Non-Reciprocal Directional Dichroism in Ni_3TeO_6

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Introduction

Enantiomers are outstanding platforms for exploring symmetry. Sugar molecules, for instance, are chiral, and as a result, rotate the polarization of light. Non-reciprocal directional dichroism extends these ideas – relying on magneto-electric excitations in ultra-low symmetry solids to create "one-way mirror" effects.[1] Prior work focused primarily on the non-reciprocal nature of the electromagnon – for instance, in $BiFeO_3$.[2] More recently, we realized that any magnetoelectric excitation should display a similar response.[3] Further, a symmetry analysis reveals that strongly spin-orbit coupled, chiral multiferroics like Ni_3TeO_6 have the potential to exhibit non-reciprocal directional dichroism in both chiral and ferrotorroidic geometries.[3] Our team is pursuing these ideas from both an experimental and theoretical perspective.

Results and Discussion

To extend our understanding of magnetoelectric coupling and other forms of entanglement, we combined pulse field techniques, optical spectroscopy, a symmetry analysis, and first principles calculations to analyze the electronic structure of Ni_3TeO_6 across the 9 and 52 T magnetic transitions [Fig. 3(b - e)].[4] Crucially, spin-orbit coupling is required to capture how the color properties evolve across these magnetically ordered phases. One consequence of spin-orbit coupling is the possibility that one (or more) of these out-of-equilibrium phases may host non-reciprocal directional dichroism. Because this effect emanates from the product of both magnetic and electronic matrix elements, magnetoelectric multiferroics like Ni_3TeO_6 are good platforms with which to explore this peculiar behavior. Further, we focused on the color properties – which have the potential to provide broadband non-reciprocal directional dichroism. Figure 3(e, f) summarizes our findings thus far. Both field and light direction were varied in these measurements. We find that while the Ni *d*-to-*d* excitations at 1.4 eV are most sensitive to the development of new phases, the color bands near 1.1 eV display the largest non-collinear effects. Moreover, the largest effects occur in the spin-flop phase - which is stable between 9 and 52 T. These large, broadband effects emanate from color band excitations are magneto-electric in nature. Modeling is on-going to fully understand the mechanism and symmetry conditions.

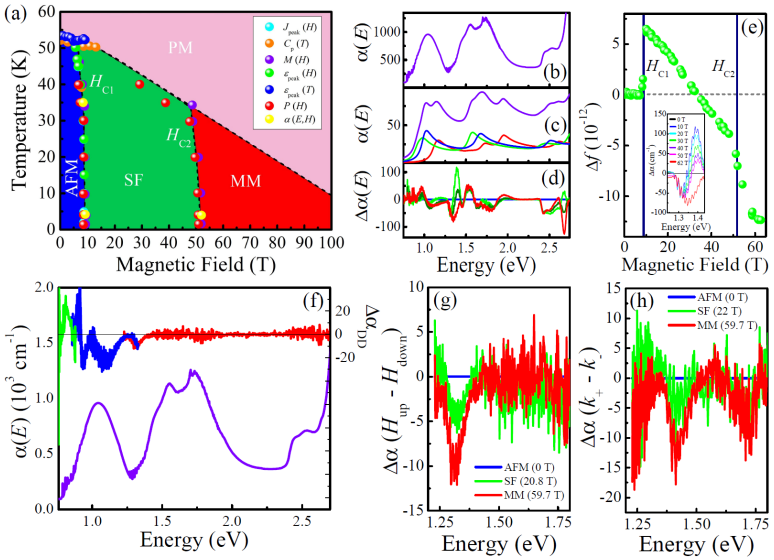

Figure 1: (a) Phase diagram on Ni3TeO6. (b, c, d) Low temperature optical absorption, theoretically predicted optical response (along with projected contributions for Ni1, Ni2, and Ni3), and magneto-optical response in the three phases (AFM, SF, and MM) up to 65 T. (e) Oscillator strength analysis across the phase boundaries. (f) Comparison of optical absorption with the non-reciprocal directional dichroism obtained by pulsing field "up" and "down". (g, h) Close-up comparison on directional dichroism switching field direction vs. that obtained switching the light propagation direction.[4]

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