Magnetoelectric coupling in Ni_3TeO_6

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Introduction

The development of materials in which 3*d* transition metals coexist with heavier centers in multicomponent oxides continues to fascinate. In these systems, the 3*d* ions contribute narrow bandwidths, robust magnetism, and strong electron-electron interactions while 4*p* and 5*d* ions exhibit strong spin-orbit coupling, larger bandwidths, and tendencies to dimerization and band inversion. This mixture of physical and chemical properties provides a superb platform for the discovery of multiferroics, hard magnets, topological insulators, superconductors, and thermoelectrics with enhanced properties. Ni_3TeO_6 attracted our attention in this regard. We recently combined high field optical spectroscopy and first principles calculations to analyze the electronic structure of Ni_3TeO_6 across the 53 K and 9 T magnetic transitions, both of which are accompanied by large changes in electric polarization,[1,2] and found that the color properties are sensitive to magnetic order due to field-induced changes in the crystal field environment, with those around Ni1 and Ni2 most affected. The recent discovery of a spin reorientation transition at 52 T [Fig. 1(a)] provides an opportunity to extend toward new states of matter sporting giant polarizations.[3]

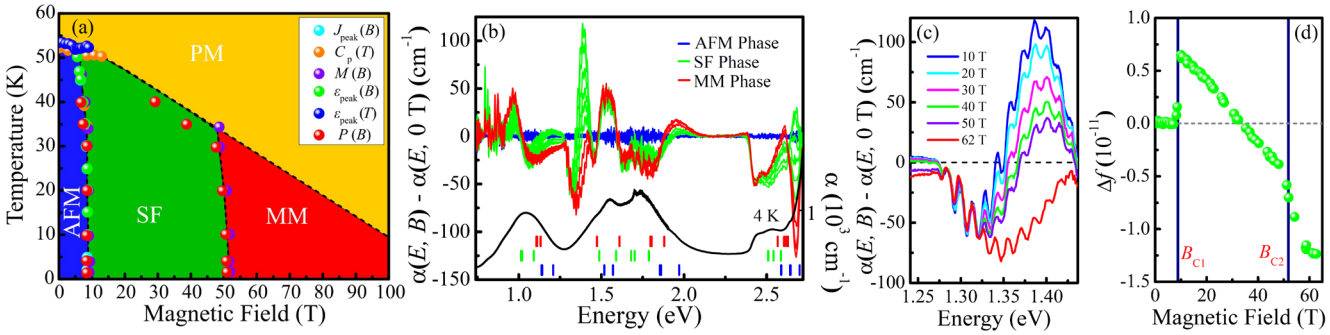

Figure 1: (a) Temperature - magnetic field phase diagram of Ni_3TeO_6 as determined by heat capacity, magnetization, polarization, and magnetostriction showing the 53 K, 9 T, and 52 transitions. Polarization develops below T_N and changes across the spin flop and metamagnetic transitions. The 52 T transition sports a giant change in polarization and huge magnetoelectric effect.[3] (b) Optical absorption, calculated d-to-d excitations for the three different Ni sites, and absorption difference spectra [$\Delta\alpha = \alpha(B) - \alpha(0\ T)$] at fields up to 65 T at 4.2 K. (c, d) Close-up view of the absorption difference spectra near the edge of the $^3A_{2g} \rightarrow {}^1E_g$ excitation and oscillator strength sum rule analysis in the vicinity of this feature.

Results and Discussion

In order to investigate spin-charge coupling across the series of magnetically-drive transitions, we measured the optical properties of Ni_3TeO_6 single crystals and compared our findings with complementary first principles calculations [Fig. 1]. Field-induced changes in the Ni *d*-to-*d* crystal field excitations are strong on the leading edge of the $^3A_{2g} \rightarrow {}^1E_g$ excitation, a finding that we trace back to local lattice distortions due to spin reorientations at 9 and 52 T that modify the optical matrix elements. These findings advance the understanding of magnetoelectric coupling in materials in which magnetic 3d centers coexist with nonmagnetic heavy chalcogenide cations.

Acknowledgements

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References

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