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Citation: AIP Conference Proceedings **1591**, 1595 (2014); doi: 10.1063/1.4873046
View online: http://dx.doi.org/10.1063/1.4873046
View Table of Contents: http://scitation.aip.org/content/aip/proceeding/aipcp/1591?ver=pdfcov
Published by the AIP Publishing

Magnetic Origin Of Dielectric Transition In $BiFeO_3$

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Abstract. Magnetic relaxation measurements at 5K, 50K and 120K on $BiFeO_3$ prepared by sol-gel auto combustion method shows stretched -exponential decay. These results shows the two factors viz, cooperative dynamics and rate of dynamics of spin, may be responsible for the low temperature magnetic-glassy behavior, concluded from bifurcation of zero field cooled (ZFC) and field cooled (FC) data of dc magnetization. Temperature dependent dielectric measurement shows a possible phase transition, seen in the dielectric-relaxation time and dielectric constant in the range 200 – 240K. Comparison of dielectric and magnetization data indicates a possibility of magneto-electric coupling.

Keywords: Sol-gel method, Magnetic relaxation, Cole-Cole plot.
PACS: 75.10.Nr, 76.60.Es

INTRODUCTION

Bismuth Ferrite (BFO) is considered as a unique prototype among all the multiferroics even though it exhibits a weak magnetoelectric coupling. Recently, a lot of attention has been focused on the low temperature studies due to various noticeable phase transitions observed in this compound[1]. Particular interest of many researchers is to understand the origin of electrical anomaly and the spin freezing behavior seen at low temperatures[2]. In this context, this study reports the results of low temperature dielectric properties and magnetic relaxation behavior in the multiferroic compound of $BiFeO_3$.

EXPERIMENTAL DETAILS

The pristine $BiFeO_3$ sample is prepared by a sol-gel auto-combustion method[3] and characterized by x-ray and neutron diffraction[4]. The magnetization measurements are carried out using Quantum Design make 9T PPMS-VSM in the temperature range of 2K to 350K. The temperature dependent impedance measurements are performed using a lock-in amplifier (SR 830) and closed cycle refrigerator (Janis) equipped with temperature controller (Lakeshore-331).

RESULTS AND DISCUSSIONS

The magnetic relaxation measurements are carried out at 5K, 50K and 125K, (temperature range over which zero-field cooled (ZFC) and field-cooled (FC) curves bifurcate) using the following protocol. The sample is cooled from 350K to the target temperature in ZFC mode and allowed enough time for temperature to stabilize. Then a 6 Tesla magnetic field is applied at the rate of 100 Oe/sec and the field is held constant for one hour. Subsequently the field is reduced to zero (at 100 Oe/sec) and the magnetization is measured continuously as function of time for 14400 secs (4 hours). Fig.1 shows the normalized M(t)/M(t=0) versus time at T=125K. It can be seen the magnetization is continuously decreasing with time and it has not stabilized even after 4 hours. The exponentially decay of magnetization with time is

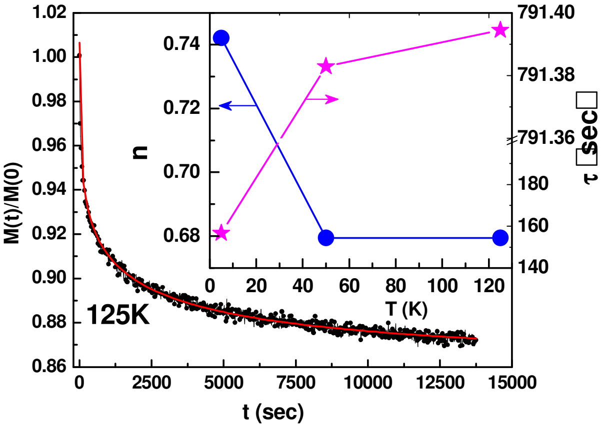

FIGURE 1. Normalized ZFC of BFO measured as a function of time at T = 125K. Inset shows the variation of relaxation component (n) and relaxation time (τ) with temperature.

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fitted to a stretched exponential Eq.1.

$$M(t) = M_0 + M_r \exp\left[-\left(\frac{t}{\tau}\right)^{1-n}\right] \qquad (1)$$

Here, M_0 and M_r represent the intrinsic magnetic moment and glassy component mainly contributing to relaxation effect, respectively. Similar fitting of the magnetic relaxation data taken at other temperatures are attempted. Both the parameters (M_0 and n) are found to decrease with decrease in temperature. The values of M_0 are M_0 (emu/gm) = 0.005 (120K), 0.0016 (50K) and 0.00089 (5K). The decrease in M_0 seems to indicate an increasing glassiness in the compound as T is lowered. Inset of Fig.1 shows the variation of relaxation component (n) and relaxation time (τ) as function of temperature. At 5K, the value of n is 0.75 indicating fractal-exponential decrease and hence showing slight glassiness of the sample. With increase in temperature this value decreases to 0.68, indicating improvement in the exponential decrease. The plot of τ is indicating relatively faster dynamics at low temperature. Hence, from these data, it is very clear that two process are responsible for the glassy behavior of BFO: a non-cooperative spin dynamics as seen from the value of 'n' and rate of spin dynamics (may be due to spin frustration), evident from the value of 'τ'.

Fig. 2 shows the Cole Cole (ε″ vs. ε′) plot obtained from impedance data in the temperature range of 300K to 20K. At room temperature, the data corresponds to a single semi-circle. As the temperature is lowered, the shape of the semicircle becomes asymmetric and only the high frequency region could be fitted to a semi-circle. With further decrease of temperature, the asymmetry increases with simultaneous decrease in the radius as well as center of the semicircle. For temperature below 200K, a second semicircle at low frequency appears. For T < 160K, the radius and center of first semicircle saturates with no further temperature induced variation. The semicircles are fitted to the following equation.

$$\left(\varepsilon' - \frac{\varepsilon_0 - \varepsilon_\infty}{2}\right)^2 + \left[\varepsilon'' + \frac{\varepsilon_0 - \varepsilon_\infty}{2}\tan\frac{\pi\alpha}{2}\right] = \frac{(\varepsilon_0 - \varepsilon_\infty)^2}{4\cos^2(\pi\alpha/2)} \qquad (2)$$

$$\varepsilon'' = \frac{\varepsilon_0 - \varepsilon_\infty}{c_s}\left(\frac{\omega\tau}{1+\omega^2\tau^2}\right) \qquad (3)$$

From the fitting of Eq (2) to the dielectric data, one obtains $\varepsilon_\infty = 0$ and α = 0. The dielectric constant ε_0 (calculated using Eq. (2)) and relaxation time τ (calculated using Eq. (3)), as a function of temperature are shown in fig 3.

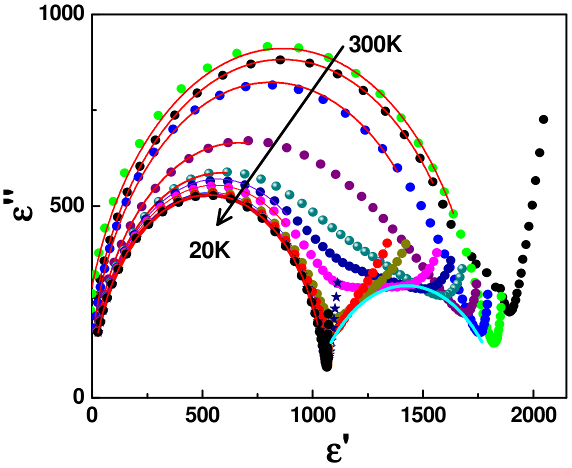

FIGURE 2. Cole - cole plot of ε″ against ε′ measured in the temperature range of 300K to 20K. Solid lines correspond to the fitting of Eq. (2) to the experimental data points.

From the thermal response of dielectric and relaxation time, the plot may be categorized under three regions as I (T > 240K), II (160 < T < 240K) and III (T < 160K). Initially, as the sample is cooled from 300K, ε_0 decreases slowly whereas τ increases and attains maxima at T ~ 240K. On further lowering T, both ε_0 and τ decreases very steeply till 200K and then slowly till 160K. Below 160K both ε_0 and τ becomes constant of temperature. The τ reflects the relaxation processes happening in the sample. This plot indicates two kinds of relaxation process (1) at high temperature (region I) and (2) at low temperature (region III), separated by a transition region II. Inset of fig. 3 shows the dc magnetization at 0.1 T with ZFC and FC plots. Interestingly, depending upon the response of ZFC and FC, the magnetization plot can also be divided into three regions of nearly same temperature range. At

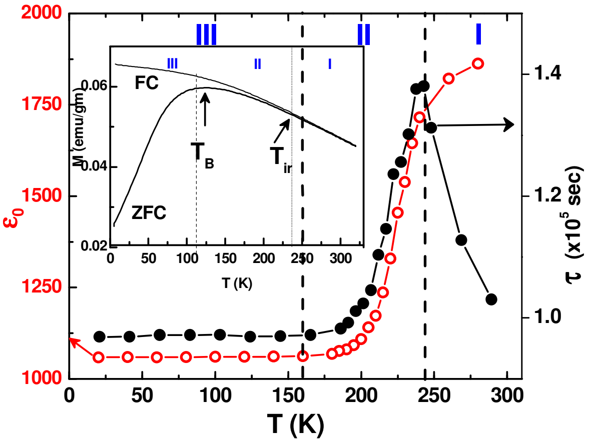

FIGURE 3. Variation of dielectric constant (open symbol) and relaxation time (solid symbol) with temperature. The lines are only guide to eye. The inset shows the temperature dependence of magnetization, ZFC and FC at 0.1T

300K, the ZFC and FC data overlaps nicely and at low temperature the two bifurcates, which starts T ~ 240K. Further, the bifurcation is found to be growing gradually towards low temperature. Bifurcation in ZFC and FC data is considered as a measure of glassy nature in the sample. Hence the transition seen in the ε_0 and τ plot may have their origin in the glassiness of the sample. There are also some reports on the possibility of magnetic glassy and coupled with weak polarization in the similar temperature range[1]. Hence, a magneto-dielectric measurement may be needed for better insight.

ACKNOWLEDGMENTS

UGC-DAE CSR, BRNS-DAE and DST, India are acknowledged for financial support in the form of the project and fellowship of authors JR and AKB. JR and AKB are also thankful to CSIR India for SRF fellowship.

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