***Thesis Title*:**

"Establishing structure-property correlation in multiferroic $Bi_{1-x}Ca_xFeO_3$ nanoceramics and solid solution with ferroelectric $PbTiO_3$ and $BaTiO_3$"

ABSTRACT

Most dielectric materials are generally insulating in nature, consist of randomly oriented dipoles and do not possess any spontaneous polarization. However, there exists interesting class of dielectric materials which have intrinsic "*spontaneous polarization*". These are termed as "***ferroelectric***". Similarly, there exists a class of materials that show spontaneous magnetization; the direction of which can be changed by application of suitable external magnetic field. Such materials are called as "***ferromagnetic***". These kinds of materials have been investigated for over a century. The next major interest in such materials was generated by the idea that systems may exist in which two types of ordering – (ferro)electric and (ferro)magnetism can coexist. These materials; where the coexistence of the ferromagnetism and ferroelectric is observed, even in the absence of external magnetic and electric fields, are now broadly classified as ***"multiferroics"*** – following the terminology given by Schmid. In principle, the term "multiferroics" can also be used to describe systems that simultaneously show the presence of at least two out of the possible four ferroic properties viz., ferroelectricity, ferromagnetism, ferroelasticity and ferrotoroidal. The use of multiferroic materials range from: spintronics, sensors, memories, capacitors, to household appliances.

$BiFeO_3$ (BFO) is perhaps the only material that is both magnetic and ferroelectric at room temperature. But the low coupling between the two order parameters limits its large scale use. Amongst the various strategies being tried to tailor increase the magneto-electric coupling, use of A-site or B-site doping has received maximum attention. The thesis deals with one such divalent ion i.e., Ca^{2+} doped $BiFeO_3$ ceramics and their solid solution with few well known ferroelectric ceramics viz., $PbTiO_3$ and $BaTiO_3$. To achieve better understanding of the materials characteristics, I have used and analyzed data from large number of experimental techniques such as: XRD, Raman, TGA/DSC, dielectric and impedance spectroscopy, SQUID-VSM and P-E hysteresis loop, magneto-dielectric, Rietveld refinement, etc. Based on these analysis, some of the major findings presented in the thesis are: **(i)** occurrence of hitherto unreported low temperature phase transitions in $Bi_{1-x}Ca_xFeO_3$ ceramics up to a certain composition, **(ii)** stabilization of dielectric anomaly near the magnetic phase transition; the origin of which lies in the "skin effect", **(iii)** transformation from antiferromagnetic to weak ferromagnetic type even at very low dopant concentration, **(iv)** significant modulation of physical properties as a function of oxygen annealing, **(v)** establishment of phase diagram for (1-y)$Bi_{0.90}Ca_{0.10}FeO_3$- (y)$PbTiO_3$ solid solution, **(vi)** Bi-relaxor type characteristics in (1-y)$Bi_{0.90}Ca_{0.10}FeO_3$ – (y)$ATiO_3$ (A=Pb, or Ba) solid solutions.

Keywords: Multiferroic, nanoparticles, spins glass, dipolar glass and dielectric relaxation.

Parts of the thesis work have been published as Journal articles (details are given in the list of publications).