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Unravelling the Magnetoelectric Coupling Mechanisms in TbMnO_3 through Fe3+ substitution

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Multiferroics, such as orthorhombic rare-earth manganites, where both magnetic and ferroelectric orders coexist and are coupled to one another, have attracted great interest. The simultaneity of magnetic and ferroelectric phases gives rise to important effects associated with the cross correlation between order parameters and external fields. A remarkable consequence is that the elementary excitations are not purely magnetic nor polar. Spin waves are mingled with the electric polarization related optical lattice modes, giving rise to the so-called electromagnons, whose spectra provide invaluable information on how magnetism couples to the electric polarization. Once the driving mechanisms are well understood, it is of great importance to aim at enhancing their coupling, the so-called magnetoelectric coupling, which is highly desired for advanced technological applications.
Because $TbMnO_3$ has been the subject of intensive research in this field, it is the ideal case-study compound. In this material, an incommensurate sinusoidal collinear order of the Mn spins occurs at $T_N = 41K$, wherein the Mn spins lie in the bc-plane ($Pbnm$ setting). Below T_{lock} = 28 K, a magnetic transition occurs into a commensurate cycloidal spin order with Mn spins lying in the bc-plane, compatible with the stabilization of an improper ferroelectric polarization along the c-axis [1]. Furthermore, a magnetic field along the b-axis rotates the cycloidal spin order into the ab-plane, and thus, the electric polarization to the a-axis [1]. Previous studies carried out in $TbMnO_3$ ceramics show that the substitution of Mn^{3+} by small amounts of the identically sized Fe^{3+} ion profoundly changes both magnetic and polar structures, altering the magnetoelectric coupling [2]. In fact, for an Fe^{3+} concentration above 5%, the multiferroic properties of the $TbMn_{1-x}Fe_xO_3$ solid solution are lost [2]. Nonetheless, as these studies were done in ceramics, anisotropic effects such as the flop of the cycloidal plane with an applied magnetic field could not be ascertained.
In this work, oriented single crystals of $TbMn_{1-x}Fe_xO_3$ with $x = 0.02$ and 0.04 were used to study the polar, dielectric and magnetoelectric properties versus temperature and magnetic field along the crystallographic directions. To further understand the effect of Mn^{3+} substitution by Fe^{3+}, THz time-domain spectroscopy as a function of temperature and applied magnetic field was performed. The obtained results will be presented emphasizing the effect of temperature and magnetic field on their physical properties for different Fe^{3+} concentrations, highlighting the contrast with previously reported studies on the unsubstituted compound [3, 4].

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[3] Y. Takahashi et al., Phys. Rev. B, 101(18), 187201 (2008)
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Summary

Topic

1. Multiferroics and ferroelectrics

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Classifica Sessioni: Afternoon Session 1