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# Relaxor behaviour and dielectric properties of BiFeO<sub>3</sub> doped Ba(Zr<sub>0.1</sub>Ti<sub>0.9</sub>)O<sub>3</sub> ceramics

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**Abstract.** Ba<sub>1-x</sub>Bi<sub>x</sub>(Ti<sub>0.9</sub>Zr<sub>0.1</sub>)<sub>1-x</sub>Fe<sub>x</sub>O<sub>3</sub> ( $x = 0-0.075$ ) ceramics are prepared using a conventional solid state reaction method. X-ray diffraction shows the presence of a single phase. Addition of Bi<sup>3+</sup> and Fe<sup>3+</sup> strongly influences the crystal structure and dielectric properties of the ceramics. The evolution from a normal ferroelectric to a relaxor ferroelectric is emphasized. Ba<sub>0.99</sub>Bi<sub>0.01</sub>(Ti<sub>0.9</sub>Zr<sub>0.1</sub>)<sub>0.99</sub>Fe<sub>0.01</sub>O<sub>3</sub> ceramic shows a relaxor behaviour at room temperature with  $\Delta T_m = 12$  K.  $P-E$  hysteresis loop of the composition,  $x = 0.007$ , shows a remanent polarization ( $P_r$ ) of  $0.5 \mu\text{C}/\text{cm}^2$  with a coercive field ( $E_C$ ) of 2 kV/cm. Raman spectra of all compounds are performed and correlated well with the X-ray diffraction and dielectric measurement results.

**Keywords.** Ceramics; relaxors; ferroelectric; piezoelectric; Raman spectroscopy.

## 1. Introduction

There has been increasing interest in relaxor ferroelectrics in the past few years owing to their interesting physical properties such as high dielectric constant and giant electrostriction (Cross 1994). Relaxor behaviour is well known in lead-based compositions such as PMN-PT, PNN-PZT, PLZT, etc (Chang *et al* 2007; Wu *et al* 2009). Nevertheless, these present a disadvantage due to toxicity and volatility of polluting substances. Therefore, much effort has been made towards investigating environment-friendly ‘lead-free’ relaxor materials. Many lead-free materials with perovskite structure such as BaTiO<sub>3</sub>, (Bi<sub>1/2</sub>Na<sub>1/2</sub>)TiO<sub>3</sub>, (Bi<sub>1/2</sub>K<sub>1/2</sub>)TiO<sub>3</sub> and KNbO<sub>3</sub> have been investigated (Ravez and Simon 2001; Takennaka *et al* 2007) in terms of their dielectric relaxation, ferroelectric phase transition and electrical properties. Out of these, BaTiO<sub>3</sub>, characterized by a ferroelectric paraelectric transition at  $T_C \sim 400$  K and a maximum of permittivity ( $\epsilon'_{r\text{max}}$ ) of about 8000, have been widely investigated (Ravez and Simon 1998; Bahri *et al* 2001; Abdelmoula *et al* 2006; Abdelkafi *et al* 2007). Ceramics with composition, Ba(Ti<sub>1-x</sub>Zr<sub>x</sub>)O<sub>3</sub>, have been found to exhibit relaxor type for  $0.26 \leq x \leq 0.40$  (Ravez and Simon 1998). In particular, the classical ferroelectric Ba(Ti<sub>0.9</sub>Zr<sub>0.1</sub>)O<sub>3</sub> exhibits a maxima of permittivity ( $\epsilon'_{r\text{max}}$ ) of about 12000 at  $T_C \sim 360$  K (Moura *et al* 2008) which is better than BaTiO<sub>3</sub>.

On the other hand, several authors have recently shown that a small amount of BiFeO<sub>3</sub> can improve the piezoelectric properties of Bi<sub>0.5</sub>Na<sub>0.5</sub>TiO<sub>3</sub>-Bi<sub>0.5</sub>K<sub>0.5</sub>TiO<sub>3</sub> (Zhou *et al* 2009), (Na<sub>0.5</sub>K<sub>0.5</sub>)NbO<sub>3</sub> (Zuo *et al*

2008), Na<sub>0.5</sub>K<sub>0.5</sub>NbO<sub>3</sub>-LiSbO<sub>3</sub> (Jiang *et al* 2009) and K<sub>0.44</sub>Na<sub>0.52</sub>Li<sub>0.04</sub>Nb<sub>0.84</sub>Ta<sub>0.1</sub>Sb<sub>0.06</sub>O<sub>3</sub> (Jiang *et al* 2010) ceramics and gives rise to relaxor behaviour in BaTiO<sub>3</sub> at low temperature (Kumar *et al* 2000).

Considering the above results, in order to obtain a relaxor material at room temperature, we have chosen to dope the classical ferroelectric Ba(Ti<sub>0.9</sub>Zr<sub>0.1</sub>)O<sub>3</sub> compound with a small amount of BiFeO<sub>3</sub> (Bi<sup>3+</sup> at A site and with Fe<sup>3+</sup> at B site simultaneously). The choice of Bi<sup>3+</sup> (substituted for Ba<sup>2+</sup>) cation is due to its  $6sp^2$  lone pair (like Pb) and such an electronic environment is favourable for the relaxor effect without the disadvantage of lead pollution. Moreover, addition of Fe<sup>3+</sup> at the B site creates a local heterogeneity with valence fluctuations. Here, we report detailed dielectric studies and structure description of  $(1-x)\text{Ba}(\text{Ti}_{0.9}\text{Zr}_{0.1})\text{O}_3-x\text{BiFeO}_3$  ( $0 \leq x \leq 0.1$ ) samples and the data have been quantitatively analysed in terms of parameters characterizing the relaxor behaviour. To investigate the phonon spectra related to the phase transition of  $(1-x)\text{Ba}(\text{Ti}_{0.9}\text{Zr}_{0.1})\text{O}_3-x\text{BiFeO}_3$  system, Raman scattering has been used in this work.

## 2. Experimental

Ba<sub>1-x</sub>Bi<sub>x</sub>(Ti<sub>0.9</sub>Zr<sub>0.1</sub>)<sub>1-x</sub>Fe<sub>x</sub>O<sub>3</sub> ( $x = 0-0.075$ ) ceramics were synthesized by a solid state reaction method using BaCO<sub>3</sub>, TiO<sub>2</sub>, ZrO<sub>2</sub>, Bi<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> powders. All these materials were dried at 393 K for 2 h, weighed, mixed for 1 h and calcined at 1283–1523 K for 10–12 h. After calcination, powders were mixed for 1 h and pressed under 100 MPa into disks of 8 mm in diameter and about 1.5 mm in thickness. Finally, the pellets were sintered in oxygen atmosphere

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between 1573 and 1693 K for 2–4 h followed by furnace cooling.

At room temperature, X-ray diffraction was used to characterize the crystal structure of  $\text{Ba}_{1-x}\text{Bi}_x(\text{Ti}_{0.9}\text{Zr}_{0.1})_{1-x}\text{Fe}_x\text{O}_3$  ceramics on Philips diffractometer using  $\text{Cu-K}\alpha$  radiation ( $\lambda = 1.5418 \text{ \AA}$ ). Dielectric measurements were performed on ceramic discs after deposition of gold electrodes on the circular faces by d.c. sputtering. The real and imaginary parts of the permittivity,  $\epsilon'_r$  and  $\epsilon''_r$ , were measured under helium as a function of both temperature (85–600 K) and frequency ( $100-2 \times 10^5 \text{ Hz}$ ) using a Wayne-Kerr 6425 component analyser. Ferroelectric hysteresis loop parameters such as remanent polarization ( $P_r$ ) and coercive field ( $E_c$ ) were estimated from the hysteresis loop observed on the screen of a 515A JEKT Ronicks oscilloscope with the aid of a home built ‘Sawyer-Tower’ circuit.

Raman spectra of sintered samples are recorded from 50 to  $800 \text{ cm}^{-1}$  in a micro-Raman Spectrometer (LABRAM HR-800), working in a backscattering configuration, equipped with an  $\text{He}^+$  ion ( $\lambda = 633 \text{ nm}$ ) laser. The spectral resolution of the system is  $3 \text{ cm}^{-1}$ .

### 3. Results and discussion

#### 3.1 Crystal structure characterization

X-ray diffraction (XRD) is used to confirm purity of phases and to determine the lattice parameters of  $\text{Ba}_{1-x}\text{Bi}_x(\text{Ti}_{0.9}\text{Zr}_{0.1})_{1-x}\text{Fe}_x\text{O}_3$  samples with  $x = 0, 0.005, 0.007, 0.01, 0.025, 0.05$  and  $0.075$ . A matching profile of XRD spectra is made using the ‘FullProf’ software (Rodriguez-Carvajal 2008).

$\text{Ba}_{1-x}\text{Bi}_x(\text{Ti}_{0.9}\text{Zr}_{0.1})_{1-x}\text{Fe}_x\text{O}_3$  crystallizes in tetragonal ( $P4mm$ ) for compositions in the range  $0 \leq x \leq 0.01$  and pseudocubic for compositions in the range  $0.01 < x \leq 0.075$ . Figure 1 shows X-ray diffraction patterns for  $x = 0.005$  and  $0.025$  as examples.

Figure 2 shows variations of the unit cell volume vs composition,  $x$ . The unit cell volume decreases with increasing  $x$ . The average of the ionic radius in the A site ( $\langle r_A \rangle$ ) ( $\langle r_A \rangle = (1-x)r(\text{Ba}^{2+}) + xr(\text{Bi}^{3+})$ ) with  $r(\text{Bi}^{3+}) = 1.38 \text{ \AA}$ ,  $r(\text{Ba}^{2+}) = 1.43 \text{ \AA}$  in twelve-coordination (Shannon and Prewitt 1976) decreases from  $1.430$  to  $1.426 \text{ \AA}$  as  $x$  increases, while the average of the ionic radius in the B site ( $\langle r_B \rangle$ ) ( $\langle r_B \rangle = (1-x)(0.9r(\text{Ti}^{4+}) + 0.1r(\text{Zr}^{4+})) + xr(\text{Fe}^{3+})$ ) with  $r(\text{Ti}^{4+}) = 0.605 \text{ \AA}$ ,  $r(\text{Zr}^{4+}) = 0.72 \text{ \AA}$ ,  $r(\text{Fe}^{3+}) = 0.645 \text{ \AA}$  in six-coordination (Shannon and Prewitt 1976) varies slightly with regard to  $\langle r_A \rangle$ , from  $0.616$  to  $0.618 \text{ \AA}$ . Thus, regular decrease of the ionic radius in the A site cation induces a smaller A site and causes a decrease in space between network of  $\text{TiO}_6$  octahedra and consequently in the unit cell volume.

#### 3.2 Dielectric properties

The dielectric measurements for the solid solution,  $(1-x)\text{Ba}(\text{Ti}_{0.9}\text{Zr}_{0.1})\text{O}_3-(x)\text{BiFeO}_3$ , show different behaviours

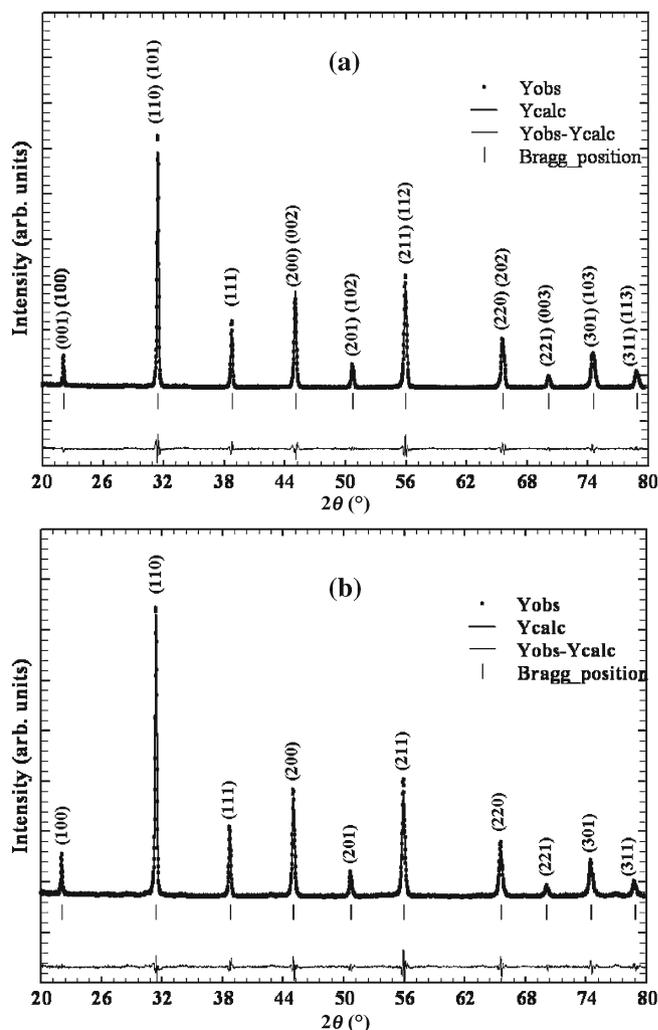


Figure 1. Observed, calculated and difference X-ray diffraction patterns of (a)  $x = 0.005$  and (b)  $x = 0.025$ .

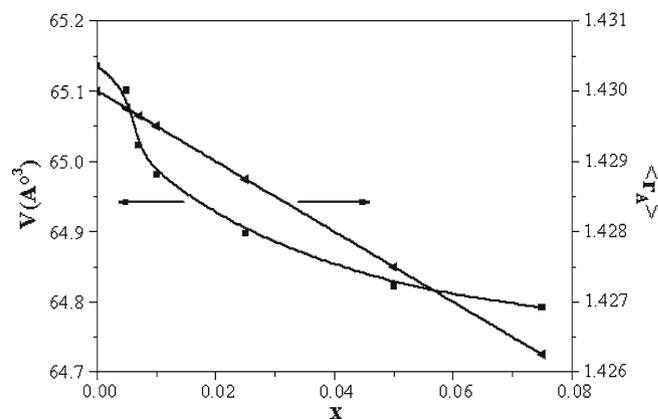
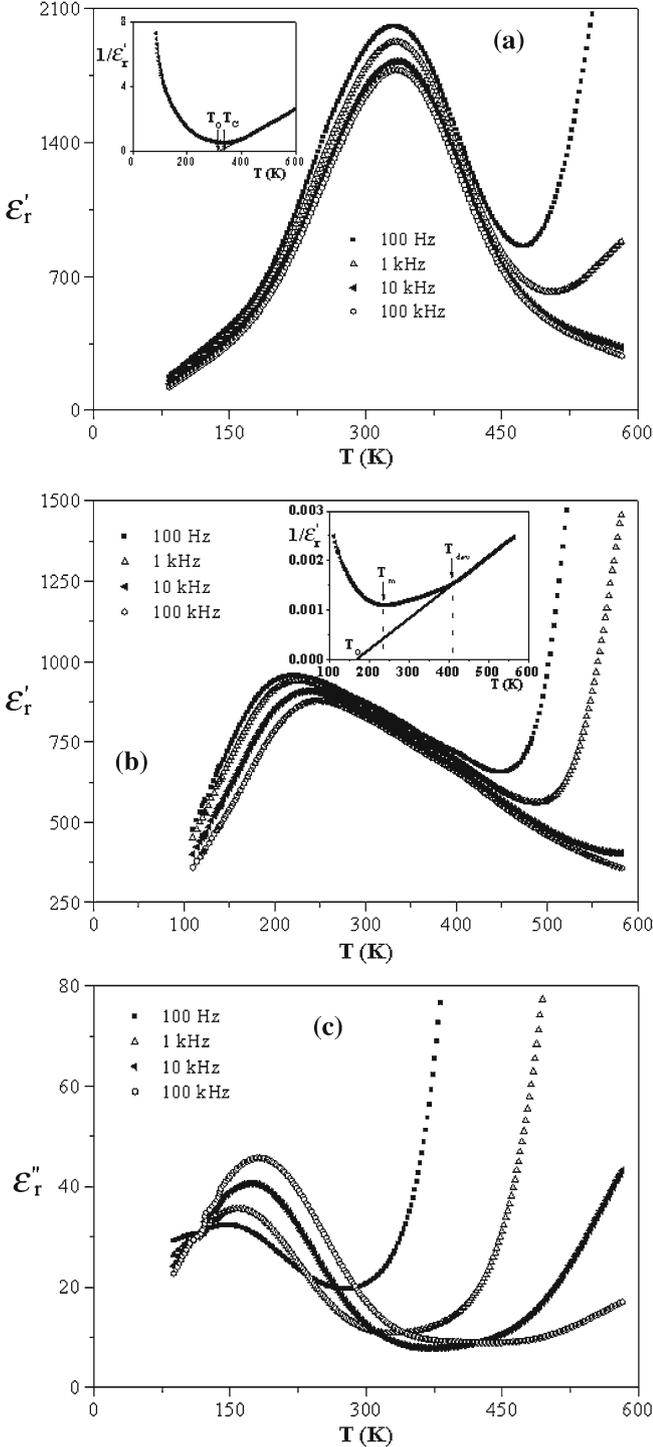


Figure 2. Variation of unit cell volume and average ionic radius ( $\langle r_A \rangle$ ) with  $x$  composition.

for different values of  $x$ . For  $x$  varying from 0 to  $0.007$ , the ceramics show classical ferroelectric behaviour with the characteristics of ferroelectric–paraelectric phase transition

(figure 3(a)) and the value of  $T_C$  is not dependent on frequency. For compositions in the range  $0.007 < x \leq 0.075$ , the system shows a relaxor behaviour, the peak of  $\epsilon'_r$  becomes very broad with strong frequency dispersion of  $\epsilon'_r$ . Figure 3(b) shows, for example, the dielectric behaviour



**Figure 3.** Temperature and frequency dependence of permittivity,  $\epsilon'_r$  and thermal variation of  $1/\epsilon'_r$  at 10 kHz for compositions: (a)  $x = 0.007$ , (b)  $x = 0.05$  and (c) temperature dependence of imaginary permittivity,  $\epsilon''_r$ , for composition  $x = 0.05$ .

for  $x = 0.05$ . The maximum value of  $\epsilon'_r$  decreases and the temperature,  $T_m$ , shifts toward higher temperature with increasing frequency (figure 3(c)).

In order to investigate the nature of the phase transition in our solid solution, we represent the temperature variation of  $1/\epsilon'_r$ . For  $0 \leq x \leq 0.007$ , temperature dependence of  $1/\epsilon'_r$  at  $10^4$  Hz shows that the Curie-Weiss law:  $\epsilon'_r = C/(T - T_0)$  is followed for  $T > T_C$ . So the phase transition for  $x = 0.007$  is of first order type (inset of figure 3(a)). For relaxor compositions ( $0.007 < x \leq 0.075$ ), there was a deviation from the Curie-Weiss law when the value of the Curie-Weiss temperature  $T_0$  was greater than that of  $T_m$ . The real part of dielectric permittivity follows the Curie-Weiss law at  $T > T_{dev}$  ( $T_{dev}$  represents temperature at which  $\epsilon'_r$  deviates from Curie-Weiss law). As an example, we present in the inset of figure 3(b), the thermal evolution of  $1/\epsilon'_r$  at 10 kHz for composition,  $x = 0.05$ . The observed deviation for  $T < T_{dev}$ , is the characteristic of dipole-dipole interactions responsible for some type of short-range order (Simon *et al* 2005).

The relaxor characteristics for some compositions are presented in table 1.  $\Delta T_m = T_m(2 \times 10^5 \text{ Hz}) - T_m(10^2 \text{ Hz})$  and the relative frequency dispersion  $\Delta\epsilon'_r/\epsilon'_r = (\epsilon'_r(10^2 \text{ Hz}) - \epsilon'_r(2 \times 10^5 \text{ Hz}))/\epsilon'_r(10^2 \text{ Hz})$  increase with compositions. As a consequence, the increase of bismuth and iron content increases the degree of relaxor behaviour in the solid solution. From table 1, we notice a relaxor behaviour that appears up to  $x = 0.01$ . This composition shows a  $\Delta T_m \sim 12$  K at room temperature and this could be the reason why it has some applications.

To describe the diffuseness of phase transition for this composition, we use a modified empirical expression proposed by Uchino and Nomura (1982).

$$\frac{1}{\epsilon'_r} - \frac{1}{\epsilon'_{rm}} = \frac{(T - T_m)^\gamma}{C}, \quad 1 \leq \gamma \leq 2,$$

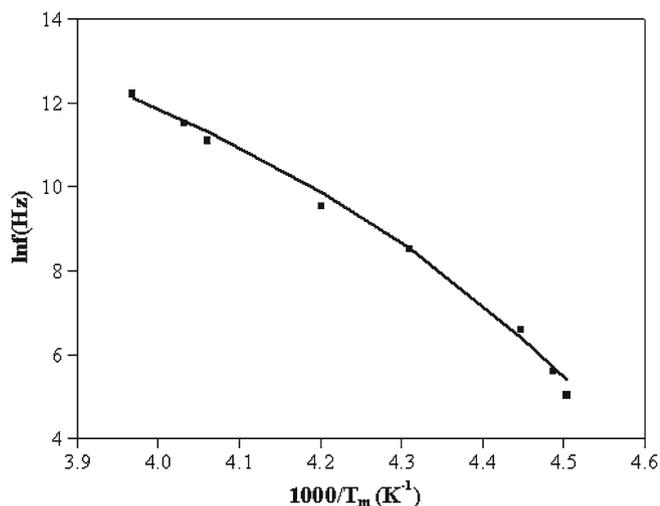
where  $C$  is a constant and  $\gamma$  value is between 1 and 2. The value of  $\gamma$  gives information on the phase transition diffuse character. The values of  $\gamma = 1.153, 1.322$  and  $1.503$  for  $x = 0.01, 0.05$  and  $0.075$  at 10 kHz, respectively. It is clear that the value of  $\gamma$  increases with increase of  $x$ . The phase transition becomes more diffuse when the quantity of  $\text{Bi}^{3+}$  and  $\text{Fe}^{3+}$  increases. This is linked to the increase of disorder in the ceramic.

**Table 1.** Values of  $T_m$ ,  $\Delta T_m$  and  $\Delta\epsilon'_r/\epsilon'_r$  characteristics for various compositions of solid solution,  $\text{Ba}_{1-x}\text{Bi}_x(\text{Ti}_{0.9}\text{Zr}_{0.1})_{1-x}\text{Fe}_x\text{O}_3$ .

$x$	$T_C$ or $T_m$ (K)	$\Delta T_m$ (K)	$\Delta\epsilon'_r/\epsilon'_r$
0.000	$T_C = 363$ K	0	0
0.007	$T_C = 334$ K	0	0
0.010	309	12	0.079
0.050	231	33	0.093
0.075	246	38	0.111

A relaxor type is generally subject to the Vogel–Fulcher relation (Vogel 1921; Fulcher 1925) which has the form  $f = f_0 \exp[-E_a/(T_m - T_f)]$ , where  $f$ ,  $E_a$ ,  $T_f$  and  $f_0$  are applied frequency, activation energy, freezing temperature and attempt frequency, respectively. In our case, compositions,  $x = 0.01, 0.05$  and  $0.075$ , also obey this relation. In figure 4, we present variation of  $\ln f$  as a function of  $1000/T$  for  $x = 0.05$  as an example. The parameters of Vogel–Fulcher relation obtained are  $E_a = 0.056$  eV,  $f_0 = 1.76 \times 10^9$  Hz and  $T_f = 181$  K. As for PMN, the fitting parameters are  $T_f = 223$  K,  $E_a = 0.043$  eV and  $f_0 = 10^{12}$  Hz (Viehland and Huang 1994). So, value of the activation energy,  $E_a$ , in our compound is more than that in PMN, indicating a higher potential barrier between two potential wells. We deduce that, in our case, the substituted atoms vibrate more easily in their sites and their displacement is more difficult from one site to another. However, the pre-exponential factor  $f_0$  is lower, reflecting that the potential well in which the elements of our material vibrate is probably wider than the case of PMN. These different characteristics related to potential well reflect different mechanisms of polarization (Cross 1987).

Various theoretical models are developed to explain the relaxor behaviour, such as superparaelectricity, dipolar glass, random-field model, etc. (Bokov and Ye 2006; Long and Ye 2007). A common point of these models is based on the local order–disorder of the crystal structure, giving rise to polar clusters embedded in the matrix (Ang et al 2000). In our case, the substitution of  $\text{Ba}^{2+}$  by  $\text{Bi}^{3+}$  and  $\text{Zr}^{4+}/\text{Ti}^{4+}$  by  $\text{Fe}^{3+}$  gives rise to composition heterogeneity. This probably promotes the presence of microregions with different local Curie points in the compound leading to diffuse phase transition in  $\text{Ba}(\text{Ti}_{0.9}\text{Zr}_{0.1})\text{O}_3$  doped with  $\text{BiFeO}_3$  and appearance of more relaxor-like behaviour. Moreover, stereochemical activity of the Bi lone pair and the large difference of ionic radii between  $\text{Ba}^{2+}$  ion and added  $\text{Bi}^{3+}$  ion can drive



**Figure 4.** Variation of  $\ln f$  with  $1000/T_m$  for a ceramic with composition,  $x = 0.05$  (symbols: experimental data and solid curve fit to Vogel–Fulcher relation).

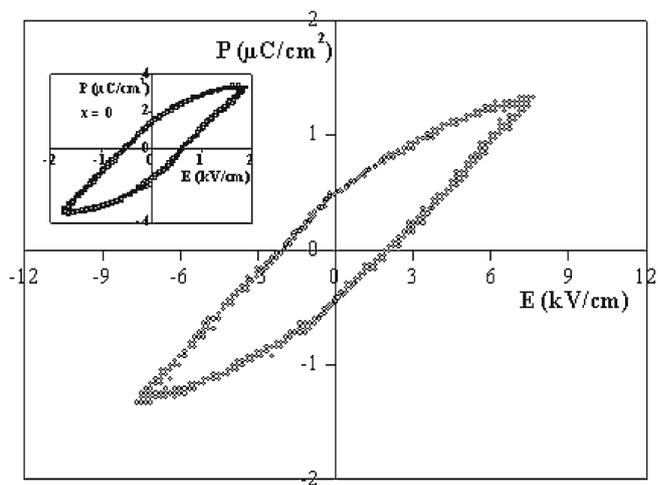
the large displacement at the off-centred A position, resulting in the formation of polar clusters. The dynamic response of this polar cluster is responsible for frequency dispersion. On the other hand, substitution of  $\text{Zr}^{4+}/\text{Ti}^{4+}$  by  $\text{Fe}^{3+}$  for site B may change the value of dielectric permittivity (Swartz and Shrout 1982).

### 3.3 Ferroelectric properties

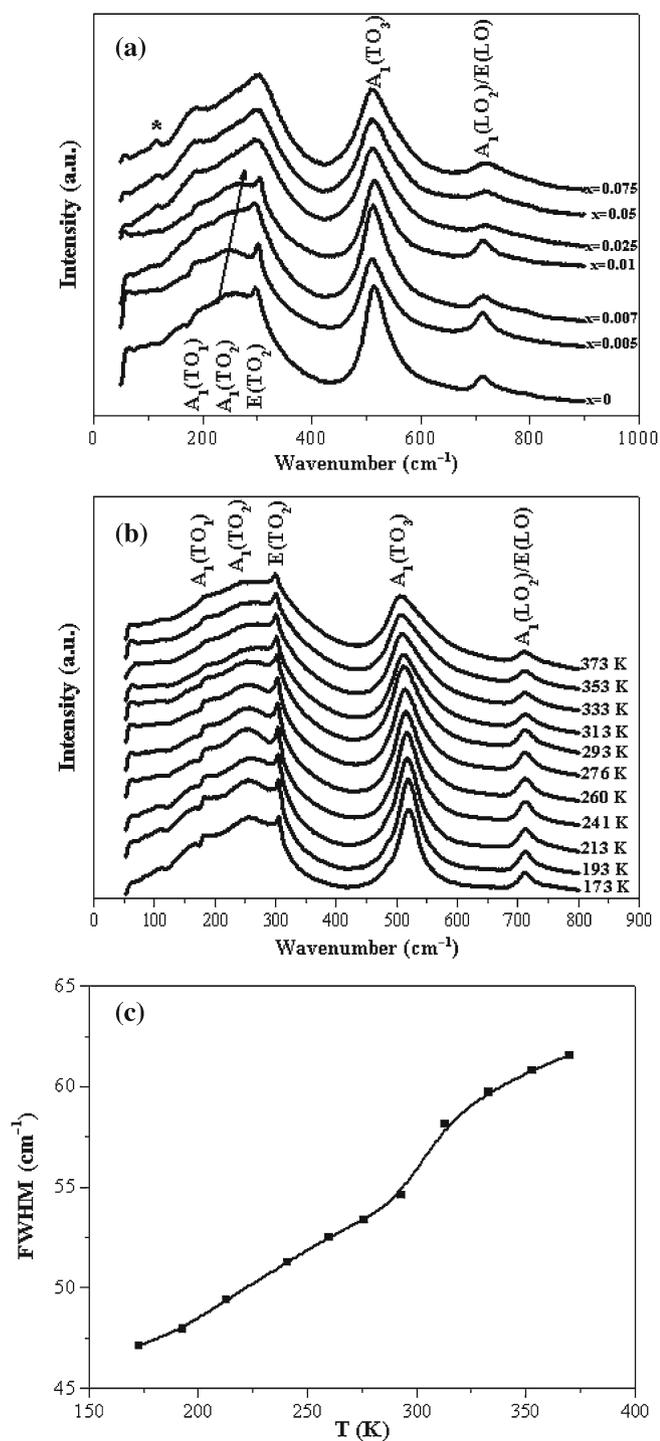
To investigate the influence of  $\text{BiFeO}_3$  doping on ferroelectric properties of  $\text{Ba}(\text{Ti}_{0.9}\text{Zr}_{0.1})\text{O}_3$ , we have measured the ferroelectric polarization as a function of electric field at room temperature for  $x = 0$  and  $0.007$  (figure 5). We note that for  $\text{Ba}(\text{Ti}_{0.9}\text{Zr}_{0.1})\text{O}_3$  ( $x = 0$ ) ceramic, the remnant polarization,  $P_r$  and the coercive electric field,  $E_C$  are  $1.5 \mu\text{C}/\text{cm}^2$  and  $0.5$  kV/cm, respectively. However, a small doping of  $\text{BiFeO}_3$  in  $\text{Ba}(\text{Ti}_{0.9}\text{Zr}_{0.1})\text{O}_3$  ( $x = 0.007$ ) decreases drastically  $P_r$  to  $0.5 \mu\text{C}/\text{cm}^2$  and increases  $E_C$  to  $2$  kV/cm. This result indicates that substituting  $\text{Ba}^{2+}$  by  $\text{Bi}^{3+}$  and  $\text{Ti}_{0.9}\text{Zr}_{0.1}$  by  $\text{Fe}^{3+}$ , increases not only the composition heterogeneity effect but also reduces the long-range ferroelectricity (Ti–O) in the compound and so promotes relaxor effect.

### 3.4 Raman spectroscopy analysis

The room-temperature Raman spectra for  $\text{Ba}_{1-x}\text{Bi}_x(\text{Ti}_{0.9}\text{Zr}_{0.1})_{1-x}\text{Fe}_x\text{O}_3$  for  $x = 0, 0.005, 0.007, 0.01, 0.025, 0.05$  and  $0.075$  ceramics are illustrated in figure 6(a). Mode assignment is done following the spectral assignment carried out by Moura et al (2008) for the ceramic,  $\text{Ba}(\text{Ti}_{0.9}\text{Zr}_{0.1})\text{O}_3$ . The Raman spectrum at room temperature for  $x = 0$  ceramic shows all bands at  $193, 250, 301, 517$  and  $720 \text{ cm}^{-1}$ . These are attributed respectively to  $A_1(\text{TO}_1), A_1(\text{TO}_2), E(\text{TO}_2), A_1(\text{TO}_3)$  and  $A_1(\text{LO}_2)/E(\text{LO})$ .



**Figure 5.** P–E hysteresis loops at room temperature for  $x = 0$  and  $0.007$  compositions.



**Figure 6.** (a) Raman spectra of Ba<sub>1-x</sub>Bi<sub>x</sub>(Ti<sub>0.9</sub>Zr<sub>0.1</sub>)<sub>1-x</sub>Fe<sub>x</sub>O<sub>3</sub> samples at room temperature, (b) Raman spectra of Ba<sub>0.99</sub>Bi<sub>0.01</sub>(Ti<sub>0.9</sub>Zr<sub>0.1</sub>)<sub>0.99</sub>Fe<sub>0.01</sub>O<sub>3</sub> ceramic at various temperatures and (c) FWHM of mode A<sub>1</sub>(TO<sub>2</sub>) as a function of temperature for Ba<sub>0.99</sub>Bi<sub>0.01</sub>(Ti<sub>0.9</sub>Zr<sub>0.1</sub>)<sub>0.99</sub>Fe<sub>0.01</sub>O<sub>3</sub>.

The simultaneous incorporation of Bi<sup>3+</sup> and Fe<sup>3+</sup> into Ba(Ti<sub>0.9</sub>Zr<sub>0.1</sub>)O<sub>3</sub> causes spectral modifications, which reflect the structural variations observed by X-ray diffraction.

Indeed, the dramatic change of Raman spectrum at  $x > 0.01$ , is marked by the disappearance of the E(TO<sub>2</sub>) mode. Interestingly, this mode is associated with the tetragonal-cubic phase transition as it will be shown elsewhere. When  $x$  increases, all bands show significant broadening connected with the disorder created on the A and B sites of Ba(Ti<sub>0.9</sub>Zr<sub>0.1</sub>)O<sub>3</sub>.

Nevertheless, a closer inspection of the spectra reveals that for  $x \leq 0.1$ , it should exhibit different local deformations, as suggested by the shift of the broad A<sub>1</sub>(TO<sub>2</sub>) mode ( $\sim 250$  cm<sup>-1</sup>) to higher frequencies (indicated by arrows in figure 6(a)). For  $x = 0.025$ , this mode appears at  $-300$  cm<sup>-1</sup>. A similar behaviour was observed by Strathdee *et al* (2011) for BaTiO<sub>3</sub>-BiYbO<sub>3</sub> system and the opposite for BaTiO<sub>3</sub>-LaYbO<sub>3</sub> system (Feteira and Sinclair 2009). So, the presence of a lone-pair of electrons from Bi<sup>3+</sup> in the Ba(Ti<sub>0.9</sub>Zr<sub>0.1</sub>)O<sub>3</sub>-BiFeO<sub>3</sub> hardens this vibration mode, which is believed to be associated with TiO<sub>6</sub> and ZrO<sub>6</sub> octahedra, in particular with the bending of Ti-O and Zr-O bonds.

Common to the  $x = 0, 0.005, 0.007$  and  $0.01$  spectra, is the presence of E(TO) mode at 301 cm<sup>-1</sup>. This spectral feature is considered by many authors as the signature for the occurrence of long-range order ferroelectricity in tetragonal BaTiO<sub>3</sub>. The absence of this feature in  $x = 0.025, 0.05$  and  $0.075$  spectra suggests the disappearance of long-range ferroelectricity in these samples, in agreement with the dielectric behaviour of these samples showing the appearance of the relaxor behaviour.

The composition,  $x = 0.01$ , represents the limit between relaxor and classical behaviour. In figure 6(b), we present the evolution of Raman spectra for  $x = 0.01$  composition at different temperatures. The width of all the modes increases as the temperature increases; this is obvious since the thermal agitation creates the disorder.

Figure 6(c) shows variation of FWHM associated with the mode A<sub>1</sub>(TO<sub>2</sub>) vs temperature for  $x = 0.01$ . An anomaly observed at 313 K corresponds to the temperature of transition,  $T_m = 309$  K at 5 kHz deduced from dielectric measurements.

#### 4. Conclusions

(1-x)Ba(Ti<sub>0.9</sub>Zr<sub>0.1</sub>)O<sub>3</sub>-(x)BiFeO<sub>3</sub> ceramics are prepared by a solid state reaction method. These compounds crystallize in tetragonal structure for  $0 \leq x \leq 0.01$  and pseudocubic for  $0.01 < x \leq 0.075$ . The material is classical ferroelectric in the range  $0 \leq x \leq 0.007$  and presents a relaxor behaviour in the range  $0.007 < x \leq 0.075$ . The Ba<sub>0.99</sub>Bi<sub>0.01</sub>(Ti<sub>0.9</sub>Zr<sub>0.1</sub>)<sub>0.99</sub>Fe<sub>0.01</sub>O<sub>3</sub> ceramic exhibits a relaxor behaviour with  $\Delta T_m = 12$  K at room temperature. This composition can be interesting for industrial applications which need a relaxor character at room temperature with non-lead-based ceramic. The vibrational Raman spectroscopy study for different compositions at room temperature correlates well with XRD and dielectric measurement results.

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