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Influence of external influences on the activation of domain walls of ferroelectric perovskites

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Abstract

Purpose: The article aims to establish the regularities of the influence of external influences (thermal and electrical) on the activation of domain walls in oxygen-octahedral-type ferroelectric materials with perovskite structure which have a wide application in electronic devices. One of the problems in this case is the stability of domain walls with respect to external influences. With this in mind, the aim of the paper is to determine the activation energy of irreversible motion of domain walls when switching the polarization of a ferroelectric by electric field in a wide temperature range.

Experimental: The value of the critical energy $W_{\max}$ of interaction of ferroelectric domains with an external electric field necessary for irreversible change of the domain structure of a ferroelectric under different thermal conditions is determined. For this purpose, the dielectric hysteresis loops $P(E)$ of samples obtained at different temperatures were analyzed. Ferroelectric materials with oxygen-octahedral perovskite structure were chosen as objects of study: $BaZrO_3/BaTiO_3$ superlattice, $Pb(Zr_{0.3}Ti_{0.7})O_3$, ceramics $(Pb_{0.96}Sr_{0.04}(Mg_{1/3}Nb_{2/3})_{0.275}(Ni_{1/3}Nb_{2/3})_{0.1}Ti_{0.375}Zr_{0.25}O_3$ and ceramics $Pb_{0.88}Ba_{0.06}Sr_{0.06}(Mg_{1/3}Nb_{2/3})_{0.37}Zr_{0.375}Ti_{0.255}O_3)$.

Conclusions: Temperature dependences of the critical energy W_{max} of interaction of ferroelectric domains with an external electric field necessary for irreversible change of the domain structure of a ferroelectric have been determined. The assumption that the energy $W_{\max}$ has the meaning of the activation energy of the domain structure of a ferroelectric at switching of polarization by an electric field, necessary for irreversible change of its domain structure, has been substantiated. It is established that the critical energy $W_{\max}$ decreases linearly with temperature in a wide temperature range below the Curie point. The obtained results have scientific and practical value, since they allow to determine the conditions of stability of the domain structure of a ferroelectric to external influences and expand the possibilities of application of the Sawyer-Tauer method for determination of activation energies of irreversible change of the domain ferroelectric structure.

Keywords: Ferroelectric perovskites, Domain walls, Dielectric hysteresis loop, Activation energy, Repolarization, Barium zirconate, Barium titanate, Lead zirconate titanate, Lead magnoniobate

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1. Introduction

For practical applications in electronic engineering and fundamental research in solid state physics and chemistry, of particular interest are ferroelectric perovskites – inorganic crystalline materials whose structure contains internal surfaces of phase interfaces with different direction of the electric polarization vector – domain walls. The presence of domain walls for ferroelectrics determines especially large values of relative dielectric constant (10^3–10^4), and also gives a contribution to dielectric losses.

Domain structures can be studied by direct (microscopy) and indirect methods (dielectric spectroscopy). The application of the latter allows us to determine the conditions of stability and activation of its domain structure, as well as to estimate the activation energy of domain walls by the dependences of the complex dielectric constant of a ferroelectric on the magnitude of external influences (temperature, pressure, electric field) [1]. Another method of indirect study of the domain structure is the measurement of dielectric hysteresis loops $P(E)$ under various external influences (temperature, mechanical, electrical). Determination of domain wall energies is also possible from first principles, which was done by the authors of papers [2] to evaluate the structure and energies of several types of domain walls for two prototypical tetragonal ferroelectric perovskites: $PbTiO_3$ и $BaTiO_3$, including charge-neutral 90° domain walls of the Ising and Ising-Bloch types, as well as 180° domain walls of different orientations.

Since real ferroelectrics contain various structure defects, the process of nucleation and motion of domain walls occurs in interaction with the “stopper” system of defects in its crystal structure [3], which affects the mobility of domain walls in their interaction with lattice defects [4]. Although the nucleation and growth of the reverse polarity ferroelectric domain is controlled by the external electric field, this process is also influenced by thermal activation [5]. Thus, the analysis of the temperature evolution of dielectric hysteresis loops allows us to obtain data on the interaction energy of domain and defect structures and the nature of its change with temperature.

Despite the numerous studies of ferroelectric perovskites, even today there are still works on studying the structure of domains, domain walls and defects in these materials, as well as the evolution of these structures under the action of electric fields [6]. It was found that point defects of the crystal lattice, in particular, oxygen vacancies, play a significant role in the stability of the domain structure of perovskites. For the ferroelectric $BaTiO_3$, the activation energy of the motion of such vacancies has a value of 0.91 eV [7].

Dislocations in ferroelectrics also cause the anchoring of domain walls and the nucleation of ferroelectric domains, which significantly affects the electromechanical properties of ferroelectrics. For example, to determine the role of dislocations in ferroelectrics, the driving force of domain walls and their interaction with dislocations have been analyzed by finite element method [8]. It was found that the needle domain originating from the dislocation nucleus causes the domain walls to be anchored, and the interaction of the domain wall with the needle domain leads to a nonlinear dependence of the critical electric field required for the breakthrough of the domain walls through dislocations on the Burgers vector [8].

On this basis, it seems relevant to investigate the influence of external influences on the activation of domain walls of ferroelectrics with perovskite structure. For this purpose, the results of the study of dielectric hysteresis loops $P(E)$ of several ferroelectric perovskites at different temperatures [9-12] are analyzed and summarized in this work, from which the interaction energies of ferroelectric domains with an external electric field in different temperature and electrical conditions are determined.

2. Experimental

To determine the critical energy required to irreversibly change the domain structure of a ferroelectric when its polarization is switched by an electric field, in this paper, based on our previous studies [13, 14], we analyze the dependence of $W(E)$ obtained from the measured ferroelectric hysteresis data $P(E)$ according to the formula:

$$W = -P \cdot E, \qquad (1)$$

where *P* is the polarization value of the ferroelectric corresponding to the electric field *E* determined from the dielectric hysteresis loop *P*(*E*).

The dependences of *P*(*E*) for several ferroelectric materials with perovskite structure in the form of films, ceramics, single crystals, superlattice were obtained at different temperatures. Experimental data presented in papers [9–12] were used for this purpose, namely:

1) Ferroelectric superlattice (multilayer epitaxial layer structure) of barium zirconate/barium titanate $BaZrO_3$/$BaTiO_3$(BZ/BT) with comparable unit cell sizes consists of 32 repeating BZ and BT layers with the thickness of individual BZ layers - 6.65 nm, and BT layers – 6.67 nm, deposited by pulsed laser deposition on a single-crystal MgO substrate. To investigate the dielectric hysteresis loops at temperatures of 523, 633, and 658 K, a modified Sawyer–Tauer scheme with conductivity compensation and an external electric field frequency of 10 kHz was used [9].

2) Lead zirconate titanate films of $Pb(Zr_{0.3}Ti_{0.7})O_3$ were obtained by sintering and crystallization of the gel-solution precursor at 923 K for 10 min. The polarization hysteresis loops of *P*(*E*) were obtained at an electric field frequency of 100 Hz in the temperature range of 293-343 K [10].

3) Ceramics $Pb_{0.88}Ba_{0.06}Sr_{0.06}(Mg_{1/3}Nb_{2/3})_{0.37}Zr_{0.375}Ti_{0.255}O_3$ (modified lead magnoniobate zirconate titanate PMN-PZT ceramics) were prepared by conventional solid-phase reaction method at a sintering temperature of 1,533 K. Polarization *P*(*E*) hysteresis loops were obtained at a frequency of 1 Hz in the temperature range from 298 to 433 K [11].

4) Ceramic samples of modified lead zirconate titanate ceramics PZT ($Pb_{0.96}Sr_{0.04}(Mg_{1/3}Nb_{2/3})_{0.275}(Ni_{1/3}Nb_{2/3})_{0.1}Ti_{0.375}Zr_{0.25}O_3$) were prepared by solid-phase reaction at a sintering temperature of 1,343 K. The polarization hysteresis loops of *P*(*E*) were measured using Sawyer-Tauer scheme in the temperature range from 300 to 433 K at a frequency of 1 Hz [12].

3. Results and discussion

The shape of the dielectric hysteresis loop is largely determined by the domain structure of the ferroelectric. And, as noted by Smolensky et al [15], the change in the direction of spontaneous polarization occurs during polarization switching not by a jump in the whole domain, but by the motion of domain walls. Domain walls are fixed on pinning centers, which in real crystals are various defects of the crystal lattice, with which domain walls interact during their motion [16]. In a weak periodic electric field, the domain walls make reversible oscillations relative to the stationary position. If the electric field exceeds some critical value E_{cr}, which is less than the coercive field E_c, the domain walls detach from the stoppers and their motion becomes irreversible. The electric field E_{cr} is characterized by the fact that its effect on the ferroelectric leads to a change in the shape of individual lines of the X-ray diagram, which is a manifestation of the change in the domain structure of the ferroelectric [13]. The value of the critical field E_{cr} can also be determined from the *P(E)* dependence [14]. For this purpose, it is necessary to determine the field dependence *W(E)* of the energy *W* from formula (1). In the phenomenological theory of ferroelectrics, expression (1) is the additional energy in the full thermodynamic potential in an external electric field:

$$\Phi = U - T \cdot S + p \cdot V - P \cdot E, \tag{2}$$

where *U* is the internal energy, *T*, *p*, *V* are thermodynamic parameters (temperature, pressure and volume), *S* is the entropy, *P* is the polarization of the ferroelectric in the electric field *E* [15].

The polarization of ferroelectrics is due to the action of several mechanisms [17], but the main contribution is given by the domain polarization mechanism. Therefore, the contribution of other polarization mechanisms will be neglected in the following.

The polarization switching process can be accompanied either by the growth of existing domains, in which the polarization vector is opposite to the applied field, through the motion of domain walls, or by the nucleation and growth of new domains with the opposite direction of the polarization vector [15]. Under conditions of constant thermodynamic parameters *T, p, V*, the change of the full thermodynamic potential in an external electric field will be caused by the polarization switching processes. In this case, the spontaneous polarization within each of the domains remains constant, and the main contribution to the macroscopic polarization of the ferroelectric is due to the rearrangement of its domain structure, which largely determines the parameters of the ferroelectric hysteresis. Based on these considerations, the value of W is predominantly determined by the interaction energy of the ferroelectric domains with the external electric field. On the example of the classical ferroelectric $BaTiO_3$ (ceramic and monocrystalline), Fig. 1 shows the dependence *P(E)* taken from the monograph [18] and the dependence *W(E)* obtained in this work. Two maximum points W_{max-} and W_{max+}, corresponding to the external critical electric fields $-E_{cr}$ and E_{cr} are emphasized on the *W(E)* dependence. As it was mentioned above, in the critical field E_{cr}, the domain structure of the ferroelectric undergoes a rearrangement [13, 14]. This means that the energy W_{max} is the critical energy of interaction of ferroelectric domains with an external electric field, at which irreversible changes in the domain structure of the ferroelectric begin under external electric influence on it. In this work, the energy dimensionality W is defined in joules per one mole of matter (J/mole), assuming that the polarization dimensionality P is C/cm^2 and the electric field strength E is V/cm. The transition of the energy dimensionality W from J/cm^3 to J/mole is done by multiplication by the molar volume V_m ($V_m = M/\rho$, where M is the molar mass, g/mole; ρ is the density, g/cm^3).

This paper analyzes the *W(E)* dependences obtained from experimental *P(E)* dependences of ferroelectric materials with perovskite structure measured at different temperatures [9–12].

For the ferroelectric superlattice $BaZrO_3/BaTiO_3$ [9], the dependences of the interaction energy of the ferroelectric domains with the external electric field *W(E)* at temperatures 523, 633, and 658 K are shown in Fig. 2. The dependence $W_{max}(T)$ is approximated by a linear dependence at temperatures 20 K below the Curie temperature:

$$W_{max} = -a_T \cdot T + b_T, \tag{3}$$

where a_T and b_T are coefficients, T is temperature. For W_{max-}: $a_T \approx 0.07$ J/mole·K, $b_T \approx 48.6$ J/mole. For W_{max+}: $a_T \approx 0.24$ J/mole·K, $b_T \approx 158.9$ J/mole.

The reason for the different values of W_{max-} and W_{max+} at $T = 523$ K for $BaZrO_3/BaTiO_3$ superlattice (Fig. 2b) is the asymmetry of the dielectric hysteresis loop arising due to the presence of an internal electric field in the sample.

Analyzing the dependence of the maximum values of switching currents on the applied field strength, Sidorkin A.S. and co-authors [9] found that the full switching curve has two sections. The initial, so-called "activation" region or the region of "weak" fields, where the specified dependence of the switching current on the applied field is close to exponential, and the subsequent linear region, i.e., the region of "strong" fields, or the

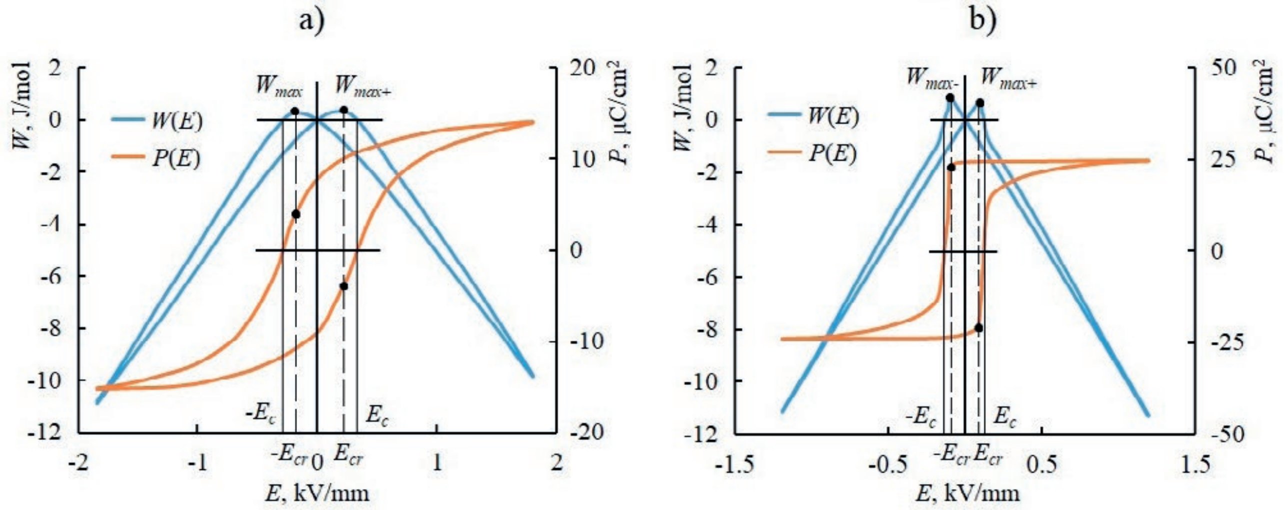

Fig. 1. Field dependence of the interaction energy of ferroelectric domains with an external electric field $W(E)$ and the dielectric hysteresis loop $P(E)$ of the ferroelectric $BaTiO_3$ in the form of ceramics (a) and a single crystal (b). E_c is the coercive field, E_{cr} is the critical field. From the experimental data $P(E)$ [18]

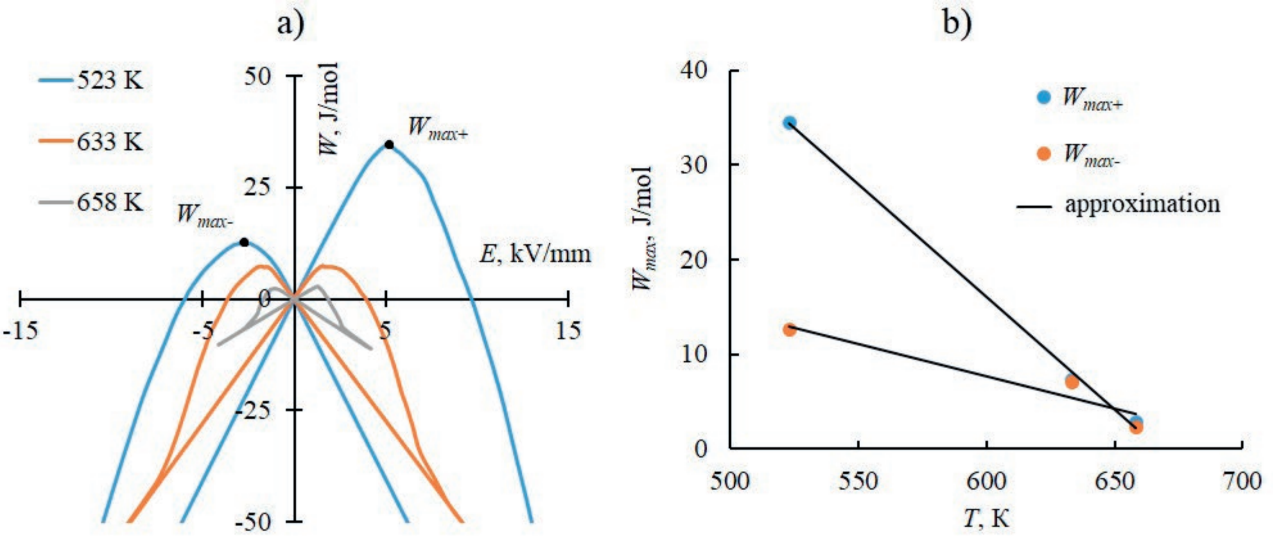

Fig. 2. Dependences $W(E)$ of the superlattice $BaZrO_3/BaTiO_3$ at different temperatures (a) and the temperature dependences of W_{max+} и W_{max-} (b), obtained using experimental data from [9]

region of "sliding", where the dependence of the maximum switching current is proportional to the field $i_{max} \approx \mu PEs/d$ (P – polarization, μ – mobility of domain walls, s – area of the top electrode, d – thickness of the sample). The boundary between the regions of activation and non-activation switching modes, the so-called E_{th} threshold or critical field, defined by the switching currents as the field corresponding to the transition of the exponential dependence into a linear one, approximately corresponds to the coercive field defined by the dielectric hysteresis loop [9].

The results of our work suggest that the critical field E_{th} determined by the authors [9] will correspond to the maximum value of the interaction energy of ferroelectric domains with the external electric field W_{max} (Fig. 2a) and the corresponding critical field E_{cr} (Fig. 1). Then the decrease in the linear law (3) of the energy value W_{max} with increasing temperature T can be associated with the growth of the domain wall mobility μ due to the decrease in the activation energy of the domain wall motion, which is caused by the weakening of the interaction of domain walls with the crystal lattice and defective environment, as well as by the decrease in spontaneous polarization as we approach the Curie temperature T_C. During polarization switching of the sample, domains of opposite sign are also nucleated, and mainly at the interfaces (grain boundaries, sample surface). But their subsequent growth is also determined by the mobility of domain walls, which mainly determines the polarization switching process.

For film samples $Pb(Zr_{0.3}Ti_{0.7})O_3$ [10], the dependences of $W(E)$ in the temperature range 293–343 K are shown in Fig. 3a. In this case, the dependences $W_{max+}(T)$ and $W_{max-}(T)$ presented in Fig. 3b, is also satisfactorily approximated by relation (3). For W_{max-}: $a_T \approx 0.29$ J/mole·K, $b_T \approx$ 134.9 J/mole. For W_{max+}: $a_T \approx 0.19$ J/mole·K, $b_T \approx$ 98.1 J/mole.

The dependences of $W(E)$ for ceramics $Pb_{0.88}Ba_{0.06}Sr_{0.06}(Mg_{1/3}Nb_{2/3})_{0.37}Zr_{0.375}Ti_{0.255}O_3$, plotted according to [11], are shown in Fig. 4a, and the $W_{max+}(T)$ and $W_{max-}(T)$ dependences, which practically coincide, are shown in Fig. 4b. Two areas can be distinguished on them. The first one corresponds to the temperature interval 298–409 K; and the second one – 409–433 K. At both sites, the dependences $W_{max+}(T)$ and $W_{max-}(T)$ are close to linear and can be approximated by expression (3). The best approximation corresponds to the following parameters. At the first site: $a_T \approx 0.04$ J/mole·K, $b_T \approx 15.7$ J/mole. At the second site: $a_T \approx 0.003$ J/mole·K, $b_T \approx 1.2$ J/mole.

As can be seen from Fig. 4b, the dependences $W_{max+}(T)$ and $W_{max-}(T)$ have two linear sections, the transition between which occurs at a temperature of 409 K. The authors of the paper [11] determined

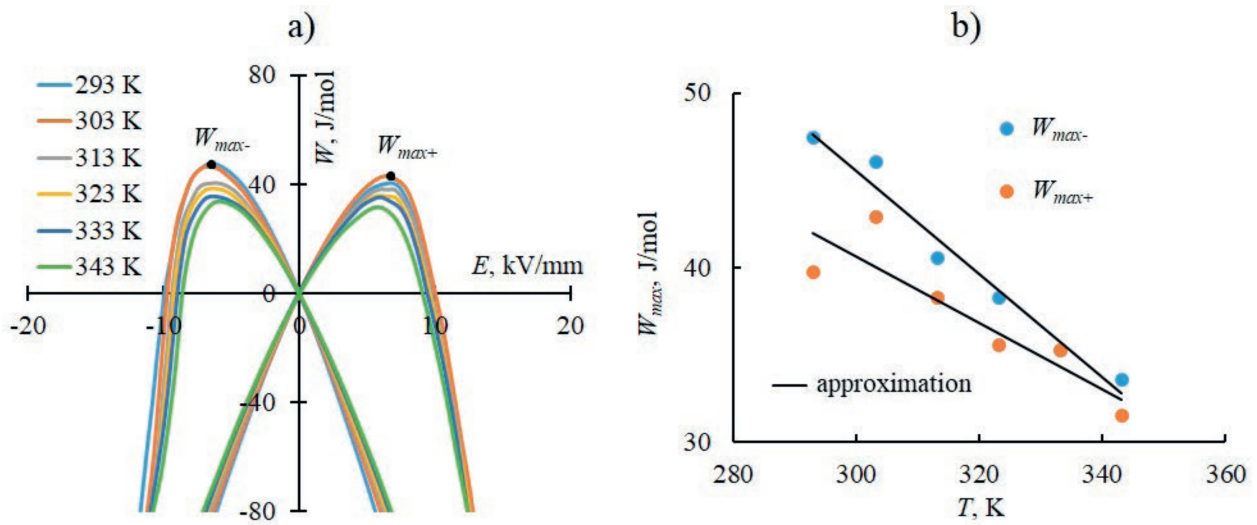

Fig. 3. Dependences $W(E)$ of thin films of $Pb(Zr_{0.3}Ti_{0.7})O_3$ at different temperatures (a) and temperature dependences of W_{max+} и W_{max-} (b), obtained using experimental data from [10]

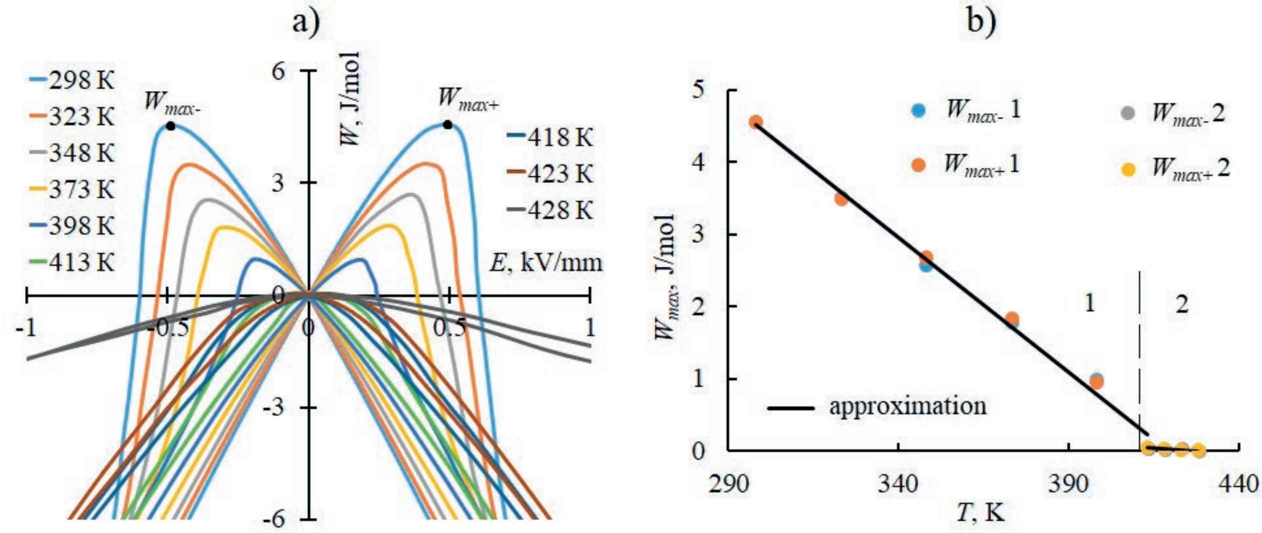

Fig. 4. Dependences $W(E)$ of $Pb_{0.88}Ba_{0.06}Sr_{0.06}(Mg_{1/3}Nb_{2/3})_{0.37}Zr_{0.375}Ti_{0.255}O_3$ ceramics at different temperatures (a) and temperature dependences of W_{max+} и W_{max-} (b), obtained using experimental data from [11]

that starting from this temperature the hysteresis dependence of $P(E)$ has the form of a double loop. This temperature practically coincides with the temperature T_{nr} (the lower index “nr” denotes the transition from normal to relaxor ferroelectric state at T_{nr} = 409 K [11]), at which the ferroelectric macrodomains decompose into microdomains – polar microspheres inherent in relaxor ferroelectrics. Such a process is characteristic of relaxor ferroelectrics [19, 20]. The questions of the formation of different types of domain structures in these materials and the influence of external influences on them are currently an urgent task and require further studies [21, 22].

As noted above, in the temperature region 298 - 409 K (site 1 in Fig. 4b), the hysteresis dependence of $P(E)$ has a form inherent to classical ferroelectrics. It can be assumed that below the temperature of 409 K polarization switching in ceramics $Pb_{0.88}Ba_{0.06}Sr_{0.06}(Mg_{1/3}Nb_{2/3})_{0.37}Zr_{0.375}Ti_{0.255}O_3$ occurs according to the laws common to ferroelectrics. Then the value of W_{max} will also have the meaning of the interaction energy of ferroelectric domains with the electric field. At the transition of the investigated sample to the relaxor state in the temperature region 409–433 K (section 2 in Fig. 4b), corresponding to the blurred ferroelectric phase transition of the first kind [15], part of the electric field energy is spent to induce the polar state, as evidenced by the double dielectric hysteresis loops, and the value of $W_{\max}$ loses the meaning of the interaction energy of ferroelectric domains with the electric field.

Analysis of $W_{\max+}(T)$ and $W_{\max-}(T)$ dependences (Fig. 5) obtained from dielectric hysteresis loops for ceramics $Pb_{0.96}Sr_{0.04}(Mg_{1/3}Nb_{2/3})_{0.275}(Ni_{1/3}Nb_{2/3})_{0.1}Ti_{0.375}Zr_{0.25}O_3$ [12] showed that they can be satisfactorily approximated by formula (3). For $W_{\max-}$: $a_T \approx 0.019$ J/mole·K, $b_T \approx 8.7$ J/mole. For $W_{\max+}$: $a_T \approx 0.021$ J/mole·K, $b_T \approx 9.5$ J/mole.

Analyzing the obtained results, it can be seen that the values of $W_{\max-}$ and $W_{\max+}$ for samples of the same composition can both differ from each other (Fig. 2b, Fig. 3b) and practically coincide (Fig. 4b, Fig. 5b). As noted above, for the $BaZrO_3/BaTiO_3$ superlattice such difference is explained by the asymmetry of the dielectric hysteresis loop as a result of the presence of the internal field of interlayer polarization. For $Pb(Zr_{0.3}Ti_{0.7})O_3$ thin films, this asymmetry indicates the presence of an internal displacement field [23]. In the case of ceramic bulk samples, however, this asymmetry is absent (Fig. 4b) or negligible (Fig. 5b).

Another issue that deserves attention is the reason for different values of $W_{\max}$ and different rates of their decrease a_T with increasing temperature for different materials. Thus, for $BaZrO_3/BaTiO_3$ superlattice and $Pb(Zr_{0.3}Ti_{0.7})O_3$ thin films, $W_{\max}$ values reach 30–50 J/mole and a_T values reach 0.2–0.3 J/mole K. At the same time, for the studied samples of modified PZT and PMN-PZT ceramics, $W_{\max}$ and a_T have values an order of magnitude smaller: $W_{\max} = 3–5$ J/mole and $a_T = 0.02–0.03$ J/mole K. Apparently, this is due to the fact that the values of the coercive field for thin-film and bulk samples also differ by an order of magnitude ($E_c \sim 10$ kV/mm and $E_c \sim 1$ kV/mm, respectively), while the values of spontaneous polarizations for the four analyzed samples do not differ so significantly and are within the range of values from 20 to 35 μC/cm^2 according to [9–12].

It is worth noting that the growth of the coercive field value with decreasing thickness of the ferroelectric samples is an experimentally established fact [24]. And the reason for this dependence is explained in the model of surface anchoring of domain walls and nucleation near electrodes [25].

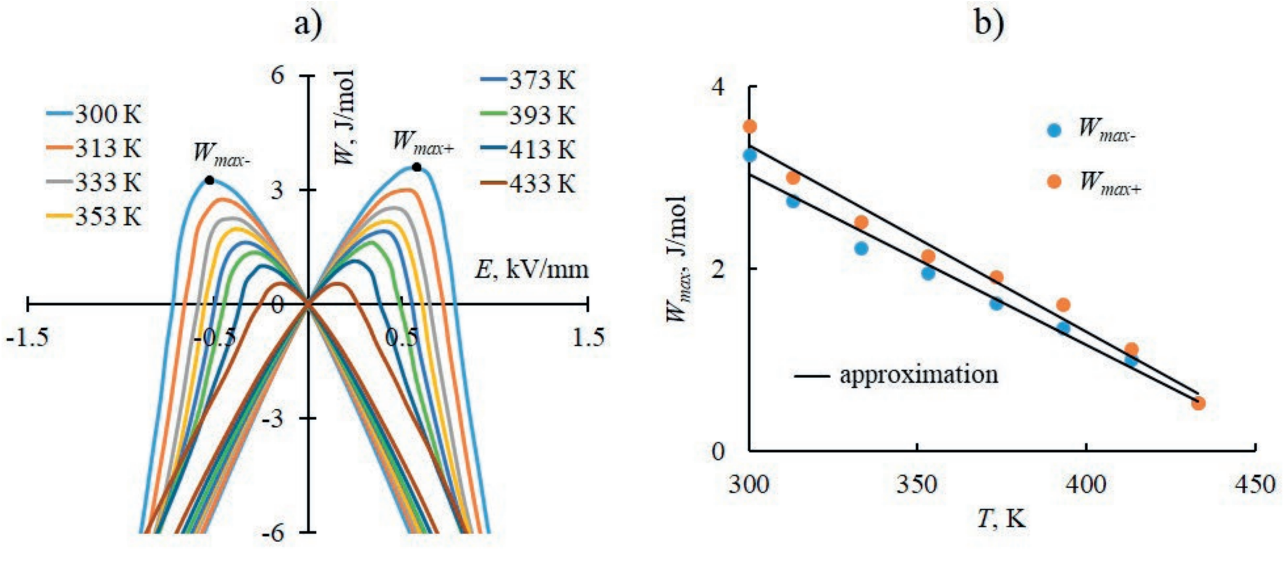

Fig. 5. Dependences $W(E)$ of $Pb_{0.96}Sr_{0.04}(Mg_{1/3}Nb_{2/3})_{0.275}(Ni_{1/3}Nb_{2/3})_{0.1}Ti_{0.375}Zr_{0.25}O_3$ ceramics at different temperatures (a) and temperature dependences of $W_{\max+}$ и $W_{\max-}$ (b), obtained using experimental data from [12]

4. Conclusions

Based on the analysis of ferroelectric hysteresis loops $P(E)$, the values of critical energies $W_{\max}$ of interaction of ferroelectric domains with an external electric field at different temperatures have been determined. In the course of the work, a reasonable assumption that the critical energy $W_{\max}$ has a physical meaning of the activation energy of the domain structure of a ferroelectric at switching polarization by an electric field, necessary for an irreversible change in its domain structure, has been put forward. It was found that in conditions of complete switching of the ferroelectric polarization, the critical energy $W_{\max}$ decreases according to a nearly linear law with increasing temperature from 300 K to the temperature lying slightly below the Curie point. The obtained results are of scientific and practical value, since they allow us to determine the conditions of stability of the domain structure of a ferroelectric to external conditions and expand the possibilities of application of the Sawyer-Tauer method for determining the activation energies of irreversible changes in the domain ferroelectric structure.

Contribution of the author

The author has independently performed all the work on the preparation of this paper.

Conflict of interests

The author declares that he has no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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