

8.1 Summary of the Work

1. All the compositions of BF-xPT ($0.20 \leq x \leq 0.50$) solid solution were successfully synthesized by sol-gel method. The average particle sizes of the powders synthesized by this method are in the range of 18nm to 2.1 μ m for all compositions. The average grain size was found to effectively growing with increasing BiFeO₃ content.
2. The controversies regarding the crystal structures of BF-xPT, for both bulk and nano particles was resolved using Rietveld analysis of high resolution synchrotron x-ray diffraction data by taking the compositions =0.5 and x=0.25, on the both side of MPB.
3. The antiferromagnetic transition temperature T_N in the tetragonal phase (x=0.50) has been found to increase drastically by over 200K on decreasing the particle. This observation is quite counter-intuitive as size reduction in both ferroelectric and magnetic materials invariably leads to a decrease in the transition temperature as per a well established scaling law of Binder. We have shown using magnetization and high resolution x-ray and neutron powder diffraction measurements that this anomalous increase occurs due to the cross coupling of the two phenomena.
4. In contrast to the magnetic transition, we have shown that ferroelectric transition temperature T_C in the same tetragonal phase has been found in this work to follow the conventional behavior. The results presented here show a significant decrease T_C with decreasing particle size. This is because the coupling effects, if any, are much weaker above the AFM transition temperature. Thus because $T_N < T_C$ in BF-0.50PT, the ferroelectric distortion affects the AFM transition due to

multiferroic coupling but the ferroelectric T_C is nearly unaffected by the paramagnetic spins.

5. We find that particle size affects the T_N of monoclinic compositions also, but in a very modest fashion. It is argued that the difference in the behavior of monoclinic and tetragonal compositions arises because of the dimensionality of the AFM phase which is nearly two dimensional in the tetragonal phase due to extremely large ferroelectric distortion along [001], whereas it is three dimensional in the monoclinic phase.
6. The last, but not the least, important finding of this work relates to the discovery of the Griffith's phase and location of the Griffith's phase transition temperature T_G in nanocrystalline monoclinic BF-xPT samples. The bulk behavior is also Griffith's like above T_N with slightly lower T_G .
7. With the help of high temperature synchrotron x-ray diffraction data for nano and bulk powders a tetragonal to tetragonal phase isostructural phase transition was observed. The ferroelectric transition temperature (T_C) decreases with size in BF-0.5PT.

8.2. Suggestions for future work

There are few open questions that remain to be investigated in BF-xPT system, as summarized below.

1. The sinterable monoclinic composition of BF-xPT should be used to prepare high density ceramic specimen with insulating characteristics using suitable dopant, such as Mn_2O_3 , so that magneto electric coupling effect can be investigated.

2. Single crystals of tetragonal phase should be grown to understand the two dimensional antiferromagnetic ordering below T_N .
3. Nano crystalline powders of BF-xPT should be investigated using Atomic pair distribution function analysis to understand the nature of disorderness, near the surface.

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