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Multiferroelectricity of corner-shared preovskite networks of Manganese and Oxygen

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Multiferroics that exhibit simultaneous ferroelectric and magnetic orders are a topic of current intense investigation both to understand how these two disparate order parameters interact and because of the promising possibility of controlling the magnetic properties electronically and vice versa [1,2]. Type-II or 'improper' multiferroics exhibit strong magnetic-ferroelectric coupling, however, the ferroelectric order parameter is more than two orders-of-magnitude smaller than robust nonmagnetic ferroelectrics such as the prototypical Ba2+Ti4+O3 for which the hybridization of the occupied oxygen p orbitals to the empty Ti d orbitals precludes the possibility of magnetic order. Type-I or 'proper' multiferroics with robust displacive-type ferroelectric order are not only rare but also they typically exhibit disparate ordering temperatures and very weak coupling between the order parameters. Recently new promising multiferroics have been discovered with strong coupling. Our work on lightly substituted magnetic Sr1-xBaxMnO3 materials synthesized with conventional fabrication techniques up to a maximum x of ~ 0.2 did not reveal ferroelectricity expected for tensile strain elongated Mn-O bonds [3]; however, expanding the Ba concentrations to higher values ($x \geq 0.45$) succeeded in achieving robust ferroelectricity [4]. These ceramics exhibit unique ferroelectricity (TF > 300 K) and G-type antiferromagnetism (TN ~ 200 K) originating exclusively from the Mn4+ (d3) cations.

By advancing elaborate synthesis processes, which are necessary to avoid the more stable hexagonal polymorphs, we were able to prepare and study structural, magnetic and ferroelectric properties [5,6] of highly strained multiferroics for x = 0.4-0.45. The classical displacive-type ferroelectric phase occurs with a polarization of several μC/cm2 when the Mn ions move out of the center of the MnO6 octahedral units. The Mn spins order below TN into a simple G-type magnetic structure while the ferroelectric order decreases dramatically demonstrating that the two order parameters are strongly coupled. A spin gap of 4.6(5) meV and the magnon density of states peaking at 43 meV characterize the ground state spin dynamics. The ferroelectric phase transition has a signature of a crossover from displacive to order-disorder type. The phonons are coupled with a central mode but contribution to is rather small. The lowest-frequency polar phonons are overdamped above TN and they exhibit pronounced softening on heating towards TC.

We have recently extended investigation of manganites to the Ti-substituted Sr1-xBaxMn1-yTiyO3 system for which ferroelectricity above 400 K and structural distortions characterizing polarization significantly exceeding that of the classical titanates were observed. The TN decreases to below 200 K and the suppression of ferroelectricity below TN is reduced, i.e., we achieved displacive-type multiferroic with large spontaneous polarization. I will describe unique properties of these materials.

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