



Colossal magnetoelectric coupling probed to ninety teslas

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Coupling between magnetism and ferroelectricity finds applications in magnetic sensing, high-frequency tunable electronics, and low-power data storage. The challenge is to find large couplings with low hysteresis. We recently showed that Ni_3TeO_6 has a record large non-hysteretic magnetoelectric coupling at 9 T. By extending our measurements to 92 teslas, we discovered an additional phase transition at 52 T, accompanied by a colossal change in polarization of $0.3 \mu\text{C}/\text{cm}^2$, among the largest magnetically-induced polarization changes ever observed.

Here we clarify the origin of the coupling at low and high fields, determining a microscopic model using a powerful combination of magnetic, electric, and structural measurements up to 92 T, along with density functional calculations. We find that the source of the non-hysteretic magnetoelectric coupling at 9 T is an unusual non-hysteretic spin flop to a canted spin structure, while the even larger magnetoelectric coupling at the high-field transition results from the flipping of two Ni spins and a halving of the magnetic unit cell.

This work has motivated additional investigations into molybdates and other related multiferroics with 3d and 4d transition metal oxide ions with a goal to find even larger magnetoelectric coupling effects.

Facilities: Pulsed-Field Facility: 65 T and 100 T magnets.

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