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Investigation of nonlinear dielectric properties in $Sr_{0.75}Ba_{0.25}Nb_2O_6$ relaxor ferroelectric thin films

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The ac and dc driving fields dependence of the dielectric permittivity for the strontium barium niobate relaxor ferroelectric thin films has been investigated. The nonlinear dielectric properties were obtained by using the measurements of the dielectric permittivity of the material as a function of the ac and dc "bias" electric field amplitude in wide frequency (100 Hz–10 MHz) and temperature (50–450 K) intervals. The results hint the existence of a true mesoscopic dielectric relaxor response in the ferroelectric thin film, which is very similar to those observed in bulk relaxor ferroelectrics. An anomalous behavior of the NL dielectric response was observed when submitted to moderate dc electric fields levels, indicating a crossover from paraelectric to a glasslike behavior on cooling the sample toward the freezing transition. The obtained results were analyzed within the framework of the models proposed in the current literature. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2828143]

I. INTRODUCTION

Over the past decades, intensive studies have been conducted for the understanding of the physical nature of the dielectric response in relaxor ferroelectrics.[1] These efforts are justified due to their several practical applications in the electroelectronic industry.[2] Indeed, such applications include multilayer capacitors, actuators, and field-induced piezoelectric transducers,[3] which often are operated at high field regimes. Thus, it is very important to investigate the physical properties of relaxors at high field levels, where nonlinear (NL) dielectric features become essential. From the fundamental point of view, these investigations are also interesting to improve the understanding of relaxor ferroelectrics, since this research may contribute to the identification of mechanisms, which govern the dielectric nature of relaxors.

In this context, several phenomenological models have been proposed to explain the physical origin of the observed relaxor features.[1,4–7] Among them, the most known models are the spherical random bond-random field[7] (SRB-RF) model and those that consider the vibration of the boundaries of the polar region (PRBV), i.e., the so-called breathing model.[6] Nevertheless, none of them has yet gained a universal acceptance due to some contradictory experimental results. Rather, the basic question of whether the relaxor state in zero electric field is a ferroelectric state broken into nanodomains under a constraint of quenched random electric fields[6] or a glass state similar to one in dipolar glasses with randomly interacting polar nano-regions in the presence of random fields[8] is still unclear. In general, it is accepted that it is not possible to get a unique answer, for the whole temperature interval, because the interactions in relaxor systems may change their nature (i.e., from individual dipoles at high temperatures to domain at low ones). Therefore, the identification of the nature of the dielectric response in relaxors requires additional experimental information, which can be obtained from the measurements of the NL dielectric response. In this way, a systematic study of the nonlinear properties of relaxors, especially in ferroelectric thin films, which have not been reported at the present, can help to increase the understanding of the microscopic mechanism of their dielectric response.

In reason of obtaining the best properties of high potential materials in the form of thin films and the possibility to use electrically controlled integrated structures based devices for high electric fields applications,[9] the strontium barium niobate (SBN) became, therefore, a material of interest with the more diversified purposes, where the NL dielectric properties have been scarcely investigated. The SBN system ($Sr_xBa_{1-x}Nb_2O_6$, or SBN$x/1-x$, for short) constitutes a solid solution for $0.75 < x < 0.25$, with an unfilled tetragonal tungsten bronze (TTB) structure (4*mm* point group) at room temperature.[10] Due to its very good photorefractive effect, SBN family is one of the most promising materials for holographic recording applications.[11] In addition, they also present high dielectric permittivity (with relaxor characteristics) and pyroelectric coefficient and high electro-optic effect, being very important for technological applications, mainly when produced in the thin film form, where thermal

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detection, sensing, and capacitive memory applications for ferroelectric random access memories (FRAMs) from SBN are also feasible.[12]

Despite their very attractive physical properties, to the best of our knowledge, no works focusing the NL dielectric properties in SBN thin films, in sense to understand their intrinsic dielectric properties have been reported in the current literature. In this work, the nonlinear dielectric properties of $Sr_xBa_{1-x}Nb_2O_6$ $(x=0.75)$ ferroelectric thin films were investigated by using the measurements of the dielectric permittivity of the material as a function of the applied ac and dc "bias" driving fields, in wide frequency and temperature ranges. The obtained results were analyzed and discussed within the framework of the models proposed in the current literature for the NL dielectric response in bulk relaxor ferroelectrics.

II. EXPERIMENTAL PROCEDURE

$Sr_xBa_{1-x}Nb_2O_6$ thin films, with $x=0.75$ $(Sr_{0.75}Ba_{0.25}Nb_2O_6$, hereafter labeled as SBN75/25), were prepared by a chemical route based on the preparation and deposition of a polymeric resin, containing the metallic formation ions of the desired SBN phase on $Pt/Ti/SiO_2/Si$ substrates.[13] The final film thickness was around 500 nm. X-ray diffraction patterns of the SBN75/25 thin films were obtained at room temperature, using a RU200B Rigaku x-ray diffractometer. For dielectric investigations, gold electrodes (0.5 mm in diameter) were sputtered on the surface of the films forming a metal-film-metal configuration. The dielectric properties were obtained by measuring the dielectric permittivity, real and imaginary components (ε' and ε'', respectively) in the frequency and temperature ranges of 100 Hz–10 MHz and 50–450 K, respectively, using an HP4194A impedance gain/phase analyzer. Two experiments were performed. First, an ac electric field was applied with variable amplitude E_{ac} (ac field level effect), up to 100 kV/cm. In the second one, a dc bias electric field with variable amplitude E_{dc} (dc field effect), up to 150 kV/cm, was superimposed on the small signal ac field. The nonlinear effect was characterized by the NL component of the dielectric permittivity $\Delta\varepsilon_{nl}$, defined as the difference of the dielectric permittivity measured at a given amplitude $E \neq 0$ and the linear (small signal) permittivity $[\Delta\varepsilon_{nl}=\varepsilon'(E\neq 0)-\varepsilon'_l]$, being $\Delta\varepsilon_{ac}$ and $\Delta\varepsilon_{dc}$ the nonlinear components according to the ac field level and dc bias field effects, respectively.[6]

III. RESULTS AND DISCUSSION

X-ray diffraction patterns (Fig. 1) and energy dispersive x-ray spectroscopy (EDS) analyses revealed a single polycrystalline tetragonal SBN phase and the expected nominal composition (SBN75/25), with any spurious or segregated additional phases. The lattice parameters $[a=12.442(3)$ Å and $c=3.9251(2)$ Å$]$ were similar to those obtained for SBN75/25 single crystals.[14] However, a sudden decrease in the *a* parameter was observed when compared to that obtained for bulk SBN with similar composition.[15] This structural characteristic hints the existence of the in-plane tensile strain on the thin film surface,[16] whose influence on the dielectric response of the SBN75/25 thin film will be discussed later.

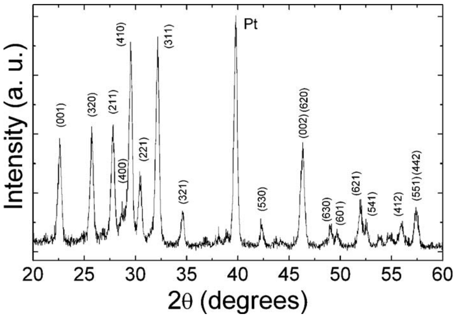

FIG. 1. X-ray diffraction patterns for the SBN75/25 thin film.

Figure 2 shows the temperature dependence of the linear (small signal) dielectric permittivity (real ε' and imaginary ε'' components) measured at different frequencies. Analogous to other classical relaxor ferroelectrics, i.e., lead magnesium niobate (PMN), the analyzed SBN75/25 ferroelectric thin film exhibits a pronounced dispersion in the complex dielectric permittivity with a very broad peak around the temperature of the maximum real dielectric permittivity $(T_m \sim 280$ K). As observed, the maximum real dielectric permittivity (ε'_m) decreases, while the maximum imaginary dielectric permittivity increases with the increasing frequency. On the other hand, the temperature of the maximum dielectric permittivity, in both cases (real and imaginary components), increases with increasing frequency. It can also be observed that the maximum of the imaginary component of the dielectric permittivity is reached at a temperature lower than that obtained for the maximum real dielectric permittivity (for a fixed frequency). As aforementioned, these behaviors are characteristics of relaxor ferroelectrics with diffuse phase transition.[1] The origin of the relaxor behavior has been studied for 40 years, but it is only in the past decade that microscopic mechanisms, with solid experimental evidence to support them, were proposed.[5–7] The relaxors do not undergo a macroscopic phase transition from the paraelectric phase into a ferroelectric state one, but they become rather "frustrated" ferroelectrics. They are complex materials where the origin of the "frustration" has been associated with compositional inhomogeneity on a nanometer scale that results from partial compositional disorder in a specific lattice site.

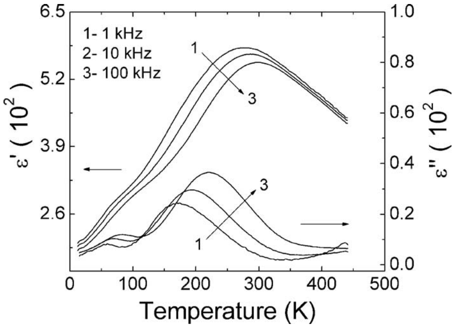

FIG. 2. Temperature dependence of the small signal (low ac electric field) complex dielectric permittivity (real ε' and imaginary ε'' components), for the SBN75/25 ferroelectric thin film.

This disorder prevents the macroscopic transformation into ferroelectric phase. A picture coherent with the experimental results is the presence of a paraelectric matrix, in which ferroelectric nanosized regions are formed upon cooling, without the evolution of a coherent macroscopic ferroelectric phase.

It is important to point out that a close inspection of Fig. 2 shows that the ε' values are about four times smaller than those reported for polycrystalline SBN75/25 (Ref. 15). Moreover, T_m (~280 K at 1 kHz) is about 40 K smaller than that reported for SBN75/25 ceramics, while the phase transition diffuseness is increased. These peculiar characteristics observed for the SBN thin films can be associated either with the grain size distribution[17] of the films or to different and/or combined extrinsic factors,[18] which, in fact, affect the dielectric response of the thin films, related to in-plane mechanical tensile, as previously reported.[17,18] Indeed, based on the results obtained for $BaTiO_3$ ceramics with different particle sizes of nanometric scales,[19] it has been reported that the suppression of the dielectric permittivity, as well as the decrease of T_m in the dielectric response of the thin films, when compared to that obtained for bulk materials, can be a consequence of the observed smaller grain size distribution of the thin films (30–50 nm).[20] Nevertheless, some works have also focused the influence of the interfaces effects.[18] In this way, it has been shown that the existence of a mechanical tensile at the film-substrate interface,[18] caused by the different thermal expansion coefficients between the film and the substrate, could also lead to an increase of the phase transition diffuseness, as well as a decrease of both ε' and T_m values for the thin films,[21] when compared to the obtained results for ceramics and single crystals.

On the other hand, as can be seen in Fig. 2, a dielectric dispersion is observed in the whole temperature interval, which depends itself on measurement frequency, showing a more intense contribution to the dispersion for temperatures around and below T_m. The slight dispersive behavior of the dielectric permittivity observed for temperatures higher than T_m could be associated with a small contribution of additional effects, which can be related to nonactive thin-film interface layers.[22] In fact, it has been shown that the relaxorlike dielectric behavior of thin film based heterostructures, mainly for temperature above T_m, can be related to the interface layer, rather than the true dielectric response of the films.[22] Thus, the real dielectric response of the thin films can be masked by the effect of the nonactive layers at the interface between the films and the substrate, which can be seen as parasitic capacitances acting in series with the inherent "bulklike" capacitance of the film.[22] This proposed "series-capacitor" model can be universally observed in ferroelectric thin film research. However, in making a correction for the parasitic capacitance, previous studies have assumed no temperature dependence of such parasitic components.

It is important to point out that although the dielectric response shows a weak dielectric dispersion above T_m, which can be associated with the contribution of parasitic capacitances as a result of a possible film-substrate interface layer, the observed dielectric dispersion for the SBN75/25 thin film above T_m is remarkably lower than the observed dispersion for some consecrated thin films obtained by other chemical and physical processing methods, which confirms the good quality and adherence of the obtained SBN75/25 thin film, when compared to those reported in the literature. In fact, such a dielectric dispersion has been always observed for temperatures above T_m. That is why, several authors have pointed out that the dielectric response of thin films can be always masked by the influence of the parasitic capacitance. In this way, such models predict a decrease of the dielectric permittivity but they do not predict shifts in the temperature of the maximum dielectric permittivity of the thin films, when compared to ceramics and single crystals. So, the obtained results for the studied SBN75/25 thin film indicate that if there exists some component of the parasitic capacitance, its contribution to the obtained dielectric response is weak, because of the weak dielectric dispersion obtained for temperatures above T_m in the present work. Therefore, it can be pointed out that the observed characteristics for the SNB75/25 thin films near the transition temperature, such as the decrease of ε' and its corresponding maximum temperature (T_m) and the increase of the phase transition diffuseness, when compared to the ceramics and single crystals results, can be associated with the small grain size distribution of the film (~50 nm),[23] as well as to an additional little component of a mechanic tensile at the film-substrate interface.

A. ac nonlinear dielectric response

One of the most widespread methods employed to investigate the dielectric properties of ferroelectric materials at high electric field regimes is the investigation of the ac (with variable amplitude) and the dc bias (superimposed to the small signal ac electric field) electric field dependence of the dielectric permittivity. In this way, in order to investigate more closely the influence of an alternate electric field, specifically on the paraelectric-ferroelectric phase transition of the SBN75/25 thin films, measurements were firstly performed up to moderate ac electric field amplitudes around the temperature of the maximum dielectric permittivity. The influence of the applied alternate electric field of variable amplitude (E_{ac}) on the temperature dependence of the dielectric response was investigated in wide temperature and frequency ranges.

Figure 3 shows the temperature dependence of the linear [Figs. 3(a)–3(c)] and nonlinear dielectric permittivities [Figs. 3(d)–3(f)], as a function of the amplitude of the applied ac electric field (E_{ac}) at the frequencies of 1, 10, and 100 kHz, for the SBN75/25 thin films. As can be observed in the Figs. 3(a)–3(c), with respect to the effect of the amplitude of the ac electric field, the linear dielectric permittivity shows a similar behavior to that obtained for ceramics and single crystal relaxor ferroelectrics.[24] That is to say, the increase of the amplitude of the ac driving field results in a change of the dielectric permittivity similar to that produced by lowering the frequency, i.e., ε' becomes larger in magnitude and ε'_m shifts to lower temperatures. On the other hand, Figs. 3(d)–3(f) (for the temperature dependence of the nonlinear dielectric permittivity, at several E_{ac} amplitudes) clearly

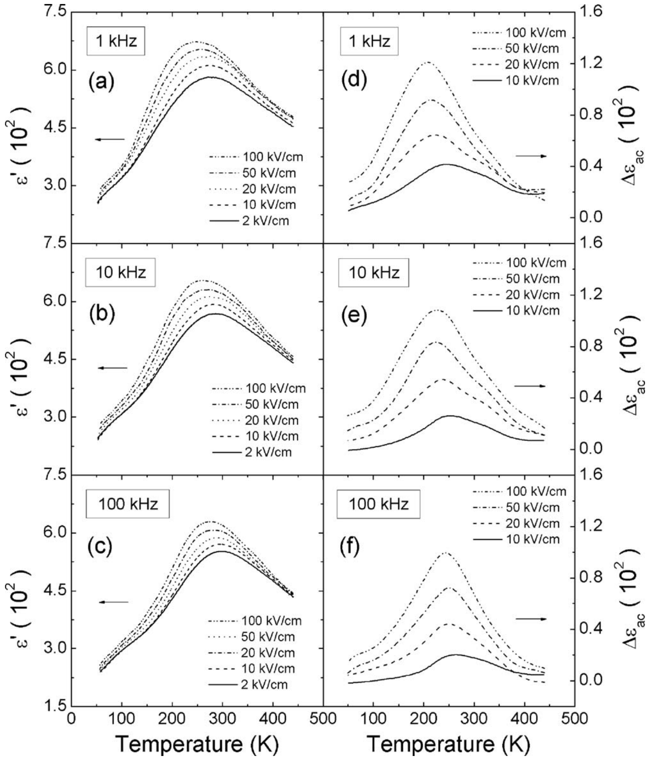

FIG. 3. Temperature dependence of the linear [(a)–(c)] and nonlinear dielectric permittivities [(d)–(f)], as a function of the amplitude of the applied ac electric field (E_{ac}), for three selected frequencies.

show that $\Delta\varepsilon_{ac}$ is always positive and passes through a maximum, which lies somewhat below T_m, the temperature of the maximum small signal dielectric permittivity (for 1 kHz, $T_m \sim 280$ K).

Figure 4 shows the temperature dependence of $\Delta\varepsilon_{ac}$ for SBN75/25 thin films, obtained at various frequencies and for the maximum amplitude of the applied ac electric field (100 kV/cm). As can be seen, the relaxor behavior prevails even when applied high levels of the ac electric fields of moderated magnitude. Results obtained in Fig. 4 clearly demonstrate that, like the linear dielectric permittivity (Fig. 2), the NL component have two regimes: A quasistatic (at high temperatures) and a frequency dispersive (at low temperatures), and for a given frequency of the applied field, the transition from the quasistatic to the dispersive regime occurs simultaneously in the linear ε' and nonlinear $\Delta\varepsilon_{ac}$ dielectric responses.

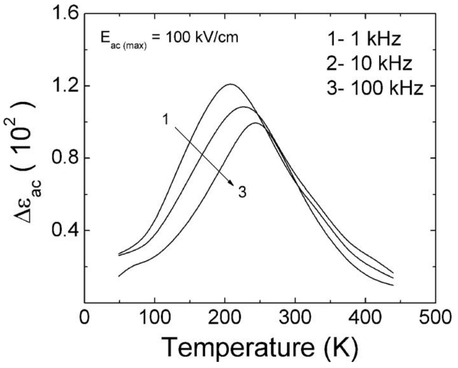

FIG. 4. Temperature dependence of the nonlinear dielectric permittivity ($\Delta\varepsilon_{ac}$) as a function of the frequency, at the maximum applied ac electric field, for the SBN75/25 thin film.

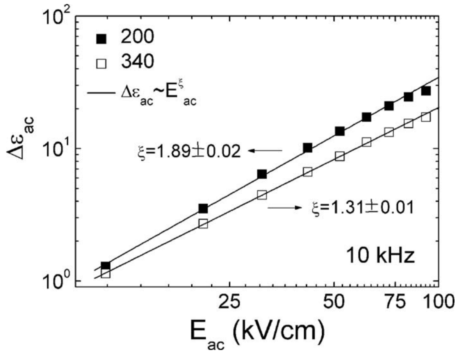

FIG. 5. Nonlinear component of the dielectric permittivity for the SBN75/25 thin films as a function of the amplitude of the ac electric field, measured at 10 kHz.

Figure 5 shows the ac electric field dependence of the nonlinear dielectric permittivity for two selected temperatures (200 and 340 K), measured at 10 kHz. As can be seen, when plotted on log-log scale, the data lie well on a straight line, which shows that the obtained results slightly deviate from a quadratic dependence between the ac electric field and $\Delta\varepsilon_{ac}$. This suggests a power-type dependence with a function $\Delta\varepsilon_{ac} \sim E_{ac}^{\xi}$. In the plot, the solid lines correspond to the fit of the power function using ξ as the adjustment parameter. Difference between the field exponents (ξ) at low and high temperatures can be clearly observed, as represented by the slopes of the lines. Therefore, results indicate that for the whole temperature range, the temperature dependence of ξ seems to obey a steplike change. At low temperatures ($T=200$ K) $\xi=1.89$, while at high temperatures ($T=340$ K) $\xi=1.31$. These obtained values are in agreement with the reported values for some other relaxor ferroelectrics,[6] which leads us to think that the observed nonlinearity in the SBN75/25 thin film corresponds with a true mesoscopic relaxor response.

The anomalous behavior obtained for the field exponent might be satisfactorily interpreted in the framework of the so-called "breathing" model,[25] where a crossover of the dynamics of polar regions boundaries is expected, and it may be responsible for the experimental results obtained for the field exponent of the NL dielectric response. This model supposes that the motion of the interphase boundary between the polar regions and the nonpolar matrix looks like breathing of the polar regions under the applied ac electric field. In this way, the ac electric field induced displacements of the boundaries of the polar regions (rather than a reorientation of the regions) control the dielectric response. According to the "breathing" model, the ergodic phase (at high temperatures) is treated as a system of polar regions embedded in a nonpolar matrix. They are elongated along the direction of the local spontaneous polarization (P_s), and the orientation does not change under the thermal agitation. The polar region pattern is determined by the spatial distribution of internal random fields, which are induced by the charge disorder and chemical inhomogeneity and can act as pinning centers.

However, due to the random distribution of the random fields, the interphase boundary of a polar region and a nonpolar matrix will not stay flat. Indeed, it will become "rough" with a short scale characteristic length (L_c), which is determined by the spatial distribution of the random fields and the elastic characteristics of the interphase boundary. If the short scale characteristic length is smaller than the total length of the polar region, the boundary will not be able to move as a whole.

Therefore, the observed crossover in the temperature dependence of the field exponent can be related to the dynamics of the interphase boundary motion and can be interpreted according to the scenario described as follows.

At low temperatures, where the field exponent is close to the classical value (quadratic), the dielectric response is controlled by the field-induced vibration of the polar region boundaries on a scale smaller than the size of the polar regions. On the other hand, at high temperatures, in the quasistatic regime, for which ξ is lower than the classical value, the dielectric response is controlled by the field-induced breathing of the polar region as a whole. This model can also explain the onset of the frequency dispersion of the real and imaginary components of the linear dielectric permittivity.

B. dc nonlinear dielectric response

Using the same samples, the effect of the dc bias electric field on the dielectric response was also investigated in the same temperature and frequency intervals. The influence of a dc bias electric field (E_{dc}), superimposed to the small signal ac electric field, on the temperature and frequency dependencies of the dielectric permittivity is shown in Fig. 6. As can be seen, the obtained results for the dc bias electric field effect showed a different behavior of ε' (near the paraelectric-ferroelectric phase transition), when compared to that observed in Figs. 3(a)–3(c) for the ac electric field level effect. That is to say, for a fixed frequency, the maximum real dielectric permittivity (ε'_m) decreases with increasing dc bias field amplitude, contrary to the observed behavior for ac electric fields nonlinear dielectric measurements, where ε'_m increases with increasing ac electric field level.

Figure 7 shows the temperature dependence of the nonlinear dielectric permittivity ($\Delta\varepsilon_{\text{dc}}$) for the SBN75/25 thin films, as a function of the frequency [Figs. 7(a)–7(c)] and dc bias electric field [Figs. 7(d)–7(f)]. As can be observed in the Figs. 7(a)–7(c), the temperature dependence of $\Delta\varepsilon_{\text{dc}}$ also demonstrates that the NL dielectric permittivity, obtained by the dc bias electric field effect, has the two above mentioned regimes, the quasistatic (at high temperatures) and a frequency dispersive observed at low temperatures,[25] similar to the obtained results for the linear dielectric permittivity (Fig. 2). On the other hand, for a fixed dc bias electric field amplitude, the maximum nonlinear dielectric permittivity becomes smaller in magnitude with the increase of the frequency, while its corresponding the temperature moves toward high temperature values. That is to say, the relaxor behavior observed in Fig. 2 (frequency dispersion near T_m) prevails even when applied dc bias electric fields of moderated magnitudes.

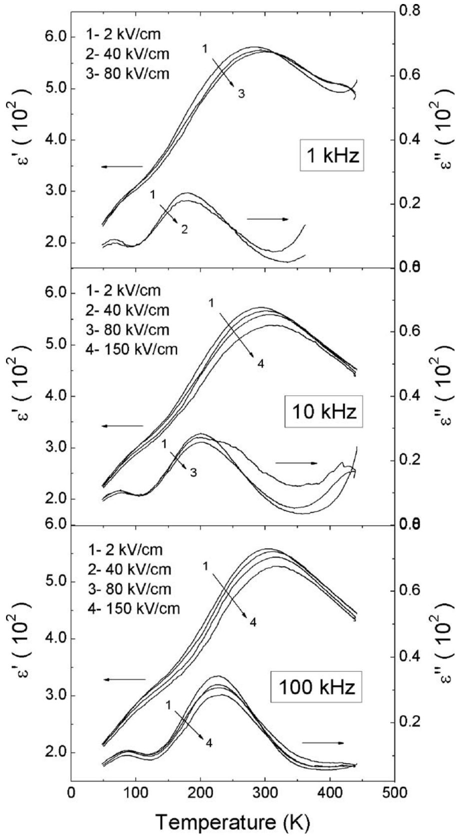

FIG. 6. Real and imaginary components of dielectric permittivity as a function of temperature for the SBN75/25 thin film at various dc "bias" electric fields, for the frequencies of 1, 10, and 100 kHz.

As observed from the obtained results, a more general description of the NL dielectric response in relaxor ferroelectrics can be carried out not only by studying the influence of an ac electric field of variable amplitude but also through the investigation of the dielectric response applying a dc bias electric field, superimposed to the small signal ac electric field. Indeed, if only small ac field is applied, the dielectric response could be isotropic because the polar regions are randomly oriented in the volume of the grain. Even at larger ac field amplitudes, the dielectric response might be isotropic because the ac electric field does not change the orientation of the local polarization vector; in fact, it only releases the interphase boundaries from the pinning centers. When consider the effect of the dc bias electric field, the external field changes the profile of the random fields and, therefore, changes the position of the interphase boundaries. On the other hand, the dc bias electric field also forces the polar regions to reorient in the direction parallel to the applied field and provides a coalescence of neighboring regions, resulting in the diminishing in the total area of the boundaries. Therefore, if both dc bias and small signal ac electric fields are simultaneously applied, the dielectric permittivity will be suppressed by the dc bias electric field and more detailed information about the nature of the relaxor behavior in ferroelectrics can be achieved.

Comparison between the plots of the Figs. 3(d)–3(f) and Figs. 7(d)–7(f) shows strong qualitative differences between both ac and dc nonlinear effects. Although the absolute val-

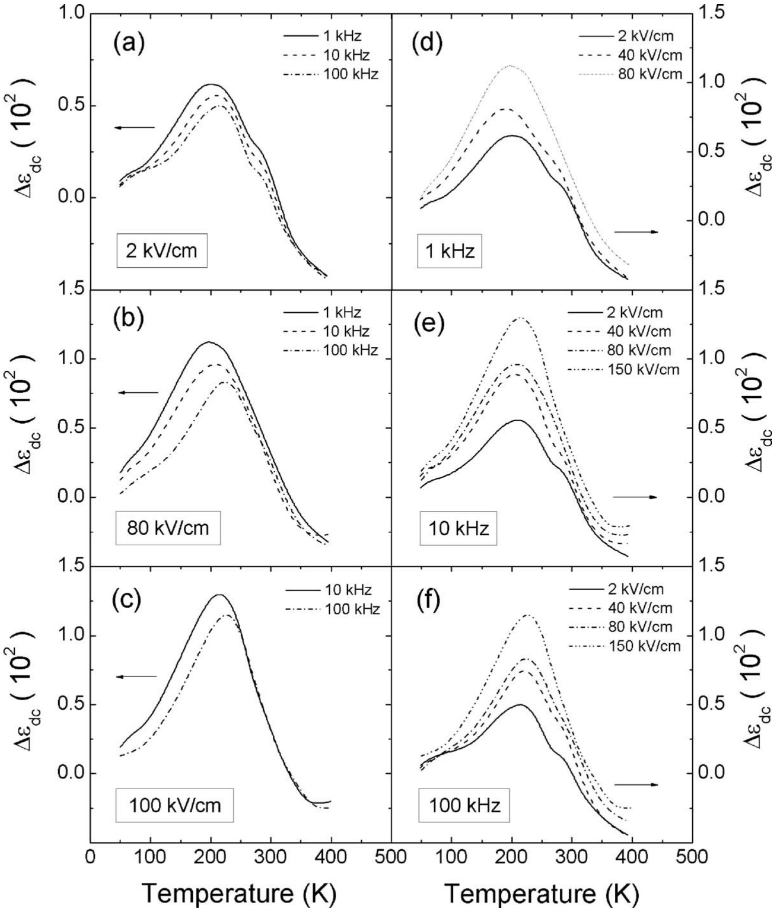

FIG. 7. Temperature dependence of the nonlinear component of the dielectric permittivity, $\Delta\varepsilon_{dc}$, for the SBN75/25 thin film, as a function of the frequency [(a)–(c)] and the dc "bias" electric field [(d)–(f)].

ues of $\Delta\varepsilon_{ac}$ and $\Delta\varepsilon_{dc}$ are in the same order of magnitude, the obtained results clearly show a sharp peak of $\Delta\varepsilon_{dc}$ at a temperature below T_m, being intensified for higher dc bias electric field amplitudes. However, as observed in Figs. 7(d)–7(f), for moderate dc bias field levels (up to E_{dc} $=40$ kV/cm), an anomalous behavior was observed at higher temperatures close to 280 K (the temperature of the maximum dielectric permittivity), which in turn disappears with the increase of the dc bias electric field magnitude. These behaviors suggest the coexistence of a glass state concomitant to a ferroelectric state, as proposed by the SRB-RF model,[7] and it may be correlated with the fact that the linear dielectric response in relaxor ferroelectric materials can be associated with existence of the random fields.[5]

The SRB-RF model can be regarded as the simplest generic semimicroscopic model of relaxor ferroelectrics where random interactions (or bonds) between polar clusters are assumed to be infinitely ranged with a Gaussian distribution, and the local random fields are similarly Gaussian and uncorrelated, so that this leads to a mean-field-type theory analogous to the case of spin[26] and dipolar glasses.[4] Thus, the model predicts the existence of the phases, namely, the spherical glass (SG) phase without long-range order and the long-range ordered polarized ferroelectric (FE) phase. By including a dc bias electric field into the model, it is possible to describe the transition from the relaxor to an inhomogeneous ferroelectric state for electric fields exceeding a critical value E_c (namely, the coercive field).[27] This transition is often referred in the literature as the field induced micro—to macrodomain or induced ferroelectric transition.[28]

The SRB-RF model predicts that the nonlinear dielectric permittivity ($\Delta\varepsilon_{dc}$) shows a sharp peak around a temperature T_f, which corresponds to the freezing temperature of the spherical spin-glass model. The mechanism of nonlinear response originally proposed within the SRB-RF model has been derived under the assumption that polar clusters are embedded in a rigid lattice; the nonlinearity effects are thus entirely due to the intrinsic nonlinearity of the rigid SRB-RF model. In fact, it qualitatively also describes a crossover behavior of the nonlinear dielectric component at a critical temperature, which is characterized by the decreasing to the increasing temperature dependence of $\Delta\varepsilon_{dc}$, when approaching the freezing temperature from above. Thus, it can be observed experimentally an anomalous behavior of $\Delta\varepsilon_{dc}$ on the high-temperature side of the peak, where the frequency dispersion is weak. This has, indeed, been found in PMN single crystal and PLZT ceramics,[29] supporting the assumption that these systems can be described as a special kind of dipolar glass, namely, the spherical glass. Namely, the scaling theory of the second order phase transition predicts that $\Delta\varepsilon_{dc}$ should vanish at the ferroelectric transition and diverges at the freezing transition in dipolar glasses, as indeed observed in the experimental results.[30]

Thus, according to the obtained results at the present work, for the SBN75/25 thin films when applying a dc bias electric field, and taking into account the above presented discussion, the observed anomalies for lower magnitudes of the applied dc electric field in the temperature dependence of $\Delta\varepsilon_{dc}$ (Fig. 7) can be a true experimental evidence of the existence of both the above mentioned states. The nonlinear dielectric permittivity showed a maximum at a temperature below the T_m, which has been related to the freezing temperature, as well as an abnormal behavior at a critical temperature near the temperature of the maximum dielectric permittivity ($\sim$280 K), as predicted by the SRB-RF model. In fact, as previously pointed out, at the critical temperature the SRB-RF model predicts a crossover from a paraelectriclike behavior at high temperatures to a dipolar glasslike behavior on approaching T_f for the temperature dependence of $\Delta\varepsilon_{\mathrm{dc}}$, showing a near zero (or a constant) value for $\Delta\varepsilon_{\mathrm{dc}}$. However, it is important to point out that the observed anomalies around 280 K for the SBN75/25 thin film, for lower dc electric field amplitudes [Fig. 7(a)], show nonzero values of $\Delta\varepsilon_{\mathrm{dc}}$ for all the analyzed frequencies, which may be associated with the influence of an extrinsic factor inherent to the thin film interface. As previously pointed out, this factor shows to be, in fact, more relevant for the dielectric response of the thin films than for the ceramics and single crystal. In particular, this behavior can be related to the weak dielectric dispersion observed for the zero field dielectric response in this temperature region, caused by possible parasitic capacitances provided by a nonactive thin film/substrate interface layer.

As can be seen, with the increase of the dc bias field' magnitude [Figs. 7(b) and 7(c)], the glass state seems to overlap to the ferroelectric one, since the critical temperature disappears and the dielectric dispersion is defined, which can be observed as well in Fig. 7(c). For this specific case, a typical Curie-Weiss behavior can be predicted for temperatures above the T_m. This result constitutes, therefore, a strong evidence of a glass state formation, with a short-range order on cooling the SBN75/25 thin films from the high temperature paraelectric phase. With a continuous increase of the dc bias electric field amplitude, for values near the coercive field, the SBN goes to a stable ferroelectric state. So that, it can be concluded that the ferroelectric state shows to be a metastable state, which can be only reached when applying strong dc electric fields and, at the same time, the glass state, related to clusters and ferroelectric domain response, prevails in this material.

IV. CONCLUSION

In summary, the nonlinear dielectric properties for SBN75/25 relaxor ferroelectric thin films were carefully determined as a function of the magnitude of ac and dc driving fields, in wide temperature and frequency intervals. Studying the influence of the ac and dc bias electric fields on the dielectric properties, it was observed that, like in the linear dielectric permittivity (small signal), the NL component has two regimes, a quasistatic and a frequency dispersive one, at high and low temperature regions, respectively, similar to those observed in bulk relaxor ferroelectrics. It was also observed that, for a given frequency, the transition from the quasistatic to the dispersive regime occurs simultaneously in the linear, ε', and nonlinear components, $\Delta\varepsilon_{\mathrm{ac}}$ and $\Delta\varepsilon_{\mathrm{dc}}$, of dielectric responses. On the other hand, it was found that the relaxor behavior may be related to the motion of the interphase boundaries of the polar regions (in correspondence with the "breathing" model), and at the same time, it seems to be hardly influenced by the coexistence of a glass state concomitant to the ferroelectric state in SBN75/25 relaxor thin films, in agreement with previous results reported in the literature. In fact, these states are intimately governed by the nonlinear measurement regime, whereas with the increase of the E_{dc} amplitude, the glass state seems to be overlapped with the ferroelectric one. Nevertheless, it was observed that the dielectric properties of the thin films may be governed by extrinsic factors (inherent to the films), related to interfacial effects, which may mask the real dielectric response of the thin films. These factors, indeed, showed to be more relevant in films than for ceramics and single crystals. Therefore, a special attention should be taken into account with respect to the processing methods for obtaining the films, in order to minimize those extrinsic factors.

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