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Frequency dielectric response of ferroelectric–magnetic ceramic composites like $PbZr_{0.65}Ti_{0.35}O_3$–$BaFe_{12}O_{19}$

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Abstract

The dielectric properties of $PbZr_{0.65}Ti_{0.35}O_3$–$BaFe_{12}O_{19}$ (PZT–BaM) multiferroic ceramic composites, obtained from the conventional solid-state reaction method, have been investigated. While there is some dielectric dispersion at low frequencies, the highly-stable values of dielectric permittivity over a wide frequency range, together with low dielectric losses ($\sim 10^{-3}$), make these composites suitable for practical radio-frequency and potential microwave device applications. Toward high frequencies, the data reveal an anomalous resonance-like phenomenon. This resonance phenomenon is connected to an over-damped resonance mechanism for the vibration of the boundaries of polar regions, associated with the ferroelectric phase. A noticeable dependence of the parameters characterizing this last dielectric dispersion with the BaM concentration is found, indicating a strong influence of the magnetic phase on the dynamics of the ferroelectric domain-wall motion.

Keywords: B. Composites; C. Dielectric properties; Ceramics

1. Introduction

The interest in studying high-frequency tunable dielectric materials has been increasing over the past decade due to the growing needs of these materials in high-speed data transmission devices. Tunable filters and resonators at microwave frequencies are a few examples [1], where high tunability (strong dependence of dielectric permittivity on an electric field 'bias') and low dielectric losses (tanδ) are important characteristics for such materials in device design. Specifically, multiferroic systems, which couple the interaction of two or more fundamental ferroic (ferroelectric, ferro/ferrimagnetic and ferroelastic) properties, have received special attention in recent years because of their multi-functionality [2]. However, despite their excellent physical properties and promissory future for novel multifunctional devices [3], only a few works have been devoted to the study of the high-frequency dielectric response of multiferroics. Additional reports on the microwave dielectric properties of single-phase multi-ferroics mostly based on $Pb(Fe_{1/2}Nb_{1/2})O_3$ (PFN), where the electric and magnetic orders are intrinsically coupled, can be found in the literature [4,5]. In fact, the first evidence of a real magneto-electric (ME) coupling in multiferroics, from microwave dielectric measurements, was carried out in PFM ceramic samples [4]. The results suggested the nature of the ME coupling to arise indirectly via a ferroelastic contribution rather than a direct coupling between the magnetic and electric orders [4]. More recently, Sobiestianskas et al. also reported microwave dielectric dispersion in a PFN thin film grown on a (001) $SrTiO_3$ substrate,

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and the observed relaxational response was associated with a possible mode-softening behavior related to the onset of polar nano-regions at a specific temperature [5].

It is now well known that better alternatives to single-phase materials are multi-phase multiferroic composites (where individual ferroelectric and ferro/ferrimagnetic phases, with high piezoelectric and magnetostrictive properties, respectively, coexist), which possess large ME response when compared to the single-phase multiferroic systems [6]. Specially, because of their high permeability values showing a rapid variation with frequency and magnetic field, ferrites are of great interest in the high-frequency region, having the possibility to be used in ferromagnetic resonance-based devices for microwave signal processing components such as resonators, filters and phase shifters [7]. However, from the experimental point of view, in most of the cases tuning the operating frequency of a ferrite device generally involves a very high-power source, which limits the miniaturization for specific components. In this way, tuning of magnetic modes can be alternatively achieved in composite systems based on ferro/ferrimagnetic and piezoelectric composites [6,8], through the interface strain produced by an external magnetic and/or electric field. When such composites are subjected to an electric field, the piezoelectric strain is transmitted to the ferrite phase, which manifests as an internal magnetic field (or an anisotropy field), leading to a mechanical strain-mediated magnetoelectric (ME) coupling at the ferromagnetic phase resonance frequency [9]. Thus, mechanical strain assisted tuning of magnetic modes in composites makes them ideal systems for studying wide-band ME interactions between the magnetic and electric subsystems, as well as their integration in miniature high-frequency devices [10], where important characteristics such as high-Q, low-insertion loss, and high out-of band rejection, can be improved.

Nevertheless, although they have promising characteristics for practical applications, the investigation of the microwave dielectric response in multi-phase multiferroic composites has been scarcely reported in the current literature. Thus, the emerging new technologies based on multiferroic composite materials, showing exceptional high-frequency activity and great useful for the manufacture of multifunctional devices with applications in global positioning systems, dielectric resonators, advanced radar systems, and others [11], require the development of new materials and better understanding of their properties.

The purpose of the present work is to investigate the dielectric response of multiferroic ceramic composites based on ferrolectric $PbZr_{0.65}Ti_{0.35}O_3$ (PZT) and ferrimagnetic $BaFe_{12}O_{19}$ (BaM) subsystems, which have not been reported before elsewhere to our best knowledge. Concretely, the influence of BaM content on the microwave dielectric properties of representative compositions in this PZT–xBaM system has been studied over a wide frequency range and the parameters characterizing, particularly, the microwave frequency dielectric dispersion have been determined after noting the recorded data to be in these cases related to the occurrence of 'over-damped' resonant processes [12]. The results are discussed within the framework of current models reported in the literature.

2. Experimental procedure

Ceramic composites based on ferroelectric $PbZr_{0.65}Ti_{0.35}O_3$ (PZT) and ferrimagnetic $BaFe_{12}O_{19}$ (BaM) were obtained following the solid-state reaction method. After synthesizing separately these phases, the composites were prepared according to the relation $PZT_{(1-x)}BaM_x$ (PZT–xBaM), where $x=0.03$, 0.04 and 0.05. Detailed information about preparation of these samples can be found elsewhere [13]. After milling, the powders were uniaxially pressed into disc-shaped samples by using 10 MPa and then sintered in closed alumina crucibles at 1250 °C for 3 h. The samples are hereafter labeled as BaM003, BaM004 and BaM005. The densities of the studied composites were obtained from the Archimedes method. Relative density values higher than 90% of the theoretical one (relative to pure PZT65/35) were obtained in all these cases, showing only a slight decrease with the increase of BaM. The (micro)structural characteristics of the materials were investigated via X-ray diffraction (XRD), by using a Shimadzu XRD 6000 diffractometer with Cu Kα radiation, and scanning electronic microscopy (SEM), by using a JEOL JSM-840 Microscope. In order to study the dielectric properties, silver electrodes were applied on the two major opposite surfaces of the samples. Low frequency (LF) dielectric measurements were performed at room temperature by using an HP4284A Precision LCR Meter, covering the frequency range of 100 Hz to 1 MHz. High frequency (HF) dielectric measurements were also carried out, again at room temperature, by using a RF Impedance/Material Analyzer HP4291A, covering the frequency range of 1 MHz to 1.8 GHz.

3. Results and discussion

Fig. 1 shows the XRD patterns obtained on the powdered ceramics for the studied PZT–BaM composites. As can be seen, presence of the perovskite-structured ferroelectric phase, with rhombohedral $R3m$ symmetry, is everywhere confirmed

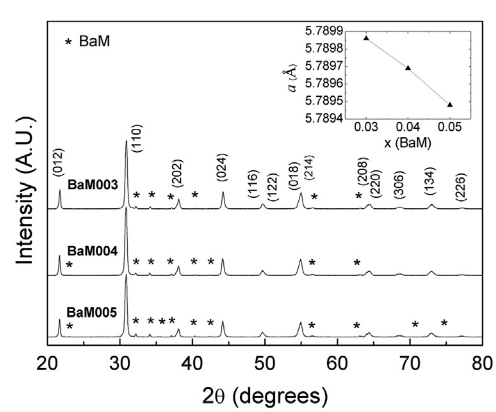

Fig. 1. X-ray diffraction patterns of the studied BaM003, BaM004 and BaM005 compositions, obtained at room temperature. Index represents the peaks corresponding to the PZT phase and symbols are the $BaFe_{12}O_{19}$ (BaM) reflections. The inset shows the variation of the PZT rhombohedral lattice-parameter with increasing the BaM content.

(indexed peaks). In addition, successful synthesis of a two-phase system is established by observation of XRD reflection peaks corresponding to the magnetic phase, with hexagonal (*P*63/*mmc*) structure, the intensity of which increases with raising the nominal content of BaM. In addition, the inset of Fig. 1 indicates that the PZT rhombohedral lattice-parameter decreases in presence of the BaM phase, the consequence of which will be later discussed in view of the dielectric response exhibited by these composites toward the microwave frequency region.

Fig. 2 shows the SEM micrographs obtained for the studied BaM003, BaM004 and BaM005 ferroelectric–magnetic composites (a, b and c, respectively). The images reveal the synthesis of dense and homogeneous microstructures (grain morphology, included) in all these cases, with a slight decrease of the grain size with the increase of the BaM concentration. That is, the estimated values of average grain size are 3.17 μm, 3.0 μm and 2.73 μm for the BaM003, BaM004 and BaM005 composites, respectively.

It is important to point out that the multiferroic characteristics of the PZT–BaM system studied here were recently confirmed through measurements of the electric field dependence of polarization (*P*–*E*) and magnetic field dependence of magnetization (*M*–*H*) hysteresis loops, as well as magneto-dielectric (MD) response [14,15]. The results reveal excellent ferroelectric and magnetic properties [14], together with an enhanced magneto-electric (ME) response [15], when comparing these materials to other typical single-phase as well as multi-phase multiferroic systems [16,17].

In the following, Fig. 3 depicts the frequency dependence of the real (ε') and imaginary (ε'') components of the dielectric permittivity ($\varepsilon^* = \varepsilon' - j\varepsilon''$) from these BaM003, BaM004 and BaM005 composites. The data correspond to measurements achieved at room temperature in the low frequency (LF) regime ranging from 100 Hz to 1 MHz. Both ε' and ε'' remain almost constant toward high frequencies (above 10 kHz), while showing a rise when decreasing the electric field frequency. The last event is in these cases associated with direct current (DC)

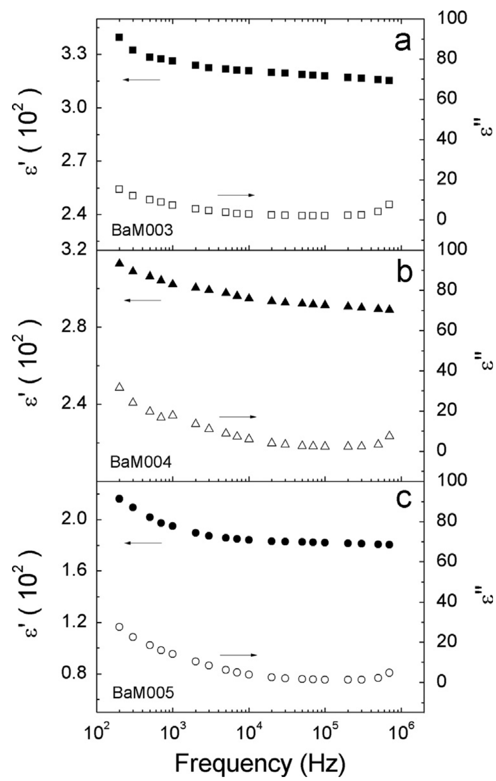

Fig. 3. Low frequency (LF) dielectric permittivity (real and imaginary components) measured on the BaM003, BaM004 and BaM005 composites at room temperature.

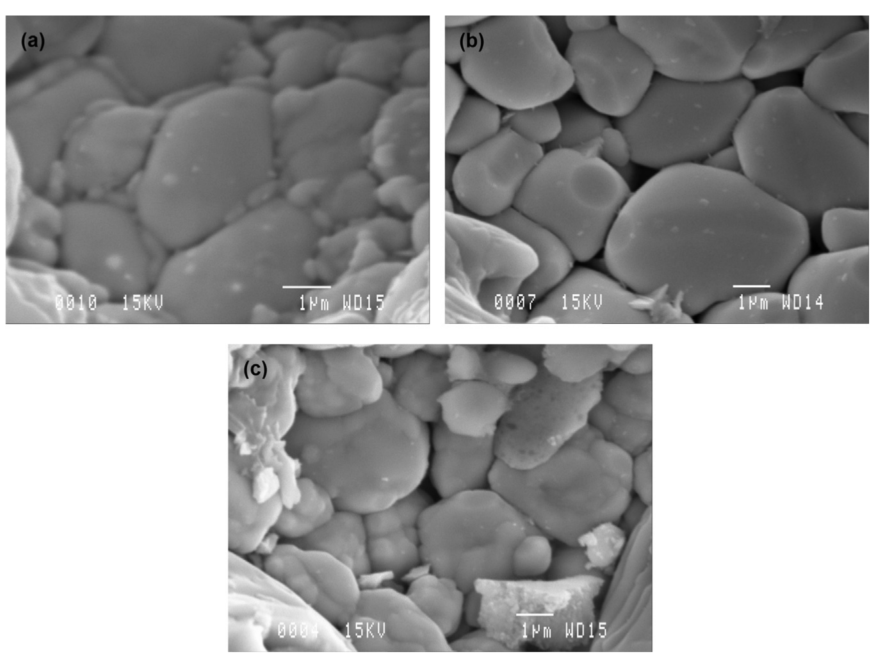

Fig. 2. Micrographs of the sintered BaM003, BaM004 and BaM005 compositions (a, b and c, respectively) studied in this work.

conduction processes (for ε'') through the materials, and Maxwell–Wagner polarization-like effects (for ε') from charge carriers being partially blocked at internal interfaces [18–22]. In particular, it is observed that ε' decreases over the entire analyzed frequency range with increasing BaM content, as it is to be expected from influence of a growing non-ferroelectric (magnetic, in this case) phase on the total dielectric properties of such a two-phase system.

To summarize, Table 1 shows the ε' and ε'' as well as dielectric loss (tanδ) values from these composites, as extracted from Fig. 3 at the frequency of 100 kHz, which should reflect the bulk dielectric properties of these materials [20–22]. As can be seen, interesting from the viewpoint of microelectronic device applications [23,24], albeit increasing with raising the BaM content, low dielectric loss values ($\sim 10^{-3}$) are observed in this composition range of PZT–BaM composites. The increase in tanδ is associated with the electrical transport process promoted by the highly-conductive magnetic phase [25,26]. That is, leakage currents related to Fe^{3+}–O^{2-}–Fe^{3+} type super-exchange as well as Fe^{3+}–Fe^{2+} type double-exchange interactions are characteristic of magnetic compounds like BaM when subjected to the action of an electric field [19,20], the effect of which can be registered in dielectric measurements. It is important to point out that the value of dielectric permittivity (ε') reported in Table 1 for the sample with the lower BaM concentration (BaM003) is higher than the values observed elsewhere in some multiferroic systems, in the same frequency range [27,28].

The variations of the real and imaginary components of the dielectric permittivity obtained in the microwave frequency region are shown in Fig. 4(a–c) for the BaM003, BaM004 and BaM005 composites, respectively. The results reveal occurrence of a dielectric anomaly in all the investigated composites toward this GHz frequency region, being typical of a resonance phenomenon. Concretely, the real component of the dielectric permittivity (ε') remains essentially flat in a wide frequency range, and then starts to increase for frequencies higher than 1×10^8 Hz. After that, ε' traverses a maximum and then decreases up to its clamped value (at frequencies approaching 1×10^9 Hz), whereas the imaginary dielectric permittivity (ε'') passes through its maximum value. The frequency corresponding to this maximum (f_R) is characteristic of the mechanism (polarization with or without resonance) responsible for energy dissipation in a material under electric field action.

At this point, we would like to state that pure PZT is known to show a ferroelectricity-related dielectric relaxation rather than resonance phenomenon toward this microwave frequency range [29], where the characteristic frequency (f_R) was found to be just

Table 1
Room-temperature real (ε') and imaginary (ε'') components of the dielectric permittivity and dielectric losses (tanδ) extracted from Fig. 3 for 100 kHz.

Sample	ε'	ε''	tanδ (10^{-3})
BaM003	318	2.12	6.67
BaM004	291	2.29	7.87
BaM005	182	1.45	7.96

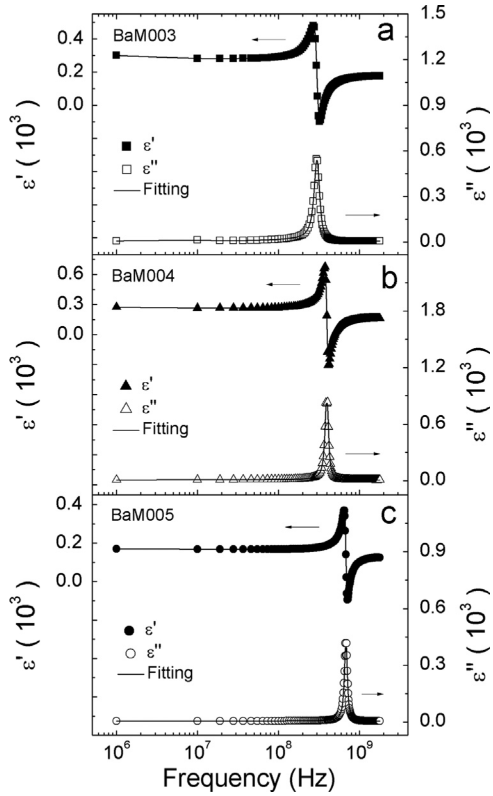

Fig. 4. High frequency (HF) dielectric permittivity (real and imaginary components) measured on the BaM003, BaM004 and BaM005 composites at room temperature.

above 300 MHz [30]. Nevertheless, results as those shown in Fig. 4 have been also found in some other situations involving the dielectric response of ferroelectric materials toward microwave frequencies [31,32]. Such a dielectric response has often been attributed to piezoelectric resonance of grains [33] and/or individual domains [34], inertial component of domain walls [35], and to the correlated hopping of off-centered ferroelectric active ions between several potential walls [36]. In all the cases, the dispersion phenomenon appears to be intimately linked to the domain state of the ferroelectric phase, because of their intrinsic excellent piezoelectric response.

A more general description of the appearance of such high-frequency dielectric anomalies has been recently retraced by Guerra et al., a description in which the vibration of the boundaries of polar regions is considered to be the mechanism responsible for the microwave dielectric dispersion processes observed in ferroelectric systems [12,37]. In this way, the observed dielectric anomalies in the microwave region is dictated by a universal mechanism involving an over-damped resonance phenomenon associated with the polar regions present in ferroelectrics [12]. Thus, either typical relaxation-like (over-damped) or resonance behaviors may be found, depending on the damping strength, which is governed by the coupling between the ferroelastic and ferroelectric contributions to the high-frequency dielectric response. Such a coupling can be controlled by applying, for instance, a mechanical stress (static pressure) parallel to the applied electric field direction, promoting a significant variation in the damping coefficient of the system. A concluding remark in this topic is that the higher the damping strength the higher the contribution to the relaxation-like (over-damped) behavior; otherwise (i.e., under low damping strength), the resonance phenomenon becomes favored.

Back then to the results depicted in Fig. 4, and considering that no dielectric anomaly is expected from the magnetic BaM phase toward this microwave frequency region, observation of a resonance phenomenon in the present materials should most likely be the consequence of the clamped-like state of the PZT phase in the PZT–BaM composites, as suggests the inset of Fig. 1, where the estimated PZT rhombohedral lattice-parameter revealed a decrease in presence of BaM. Taking into account all these remarks, the dielectric spectra measured for BaM003, BaM004 and BaM005 were here fitted following a damped harmonic oscillator model [12], in which the frequency dependence of the complex dielectric permittivity ($\varepsilon^{*}=\varepsilon'-j\varepsilon''$) is described according to following equations:

$$\varepsilon' = \varepsilon_{\infty} + \frac{\Delta\varepsilon\omega_R^2\left(\omega_R^2-\omega^2\right)}{\left(\omega_R^2-\omega^2\right)^2+\gamma^2\omega^2} \quad (1)$$

$$\varepsilon'' = \frac{\Delta\varepsilon\omega_R^2\gamma\omega}{\left(\omega_R^2-\omega^2\right)^2+\gamma^2\omega^2} \quad (2)$$

Here, the parameter $\Delta\varepsilon$ is the dielectric strength ($\Delta\varepsilon=\varepsilon_0-\varepsilon_\infty$, where ε_0 is the low frequency, or static, dielectric permittivity), ε_∞ represents the high-frequency contribution to the dielectric permittivity, ω is the angular frequency ($\omega=2\pi f$, where f is the measurement frequency) and γ is the damping coefficient. Results from the permittivity data fitting are shown in Fig. 4 using solid lines. The parameters characterizing the dielectric dispersion, as obtained from the data fitting processes, are summarized in Table 2 for these PZT–BaM composites.

As can be concluded from Table 2, the magnetic BaM component has a noticeable influence on the high-frequency dielectric response of the studied PZT–BaM composites, through affecting directly the dynamics of the ferroelectric domain-wall motion in the ferroelectric PZT phase. The values obtained for the damping coefficient are in the order of those values found in parent ferroelectric systems with similar resonance-like dielectric response [12,31]. In the context of the present report, taking into account the resonant dielectric nature exhibited by these PZT–BaM composites, the variations observed for the characteristic frequency (f_R) could be explained on the basis of the microstructural-related grain size evolution shown by these materials. That is, as reported elsewhere [33], the dielectric resonance sometimes observed in such materials over the microwave region can be attributed to the coupled resonance of piezoelectric grains of different size and orientation, relative to the measuring electric field. In this way, the high frequency dispersion can be related to a change in the permanent net dipole moment as well as creation of a mechanical deformation, which propagates at the velocity of sound. For dimensions of individual grains scaling in the order of 1–10 μm, as in this work, and considering that the speed of sound is in the order of 10^5 cm/s, then resonance frequencies values in the 100 MHz to 1 GHz range should be expected [33].

Table 2
Room-temperature high frequency (microwave) dielectric dispersion parameters: dielectric strength, characteristic frequency and damping coefficient ($\Delta\varepsilon$, f_R and γ, respectively) for the BaM003, BaM004 and BaM005 composites, obtained from fitting the curves in Fig. 4.

Sample	$\Delta\varepsilon$	f_R (MHz)	γ (10^9 s^{-1})
BaM003	123	380	8.3
BaM004	113	470	7.6
BaM005	45	790	5.2

Furthermore, a previous investigation on $LiNbO_3$ ceramics, which revealed a high-frequency dielectric dispersion closer to a true resonance rather than proper relaxation-like behavior, reported on a phenomenological model that considers an equivalent-circuit approach for grain resonance, predicting successfully the dielectric spectrum shown by such $LiNbO_3$ materials [38]. According to this model, which considers a series and parallel branches configuration representing the mechanical damping of vibration and the clamped high-frequency response, respectively, the resonant frequency is predicted and verified to be dependent on the inverse of the grain-size (d), that is to say, $f_R \sim 1/d$. Accordingly, the smaller the grain-size the higher the resonant frequency, as also noted in the present work, where the average grain-size in these PZT–BaM composites decreases with the increase of the BaM content, whereas the characteristic frequency increases. Therefore, the obtained results for the PZT–BaM composites studied here appear to follow well the prediction from the proposed theoretical model [38].

In summary, from both the fundamental and practical viewpoints, it is important to point out that distinguishing between and optimizing relaxation-like and/or resonance contributions to the overall dielectric response of ferroelectric–magnetic composites are very important steps to advance the physical understanding and ability to engineering of such material properties. This is imperative because it is to be realized that the electrical and magnetic properties of such composite bodies can be tuned with the application of an appropriate electric and/or magnetic field. The influence of external factors such as temperature and/or applied mechanical stresses in presence of either a magnetic or electric field is expected to provide additional keys for understanding the microwave dielectric behavior of such materials, keeping in mind their promising integration in high-performance frequency agile materials, components and subsystems that can significantly enhance the performance of radio-frequency (RF) systems. Further studies are in progress in order to closely treat these aspects.

4. Conclusions

The dielectric properties of PZT–BaM multiferroic composites were investigated in a wide frequency range. The ceramic samples were obtained by applying the conventional solid-state reaction method. A low-frequency dielectric dispersion was observed, revealing relatively high dielectric permittivity with stable values in a wide frequency range, as well as very low dielectric losses ($\sim 10^{-3}$), when compared to other typical multiferroic systems. This makes PZT–BaM composites good candidates for specific and practical electro-electronic applications. The high-frequency dielectric results revealed incidence of a resonance-like dielectric dispersion whose characteristics fit well with an over-damped resonance mechanism for the vibration of the boundaries of the ferroelectric polar regions. The strong dependence of the parameters associated to such dielectric dispersion with BaM concentration is indicative that the magnetic phase roughly influences the dynamics of the domain-wall motion of the ferroelectric phase. These insights provide additional features for the use of such composites in high-frequency multiferroic-based devices, where the influence of either magnetic or electric external fields may be considered.

Acknowledgments

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