Chemical Tuning of the High-Energy Magneto-Dielectric Effect in a Frustrated Kagome Lattice Material

Musfeldt, J.L., Rai, R.C., Cao, J., Vergara, L.I., Brown, S. (Tennessee), Kasinathan, D. (Davis), Singh, D.J. (ORNL), Lawes, G. (Wayne State), Rogado, N., Cava, R.J. (Princeton), and McGill, S. (NHMFL)

Introduction

Magnetic frustration and competing interactions in Kagome lattice materials give rise to complex magnetic field - temperature phase diagrams in the $M_3V_2O_8$ system (M = Ni, Co, Mn). In $Ni_3V_2O_8$, the low temperature incommensurate state displays both ferromagnetism and a finite polarization. One manifestation of magneto-electric coupling is a large static magneto-dielectric contrast. Sizable magneto-dielectric effects can also extend to relatively high energy in complex oxides due to spin-lattice-charge coupling. This work was done using a combination of optical spectroscopy, first principles calculations, and energy dependent magneto-optical measurements to elucidate the electronic structure and to study the phase diagram of $Ni_3V_2O_8$. The variable field work was carried out at the NHMFL.

Results and Discussion

We recently discovered that sizable magneto-dielectric contrast can extend to relatively high energy in complex oxides. The effect can be positive or negative depending on the energy (Fig.1). In $Ni_3V_2O_8$, the intermediate gap, local moment character favors the observed field-induced modification of local structure and resulting high-energy dielectric contrast. Since high-energy magneto-dielectric contrast is a spectroscopic rather than capacitance measurement, dead layer contact and "leaky" magneto-resistance issues are avoided. Structure-property trends in the M = Ni, Co, Mn isostructutral series are extremely interesting. The magneto-dielectric contrast is tunable depending on the chemical identify of the transition metal center (Fig. 1), an effect that we attribute to a combination of lattice stiffness and hybridization effects. Complementary vibrational measurements on $Ni_3V_2O_8$ single crystals reveal changes in magneto-elastic coupling between the nearly degenerate magnetic ground states and pinpoint the lattice displacements that are most important for establishing the state with magneto-electric cross-coupling. A softer lattice in the Co analog compound works to reduce the high energy magneto-dielectric contrast (because the lattice relaxes at high temperatures and does not couple to the low temperature magnetic transitions) and remove the interesting magneto-electric phase from H-T phase space.

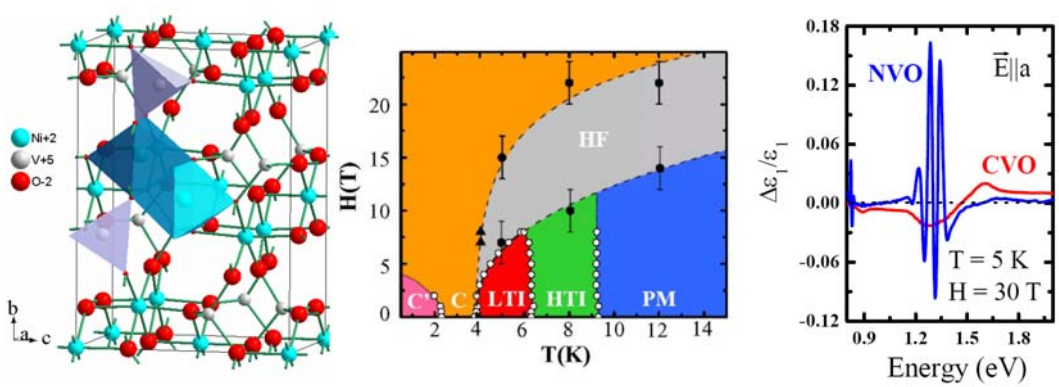

Figure 1: Crystal structure, H-T phase diagram, and high energy magneto-dielectric contrast for $Ni_3V_2O_8$ compared with that of the Co analog, Refs. 1 and 2.

Acknowledgements

We thank the U.S. Department of Energy for support of this work.

References

[1] Rai, R.C., *et al.*, Phys. Rev. B, **74**, 235101 (2006).
[2] Rai, R.C., *et al.*, Phys. Rev. B, **76**, 174414 (2007).