

Research Article

Electrically induced and controlled piezoelectricity

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Abstract: The quasi-linear electromechanical effect, which resembles piezoelectricity, can be induced by strong electric field in any solid dielectric. This effect is defined as linearized electrostriction, which is observed in the biasing electric field and is proportional to the square of the dielectric permittivity. The present work analyses physical mechanisms of the occurrence of such an effect in the piezoelectrics, the paraelectrics and the relaxor ferroelectrics, taking into account the inertia of electromechanical response. Hysteresis-less electromechanical control of deformation is used in actuators and tuneable microwave devices. The efficiency of the induced piezoelectricity is maximal in dielectrics with high permittivity, while the efficiency is ensured by mechanisms of low-inertia quasi-elastic polarization. To assess the maximum performance of piezoelectric actuators made of different materials, the method of dielectric spectroscopy is used.

Keywords: piezoelectric effect; electrostriction; paraelectrics; relaxor ferroelectrics; induced piezoelectric effect; dielectric spectroscopy

1. Introduction

The application of electric field to dielectrics always affects its electrical and mechanical properties to some extent, increasing the interdependence between them, i.e., making the electromechanical coupling stronger. At the microscopic level, this means that under the influence of the electric field the polarizability of dielectric changes significantly (it does not remain as constant parameter, independent of electric field). The reason for this is that strong field changes inter-atomic bonding of dielectric, leading to changes in polarizability. This gives rise to nonlinearity of dielectric polarization and, as a result, opens up the possibility of electrical control of many parameters that are usually considered independent of the electric field.

At the same time, polar dielectrics and, especially, ferroelectrics and related materials are much more sensitive to the action of the electric field: under its influence the electromechanical coupling in material structure changes, which affects the permittivity, and, therefore, all other polarization-related properties of dielectric [1]. The controllability of dielectric parameters is due to change in the rigidity of the atomic bonds under the influence of the electric field which reduces the polarizability of the dielectric, as well as all the parameters determined by it; in same way, it influences the restriction in freedom of reorientation of polar complexes or ferroelectric domains [2]. The degree of impact of the electric field on the dielectric parameters (including piezoelectric properties) is determined by the value of the dielectric permittivity ϵ . As shown below, electrically induced piezoelectric activity is proportional to ϵ^2 . In most dielectrics $\epsilon \leq 10$, and the influence of the electric field on the main parameters of the dielectric is practically negligible. However, in the dielectrics possessing $\epsilon = 10^3 - 10^5$, electrically induced and controlled effects can be so great that they can even exceed the conventional effects in efficiency [3]. Therefore,

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noticeable electric control of electromechanical properties is possible only in dielectrics with large permittivity. The physical mechanisms of electrically induced or controlled piezoelectric effects can be different in piezoelectrics, paraelectrics and relaxor ferroelectrics, although all of them can be applied to control the electromechanical coupling, owing to the electric-field sensitivity of the material's elastic compliance or the ferroelectrics domains orientation.

2. Mechanisms of electrostriction and piezoelectric effect

Since quite various mechanisms of quasi-linear electromechanical effect and controllable piezoelectric effect will be considered below, it is appropriate to start first with elementary models of electrostriction and conventional piezoelectric effect, so that we can indicate later which features of these models make it possible to get electrical control using piezoelectricity.

Electrostriction in ionic crystals considered in this work is due to the asymmetry of the electron density distribution along interatomic bonds. In turn, this feature is due to the difference between neighboring ions in the characteristics of their electronic shells, which, in particular, manifests itself as electronegativity. An ion with increased electronegativity displaces shared electrons towards itself, so its working charge becomes more negative, and an ion with less electronegativity acquires, accordingly, an increased positive charge. The mutual displacement of ions, forced by an external field, leads to a redistribution of the existing stable electron density in the interionic space, which leads to the appearance of a restoring force, which, after turning off the field, returns a stable unpolarized state. In centrosymmetric (non-polar) ionic structures, the crystal elongates along an applied field of any polarity with a corresponding compression in the transverse direction (Poisson's ratio).

Electrostriction always accompanies electric polarization of a solid dielectric, and it can be adequately represented by the ionic lattice polarization in one-dimensional model. The stability of ionic lattice is based on the interaction of cations and anions, and its non-polarized state is shown in Fig. 1(a). Under an external electric field E , Fig. 1(b),1(c), inter-ionic displacement leads to lattice polarization, characterizing by induced electric moment $P(E)$, which is an odd function, being linear, see Fig. 1(d). The polarization is accompanied by electrostriction, Fig. 1(e), which is the even function, being quadratic. Note, that the quadratic effect only reflects the fact that electric charges arrangement in lattice is centrosymmetric (both in the covalent crystal and in the cubic ionic crystal). In the case of nonlinear polarization, the electrostriction of centrosymmetric dielectric is described in a more complex way, but it is also an even function, represented by series $x(E) = RE^2 + R'E^4 \dots$ where R and R' are the electrostriction coefficients.

As it will be shown below, it is electrostriction that underlies the possibility of electric controllability of piezoelectric effect.

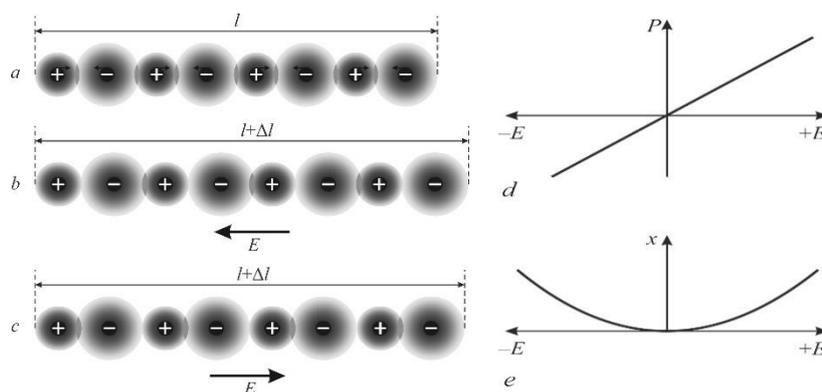


Figure 1. Modelling of ionic lattice polarization: *a* – balanced chain of negative and positive ions; *b, c* – influence of electric field, *d* – induced by electric field polarization P , *e* – induced by electric field strain x ; the shaded areas approximately indicate the distribution of electronic density in the outer shells, while the volume of negative ions is larger since they contain valence electrons

The piezoelectric effect is a characteristic of hybrid ionic-covalent crystal, the internal polarization of which usually leads to a non-centrosymmetric crystal structure, and the latter predetermines both piezoelectric and pyroelectric effects. It also can be presented by the model of positive and negative ions, but only in the case when they have hybrid (mixed) bonding, Fig. 2. The classical piezoelectric effect is possible only in polar (non-centrosymmetric) structures. The nature of internal polarity is complex, but certain assumptions can be made regarding it, again, as in the case of electrostriction, based on the phenomenon of electronegativity. In some ionic

compounds, the difference in the electronegativity of neighboring ions becomes so significant that even without the application of an external field, the ions combine into stable polar groups: in the simplest case, into dipoles, as shown in Fig. 2, but sextupoles and octupoles are also possible [2]. It can be assumed that the main reason for internal polarity in non-centrosymmetric crystals is the inhomogeneous distribution of the electron cloud density along the interionic bond. Thus, we can conclude that the internal polarity observed in non-centrosymmetric crystals arises due to structural compensation for differences in the characteristics of the electronic shells of neighboring ions.

Note that in simplified model, under the influence of an electric field, the usual linear polarization also appears; however, against the background of a large converse piezoelectric effect, the small effect of electrostriction is almost invisible in the crystals possessing permittivity $\epsilon \approx 5-10$, which is typical for ordinary crystals. We can conclude that the polar state in crystals is ensured by the fundamental properties of hybrid ionic-covalent mixed structure, and particularly by the different electronegativity of the nearest ions [3].

In Fig. 2 (as in Fig. 1), one-dimensional model is shown, but for a polar crystal this time. The one-dimensional simplified model has length l and stocked by electrodes at ends. Figure 2 conventionally shows polar-sensitive connections. Mechanical stress $+X$ leads to increase in length of crystal by amount Δl shown in Fig. 2(b); in this case, electric moment (polarization) is generated mechanically, being compensated by electric charges located on the electrodes. Figure 2(c) shows how mechanical compression $-X$ of the model chain changes the configuration of the electronic shell. It is important to note that polarization linearly depends on the magnitude of mechanical stress, Fig. 2(d). When the sign of the mechanical stress changes, crystal contracts, which is accompanied by a change in sign of the electrical polarization and a change in the signs of the compensating charges. As seen from Fig. 2(d), the dependence of polarization on mechanical stress is linear.

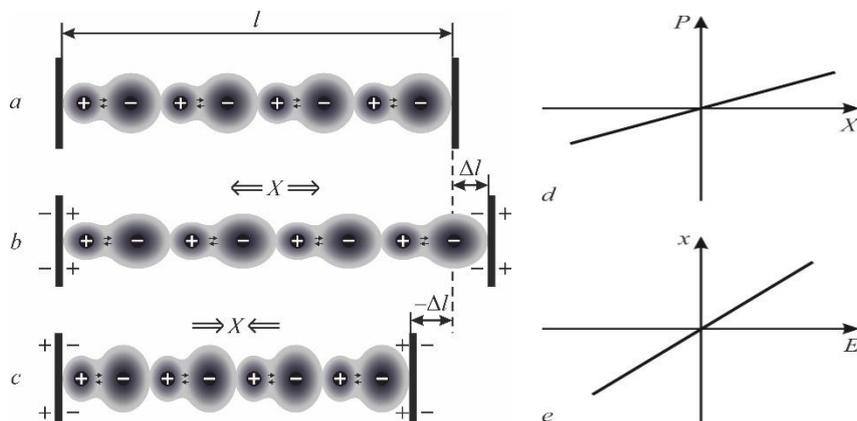


Figure 2. Model of piezoelectric effect in polar crystal: *a* – in absence of electric field; *b* – applied electric field coincides with direction of own polarity; *c* – electric field is opposite to direction of own polarity; *d* – direct piezoelectric effect; *e* – converse piezoelectric effect, Δl shows the change in the length of the model chain under the influence of mechanical stress X both in tension and compression

The model shown in Fig. 2 also allows to consider the converse piezoelectric effect, if one assumes that a longitudinal deformation occurs in externally applied electric field: $x = d \cdot E$. It also depends linearly on the electric field strength, as shown in Fig. 2(e). The model under consideration can also describe the volumetric piezoelectric effect, which is important for use in pressure sensors. The fact is that compression-tension leads to almost the same linear deformation $\Delta l/l$, and any changes in the transverse dimensions can be considered electrically neutral.

Shown in Fig. 2, the hybrid ionic-covalent bonding model is a characteristic property of the polar crystals discussed here, which naturally exhibit a piezoelectric effect (i.e., a linear electromechanical effect) without the application of an external electric field. An example would be the tuning characteristic of lithium niobate crystal, Fig. 3(a), widely used in electronics in various piezoelectric devices. Thus, it can be considered that the intrinsic polarity of a crystal is such that it replaces the application of an external electric field. The polar crystal shown corresponds to so-called linear pyroelectric, which can exhibit both a linear dependence of polarization when mechanical stress is applied (piezoelectric effect), and a pyroelectric effect when the temperature of the crystal changes. It is appropriate to note that pyroelectrics can also be nonlinear: it is the ferroelectrics, in which case the electromechanical effect will also be nonlinear, as can be seen in Fig. 3(b).

Stable structure of a linear pyroelectric or piezoelectric, Fig. 2(a), rigidly maintains the direction of its own polarity (usually until the crystal melts), but still allows electrical control of the piezoelectric properties, though within very small limits, Fig. 3(a). Nevertheless, this control can be successfully applied, for example, in piezoelectric delay lines and controlled filters.

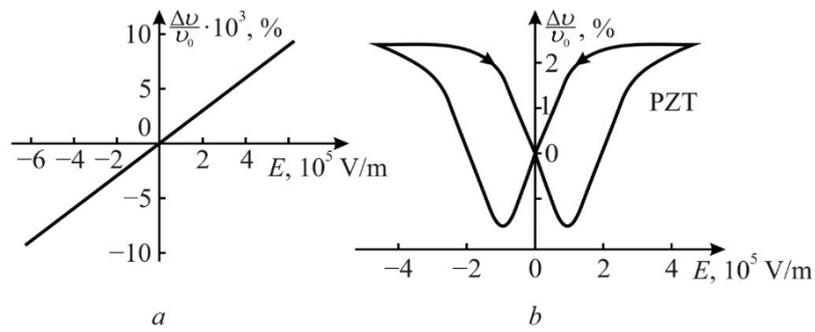


Figure 3. Electrically controlled piezoelectric effect shown as relative change in speed of sound; *a* – lithium niobate crystal; *b* – non-polarized ferroelectric ceramics

The unstable (nonlinear) structure of piezoelectric-ferroelectrics allows, firstly, the possibility of changing the direction of their own polarity, and, secondly, electrical control of piezoelectricity within very wide limits, but usually with pronounced hysteresis, Fig. 3(*b*), which is undesirable when using electronic devices. Note that the mechanism for controlling the speed of sound by an electric field in this case is complex. An applied electric field changes the elastic compliance as domain fusion and consolidation increases it. In addition, the controlling field increases internal stresses in ceramic crystallites due to electrostriction. As a result, under the influence of an electric field, the dielectric constant of the ferroelectric decreases. Hysteresis (i.e., the aftereffect shown in Fig. 3(*b*)) is due to the tendency of ferroelectric domains to maintain the acquired orientation.

Therefore, for controllable piezoelectricity, three possibilities are visible:

First, in a stable polar crystal, an external controlling field can increase or decrease the rigidity of bound ions pairs, thereby changing crystal's elasticity, and, consequently, piezoelectric modulus.

Secondly, a quasi-polar structure like shown in Fig. 2(*a*) might be artificially created from the non-polar crystal (shown in Fig. 1(*a*)) by applying a strong biasing electric field, which, in addition, can be used to control piezoelectric parameters.

Thirdly, in softly polarizable polar crystals the external field can change the direction of their own polarity (ferroelectric); in this case, significant control of all piezoelectric parameters can be obtained, though accompanied by a pronounced hysteresis.

However, as for paraelectrics and relaxor ferroelectrics, compromise solutions can be obtained, the physical mechanisms of which are discussed in this work. The possibility of hysteresis-free control of large deformations will be discussed below.

3. Creating the polar properties artificially

As already mentioned, electrically induced piezoelectricity can be obtained in any ionic and covalent crystals by applying a strong bias electric field. In this case, the induced linear polarization $P = \epsilon_0(\epsilon - 1)E$ is accompanied by a quadratic electrostriction effect $x(E) = RE^2$, Fig. 1(*d*). This is quite different from the polar crystals, because in them the field-induced strain depends linearly on the applied field, Fig. 2(*e*). This can be seen in Fig. 1(*d*): when the electric field changes its sign, the electrostriction does not change its sign, unlike piezoelectric.

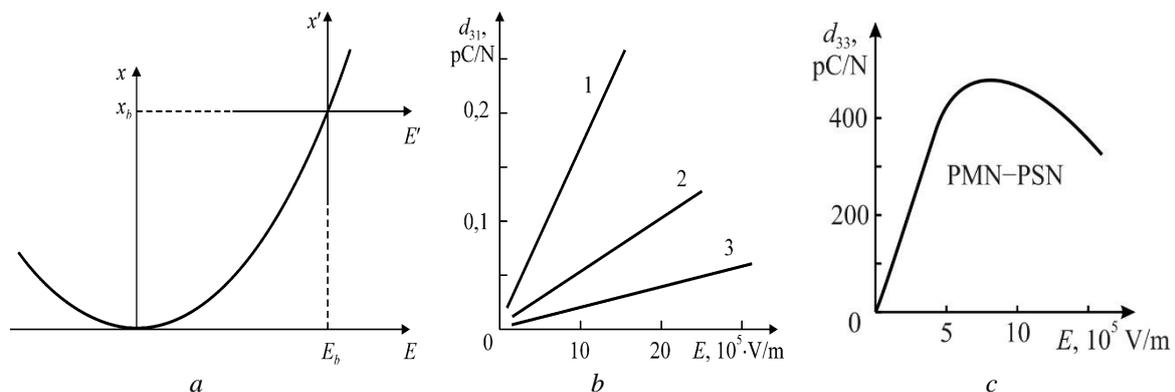


Figure 4. Electrically induced polarity in dielectric:

- a* – on parabolic electrostriction curve $x \sim E^2$ near bias field E_b a quasi-linear dependence $x'(E')$ imitates piezoelectric effect;
- b* – induced transverse piezoelectric effect in dielectrics SrTiO₃ with $\epsilon = 300$ (1); CaTiO₃ with $\epsilon = 150$ (2); TiO₂ with $\epsilon = 100$ (3);
- c* – induced longitudinal piezoelectric effect in diffused phase transition ceramics

A distinctive property of polar crystals is their ability to exhibit linear electromechanical effect (piezoelectricity), which can be represented as linearized electrostriction, Fig. 4(a). The point is that the initial symmetry of any crystal changes under the influence of the biasing field E_b due to additional deformation x_b . When the displacement field is maintained, an artificially created polar (non-centrosymmetric) structure appears; this can be seen on the right-hand section of the electrostriction parabola, which imitates the quasi-linear dependence of the deformation on the electric field (which is the main characteristic of piezoelectrics). An alternating electric field \tilde{E} , if applied in addition to the biasing field E_b , would cause an almost linear deformation: $x = d'\tilde{E}$. This quasi-piezoelectric effect, supported by a strong biasing field and representing linearized electrostriction Q_e , can only be significant in the dielectrics possessing a high permittivity, as the effect is proportional to ϵ^2 . Among such dielectrics, paraelectrics and relaxor ferroelectrics should be noted, Fig. 4(b,c).

To quantify the effects discussed here, the field-induced deformation can be expressed by a rapidly convergent series in even degrees of polarization: $x = Q_e P^2 + Q'_e P^4 + \dots$ [4, 5]. If the electric field is not very strong, then it is enough to take into account only the first term of this series. This need arose in connection with the extremely high electromechanical effect in the relaxor ferroelectrics, which can be seen, for example, from the nonlinear dependence of the induced piezoelectric modules in Fig. 4c for PMN. However, further calculation of this “piezoelectric modulus” given in the work is limited only by the quadratic effect, since taking into account the term $Q'_e P^4$ leads to unjustifiably complex expressions. To describe the piezoelectric activity, it is needed to distinguish two polarization components, one of them (P_b) being due to the action of the biasing field (the control field), which induces the polar properties causing piezoelectric activity, while the second component (\tilde{P}) describing the actual dynamic application of induced piezoelectric effect:

$$x = Q_e (P_b + \tilde{P})^2 = Q_e P_b^2 + 2Q_e P_b \tilde{P} + Q_e \tilde{P}^2.$$

It should be borne in mind that parametric interactions are not taken into account here (this is a separate problem), which comes down to condition $P_b \gg \tilde{P}$. In this case, the last term of this series can be neglected. It is also assumed that the polarization that excites the piezoelectric response is a faster changing signal compared to the slow changing control voltage. This gives grounds to consider the value P_b of induced piezoelectric effect as a constant ($x_b = Q_e P_b^2$). Therefore, the “basic” electrostriction obtained in the biasing field, against the background of an alternating field, appears to be a constant value, which allows us to calculate the “piezoelectric” effect: $x = 2Q_e P_b \tilde{P} = d \cdot \tilde{E}$, where d is the piezoelectric modulus induced by the displacement (i.e., biasing) field.

Obviously, the value of this parameter can be controlled electrically. For further calculations it is necessary to take into account that in the paraelectrics and relaxor ferroelectrics $\epsilon \gg 1$, so we can get

$$P_b = \epsilon_0(\epsilon - 1)E_b \approx \epsilon_0\epsilon E_b; \quad \tilde{P} = \epsilon_0(\epsilon - 1)\tilde{E} \approx \epsilon_0\epsilon \cdot \tilde{E}; \quad \tilde{x} = d \cdot \tilde{E} \approx 2Q_e \epsilon_0^2 \epsilon^2 E_b \tilde{E}$$

The field that induces piezoelectric effect can be denoted as $E_b = E$, thus we can write the following expression for the piezoelectric module:

$$d = 2Q_e \epsilon_0^2 \epsilon^2 E$$

From the resulting expression it becomes clear that the quasi-piezoelectric effect, induced by the displacement field, is due to electrostriction. The artificially obtained linear electromechanical effect is characterized by a quasi-piezoelectric modulus, which is proportional to the magnitude of the displacement field (i.e., the control field) and the square of the dielectric permittivity. It is for this reason that the electrically induced linear electromechanical effect is more pronounced with a higher permittivity, Fig. 4(b) and Fig. 5. Measurements of transverse piezoelectric modules were carried out at radio frequencies using the dynamic classical “resonance - anti-resonance” method with the need to isolate the control voltage circuit from the studied resonant circuit. The ceramic samples were disks with a diameter of about 10 mm and a thickness of about 0.3 mm with silver electrodes. The longitudinal piezoelectric modulus in relaxors was determined by the usual dilatometric method in a quasi-static mode. All this is included in the text as amendments to the article. In fact, our measurements have shown in the dielectrics with permittivity of $\epsilon \sim 100-300$ the electrically induced piezoelectric modulus has a value of $d_{31} \sim 0.3$ pC/N, but in the relaxor ceramics with a diffuse phase transition, where $\epsilon \sim 30000$, in field $E_b = 10^6$ V/m, the artificial piezoelectric modulus is a thousand times greater: $d_{31} \sim 500$ pC/N, which corresponds to PZT ceramics [3].

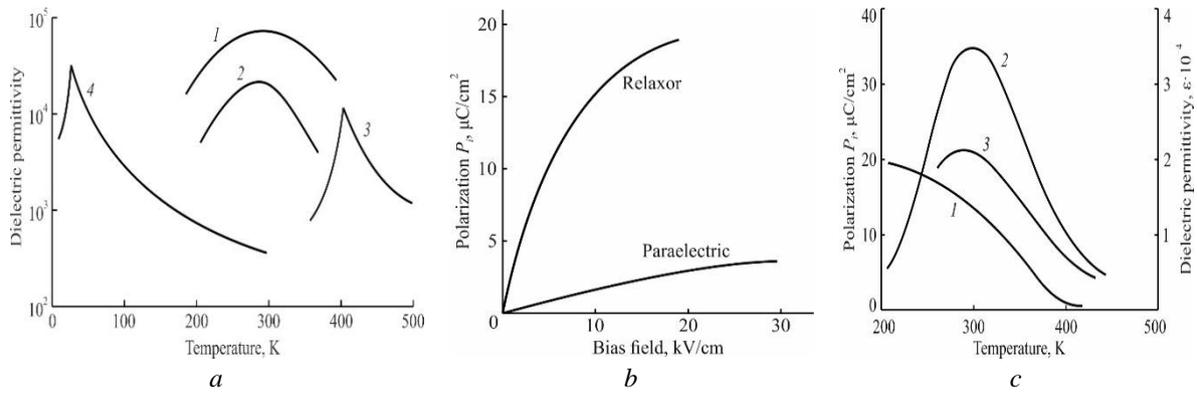


Figure 5. Paraelectric and relaxor characteristics:

- a* – permittivity in relaxor PMN ($\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$) (1), ferroelectrics $\text{Ba}(\text{Ti,Sn})\text{O}_3$ (2) and BaTiO_3 (3), paraelectric SrTiO_3 (4);
b – field-induced polarization P_i in PMN and in BST = $\text{Ba}(\text{Ti}_{0.6}\text{Sr}_{0.4})\text{O}_3$;
c – main PMN characteristics: 1 – polarization in by $E_b = 10$ kV/cm; 2 – permittivity without bias filed; 3 – permittivity in bias field [6, 7]

The dielectric permittivity of the basic materials used to obtain the controlled piezoelectric effect is compared on a logarithmic scale in Fig. 5(a); at that, the permittivity of the relaxor is an order of magnitude greater than in the ferroelectric solid solution based on barium titanate used in the paraelectric phase. Figure 5(b) indicates that the magnitude of the electrically induced polarization of the relaxor ferroelectric PMN is significantly greater compared that of the paraelectric BST, and it is this difference that is the reason why a relaxor ferroelectric produces a much greater linear electromechanical effect than a paraelectric. But, as will be shown below, in the BST this effect is less inertial, which is due to a difference in the polarization mechanisms. The electrically induced piezoelectric effect is interesting because it can explain the microscopic nature of the electromechanical coupling. Moreover, in a relaxor ferroelectric the electrically induced polarization P_i depends on temperature (like ferroelectrics spontaneous polarization P_s), as it can be seen in Fig. 5(c). Therefore, under the influence of an electrical biasing field the relaxors also exhibit an electrically induced pyroelectric effect.

The relaxor ferroelectrics have an undeniable advantage for use in actuator devices [6]. Really, in some of PMN-based relaxors with $\epsilon \approx 40000$ [8], the artificial piezoelectric modulus reaches $d = 2000$ pC/N, which is much greater than in the best piezoelectric ceramics of PZT type, whereas the electromechanical effect in the relaxors is practically free of hysteresis, Fig. 6(b).

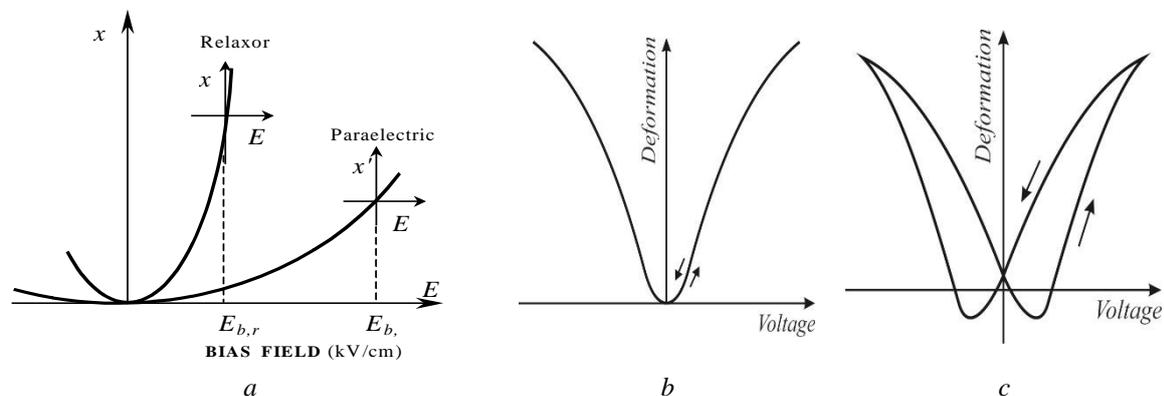


Figure 6. Electrically induced piezoelectricity as $x(E_b)$ -dependence:

- a* – paraelectric and relaxor comparison;
b – PMN based relaxor; piezoceramics PZT-8

Below we list several physical effects that determine the electrical control of the parameters of piezoelectric devices based on linear electromechanical effects induced by an electric field:

- When a strong electric field is applied to a non-polar dielectric, a non-centrosymmetric structure is forced to arise, exhibiting piezoelectric activity. In this case, the electric field changes the elastic properties of the crystal and, as a result, it affects the speed of sound, Fig. 3(a). Accordingly, the electric field changes the natural frequency of the field-activated artificial piezoelectric resonator made of a non-polar dielectric (like Fig. 4(b)). In ordinary dielectrics this effect is very small, but in such dielectrics as rutile, with $\epsilon = 100$, or strontium titanate ($\epsilon = 300$) such control can be very useful in electronics due to a high quality factor and instant response.

- A change in the elasticity of a crystal (its Young's modulus) by an external electric field leads to electrical control of the speed of sound and, in principle, can be observed in all piezoelectrics (such as quartz, potassium dihydrogen phosphate group, etc.), but this effect is more significant in crystals with increased dielectric permittivity (lithium niobate, silicosilenite), Fig. 3(a). In conventional piezoelectrics, due to very strong interatomic bonds, this effect is small, but allows, for example, to control delay lines. The high electrical and mechanical quality factor of these crystals is promising for their use in acoustoelectronics, for example, in pulse convolution processors.

- In ferroelectrics, under the influence of an electric field, polarization occurs with domain orientation. In addition, the applied electric field affects both the speed and attenuation of sound waves, which also has a noticeable effect on the properties of piezoelectric devices, Fig. 3(b). Despite the high efficiency of such control, its disadvantages are the phenomena of hysteresis and the relatively low speed of regulation. Its reason is the inertia of repolarization of ferroelectric domains.

- An important case of electrically induced piezoelectric effect is the excitation of piezoelectricity in paraelectrics. The physical mechanism for such control is that the electric field affects the frequency of transverse optical (“soft”) phonons of lattice vibrations, which has a significant impact on all properties of the paraelectric. The resonant frequency control range of piezoelectric resonators obtained in paraelectrics is about a few percents, but the quality factor of the devices remains quite high.

To conclude, the induced quasi-linear electromechanical effect can be described by the quasi-piezoelectric modulus $d = 2Q_e \epsilon_0^2 \epsilon^2 E$, where Q_e is the electrostriction coefficient. In dielectrics with high permittivity $\epsilon \sim 10^4$ - 10^5 this induced effect can be “giant”. Obviously, when a biasing electric field is applied, induced polarity can occur in any crystal, but in the dielectrics with high dielectric permittivity the induced piezoelectric effect can even outperform the efficiency of piezoelectricity of most materials that are used in practice.

4. The response time of electromechanical effects

Electrically induced piezoelectricity is determined by polarization processes, which, in turn, are characterized by dielectric permittivity. Therefore, it is advisable to evaluate the inertial properties of electromechanical control, using the frequency dependence of permittivity: $\epsilon(\omega)$. In the frequency regions where dielectric dispersion is observed the polarization is established with a time delay, which means that the inertia of the piezoelectric effect manifests itself.

The given judgment about the frequency limit of the use of the piezoelectric effect also applies to ordinary piezoelectrics and at ordinary electric field values. Fig. 3 shows two typical cases of changes in the speed of sound due to electromechanical action for different piezoelectrics: linear and nonlinear.

- **Linear piezoelectrics**, are firstly illustrated by LiNbO_3 crystal in Fig. 7(a), it has $\epsilon \sim 40$. In fact, it is a ferroelectric, though exposed to temperatures much below its Curie point, so its domains are not moveable (i.e., a “frozen” ferroelectric). In this crystal, as seen in Fig. 3(a), the change of sound velocity v under bias field influence is small (fractions of a percent), but it is sufficient for the use in the control of the SAW devices. This property shows controllability of $\Delta v/v(E)$ characteristic, which indicates that the strength of intra-crystalline bonding in crystal LiNbO_3 significantly exceeds the impact of the control field. In this case, the speed of electromechanical reaction is limited only by the structural elements of the piezoelectric device, since at the microscopic level it is determined only by natural frequency of crystal lattice vibrations, so that the relaxation time does not exceed nanosecond, as dielectric dispersion begins only after a frequency of 1000 GHz, Fig. 7(a).

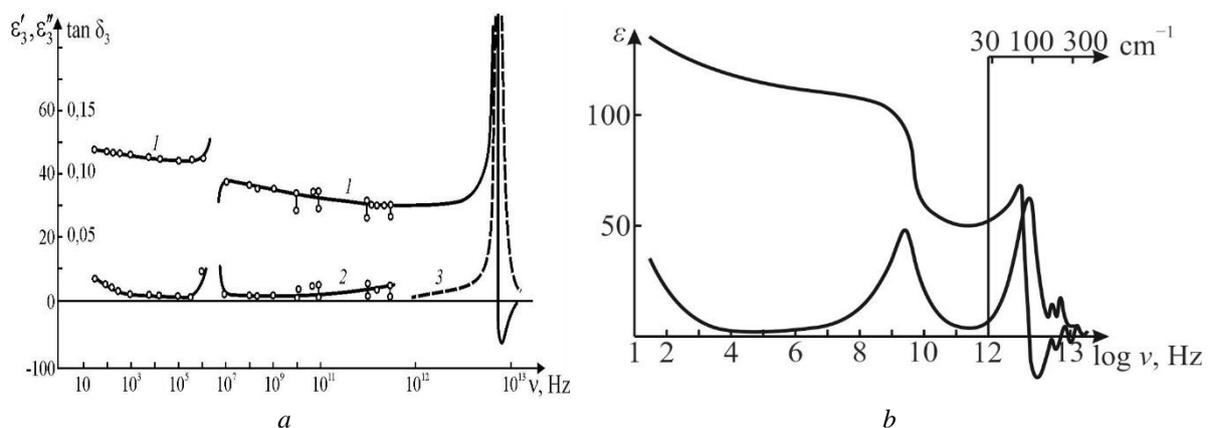


Figure 7. Dielectric spectrum of two piezoelectrics at ~300 K:
a – polar direction of lithium niobate (LiNbO₃) crystal: permittivity ϵ_3' (1), losses $\tan\delta_3$ (2), and ϵ_3'' (3);
b – lead titanate (PbTiO₃) ceramics

Another illustrative example of linear piezoelectrics is polycrystalline lead titanate, Fig. 7(b). In a weak electric field, the piezoelectric reaction of ceramic materials is already limited by a frequency of around 1 GHz, as can be seen from the dielectric dispersion plot. The speed of parameters control in piezoelectric resonant device usually not exceeds the frequency of ~ 100 kHz. This is due to delay in the reaction of polar clusters located in the vicinity of domain boundaries.

Thus, in piezoelectric crystals, the speed of electromechanical processes is not limited by any polarization mechanisms, but is determined by the inertia of natural mechanical vibrations of the given piezoelectric device. In piezoceramics, which are, in fact, polarized ferroelectrics, the boundaries of crystallites and domains in which polar clusters are localized lead to dielectric dispersion and losses, limiting the working frequency to the microwave range, which usually significantly exceeds the frequency of mechanical resonances of the devices.

• **Non-linear piezoelectrics** are usually the polycrystalline ferroelectrics, which exhibit very effective electrical control of the speed of sound, as shown in Fig. 3(b) for non-polarized ceramics. The latter materials usually are in nature “pliable” ferroelectrics with $\epsilon \sim 1000$. This means that the strength of the intra-crystalline bonding in this case can be comparable to the influence of the electric field. The large change in the speed of sound is due to the reorientation of ferroelectric domains, which also explains the pronounced hysteresis of the control characteristics.

The inertia of electrical control of electromechanical properties in a strong electric field is assessed by dielectric spectroscopy, see Fig. 8, Curves 2 and 3 in Region I. In this context, Curves 3 describe the effective dielectric permittivity ϵ'_{ef} and loss factor ϵ''_{ef} obtained by averaging the dielectric response per one period of the alternating field, and Curves 2 show the dielectric permittivity ϵ' and loss factor ϵ'' measured in a weak electric field of more increased frequency, but with simultaneous domain switching by a strong low-frequency field.

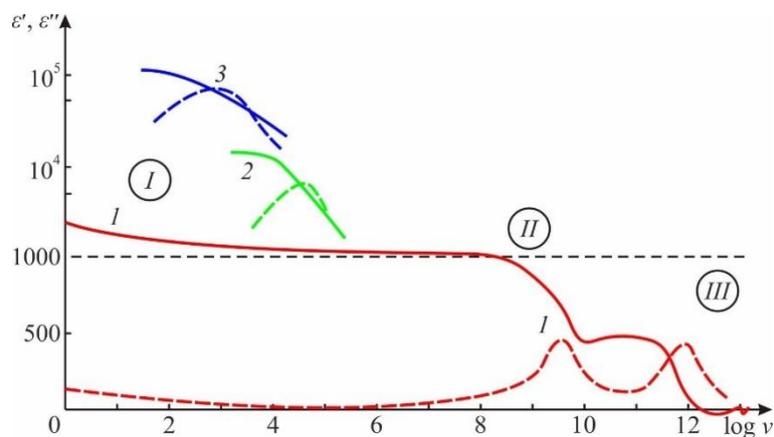


Figure 8. Dielectric permittivity dispersion in the ferroelectric phase of BaTiO₃,

where $\epsilon'(\nu)$ dependence is shown by solid curves while $\epsilon''(\nu)$ dependence is shown by dotted curves);

I – low frequencies, strong electrical fields, **II** – microwave region, weak fields; **III** – infrared range frequencies, weak fields

Compared with the study in weak electric field, shown in Regions II and III, the permittivity in strong fields in Region I is two orders of magnitude higher, while its dispersion frequency is a million times lower. From the data obtained by measurements in strong fields, it follows that the control of domain reorientation, which leads to hysteresis dependences (such as those shown in Fig. 3(b)), is possible only up to the frequencies of the order of kilohertz. (However, in the thin films the maximal frequency of domain switching increases to the order of megahertz.) Note that in strong electric fields several mechanisms of polarization in ferroelectrics are involved, whereby faster polarization occurs in the polar clusters localized in domain walls. As can be seen in Fig. 8, Region I Curves 2, the contribution of this mechanism is an order of magnitude smaller than that of Curves 3, and it can be measured using high-frequency filtering of the response during simultaneous switching of the domains at low frequencies.

Therefore, the use of nonlinear piezoelectrics (such as ferroelectrics) as electromechanical devices looks inappropriate not only due to the strong hysteresis, but also due to the inertia of domain switching.

• **Paraelectrics** permittivity exhibits practically no dispersion down to millimeter waves. As can be seen from barium titanate temperature dependence of permittivity, Fig. 9(a), strongly pronounced dielectric dispersion in both single crystals and ceramics occurs only below Curie point, while above it (in the paraelectric phase), permittivity decrease with frequency, and, accordingly, the increase in losses begin only at frequency of about 75 GHz.

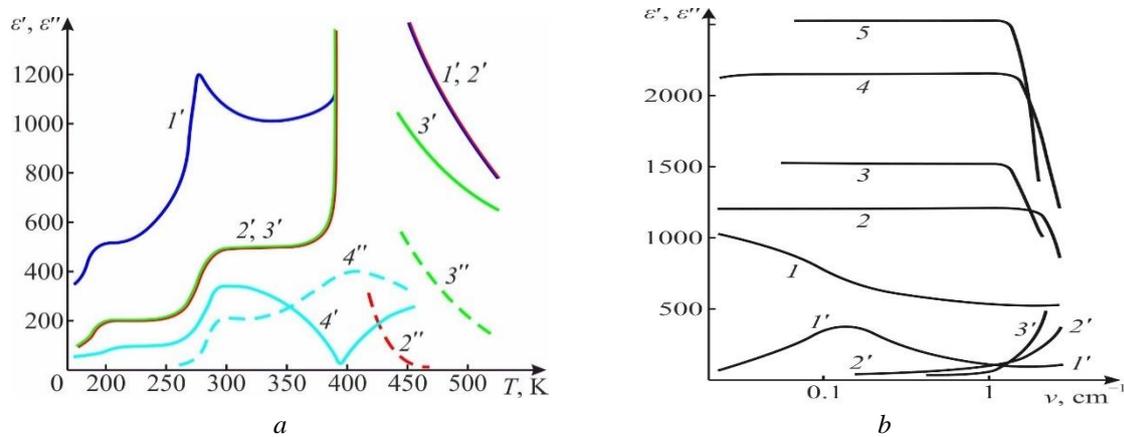


Figure 9. Dielectric dispersion in ferroelectrics and paraelectrics:

a – temperature dependence of barium titanate permittivity ϵ' and loss factor ϵ'' at different frequencies:

1' – ϵ' at 1 MHz; 2', 2'' – ϵ', ϵ'' at 9,4 GHz; 3', 3'' – ϵ', ϵ'' at 75 GHz; 4', 4'' – ϵ', ϵ'' at 340 GHz [24];

b – frequency dependence of ϵ' (1-5) and ϵ'' (1'-3') at different temperatures:

1, 1' – polycrystal at 22 °C, 2, 2' – polycrystal at 200 °C, 3, 3' – single crystal at 200 °C,

4 – polycrystal at 152 °C, 5 – single crystal at 155 °C [4]; frequency is indicated in reverse wavelengths: $1 \text{ cm}^{-1} = 30 \text{ GHz}$

This type of dispersion, illustrated in Fig. 9 using barium titanate as example, is typical for all ferroelectrics of the oxygen-octahedral type, which are commonly used in electromechanical drives and other piezoelectric devices. From the analysis of dielectric dispersion, it follows that the polarization processes in paraelectrics (which are due to the “soft mode” of vibrations of the perovskite crystal lattice) do not exhibit noticeable inertia of electromechanical properties down to millimeter waves. In this case, the electrically induced piezoelectric effect remains large and free of hysteresis. However paraelectrics, in principle, are not thermally stable materials, although, when using them, compromise solutions can be found (for example, thermal stability increases significantly in paraelectric films).

• **Relaxor ferroelectrics** have large permittivity over a wide temperature range; this property is very important for their application in electronic devices (micromechanics and microdrivers). The point is that controllable displacement, produced by relaxor actuator, is determined by the value of its permittivity. To assess the limitations on the performance of relaxor ferroelectrics in electronic devices, the frequency dispersion of the relaxor dielectric permittivity was studied in a broad frequency range, Fig. 10. It has been established that the dominant contribution to the permittivity of relaxor ferroelectric comes from the electromechanical coupling of different polar clusters [4]. So, the speed of response in relaxor based devices is determined by the speed of sound in relaxor ferroelectrics.

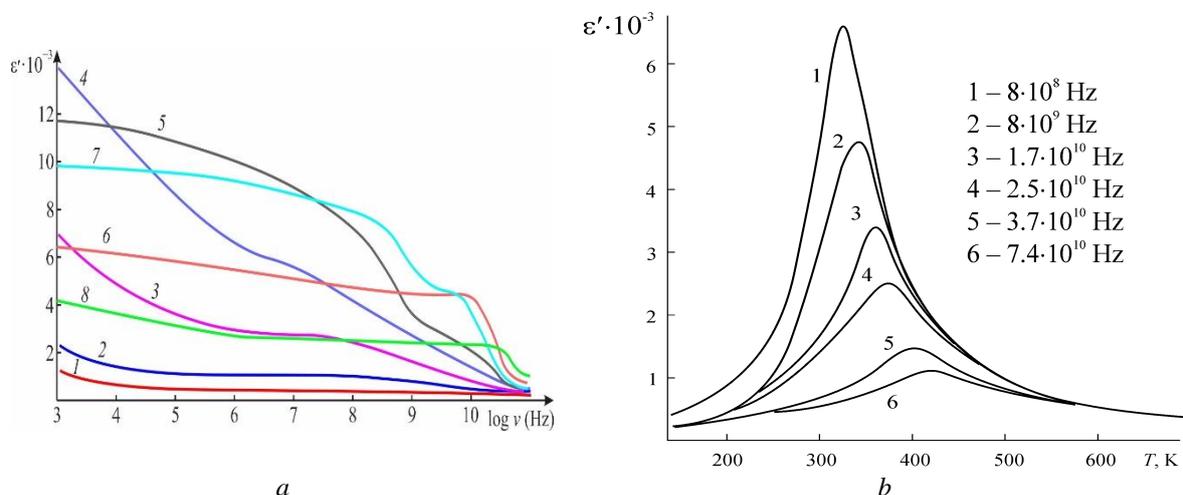


Figure 10. Dielectric spectra of PMN crystals:
a – permittivity at temperatures:
1 – 140 K, 2 – 200 K, 3 – 240 K, 4 – 270 K, 5 – 290 K,
6 – 300 K, 7 – 320 K, 8 – 360 K, 9 – 400 K, 10 – 500K;
b – permittivity temperature dependence at high frequencies [4, 7]

The broad temperature maximum of permittivity has two main contributions, listed just below; based on the above observations on the relaxor ferroelectrics, one can speculate about the physical mechanisms behind them.

- The low-frequency ($1-10^5$ Hz) contribution to the broad ϵ -maximum is usually explained by the reorientation of dipole moments of polar microregions. This is accounted for by the strong electromechanical coupling of the “soft-polarizable” polar regions, whereby electrostriction turns into piezoelectric effect.

- The high-frequency (10^8-10^{10} Hz) contribution to the dielectric permittivity is commonly explained by the motion of the boundaries of polar nanoregions. It resembles the microwave dispersion of permittivity in ferroelectrics visible in Fig. 7(b) and Fig. 8(a), which the latter owe to their multi-domain structure, whereas in the present case dielectric dispersion is accounted for by “frozen paraelectricity” in the boundaries of polar nanoregions [4].

The “piezoelectric modulus” of relaxor ferroelectrics is tens of times greater than that of inertia-free paraelectrics, and their increased inertia is not an obstacle to their use in actuators. A particularly large deformation under the influence of an electric field is observed in $\text{PbZn}_{1/3}\text{Nb}_{2/3}\text{O}_3-4,5\%\text{PbTiO}_3$ (PZN-4,5%PT) crystals [8, 9]. For such a crystal, the electrically controlled deformation is 10 times higher than the same parameter for the widely used PZT-8 piezoelectric ceramics. But the biggest advantage of relaxor ferroelectrics is the almost complete absence of hysteresis in their control characteristic, Fig. 6(b). Sometimes these properties are described as “giant” electrostriction [10, 11].

Thus, the response time of devices based on relaxor ferroelectrics is determined by dielectric dispersion mechanisms. The electromechanical contribution to the dielectric permittivity can be the dominant factor for the speed of response, and it is determined by the speed of sound in the relaxor ferroelectric, and thus the operating rate of the respective devices depends on the size of the relaxor element used [12].

The possibilities of obtaining and technically applying the electrically induced piezoelectric effect have been successfully realized [3], and the materials used for this purpose continue to be improved [11]. The purpose of this work is to clarify the physical concepts of the nature of this effect, especially in paraelectrics and relaxor ferroelectrics. The physical mechanisms that make it possible to obtain an effective electromechanical effect are very diverse and, in the case of relaxor materials, can act simultaneously. To understand their differences, the dielectric spectroscopy method is used, which also allows us to determine the limits of applicability of the induced piezoelectric effect at various frequencies.

Electrically controlled piezoelectric actuators are used in robotics, mechanical engineering, instrumentation and electronics. They provide high precision of movement, which is especially important for propulsion, in tunnelling and atomic force microscopes, where high (atomic order) resolution is required. A characteristic feature of the electrically induced piezoelectric effect is the unique possibility of the occurrence of piezoelectric resonance (or other manifestations of piezoelectric activity) exclusively during the action of the control voltage. The fact is that the inertia of control by electrically induced deformation is determined by the dispersion of the dielectric permittivity in paraelectrics or relaxor ferroelectrics.

5. Conclusions

In any solid dielectric, the electric field leads to electrical polarization, which is certainly accompanied by electrostriction. Usually this quadratic effect is very small, but in paraelectrics and relaxor ferroelectrics, which have a large dielectric permittivity, under the influence of the biasing field the electrostriction is converted into induced piezoelectricity, which can be even greater than the usual piezoelectric effect. The main reason for these effects is that electrostriction changes the symmetry of any isotropic dielectric, which becomes, as it were, “polar” (i.e., non-centrosymmetric). In this case, the polar-active structure, both naturally existing in the crystal and induced in a non-polar material, can be controlled by changing the biasing electric field. The present work examines the influence of electric polarization mechanisms on the magnitude and controllability of electromechanical effects and their inertia, for which the method of dielectric spectroscopy is used. In piezoelectric crystals, the speed of electromechanical processes is not limited by polarization mechanisms, but in piezoceramics they are limited at the microwave range. The use of nonlinear piezoelectric-ferroelectrics as electromechanical devices (despite large electromechanical response in strong fields) seems inappropriate not only because of strong hysteresis, but also because of the inertia of domain switching, which for bulk samples is limited by the kilohertz

frequency, and for films – by several megahertz. In paraelectrics, the magnitude of the induced electromechanical effect and its electrical controllability are much lower, while they are practically inertia-free, whereas their use is possible only in a narrow temperature range. Relaxor ferroelectrics are capable of providing a large and practically hysteresis-free effect, which, however, is significantly reduced at kilohertz and especially at megahertz frequencies.

Conflict of interest

There is no conflict of interest for this study.

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