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Effects of 18 O isotope substitution in multiferroic RMnO3 (R=Tb, Dy)

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Multiferroic materials demonstrate desirable attributes for next-generation multifunctional devices as they exhibit coexisting ferroelectric and magnetic orders. In type-II multiferroics, coupling exists that allows ferroelectricity to be manipulated via magnetic order and vice versa, offering potential in high-density information storage and sensor applications. Despite extensive investigations into the subject, questions of the physics of magnetoelectric coupling in multiferroics remain, and competing theories propose different mechanisms. The aim of this investigation was to study changes in the statics and dynamics of structural, ferroelectric and magnetic orders with oxygen-18 isotope substitution to shine light into the coupling mechanism in multiferroic RMnO3 (R=Tb, Dy) systems.

We have performed Raman spectroscopy on 16O and 18O-substituted TbMnO3 single crystals. Oxygen-18 isotope substitution reduces all phonon frequencies significantly. However, specific heat measurements determine no changes in Mn3+ (28 and 41 K) magnetic phase transition temperatures. Pronounced anomalies in peak position and linewidth at the magnetic and ferroelectric phase transitions. While the anomalies at the sinusoidal magnetic phase transition (41 K) are in accordance to the theory of spin-phonon coupling, further deviations develop upon entering the ferroelectric phase (28 K). Furthermore, neutron diffraction measurements on 16O and 18O-substituted DyMnO3 powders show structural deviations at the ferroelectric phase transition (17 K) in the order of 100 fm. These results indicate that structure is actively involved in the emergence of ferroelectricity in these materials.

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