A.6: Correlation of the physical properties in bulk $BaFeO_{3-\delta}$ with structural properties studied at the ADXRD beamline, Indus-2

Recently there has been a quest for materials having high dielectric permittivity, due to their technological importance. The materials exhibiting giant dielectric permittivity (GDP) are found to exhibit high dissipation factor (tan) which limits their practical applications. Doped $BaTiO_3$, $CaCu3Ti_4O_{12}$ (CCTO) with GDP of ~5000 and ~10^5 respectively are the potential materials reported in the literature. Apart from GDP, there are reports on the simultaneous existence of GDP plus magneto-dielectric effect (MD) in charge ordered manganites, ferrites etc. A combination of these two effects in the same material can be of technological importance.

$BaFeO_3$ is one such interesting perovskite, with Fe in 4^+ state that exhibits charge disproportionation and antiferro-magnetism at low temperatures. Hexagonal $BaFeO_{3-\delta}$ which is the most stable phase is reported to have large electrical resistivity (T<200K) comparable to that of dielectric materials. This is in contrast with other Fe^{4+} oxides commonly exhibiting a tendency towards metallicity as well as ferromagnetism. However, dielectric studies on bulk $BaFeO_3$ and their possible correlation with the magnetic and magneto-transport properties are rare. Keeping this in mind the present study was undertaken with an intension to study the dielectric and magnetic properties of $BaFeO_{3-\delta}$ and their possible correlations.

Polycrystalline sample of $BaFeO_{3-\delta}$ was prepared by standard solid-state reaction route. The stoichiometric mixture of high purity $BaCO_3$ and Fe_2O_3 were calcined at various temperatures between 900-1100°C with intermediate grinding, followed by sintering at 1250°C for 12 hours. Sintered pellets were kept in oxygen flow at 900°C for 24 hours. The sample was characterized using x-ray diffraction (XRD) at 13 keV and X-ray absorption near edge structure (XANES) measurements at beamline (BL-12) on Indus-2 Synchrotron source. XRD measurements confirm the hexagonal phase of the prepared samples and XANES measurements indicates that Fe present in the sample is in mixed valence Fe^{3+} and Fe^{4+} state.

Temperature dependent (150-300K) dielectric measurements performed in the frequency range of 100 Hz to 1 MHz, shows a large dielectric permittivity (DP) of more than $2x10^7$ at 300K and 10 kHz, with relatively large dissipation factor of ~20. This value of DP observed for the studied sample is large as compared to CCTO, which is believed to have the highest DP of the order of 10^5, in the same temperature and frequency range. Impedance spectroscopy analysis carried out in order to understand the contributions due to grain and grain boundaries to the DP, shows an anomaly in grain resistance at ~168K. Analysis of magnetization data as a function of temperature gives a negative value of T_c, indicating a presence of antiferromagnetic interaction in the sample. However, bifurcation in the field-cooled and zero field-cooled data indicates magnetic in-homogeneity in the sample. Magneto-resistance (MR) measurements as a function of temperature at 8 Tesla, shows a peak at ~180K with a positive value of 10%.

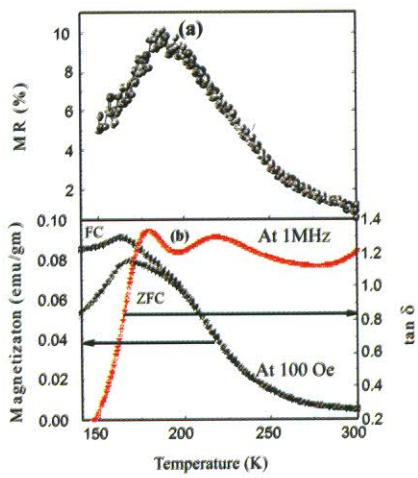

Fig.A.6.1: Simultaneous plot of (a) magneto-resistance (Top) and (b) magnetization data (left) and the dissipation factor (right) clearly shows the transition temperature in magnetization data and peak in magneto-resistance coincides with the temperature at which a sudden drop in the dissipation factor transpires.

A correlation of electrical and magnetic properties of the sample is shown in Fig. A.6.1. It is clear that the onset of magnetic transition, decrease in dissipation factor and peak in MR are close in temperature. It should also be noted that the anomaly in grain resistance also appears around the same temperature. Since all the observed changes in magnetic and electrical properties are close in temperature, the origin of these changes appears to be the same. Hence the possibility of magnetic and dielectric correlations in the studied sample cannot be ignored.

Thus, the structural, magnetic and dielectric properties of $BaFeO_{3-\delta}$ have been studied. The sample exhibits a large dielectric permittivity comparable to that of doped $BaTiO_3$ and CCTO. The origin of high dielectric permittivity appears to be governed by magnetic state of the sample. Obtained results indicate that the charge/orbital and spin degrees of freedom are possibly coupled and may lead to possibility of magneto-dielectric coupling in such materials.

(For details, refer to: Archna Sagdeo et al., Appl. Phys. Lett. 105(4), 042906 (2014))

Reported by:
Archna Sagdeo (archnaj@rrcat.gov.in) and Tapas Ganguli