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Effects of ^{18}O isotope substitution in multiferroic RMnO_3 ($R=\text{Tb, Dy}$)

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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 RMnO_3 ($R=\text{Tb, Dy}$) systems.

We have performed Raman spectroscopy on ^{16}O and ^{18}O -substituted TbMnO_3 single crystals. Oxygen-18 isotope substitution reduces all phonon frequencies significantly. However, specific heat measurements determine no changes in Mn^{3+} (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 ^{16}O and ^{18}O -substituted DyMnO_3 powders show structural deviations at the ferroelectric phase transition (17 K) in the order of 100 fm in the b direction. The $Pbnm$ space group is centrosymmetric and therefore does not allow ferroelectricity via atomic displacements, however our Reitveld analysis for the subgroup $P2_1$ shows significant displacements and polarisation along b that is comparable to the experimental value, making it the most promising candidate for ionic displacement induced polarisation in DyMnO_3 . These combined results demonstrate that structure is an important consideration in the emergence of ferroelectricity in these materials.

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