The Quest for new Room Temperature Multiferroic Thin Films

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Multiferroic materials that exhibit both ferroelectric and magnetic properties continue to generate great interest for a variety of applications. However, obtaining materials with ferroelectric and magnetics properties that are sufficiently strong and robust at or above room temperature for potential integration into novel devices remain elusive. In addition, novel integrated devices stipulate that these materials be produced in the form of thin films of high quality. Fortunately thin films, in particular when synthesized via deposition processes far from equilibrium, do provide for a number of parameters controlling the synthesis, such as, strain engineering for epitaxial films, which plays an important role and provides a convenient parameter to tune the properties and enable the existence of otherwise metastable phases. As a result, several strategies have been pursued in the quest of thin films of novel multiferroic materials with good multiferroic properties at room temperature.

Replacing every second atom of Fe with an atom of Cr in $BiFeO_3$ (BFO) - until recently the only single phase material known to be multiferroic at room temperature - changes the weak ferromagnetism of BFO stemming from slightly canted spins antiferromagnetically coupled, into ferrimagnetic Bi_2FeCrO_6 (BFCO) with order of magnitude larger magnetic properties without affecting much its ferroelectric properties. The recent developments in the growth and characterization of BFCO epitaxial thin films[1,2] as well as our current understanding of their properties will be briefly presented[3].

Composite materials with one component ferroelectric and a second component magnetic above room temperature is another route to engineer a material that exhibit multiferroic properties at room temperature. The coupling is ten mostly elastically mediated at the boundaries between the two phases. An example of such a system is the maghemite(γ-Fe_2O_3)/BFO composite epitaxial films spontaneously forming when synthesized in the right conditions. Another similar system spontaneously forming a multiferroic composite in the bulk is the tetragonal tungsten bronze $Ba_2LnFeNb_4O_{15}$ (TTB-Ln) / $BaFe_{12}O_{19}$ / $LnNbO_4$, with Ln = Nd, Sm and Eu[4].

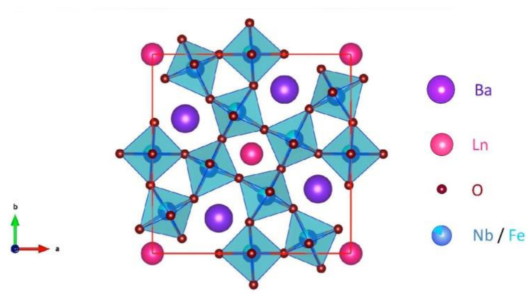

Figure 1 Crystal structure of $Ba_2Ln\ Fe_4O_{15}$

Using TTB-Ln bulk ceramic as targets, epitaxial thin films of $Ba_2LnFeNb_4O_{15}$ (Ln = Eu, Sm, and Nd) have been successfully obtained on $SrTiO_3$(100), MgO(100) and $Gd_3Ga_5O_{12}$(100), and the optimum conditions of deposition established. A structural analysis shows that the lattice parameters are slightly smaller than those observed in the bulk[1], indicative of a compressive stress buildup in the films during the deposition. Ferroelectric macroscopic P-E hysteresis loops as well as microscopic electromechanical behavior probed by piezoelectric force microscopy have demonstrated the existence of a spontaneous polarization at room temperature in TTB-Ln thin films.

A third system that is magnetoelectric and potentially multiferroic at room temperature is ε-Fe_2O_3 (epsilon ferrite), a metastable phase of iron (III) oxide with orthorhombic structure, intermediate between maghemite (γ-Fe_2O_3) and hematite (α-Fe_2O_3). Epsilon ferrite is ferrimagnetic (T_C = 510 K). It exhibits a polar distortion (non-centrosymmetric space group $Pna2_1$,) and a large magnetic anisotropy at room temperature, as well as a magnetoelectric coupling[5-7]. Moreover, the material is characterized by a ferromagnetic resonance (FMR) frequency in the THz range in the absence of magnetic field and at room temperature, which is of interest for short-range wireless communications (e.g. 60GHz WiFi) and ultrafast computer non-volatile memories[8-10]. It has been recently shown that thin films of metastable ε-Fe_2O_3 can be stabilized by epitaxial strains.[11]

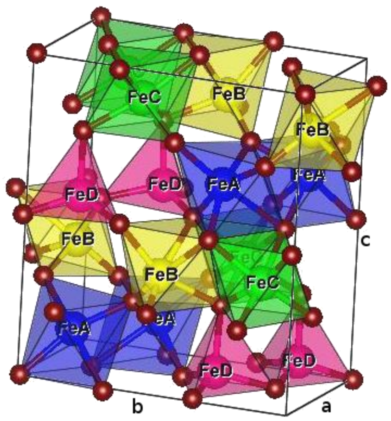

Figure 2 Crystal structure of ε-Fe_2O_3

Epitaxial thin films of ε-Fe_2O_3 and ε-$Al_xFe_{2-x}O_3$ grown by Pulsed Laser Deposition on different single crystalline substrates have been investigated, in order to understand the role of the Al-doping and of the compressive epitaxial strain in the epsilon phase stabilization as well as on the ferroelectric and magnetic properties. While detailed X-ray diffraction studies evidence multiple growth orientations, piezoelectric and ferroelectric properties were analyzed both at the macroscopic level and locally at the nanoscale by Piezoresponse Force Microscopy (PFM) and magnetic properties were characterized using a Vibrating Sample Magnetometer (VSM) and Magnetic force Microscopy (MFM), verifying that the synthesized epitaxial thin films indeed exhibit a high magnetic anisotropy.

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