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Strain Engineering of Multiferroic Thin Films

<u>Christophe J.M. Daumont</u>¹, B., Peter Fonteijn¹, B. Noheda1 and Gustau Catalan^{1,2}.

¹Solid State Chemistry Laboratory, Materials Science Centre, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands. ²Department of Earth Sciences, University of Cambridge,

Downing Street, Cambridge CB2 3EQ, United Kingdom.

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Multiferroics are materials exhibiting ferroelectricity and magnetism simultaneously. From the applications point of view they are also very interesting since they present the possibility to manipulate their electric and magnetic moments by magnetic and electric fields, respectively, if the two order parameters are coupled. While efforts have been devoted to search and design new multiferroics, the nature of the coupling between ferroelectricity and magnetism is still under investigation and it greatly differs from system to system[1]. Using epitaxial growth, the properties of multiferroic thin films can be engineered by tuning the strain imposed by the substrate. Among the known multiferroics, the perovskite oxides, such as BiFeO₃ or TbMnO₃, are very interesting candidates for epitaxial growth because of their small lattice mismatch with, for instance, SrTiO₃ substrates. Using the Pulsed Laser Deposition technique (PLD), epitaxially grown BiFeO₃ and TbMnO₃ thin films on SrTiO₃ substrates have been reported with thicknesses of tens of nanometers [2]. Recently, ultrathin films of BiFeO₃ have been also reported down to 5nm [3], but the characterization of such thin layers by means of standard X-ray diffraction is not easy because of the limited diffracted intensity. As far as we know, there are no reports of neither fully-strained TbMnO₃ films nor of TbMnO₃ films below 100 nm. Growth and characterization of ultrathin films is not only of fundamental importance, but it is also significant in the context of multilayer growth, in order to gain control of the behavior and thickness limitations of the individual layers, which are usually very thin. Here we report the successful growth of epitaxial ultrathin (5 nm) BiFeO₃ and TbMnO₃ films on SrTiO₃ single crystals via RHEED-assisted PLD and their structural characterization using synchrotron diffraction in grazing-incidence geometry, along with highresolution laboratory X-ray diffraction. Atomic Force Microscopy was used to investigate the surface of the films. Thickness dependence and the strain relaxation of the grown films is also reported. We show that for thicknesses below ~50nm, BiFeO₃ thin films are tetragonal and that above that thickness they start to relax via a monoclinic distortion, still different from the fully relaxed (bulk) rhombohedral phase.

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