

Poster-2-2

Strain dependent multiferroicity in TbMnO₃ thin films observed by infrared ellipsometry

Jessica Ruffiner,¹ Laurent Bugnon,¹ Alexander Ferguson,¹ Lucile Chassat,¹ David P. Santos-Cottin,¹ Kenta Shimamoto,² Christof W. Schneider,² Yurii G. Pashkevich,^{1,3} Christian Bernhard,¹ and Premysl Marsik¹

¹ Department of Physics, University of Fribourg - Fribourg, Switzerland

² Laboratory for Multiscale Materials Experiments, Paul Scherrer Institut - Villigen-PSI, Switzerland

³ O. O. Galkin Donetsk Institute for Physics and Engineering of NASU - Kyiv, Ukraine

We investigated three TbMnO₃ thin films (200 nm, 91 nm, and 44 nm) on *b*-cut YAlO₃ substrates using infrared ellipsometry at temperatures between 7 K to 300 K. Shimamoto *et al.* grew the films by PLD and reported that strain modifies the multiferroic order in the films and their ferroelectric polarization reaches $2 \mu\text{C cm}^{-2}$ along the *a*-axis. This is significantly larger than the bulk polarization of $0.06 \mu\text{C cm}^{-2}$ oriented along the *c*-axis.

Eight of nine *a*-axis and seven of eight *c*-axis phonon modes were resolved by infrared ellipsometry in all films, along with two longitudinal optical phonons along the out-of-plane (*b*) direction. The 200 nm film exhibits bulk-like phonon behavior. As thickness decreases, most modes systematically blue shift, indicating increased strain. Since YAlO₃ has smaller lattice constants than bulk TbMnO₃, theoretical in-plane compressive strains of -2.1% along the *a*-axis (5.180 Å vs. 5.293 Å) and -0.4% along the *c*-axis (7.371 Å vs. 7.403 Å) are expected, increasing interatomic restoring forces and explaining therefore the observed phonon hardening.

The *Mn-O* stretching/bending mode near 340 cm^{-1} splits in the two thinnest films, but remains unsplit in the thickest. The splitting occurs at higher temperatures and is more pronounced in the thinnest sample. In the orthorhombic structure of TbMnO₃, no degenerate phonons are expected; however, a Raman-active mode exists near 340 cm^{-1} . The activation of the Raman mode in the infrared spectra arises from symmetry breaking due to the induced polarization or zone folding effects related to the magnetic supercell.

[1] K. Shimamoto, S. Mukherjee, S. Manz, et al., Sci. Rep. 7, 44753 (2017).