# Dielectric and optical properties of transparent ceramics $K_{0.5}Na_{0.5}NbO_3 - 0.02\,Ba_2NaNb_5O_{15}$

© L.S. Kamzina

Ioffe Institute,
St. Petersburg, Russia

E-mail: ASKam@mail.ioffe.ru

The relationship between depolarization temperature $T_d$ and polymorphic phase transition temperature $T_{F-R}$ in poorly studied relaxor hot-pressed ceramics $(K_{0.5}Na_{0.5}) \cdot NbO_3 - 0.02Ba_2NaNb_5O_{15}$ was studied. Dielectric and optical properties of polarized samples were measured for this purpose. It was found that the temperatures $T_d$ and $T_{F-R}$ do not match. The obtained results are discussed in terms of the degree of the phase transition diffuseness and the size of the polar regions. It is suggested that the relative position of the depolarization temperature and the temperature of transition of the polarized sample to the relaxor phase $T_{F-R}$ does not depend on the type of phase boundary (polymorphic or morphotropic), but is related only to the size of the polar regions.

**Keywords:** lead-free transparent ceramics, polymorphic phase transition, relaxors.

## 1. Introduction

Ferroelectric lead-containing materials with a perovskite structure, such as lead zirconate titanate $Pb(Zr_xTi_{1-x})O_3$ (PZT) [1], lead magnoniobate $Pb(Mg,Nb)O_3$ (PMN) [2] and solid solutions based on them feature excellent piezoelectric properties, but their use in appliances causes serious environmental and health problems because of their toxicity. Lead-free ceramics based on $(KNa)NbO_3$, which is a mixture of solid solutions of $KNbO_3$ and $NaNbO_3$, is one of the most studied ferroelectric systems with a perovskite type structure [3]. $(KNa)NbO_3$ has the best dielectric and piezoelectric properties in the area of composition $(K_{0.5}Na_{0.5})NbO_3$ (KNN). The following transitions are observed in KNN with an increase of temperature: transition from rhombohedral (R) to orthorhombic phase (O) at $T_{R-O} \approx 153\,K$, transition from orthorhombic to tetragonal phase (T) at $T_{O-T} \approx 473\,K$ and transition from tetragonal to cubic phase (C) $T_C \approx 673\,K$ (Curie temperature $T_C$) [4,5].

The authors in numerous early works associate the best piezoelectric properties of the KNN composition, compared with other $(KNa)NbO_3$ compositions, with the presence of a morphotropic phase boundary (MPB) between the orthorhombic and tetragonal phases [6]. Although the presence of this MPB still raises controversy in the literature [7,8], the possibility of improvement of the piezoelectric properties of KNN by introducing impurities is an indisputable fact [9,10].

The disadvantages of this composition are the relatively small value of the piezoelectric constant (pure ceramics $d_{33} \approx 80\,pC/N$), and the mechanical quality factor $Q_m$ is also too low for a number of industrial applications (below 300). These disadvantages can be easily eliminated by doping or

by small additives of another component ($d_{33}$ can be significantly increased to $d_{33} \approx 416\,pC/N$ and higher [11,12]). The temperatures $T_{O-T}$ and $T_C$ decrease with the introduction of additives, and $T_{R-O}$ increases and shifts to room temperature with an increase of the additive content.

The authors of [13–15] associate a significant increase of the piezoelectric constant with the introduction of additives with the formation of a multiphase structure (the coexistence of many phases (O−T, R−O−T or R−T) instead of the pure O-phase in KNN. A complex domain structure is observed in the multiphase system, including micron domains ($200\,nm-5\,\mu m$), nanodomains ($1-200\,nm$) and polar nanoregions (PNR) ($1-10\,nm$), which plays a crucial role in improving the piezo properties of compositions at MPB. Moreover, the coexistence of many phases results in the destruction of the long-range order of the ferroelectric phase, prevents the formation of long-range order domains and induces the formation of nanodomains and even PNR [15]. Such a variety of domain structures with regions of different sizes is one of the reasons for the relaxor properties of solid solutions based on KNN.

The authors in [14] examining ceramic solid solutions of KNN with $SrZrO_3$ and $Bi_{0.5}Ag_{0.5}ZrO_3$ by an electron microscope found the PNRs in them embedded in a matrix of ordered ferroelectric domains with long-range order. Compositional inhomogeneities and defects are the main causes of phase coexistence [16].

It should be noted that the causes of the relaxor properties of solid solutions based on KNN differ from classical relaxors.

Two types of PNRs are usually observed.

PNRs of type I are embedded in a nonpolar matrix, and the relaxor behavior takes place in case of

paraelectric−ferroelectric phase transition. type I is widely observed in PMN and PZT.

PNRs of type II are embedded in a polar matrix, which, in addition to them, demonstrates an abundance of ordered ferroelectric domains with long-range order. The relaxor behavior of type II manifests itself both in case of ferroelectric−ferroelectric phase transitions, which occurs at sufficiently high temperatures, and in case of paraelectric−ferroelectric phase transitions. Type II is usually observed in relaxor-ferroelectric solid solutions such as $PbMg_{1/3}Nb_{2/3}O_3−PbTiO_3$ (PMN−PT) and $PbZn_{1/3}Nb_{2/3}O_3−PbTiO_3$ (PZN−PT).

A variety of phases is observed already at room temperature in solid solutions based on KNN ceramics and the coexistence of long-range order domains with nanoregions and PNR. Thus, the relaxor behavior of ceramics occurs already during the transition between the ferroelectric-ferroelectric phase ($T_{R−O}$) at room temperature, which differs from the relaxor behavior of the above two types. Nanodomains in ceramic solid solutions based on KNN are induced both due to the coexistence of many phases and because of the destruction of the long-range order. This differs from PZT ceramics, in which nanodomains are formed only due to the coexistence of R−T-phases.

Nanodomains have a size of 20−200 nm in non-polarized ceramics based on KNN and a complex configuration in the form of bands, „herringbone patterns", watermarks, etc. [14,15,17]. The configuration of nanodomains is significantly simplified after polarization. The main part of the nanodomains has the form of bands of size of 20−100 nm. The simplification of the nanodomain configuration is caused by the disappearance of the domain wall 180° and the switching of domains other than 180° [17,18]. The nanodomains gradually shrank with an increase of temperature, there was a gradual depolarization of the sample and a transition to the relaxor phase.

However, the choice of additives should be approached with caution. For instance, some additives can cause a rapid decrease of the Curie temperature due to a discrepancy in valences and a nonlinear dependence of the Curie temperature on the component composition [19].

Some of the best components are ferroelectric compounds with a tungsten bronze structure, for example, $Ba_2NaNb_5O_{15}$ (BNN). They have excellent electro-optical and nonlinear optical properties. A small addition of this compound can significantly improve the piezoelectric properties of KNN. The authors of [20,21] associate the shift of the temperature of transition from the orthorhombic to the tetragonal phase towards room temperature with the polymorphic phase transition (PPT). This transition, like MPT, presupposes the coexistence of phases. The phase boundary in case of PPT between the tetragonal and orthorhombic phases (PPB) is not vertical (as, for example, MPB), but reveals a strong temperature dependence. When heated above the temperature $T_{F−R}$ (temperature $T_{O−T}$ for an unpolarized sample) the polarized sample undergoes

transition from the ferroelectric orthorhombic phase to the relaxor phase ($T_{F−R}$).

Information about the structure of the phase at room temperature in ceramics KNN−$x$BNN is extremely contradictory in the literature. It was shown in [22] that ceramics KNN−$x$BNN containing $x \leq 0.025$ is pure perovskite at room temperature with orthorhombic symmetry, and the coexistence of orthorhombic and rhombohedral phases is observed only in compositions $0.05 \leq x \leq 0.075$. At the same time, the authors of [23], studying the X-ray properties of ceramics KNN−$x$BNN with a lower composition than $x = 0.025$ ($x = 0.013$ and 0.015) came to the conclusion that a mixture of orthorhombic and rhombohedral phases is observed at room temperature even in these compositions, and the transition temperature $T_{RisO}$ for the composition KNN−0.015BNN $\approx 243$ K. In later papers [13−15], the authors, using a variety of more sensitive research methods, came to the conclusion that the structure of ceramic compounds KNN−$x$BNN with PPT is more complex at a room temperature and constitutes a mixture of phases R−O−T.

Unfortunately, there are very few papers devoted to solid solutions of KNN−$x$BNN in the literature [22,23]. Only elastic and piezoelectric properties were mainly discussed in these papers. There is absolutely no information about the relationship between the depolarization temperature of pre-polarized samples of $T_d$ and the temperature of transition from ferroelectric to relaxor phase $T_{F−R}$ in samples with polymorphic phase transition. As is known from the literature and our work, these temperatures may coincide with each other in a number of relaxors, such as, for example, PZT, PMN [24], and may differ, for example, in such relaxors as PZN, solid solutions of PZN−PT, $Na_{1/2}Bi_{1/2}TiO_3$ (NBT) and NBT solid solutions with $BaTiO_3$ (NBT−BT) [25−30].

There is no single point of view on the observed difference of temperatures $T_d$ and $T_{F−R}$ in a number of relaxors. For instance, the authors of [26] associate the difference in temperatures in solid solutions of NBT−6BT with the fact that depolarization in them is a two-stage process. At the first stage, the heating of the polarized sample above $T_d$ destroys only macroscopic polarization, but the relationship of local dipoles within the domains is not lost. In the second stage, the domains disintegrate into polar nanoregions (PNR) when the material is heated to a temperature of $T_{F−R}$ and higher. The authors of [29], studying ceramic samples of $Pb_{0.99}[Zr_{0.45}Ti_{0.47}(Ni_{0.33}Sb_{0.67})_{0.08}]O_3$, explain the loss of polarization at lower temperatures $T_d$ than the long-range order decay at temperature $T_{F−R}$, by the existence of depolarizing fields and stresses caused by mismatch of deformation at grain boundaries due to different crystallographic grain orientations and distortion of structures. In our work [28], studying single-crystal solid solutions of PMN−$x$PT, PZN−$x$PT, NBT−$x$BT, we assumed that the coincidence or difference of temperatures is related to the degree of diffuseness of the phase transition and PNR sizes. These temperatures coincide in relaxors with the highest degree of phase transition diffuseness and

small PNR sizes. Not only macroscopic polarization is destroyed in such compositions at a temperature of $T_d$ when a polarized sample is heated, but the interconnection of local dipoles within the domains is also lost due to the small PNRs formed. These temperatures do not coincide in relaxors with a lower degree of phase transition diffuseness and large PNR sizes. The information available in the literature regarding the relative positions of temperatures $T_d$ and $T_{F-R}$ in various relaxors was provided only for compounds with morphotropic phase transition (MPT).

There is absolutely no information about the behavior of these temperatures in compounds with polymorphic phase transition, which include ceramics KNN$-x$BNN. In this regard, it is interesting to study the relationship between temperatures $T_d$ and $T_{F-R}$ in poorly studied relaxor ceramics KNN$-x$BNN. The transparent ceramics KNN$-0.02$BNN will be examined for this purpose, the dielectric and optical properties of polarized samples will be studied. Optical research methods are a good addition to dielectric methods, since they are more sensitive, especially for studying the changes of sizes of inhomogeneities in case of phase transitions.

## 2. Examined samples and experimental procedure

Transparent ceramics KNN$-0.02$BNN were prepared from ordinary materials. The hot pressing method was used. The input materials were pressed into tablets, which were then sintered in an oxygen atmosphere. The sintered tablets were subjected to hot pressing under pressure. The tablets were polished to a thickness of 0.5mm for optical measurements. A He$-$Ne laser was used for measurements. Dielectric measurements in the absence of an electric field and with the application of an electric field were carried out at a frequency of 1 kHz in the temperature range 298$-$650 K. The sample was first heated without a field to temperatures exceeding the temperature $T_{\max e}$ by $\sim 50$ K and was held at this temperature for 10$-$15 min to eliminate the memory effects in the samples associated with the application of an electric field. The samples were cooled without a field to room temperature after annealing.

## 3. Experimental results and discussion thereof

Figure 1 (curves *1* and *2*) shows the temperature dependences of the dielectric permittivity $\varepsilon$ for transparent ceramics KNN$-0.02$BNN, taken in case of heating in the absence of an electric field (curve *1*) and after application of the electric field of 10 kV/cm at a temperature of 383 K for 1 h (curve *2*).

Two phase transitions are clearly visible on both curves: one transition at temperature of $\sim 493$ K from orthorhombic to tetragonal phase (polymorphic transition $T_{O-T}$), the other transition at Curie temperature of $\sim 623$ K. In addition,
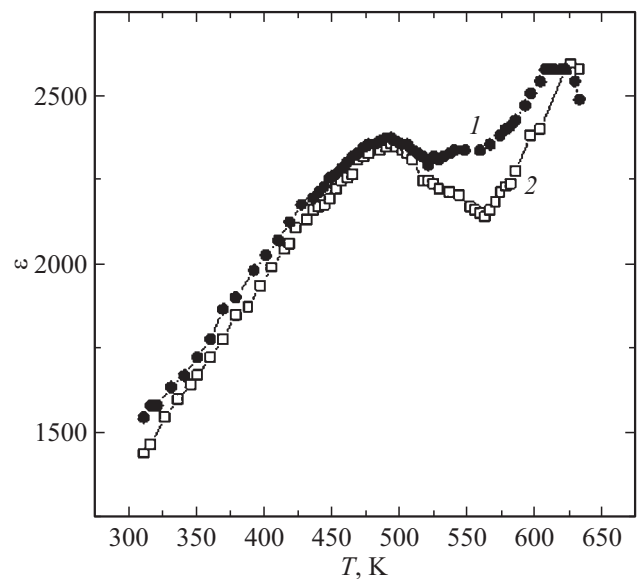


**Figure 1.** Temperature dependences of the dielectric permittivity $\varepsilon$ for ceramics KNN$-0.02$BNN taken in case of heating: curve *1* — in nonpolarized sample, curve *2* — in the absence of a field after the application of the fields of 10 kV/cm at a temperature of 383 K for 1 h.

the third anomaly in the form of a barely noticeable inflection in the temperature range of 433$-$443 K, which may correspond to the depolarization temperature $T_d$ is also noticeable on the curve *2* taken in a partially polarized sample.

We applied an electric field of 10 kV/cm at a temperature of 493 K with an exposure time of 1 h for a more complete polarization of the sample. The sample was cooled after that in the same field to room temperature, then the field was turned off and temperature measurements $\varepsilon$ were carried out in the absence of an electric field. Unfortunately, it was not possible to apply large fields to transparent ceramics which is necessary for the complete polarization of the sample. The polarization temperature of 493 K was not chosen by chance, because the polarization conditions (especially the polarization temperature) are an important factor for KNN ceramics and solid solutions based on it. This is because the phase transition at temperature $T_{O-T}$ is a polymorphic phase transition (PPT). This means that the polymorphic phase boundary (PPB) is not vertical in the phase diagram temperature$-$composition and shows a strong temperature dependence. Therefore, a temperature near the phase transition temperature [30–32] is usually chosen for the polarization of KNN. It should be noted that the polarization temperature does not affect MPB in case of a vertical MPB.

Figure 2 (curves *1$-$3*) shows the changes of $\varepsilon$ in the anomaly region of 433$-$443 K in the absence of an electric field (curve *1*) and in the field of 10 kV/cm applied at different temperatures (curves *2* and *3*).
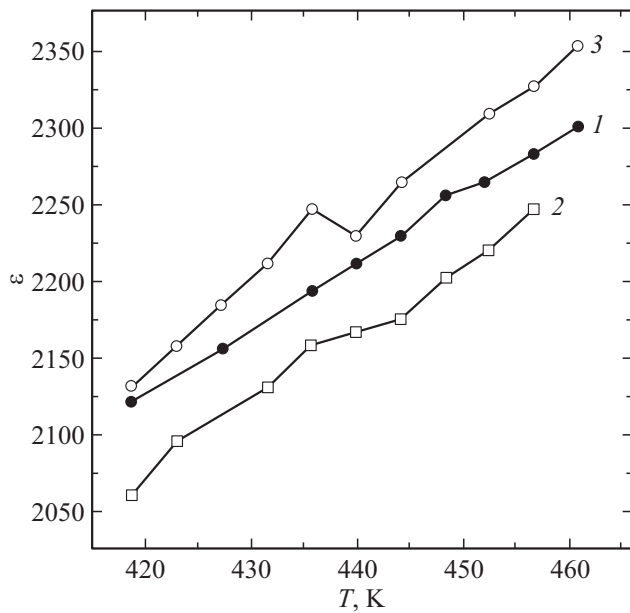
**Figure 2.** Dielectric permittivity $\varepsilon$ of ceramics KNN$-$0.02BNN as a function of temperature in the region of 418$-$458 K, taken during heating in the absence of a field and after polarization in different electric fields: curve *1* — 0 kV/cm, curve *2* — after application of the field of 10 kV/cm at a temperature of 383 K, curve *3* — after application of the field of 10 kV/cm at a temperature of 493 K.

It is clearly seen from Figure 2 that no anomalies are observed in this temperature region in the absence of an electric field (curve *1*), whereas an anomaly of $\varepsilon$ is observed in a partially polarized sample in the region of 433$-$443 K. The anomaly is more clearly observed in the case of the application of a field at a temperature close to $T_{O-T}$ (curve *3*). A small peak is located at $\sim$ 50 K below the polymorphic transition temperature $T_{O-T}$ and may be associated with the depolarization temperature $T_d$.

We performed optical measurements to confirm the assumption that the depolarization temperature exists below the temperature $T_{F-R}$. We used measurements in crossed Nicol's prisms since the changes of transmittance in a partially polarized sample may be insignificant and difficult to notice against a background of high transmittance. The sample was partially polarized in a field of 8 kV/cm at a temperature of 383 K for 1 h for these measurements. The sample in this field was cooled to room temperature, the field was turned off and temperature measurements of optical transmittance were performed. Figure 3 shows the measurements of $\Delta n$ in the absence of an electric field (curve *1*) and after the application of the field (curve *2*).

Despite the fact that the change of optical transmittance is associated only with a change of the size of inhomogeneities and occurs over a wide temperature range, it may suggest phase transition in which these changes occur. The increase of $\Delta n$ on the curve *1* as the temperature rises and approaches the phase transition temperature $T_{O-T} \approx 493$ K

($T_{F-R}$ in polarized the sample) indicates an increase of the number and size of regions of the tetragonal ferroelectric phase. The number and sizes of tetragonal regions decrease, and $\Delta n$ sharply drops after the transition above 493 K when approaching the Curie temperature. $\Delta n$ is practically zero in the cubic phase. It should be noted that, as distinct from dielectric measurements, another minor anomaly is clearly visible on the temperature dependence $\Delta n$ in the absence of an electric field in the temperature range of 433$-$443 K, which is below the transition temperature $T_{O-T}$ and may be related to the depolarization temperature $T_d$.

A decrease of $\Delta n$ in the depolarization temperature region and a further increase of $\Delta n$ at the approach to the temperature $T_{F-R}$ is clearly seen on the curve *2* taken after the application of an electric field. The decrease of $\Delta n$ is associated with the destruction of the macroscopic polarization, which occurred only in part of the volume of the polarized sample, while the rest of the sample remains unpolarized because of a small polarizing electric field. The electric fields used in the study do not result in the complete polarization (monodomain state), but lead only to the partial polarization of the sample, and a long-range order occurs in a part of the sample volume. Unfortunately, significant dielectric losses in the sample did not allow applying large electric fields. The long-range order is destroyed after the temperature $T_{F-R}$, the sample undergoes transition to the relaxor phase.

It can be seen from the above dependencies (Figures 1$-$3) that the temperatures $T_{F-R}$ and $T_d$ in the studied ceramics differ from each other. A similar difference of temperatures $T_{F-R}$ and $T_d$ was observed in single-crystal samples of PZN, NBT and their solid solutions [25,28$-$30].

In our previous papers [28,30], examining single crystal samples of PZN, NBT and their solid solutions, we came to the conclusion that the difference of temperatures $T_d$
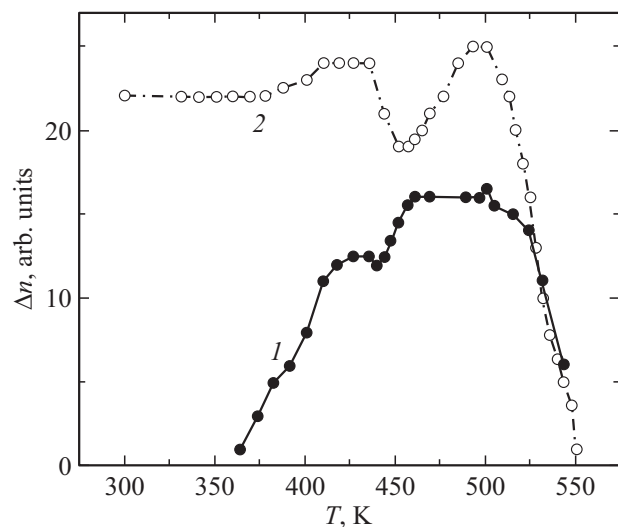


**Figure 3.** The change of birefringence $\Delta n$ with temperature in the absence of an electric field (curve *1*) and after application of a field of 8 kV/cm at a temperature of 383 K for 1 h (curve *2*).

and $T_{F-R}$ is observed only in those relaxors, in which there is a blurred phase transition to a ferroelectric state in the absence of an electric field, and this is attributable to the size and concentration of PNR (size $\sim 100-200$ nm).

A spontaneous diffuse transition to a ferroelectric state is also observed in the ceramic KNN$-$0.02BNN studied in this paper in the absence of an electric field at a temperature of $T_{O-T}$. The domains of the ferroelectric tetragonal phase appear at a temperature of $T_C$ when ceramic KNN$-$0.02BNN is cooled from the paraelectric phase, then they grow very quickly because of a decrease of the volume of the cubic phase and most of the sample passes into a mixed phase below the temperature $T_{O-T}$ in which R$-$O$-$T-regions coexist, but in general the macrostructure remains cubic.

It is well known that the domain size in ferroelectric materials is closely related to the corresponding grain size [33,34]. The grain size in hot-pressed ceramic KNN$-$0.02BNN is $\sim 200$ nm [14,17], therefore, the average domain size is also $\sim 200$ nm. The temperature discrepancy between $T_d$ and $T_{F-R}$ in the studied composition, as in solid solutions of PZN and NBT, may be related to the sizes of the domains, which result in a two-stage phase transition. In this case $T_d$ does not necessarily include a complete loss of the polarization state of the material.

## 4. Conclusion

The effect of an electric field on phase transitions in poorly studied lead-free hot-pressed ceramic KNN$-$0.02BNN is studied in this paper. This ceramics has a number of features compared to both lead-containing solid solutions and lead-free NBT-based solid solutions. Firstly, ceramic KNN$-$0.02BNN undergoes a polymorphic phase transition (PPT) between orthorhombic and tetragonal phases ($T_{O-T}$), whereas morphotropic phase transition (MPT) is observed in the above compositions. The phase boundary of PPT is not vertical (as, for example, the morphotropic phase boundary in PZT, solid solutions with PMN and a number of other compounds), but reveals a strong temperature dependence. Secondly, the relaxation behavior in this ceramics is observed already at room temperature during the phase transition between rhombohedral and orthorhombic ferroelectric phases ($T_{R-O}$). Thirdly, nanodomains in ceramic solid solutions based on KNN are induced both because of the coexistence of many phases (R$-$O$-$T) and because of the destruction of the long-range order. This differs from the ceramics PZT, in which nanodomains arise only in the result of the coexistence of R$-$T phases.

The mutual position of depolarization temperatures was studied in such ceramics with PPT for the first time ($T_d$) and ($T_{O-T}$) ($T_{F-R}$ — the temperature of transition to the relaxor state in a polarized sample). A difference of these temperatures was found, which is associated with the two-stage transition of the polarized sample to the relaxor phase: the macroscopic polarization is lost at $T_d$, and then the final

transition to the relaxor phase occurs at $T_{F-R}$. A assumption is made that the reason is a lower degree of diffuseness of the phase transition and larger PNR sizes. The polarized sample also undergoes a gradual transition to the relaxor phase consisting of two stages because the beginning and end of the transition do not coincide.

A similar pattern was observed in solid solutions with MPB with sufficiently large PNR sizes.

A conclusion is made that the relative position of the depolarization temperature and the temperature of transition of a polarized sample to the relaxor phase does not depend on the type of phase boundary (polymorphic or morphotropic), but is related only to the degree of diffuseness of the phase transition and the size of the polar regions.

## Conflict of interest

The authors declare that they have no conflict of interest.

## References

[1] Y. Deng, J.L. Wang, K.R. Zhu, M.S. Zhang, J.M. Hong, Q.R. Gu, Z. Yin. Mater. Lett. **59**, *26*, 3272 (2005).

[2] F. Li, M.J. Cabral, B. Xu, Z. Cheng, E.C. Dickey, J.M. LeBeau, J. Wang, J. Luo, S. Taylor, W. Hackenberger, L. Bellaiche, Z. Xu, L.-Q. Chen, T.R. Shrout, S. Zhang. Science **364**, *6437*, 264 (2019).

[3] K. Wang, B. Malič, J. Wu. MRS Bull. **43**, *8*, 607 (2018).

[4] N. Ishizawa, J. Wung, T. Sakakura, Y. Inagaki, K. Kakimoto. J. Solid State Chem. **183**, *11*, 2731 (2010).

[5] L. Egerton, D.M. Dillion. J. Am. Ceram. Soc. **42**, *9*, 438 (1959).

[6] V.J. Tennery, K.W. Hang. J. Appl. Phys. **39**, *10*, 4749 (1968).

[7] Y.J. Dai, X.W. Zhang, K.P. Chen. Appl. Phys. Lett. **94**, *4*, 042905 (2009).

[8] J. Tellier, B. Malič, B. Dkhil, D. Jenko, J. Cilensek, M. Kosec. Solid State Sci. **11**, *2*, 320 (2009).

[9] L. Wu, J.L. Zhang, C.L. Wang, J.C. Li. J. Appl. Phys. **103**, *8*, 084116 (2008).

[10] Y. Kang, Y. Zhao, R. Huang, Y. Zhao, H. Zhou. J. Am. Ceram. Soc. **94**, *6*, 1683 (2011).

[11] C. Montero-Tavera, M.D. Durruthy-Rodríguez, F.D. Cortés-Vega, J.M. Yañez-Limón. J. Adv. Ceram. **9**, *3*, 329 (2020).

[12] M.-H. Zhang, K. Wang, J.-S. Zhou, J.-J. Zhou, X. Chu, X. Lv, J. Wu, J.-F. Li. Acta Mater. **122**, 344 (2017).

[13] C. Shi, J. Ma, J. Wu, K. Chen, B. Wu. Ceram. Int. **46**, *3*, 2798 (2020).

[14] W. Yang, P. Li, F. Li, X. Liu, B. Shen, J. Zhai. Ceram. Int. **45**, *2 Part A*, 2275 (2019).

[15] X.-X. Sun, J. Zhang, X. Lv, X.-X. Zhang, Y. Liu, F. Li, J. Wu. J. Mater. Chem. A **7**, *28*, 16803 (2019).

[16] J. Frantti, S. Ivanov, S. Eriksson, H. Rundlöf, V. Lantto, J. Lappalainen, M. Kakihana. Phys. Rev. B **66**, *6*, 064108 (2002).

[17] X. Lv, J. Wu. J. Mater. Chem. C **7**, *7*, 2037 (2019).

[18] X. Lv, X. Zhang, J. Wu. J. Mater. Chem. A **8**, *20*, 10026 (2020).

[19] V.A. Isupov. Physica Status Solidi A **181**, *1*, 211 (2000).

[20] S. Zhang, R. Xia, T.R. Shrout, G. Zang, J. Wang. J. Appl. Phys. **100**, *10*, 104108 (2006).

[21] Y. Shiratori, A. Magrez, C. Pithan. J. Eur. Ceram. Soc. **25**, *12*, 2075 (2005).

[22] X. Chen, Y. Wang, J. Chen, H. Zhou, L. Fang, L. Liu. J. Am. Ceram. Soc. **96**, *11*, 3489 (2013).

[23] K. Yoshida, K. Kakimoto, M. Wei, S.J. Rupitsch, R. Lerch. Jpn J. Appl. Phys. **55**, *10S*, 10TD02 (2016).

[24] V. Bobnar, Z. Kutnjak, R. Pirc, A. Levstik. Phys. Rev. B **60**, *9*, 6420 (1999).

[25] X. Tan, E. Aulbach, W. Jo, T. Granzow, J. Kling, M. Marsilius, H.J. Kleebe, J. Rödel. J. Appl. Phys. **106**, *4*, 044107 (2009).

[26] E. Sapper, S. Schaab, W. Jo, T. Granzow, J. Rödel. J. Appl. Phys. **111**, *1*, 014105 (2012).

[27] Y. Hiruma, H. Nagata, T. Takenaka. J. Appl. Phys. **105**, *8*, 084112 (2009).

[28] L.S. Kamzina. Phys. Solid State **64**, *6*, 654 (2022).

[29] A.B. Kounga, T. Granzow, E. Aulbach, M. Hinterstein, J. Rödel. J. Appl. Phys. **104**, *2*, 024116 (2008).

[30] L.S. Kamzina. Phys. Solid State **64**, *11*, 1754 (2022).

[31] H. Du, W. Zhou, F. Luo, D. Zhu, S. Qu, Z. Pei. Appl. Phys. Lett. **91**, *20*, 202907 (2007).

[32] J. Wu, D. Xiao, Y. Wang, W. Wu, B. Zhang, J. Zhu. J. Appl. Phys. **104**, *2*, 024102 (2008).

[33] M. Eriksson, H.X. Yan, G. Viola, H.P. Ning, D. Gruner, M. Nygren, M.J. Reece, Z. Shen. J. Am. Ceram. Soc. **94**, *10*, 3391 (2011).

[34] W.W. Cao, C.A. Randall. J. Phys. Chem. Solids **57**, *10*, 1499 (1996).

*Translated by A.Akhtyamov*