Magnetoelectric coupling in Ni₃TeO₆

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We measured the optical and magneto-optical properties of Ni_3TeO_6 and combined our findings with complementary electronic structure calculations to understand magnetoelectric coupling in a system with strong spin-orbit coupling. We find striking changes in the polarized optical response through the 9 T spin flop transition that we understand in terms of microscopic nature of the excitations and the development of a substantial polarization. Precursor effects on approach to the spin flop as well as evidence for short range ordering above the 54 K Neel temperature are also observed. The high field response of Ni_3TeO_6 continues to change beyond 9 T, evidence that another magnetically-driven transition is likely to occur at fields higher than 35 T. These findings advance the understanding of magnetoelectric coupling in 4*d*containing materials.