Evidence for spin glass transitions in multiferroic (1-x)BiFeO$_3$-xBaTiO$_3$ solid solutions

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Magnetolectric (ME) multiferroics offer the possibility of controlling the electric (magnetic) polarization by applying a magnetic (electric) field and have attracted significant interest in view of the interesting physics of coupling between magnetic and ferroelectric order parameters and potential technological applications in several multifunctional devices [1-3]. BiFeO$_3$ is unique amongst various magnetoelectric multiferroics, as its ferroelectric and magnetic transition temperatures (Tc~1103 K, TN~ 643 K) are well above the room temperature [1-3]. (1-x) BiFeO$_3$-xBaTiO$_3$ (BF-xBT) solid solutions have received considerable attention in recent years due to large ferroelectric polarization, large remnant magnetization, linear magnetoelectric coupling and highest depolarization temperature (Td) for piezoelectric applications and considered to be alternative to toxic lead (Pb) containing piezoelectric ceramics like Pb(ZrxTi1-x)O$_3$ (PZT) and (1-x)Pb(Mg1/3Nb2/3)O3-xPbTiO3 (PMN-PT). However, the ground state of neither BiFeO$_3$ nor BF-xBT is still not clear. We present here results of Rietveld analysis of x-ray and neutron powder diffraction data in conjunction with results of magnetization, AC susceptibility and specific heat measurements. Our results reveal presence of two spin glass phases below room temperature both of which coexist with the long range ordered (LRO) antiferromagnetic (AFM) phase formed well above room temperature at the Neel transition temperature. It is shown that both the spin glass transitions are accompanied with decrease in the ordered magnetic moment and strong magnetoelastic coupling suggesting that the LRO-AFM and spin glass transitions occur on the same magnetic sublattice in agreement with theoretical predictions for Heisenberg systems. We also present a magnetic phase diagram of the BF-xBT system.


Keywords: Magnetolectric Multiferroic, BiFeO$_3$, Solid Solutions