm) for an orthorhombic film measured in H = 1000 Oe (red line, H applied out of plane of the film, along the c axis; black line, H applied in the film (ab) plane) , ZFC dotted lines, and FC solid lines.

Acknowledgements

A portion of this work was performed at the National High Magnetic Field Laboratory, which is supported by National Science Foundation Cooperative Agreements No. DMR-1157490 and No. DMR-1644779, and the State of Florida. A portion of this research used resources at the High Flux Isotope Reactor, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory. Use of the Advanced Photon Source was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357. G.M. and D.R. were supported by the National Science Foundation, under Grant No. DMR-1455264

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