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Induced domains and periodic domain structures in electrically and magnetically ordered materials

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Contents

Abstract. The state of the art of studies of the formation of domain structures is reviewed. Various methods of ferroelectric and magnetic domain formation are discussed, including putting single crystals into electric, magnetic, optical, and acoustic fields as well as exposing them to thermal annealing or changing their chemical composition. It is demonstrated that the realization of induced domain structures has already led to new frequency multiplication and ultrasound generation methods as well as to those for the parametric conversion of acoustic and optical beams.

1. Introduction

Research interest in extended structures with periodically varying values of the optical, elastic, and other physical parameters emerged at the beginning of the 1960s in connection with the possibility of using such structures to generate light and acoustic radiation or convert the radiation frequency [1, 2]. Later, it was found that periodic or quasi-periodic structures consisting of metallic or semiconducting layers possess several extraordinary physical properties, e.g., localization of electronic states [3 – 6]. Since for domains with antiparallel polarizations the odd-rank tensors that describe various physical properties have opposite signs, the nonlinear optical, electrooptical, magnetooptical, magnetoelastic, piezoelectric, and pyroelectric characteristics of such structures differ from those of single-domain samples [7 – 11].

The first real application of domain structures was probably the use of magnetic-bubble lattices in storing and processing information [11].

Natural domain structures were the first to be studied in experiments in frequency conversion of laser light [12, 13], diffraction of light beams [14, 15], and the generation and conversion of acoustic waves [16, 17]. However, the use of grown domain structures was found ineffective because of the wide spread of their characteristics, so that up to the mid-1980s the main materials used in opto- and acoustoelectronics were single-domain. Moreover, it was found that the use of grown domain structures leads to additional scattering of light and acoustic beams, broadening of the range of the control voltage needed for phase rotation, and changes in the speed of ultrasonic waves.

The breakthrough came in the second half of the 1980s, when new ways of forming sufficiently perfect periodic domain structures in a number of oxygen-octahedral ferroelectrics ($LiNbO_3$, $LiTaO_3$, $KTiOPO_4$, $BaTiO_3$, and other similar materials) were discovered [18 – 23]. A periodic domain structure consists of domains with inverted polarization with respect to the uniform spontaneous polarization that existed initially and of domains that retain the direction of spontaneous polarization. One of two types of periodic domain structures is usually formed: with 'head to tail' (Fig. 1a) or with 'head to head' (Fig. 1b) orientation of the domains. Lately, considerable progress has been made in forming periodic domain structures with domain sizes ranging from several micrometers to several millimeters, with the most recent results showing that it is possible to form domains of nanometer sizes [24, 25].

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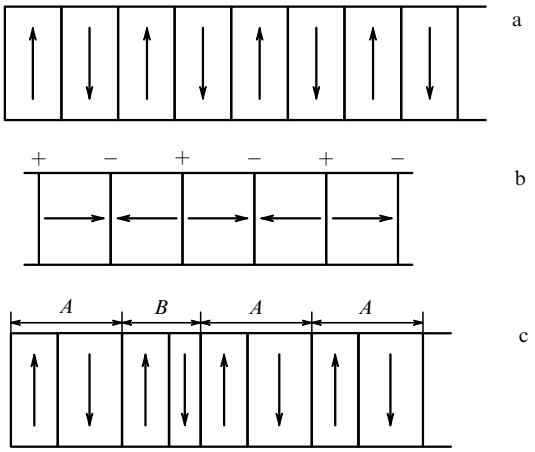

Figure 1. Different domain structures: (a) 'head to tail' and (b) 'head to head' periodic structures, and (c) a quasi-periodic structure.

The first periodic domain structures formed in ferroelectrics were used, as was proposed earlier by Armstrong et al. [1] and Seavey [2], to convert coherent light into the second harmonic [26 – 28] and, later, to generate and convert acoustic vibrations [29 – 33]. In both cases the efficiency of conversion via periodic domain structures was found to be much higher than when the same single-domain crystals without periodic domain structures were used.

Earlier studies of magnetic substances acting as transducers of various types of waves dealt primarily with single-domain samples [34] (the same was true in the case of ferroelectrics). Only later, theoretical studies of domain structures and the possibilities of using such structures for practical applications began [35]. During recent years, various mechanisms of formation in magnetic substances of dynamical domain structures by the field of an ultrasonic wave have been elaborated [36 – 39]. Further experiments will certainly make it possible to study other nonlinear effects in greater detail. For instance, we believe that the possibility of magnetically and thermally controlling the domain parameters in multilayer thin-film magnetic structures [6] and magnetic semiconductors [40] with giant magnetoresistance is very promising.

At the same time, studies began of quasi-periodic domain structures (such structures also became known as quasi-periodic superlattices) [41] in which the building blocks are 180° domains differing somewhat in size (Fig. 1c). In such systems the phase matching of the first and second harmonics depends on the difference in domain sizes, with the result that the generation of the second optical or acoustic harmonic may occur over a broader frequency range. Hence, the periodic and quasi-periodic structures may be considered an analog of one-dimensional quasicrystalline structures with the same diversity of extraordinary physical properties [42, 43].

We introduced the concept of induced domains in an attempt to focus attention on the possibility of creating domains of specific configurations via external actions and in this way forming periodic structures in which such domains are the building blocks. Moreover, the induced domains and periodic domain structures are dynamical, i.e., they have the capacity for undergoing spatial and temporal transformations depending on the intensity and nature of the external agents. It is these induced domains that are the most interesting entities for use in various new electronic devices.

Thus, the results of numerous studies of induced domains and the possible applications suggest that a new avenue of research has developed that combines the investigation of the various properties of electrically and magnetically ordered materials used in nonlinear optics and acoustics. The need for a review stems from the fact that the number of studies devoted to the new ways in which periodic domain structures are formed and to the various applications is rapidly growing while no generalizing papers in this field have appeared so far.

2. Methods of formation of induced domains and periodic domain structures

A domain structure that spontaneously emerges in ferroelectric or magnetic substances is usually in equilibrium and corresponds to a minimum in the crystal's energy. When an external field is introduced, the domain structure changes due to the growth of domains oriented along the field. At a certain critical value of the field, the structure is transferred to the single-domain state. The field evolution of the domain structure constitutes the basis for most methods of formation of domains and domain structures with specific parameters.

2.1 Formation of ferroelectric domains in electric fields

As is known (see Ref. [44]), the formation of 180° domains in oxide ferroelectrics occurs because of the noncentrosymmetric arrangement of metal ions (e.g., Li and Nb for lithium niobate) in relation to the sublattice of oxygen anions, with the direction of displacement of the cations determining the direction of the polarization vector in the domain (Fig. 2). Thus, polarization switching occurs when the ions are shifted from one noncentrosymmetric position to another noncentrosymmetric position along the spontaneous-polarization axis. Hence, all methods known to date can be divided, according to the polarization-switching process, into two groups: those that use external electric fields, and those that use the gradients of the internal magnetic fields.

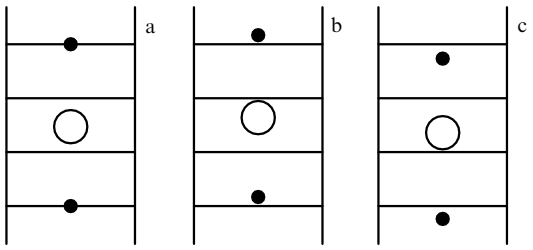

Figure 2. Fragment of the structure of a lithium niobate single crystal: (a) paraphase, (b) positive domain, and (c) negative domain; ○ niobium ions and ● lithium ions.

2.1.1 Application of external fields. The study of the behavior of ferroelectrics in strong electric fields began in the 1950s, when Merz [45] detected electrically induced polarization in barium titanate. Later, Kovalevich et al. [46] detected, via the Barkhausen effect, a change in the direction of polarization in single-domain crystals of lithium niobate, with the field strength needed for polarization switching dropping from 10^6 V cm^{-1} at room temperature to 10^4 V cm^{-1} at $T = 150\,^{\circ}$C. In more recent works [47 – 50], external electric

fields were used to form extended periodic domain structures with domain sizes of several microns.

The method of formation of periodic domain structures consisted in depositing periodically spaced metallic electrodes on the surface of thin samples (thickness $d < 1$ mm) perpendicular to the polarization axis C (Fig. 3) [47]. When an electric field whose magnitude exceeded that of the polarization field and whose sign was opposite to that of the polarization field was applied to the electrodes, an inverted domain structure of the head-to-tail type was formed, with the domain depth proportional to the time of action and the strength of the applied field. The domain walls were parallel to the C axis. This method was used to form periodic domain structures in crystals of lithium niobate, lithium tantalate, and barium titanate. Since polarization switching in such rigid structures as lithium niobate and lithium tantalate requires the use of fields whose strength at room temperature is $E \sim 10^6$ V cm^{-1}, some researchers induced polarization switching at lower fields by heating the crystals using, say, the radiation from pulsed lasers. However, the most effective approach is to apply electric field pulses through a system of electrodes. As established in the experiments conducted by Merz [45] and Zhu et al. [50] (Fig. 4), the dependence of the polarization-switching time t_s on the electric field strength E exceeding the coercive field E_c is given by one of the following formulas:

$$t_s \sim \exp\left[-\alpha(T)E\right] \tag{1}$$

(for small values of E) or

$$t_s \sim \frac{\beta(T)}{E - E_c} \tag{2}$$

(for large values of E), where $\alpha(T)$ and $\beta(T)$ are coefficients determined by the temperature at which polarization switching occurs.

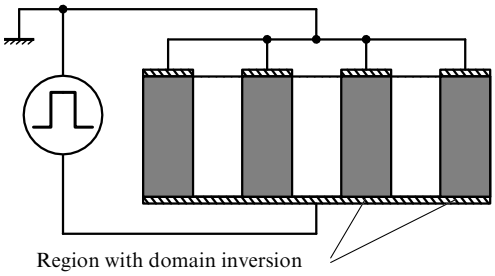

Figure 3. Formation of a periodic domain structure in a pulsed electric field.

The minimum sizes of domains of the head-to-tail type formed by applying pulsed electric fields perpendicular to the spontaneous-polarization field amount to about 2–10 μm in plates with a thickness up to 0.5 mm. These sizes are determined by the possibility of creating a system of metallic electrodes in ways developed earlier for interdigital transducers of surface acoustic waves. The main difficulty in dealing with high-coercivity materials, which amounts to the possibility of electric breakdown of the space between the electrodes in the applied field, was overcome by using liquid electrolytic contacts [51].

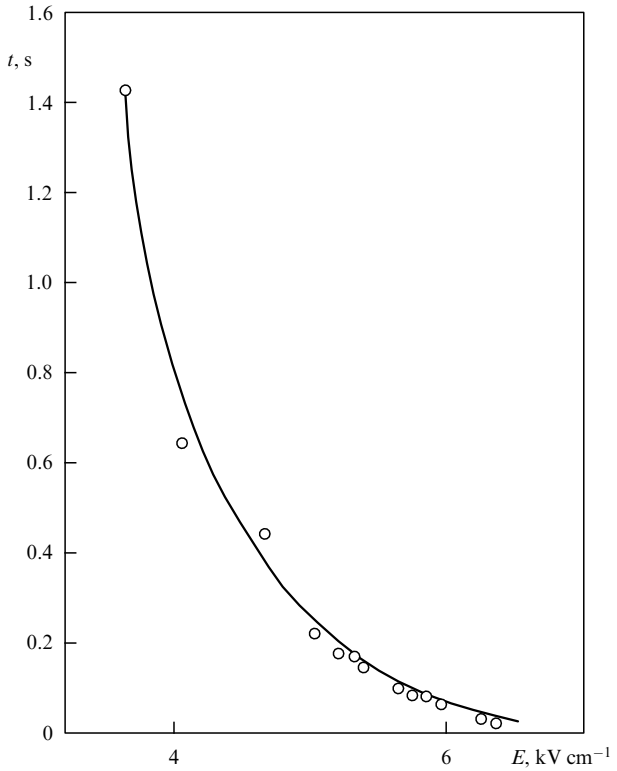

Figure 4. Polarization switching time of a lithium niobate single crystal as a function of the strength of the external electric field.

2.1.2 Formation of periodic domain structures in internal fields. The first regular domain structures were obtained in ordinary technological processes used to form single-domain samples both directly in the process of growing using the Czochralski method and by heat treatment after growth. A detailed history of the formation of domain structures can be found in Aleksandrovskiĭ's review [13]. In the first method, the domain structure was formed in the process of cooling to the Curie point (T_C) of a melt containing spatial gradient concentrations of impurity ions (e.g., yttrium) [15, 52], with the structure's period determined by the gradient distribution of the impurity. In the second method, to create a region with inverted polarization, heating to temperatures close to T_C was combined with diffusion of titanium ions or protons into the crystal or, on the other hand, with out-diffusion of oxygen ions or lattice-forming ions from the crystal [30, 53].

The method of forming a periodic domain structure by scanning the surface of the ferroelectric with a narrow electron beam is, in a certain sense, close to that just described [54]. The beam of a scanning electron microscope was used to form micron-sized domains.

A general feature of these methods is the production of a large number of free electrons, which form a space-charge region. The electric field generated in this process is in opposition to the spontaneous-polarization field and is responsible for polarization inversion, especially at high temperatures.

The microscopic model of free-electron production has been developed most thoroughly for $LiNbO_3$ and is based on the diffusion of ions of oxygen and lithium from the crystal at high temperatures [23]. Here, the emerging vacancies are substituted either by the structural niobium ions or by

impurity ions (yttrium, titanium, etc.). In both cases the difference of the charge states of the diffusing ions and those of the structural or impurity ions leads to the production of free electrons from vacancy centers, which form a charge compensation of the substitution process. The electrons, with a high concentration near the surface, then diffuse into the bulk of the crystal and generate the field E_d of the space charge. When there is bidirectional diffusion, the field E_d is antiparallel to the spontaneous-polarization field near the $+C$ polar surface, where polarization switching occurs.

The value of the electric field generated by the free electrons can be written as

$$E_d = \frac{kT}{q}\left(\frac{\Delta N}{N} + n\right), \tag{3}$$

where ΔN is the gradient of the charged centers N along the polarization axis, q is the carrier charge, and n is the electron concentration.

Qualitative estimates of E_d made on the basis of the experimental data on the profile of N in lithium niobate have shown [53] that at 1100 °C the induced electric fields may be as high as 200 – 300 V cm^{-1}, which is quite sufficient for polarization switching to occur even in ferroelectrics with a high coercive field. For instance, for $LiNbO_3$ the polarization-switching field at such a temperature is only 5 V cm^{-1} [23]. The depth of the inverted domains formed by this method may reach 500 μm. A similar model is valid for other oxide ferroelectrics.

2.2 Light-induced domains and periodic domain structures in ferroelectrics

Soon after the discovery in 1966 of a light-induced change of the refractive index in a number of ferroelectrics, which became known as the photorefractive effect [55], the first photorefractive (or, alternatively, holographic) gratings were produced [56]. Such gratings are formed when a crystal surface is irradiated with two interfering light beams and consist of alternating stripes differing in their electric-field gradients and, due to the electrooptical effect, in the gradients of the refractive index. Light-induced fields, especially in crystals containing impurity ions with a variable valence can be as high as 2×10^5 V cm^{-1} and can generate induced variations of the refractive index as high as 10^{-3}. However, holographic gratings have found only limited application in converting light and acoustic waves [57, 58] and in recording data in optical or acoustic form [59, 60]. Their main drawback is that the record can easily be erased by reirradiation or heating above 180 °C. Hence special methods of fixation of such gratings and holograms based on the gratings have been developed. Since the fixation method presupposed the use of light irradiation in the 120 – 150 °C temperature range and the strength of the induced fields reached values in the 10^4 – 10^5 V cm^{-1} range, individual light-induced domains were probably formed in some of the experiments (e.g., see Ref. [61]). Phenomenologically, the formation of inverted domains was explained thus: the impurity-ion field generated as a result of spatial displacement of light-excited electrons creates a new orientation of polar complexes, which leads to local polarization switching of the formerly single-domain sample. Domain formation was also detected by Micheron and Bismuth [62], who, in addition to irradiating $SrBaNb_2O_6$ and $BaTiO_3$ crystals with light, applied an electric field whose direction was opposite to that of the spontaneous-polarization field. At that time (the 1970s), only the lack of reliable methods of detecting domains made it impossible to corroborate the earlier experimental facts with a high degree of accuracy.

Convincing proof of the formation of light-induced domains and domain structures in ferroelectrics was obtained much later, in the 1990s [63 – 66]. The first periodic domain structures were formed in barium titanate crystals by Cudney et al. [63], who used two interfering laser beams (both of $\lambda = 488$ nm) and applied a field whose strength was below that of the coercive field. The result was a periodic domain structure with domain sizes of roughly 20 – 25 μm, which corresponded to the period of the interference optical structure. The size of the domains and the perfection of the new structure were later corroborated by successful generation of the second laser harmonic [67]. The same laser-electric method was used to form a periodic domain structure in a $Sr_{0.25}Ba_{0.75}Nb_2O_6$ single crystal [65].

The formation of inverted domains can be explained by a model in which the field of the photoexcited charges is compensated by the fields of the domain walls [66]. In the course of photoexcitation, the charges localized on the impurity or lattice ions drift in the applied field E and recombine on other ions, creating a spatial redistribution of charges (Fig. 5)

$$\rho_d(E) = \rho_0(E) \sin \frac{2\pi z}{D}, \tag{4}$$

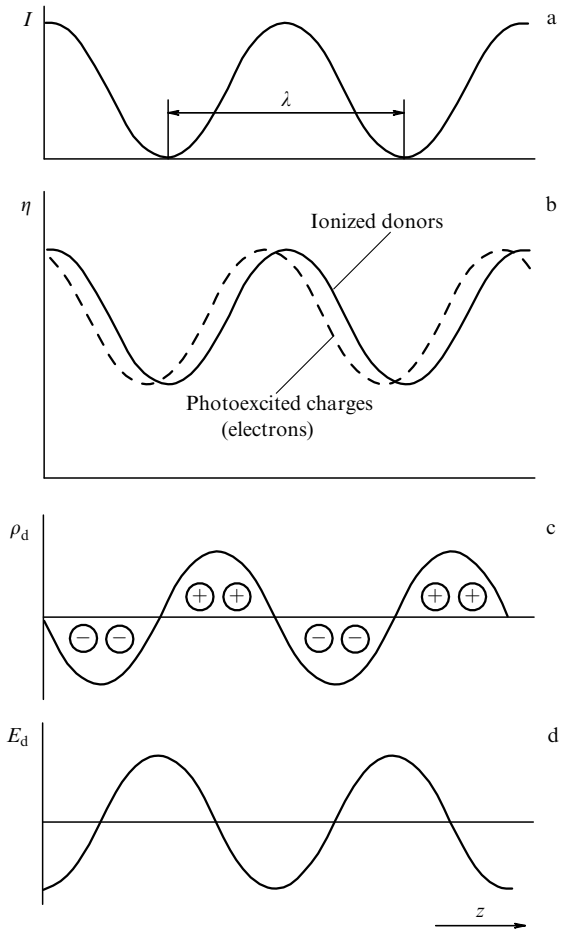

Figure 5. Space distributions of (a) the intensity of the light beam, (b) photoexcited charges, (c) charged centers, and (d) the electric field of the charges.

where ρ_0 is the charge density depending on the state of the ions and the strength of the applied field, z is the distance along the polarization axis, and D is the period of the interference bands.

This charge distribution generates a space-charge field E_d. When the total field strength $E + E_d$ reaches the value of the polarization-switching field E_c, domain centers with inverted polarization begin to form. As the primary domains coalesce into macrodomains with inverted polarization, the formation of domain walls begins and of domain charges with a density ρ_b that can be defined as $\rho_b = \Delta P_s$, where P_s is the spontaneous polarization. In a single-domain crystal $\Delta P_s = 0$, since spontaneous polarization is homogeneous. In a polydomain crystal, the magnitude of P_s changes abruptly from one domain to another.

The orientation of the domain walls in the process of nucleation and coalescence depends on the interaction between the emerging domain structure and the vector of the space-charge field E_d. It is assumed that the stable state of the boundary charges spatially follows the free-charge distribution, so that in a polydomain sample there is almost a perfect compensation between the free charges and the boundary charges:

$$\rho_b(k) = -\sigma\rho_d(k)\,, \tag{5}$$

where σ is the degree of compensation; $\sigma < 1$.

It has been found that when a domain structure is formed in barium titanate crystals, σ reaches values of roughly 0.8–0.9.

The need to apply an electric field in crystals with high conductivity is chiefly determined by the mechanism of photoexcited charge transfer. In high-resistance ferroelectrics such as lithium niobate, the main mechanism of such transfer is not drift but the photogalvanic mechanism, which does not require an additional external field. Hence, the inversion of spontaneous polarization can, theoretically, be achieved at the expense of the light-induced field E_d alone. This assumption was corroborated by experiments after the discovery of inverted microdomains in the form of needles that align themselves along the polarization axis as a result of irradiating the lithium niobate surface with a laser beam [68]. Microdomains were obtained in the single crystal $LiNbO_3 : 10^{-2}$ at.%Fe by solely irradiating the sample with a laser beam with $\lambda = 0.53$ μm at 150 °C [69]. The need to raise the temperature is related to the fact that E_c reduces from 10^6 V cm^{-1} at $T \sim 20$ °C to 3×10^4 V cm^{-1} at 150 °C. When the light spot was a narrow (0.2–10 mm) band, the width of the inverted domain was 150–200 μm and followed the shape of the region irradiated with the laser beam.

The microscopic model of generation of a light-induced field that is sufficiently strong to initiate polarization switching in a ferroelectric follows from the manner in which impurity ions with variable valence (Fe, Cr, Mn, Cu) are incorporated into the crystal lattice [68, 69]. The model has been developed most thoroughly for iron ions constituting the main component uncontrollably entering into ferroelectric crystals and determining the magnitudes of the light-induced fields of the space charge [70, 71]. It is common knowledge that the Fe^{2+} and Fe^{3+} ions occupy the same positions in the crystal lattice, e.g., substituting for lithium ions in $LiNbO_3$. Due to the conditions of charge compensation, the iron ions are shifted somewhat along the polarization axis relative to the positions of lithium ions. The Fe^{2+} ions are donor centers and the Fe^{3+} ions, acceptor centers. As a result of laser irradiation, electrons are photodetached from Fe^{2+} ions. The photogalvanic field shifts the electrons along the polarization axis, which are then captured by Fe^{3+} ions. This leads to a spatial redistribution of the charges of the iron ions, which results in the emergence of a strong galvanic field directed opposite to the spontaneous-polarization field. When the ratio of the numbers of acceptor and donor centers is optimal, the increase in the strength of the induced field is one to two orders of magnitude larger than in pure ferroelectrics, with the overall concentration of impurity centers being no higher than 10^{19} cm^{-3}.

2.3 Ferroelectric periodic domain structures in the field of an acoustic wave

Late in the 1970s, studies began of the nonlinear interaction between acoustic waves and photoexcited electrons in photorefractive crystals with a large piezoelectric effect [72, 73]. Soon after, it was discovered [58] that the electric field accompanying the acoustic wave changes the space distribution of the electrons. A photorefractive grating was recorded on a $LiNbO_3$ crystal by irradiating the crystal simultaneously with a homogeneous light beam and a standing acoustic wave [74, 75]. Later, a similar grating was written by irradiating the crystal with X-rays [76]. Although in these studies no regular domain structures were discovered, the possibility of detecting such structures was becoming more and more real. The basic difficulty consisted in creating such gradients of the electric fields generated by the field of the acoustic wave that would be strong enough to form domains with inverted polarization. The main reason for the unsuccessful attempts was that the experiments were done at room temperature, with the acoustically induced gradients of the electric field being not strong enough to initiate polarization switching.

Only when the ratio of the ion concentrations was optimal, $Fe^{2+}/Fe^{3+} \sim 0.3$, and the temperature was in the 130–150 °C range did it become possible to form a periodic domain structure in lithium niobate using a standing surface acoustic wave with a relative-deformation amplitude of order 10^{-4} and simultaneous exposure to a homogeneous laser beam with $\lambda = 0.53$ μm [77]. The period of the periodic domain structure corresponded to the wavelength of the standing surface acoustic wave. Since the surface acoustic wave propagates along the polarization axis, a head-to-head structure was formed with domain sizes of about 35 μm. The depth of the domains measured from the surface was 20–30 μm. The domains proved to be fairly perfect, and the resulting periodic domain structure was used for laser generation of acoustic waves.

The mechanism of formation of periodic domain structures in the field of an acoustic wave is almost the same as in the optical method described above, and the acoustic field only shifts the photoelectrons to an antinode of the standing wave [78].

2.4 Quasi-periodic domain structures

The possibilities of using quasi-periodic structures consisting of a certain domain sequence in ferroelectrics were first discussed by Zhu et al. [41, 79]. Actually, such quasi-periodic structures are similar to a one-dimensional quasicrystal with incommensurate domains alternating in a certain order described by the Fibonacci sequence [43]. The quasi-periodicity in the Fibonacci sequence can be realized for blocks A

and B in Fig. 1c according to the following rule:

$$S_j = \frac{S_{j-1}}{S_{j-2}}\,, \qquad j \geqslant 3\,,$$

or

$$S_1 = |A|\,; \qquad S_2 = |AB|\,; \qquad S_3 = \left|\frac{AB}{A}\right| \ldots \tag{6}$$

Here, the ratio of the domain sizes d_A and d_B must satisfy the 'golden section' condition:

$$\frac{d_A}{d_B} = \tau = \frac{\sqrt{5}+1}{2}\,. \tag{7}$$

In this case, the wave vector of the quasi-periodic domain structure is

$$k = 2\pi\,\frac{m+n\tau}{D}\,, \qquad m,n = 1,2,3\ldots, \qquad D = \tau d_A + d_B\,. \tag{8}$$

A periodic domain structure has the most prominent properties needed, as we will show shortly, for phase wave matching [80, 81].

The simplest way to ensure the formation of quasi-periodic structures in ferroelectrics is to apply pulses of an electric field to a system of electrodes arranged in a given sequence.

2.5 Domain structures in magnetic materials

When there are no external magnetic fields, magnetic materials usually have a spatial domain structure. In uniaxial crystals, with one direction of easy magnetization ('easy axis'), the magnetizations of the neighboring domains are antiparallel. In multiaxial crystals, the domain magnetizations can make angles that differ from 180° and correspond to the angles between the various directions of easy magnetization. In some magnetic materials, the spontaneously formed domain structures have more complicated configurations, e.g., those of a 'labyrinth,' 'herringbone,' and 'honeycomb' [82].

As noted earlier, the simplest way to control the domain structure is to apply a magnetic field. After the field is switched off, the magnetic domain structure induced by it is partially retained and, hence, a metastable structure emerges.

For instance, in multiaxial antiferromagnets, whose characteristic feature is spontaneous magnetization in the form of 180° and 90° domains, application of an external field transforms 90° domains into 180° domains [83]. Since the magnetization process exhibits hysteresis, there is a region of magnetic field strengths in which there is a metastable state only for a 180° domain structure of a definite configuration.

The above method of forming domain structures has been used for thin magnetic films with a thickness of less than 1 μm. A fundamental property of such films is magnetic anisotropy, which determines the type of magnetic structure. In films whose magnetic anisotropy is perpendicular to the film surface, round magnetic bubbles can be formed by applying a magnetic field along the easy axis. Within a certain range of strengths of the applied magnetic field, there emerge equilibrium magnetic bubbles, whose movement along the film is caused by the action of a magnetic-field gradient [11].

In recent years, there has been an upsurge of interest in ultrathin magnetic films with thicknesses of several atomic layers. Various size effects manifest themselves in such films. For instance, for multilayer magnetic structures consisting of alternating ferromagnetic (Fe, Co) and nonmagnetic (Cr, Cu) metallic layers, Baibich et al. [5] found that there is an indirect exchange interaction between the electrons in different ferromagnetic layers. The interaction oscillates with the thickness of the nonmagnetic layer and leads either to collinear ferromagnetic ordering of magnetization or to collinear antiferromagnetic ordering in the adjacent magnetic layers. Such a structure with the nature of magnetic ordering that changes from layer to layer can be interpreted as a system of magnetic domains.

For the more complicated case of a multilayer structure consisting of alternating ferro- and antiferromagnetic layers, Morozov and Sigov [6] found that splitting of the monatomic layers near the boundary into domains becomes advantageous energywise. Inside each domain, the magnetization is either parallel or antiparallel, depending on the nature of the exchange interaction. Thus, an extended monolayer domain structure can arise in a multilayer magnetic structure. The emergence of domain walls mixes the spin states of the electrons and contributes considerably to magnetoresistance. The external magnetic field that causes reorientation of the magnetizations from antiferromagnetic to ferromagnetic changes the resistance by hundreds of percent even at room temperature.

3. Kinetics and mechanisms of formation of induced domains and periodic domain structures

3.1 Induced domains in ferroelectrics

Domain formation in ferroelectrics has been studied most thoroughly by applying pulsed electric fields with amplitudes exceeding the coercive field E_c. Observations of polarization switching have been carried out by studying the temporal dependence of the coercive fields, the polarization switching currents [47–49] and directly, by using scanning tunneling and scanning atomic-force microscopes [25]. It was found that the inversion of the initial domain state initiated by applying an external field E whose direction is opposite to that of the spontaneous-polarization field is a multistage process. First, domains in the form of sharp nanometer-sized needles form near the electrodes, then the domains align themselves over the bulk of the crystal, and finally reach the lateral surfaces of the crystal. At the same time, the domains coalesce, reaching several microns in size. Depending on the duration and intensity of the electric-field pulses, one can observe both the formation of separate nano- and micrometer-sized domains and the formation of different types of quasi-periodic structure. For instance, in lithium niobate, stripe domains with a width smaller than 100 nm and chains of round and triangular domains 30–50 nm in diameter with a linear density of 10^4 mm^{-1} emerge.

The way of formation of primary nuclei also depends on the structure of the metallic electrodes. When homogeneous electrodes are deposited on opposite surfaces perpendicular to the polarization axis, the inverted domains first randomly are formed near the surface and then protrude into the bulk in the form of needles or triangles. For the periodic electrode structure, primary domains in the form of pointed wedges first are formed at the polar surface (0001) along the edges of the electrodes (Fig. 6a) [25]. Then, new domains penetrate the structure in polar and lateral directions (Fig. 6b) and occupy the entire area under the electrodes (Fig. 6c); at this stage, the

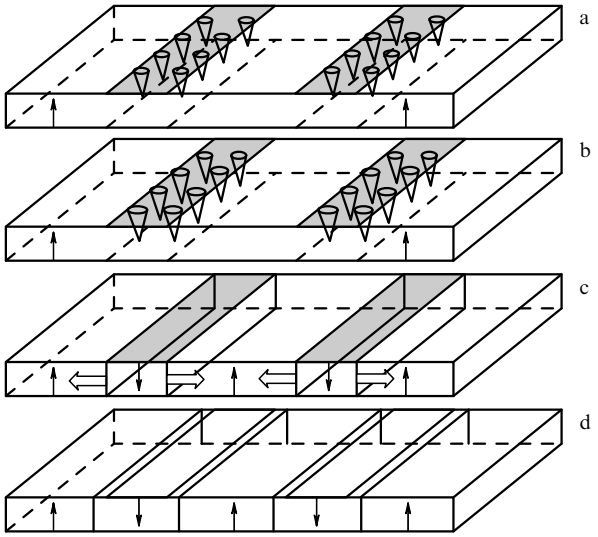

Figure 6. Formation of a periodic domain structure in a ferroelectric placed in an electric field.

domain size is determined by the surface area of the electrodes. At the last stage, the flat domain walls of the macrodomains that have formed move away from the electrodes (Fig. 6d). The lateral shift stops because of a decrease in the strength of the local polarizing field beyond the electrodes.

A manifestation of domain formation in an electric field is the temporal variation of field magnitude needed for polarization switching. The experimental data of Mizuuchi and Yamamoto [47], Chao et al. [48], and Wang et al. [49] suggest that after the condition $E \geqslant E_c$ is met, the direction of spontaneous polarization changes and, at the same time, an internal field is generated. This field is formed by the depolarizing field E_{dep} and the space-charge field E_d. The depolarizing field in a ferroelectric is caused by the presence of surface charges and hinders the domain growth:

$$E_{dep} = \frac{2L_D P_s}{\varepsilon\varepsilon_0 L}\,, \tag{9}$$

where L_D and L are the Debye and crystal lengths, respectively.

The space-charge field E_d is associated with several dynamical processes. First, with the spatial shift along the polarization axis of the crystal-lattice-forming ions in the polarization-switching field (see Fig. 2). Second, with the redistribution of free charge carriers and their possible capture by structural or impurity centers. Third, with the spatial redistribution of impurity ions of the iron group, which are usually present in crystals of oxide ferroelectrics. The low mobility of ions and electrons in low-conductivity ferroelectrics determines the sufficiently long relaxation time of the field E_d. For oxide ferroelectrics, the relaxation time τ of the field E_d varies from a few seconds to several thousand seconds.

The field E_d to a large measure compensates the coercive field. Thus, the formation of microdomains is due to the action of the total local electric field

$$E_\Sigma(\mathbf{r}_i, t) = E + (E_d - E_{dep})\,, \tag{10}$$

where $\mathbf{r}$ is the local coordinate.

Qualitatively, polarization switching can be described by the following equation:

$$E_\Sigma = E_c - E_d \exp\left(-\frac{t}{\tau}\right). \tag{11}$$

Partial compensation of the coercive field leads to the practical conclusion that it is possible to control domain formation by applying weak electric fields in the time interval when there is a strong internal field.

The most complicated and yet-to-be resolved question is which centers are the nuclei of domains. For crystals containing impurity ions, the most probable event is nucleation near impurity centers that are strongly coupled to the lattice environment and are characterized by an intrinsic gradient of electric fields in opposition to the spontaneous-polarization field. Among such ions are Fe^{2+}, Mn^{3+}, Cr^{2+}, Cr^{4+}, and a number of other ions of the iron group. Since the spontaneous-polarization fields surrounding these ions are partially compensated by the intrinsic field of the ion, the conditions for polarization switching are less stringent. Nucleation can also occur near such structural ions that are substitutes for other structural ions in the lattice for crystals of congruent composition. For instance, for lithium niobate such ions are niobium ions substituting for lithium ions.

3.2 Dynamical magnetic domain structures

As shown theoretically by Kabychenkov and Shavrov [36, 84], acoustic waves can induce dynamic periodic domain structures in homogeneously ordered magnetic materials, with the width of the domains and domain walls determined by the wavelength of the acoustic wave and the amplitude of that wave. The reason for this is the appearance of an additional uniaxial magnetic anisotropy caused by magnetostriction. This effect should manifest itself most vividly near easy-axis–easy-plane type orientational magnetic phase transitions. When the amplitude of the acoustic wave is sufficiently large, there are periodic variations of the value and sign of the magnetic anisotropy constant, which lead to easy-plane anisotropy in the compression region and to easy-axis anisotropy in the expansion region (provided that the magnetostriction constant γ is positive). If the anisotropy energy is much higher than the exchange energy, a sample that was earlier magnetically homogeneous will 'split' into separate layers with alternating direction of magnetization: along and across the direction of propagation of the acoustic wave. When a traveling wave propagates in the magnetic substance, the induced domain structure moves along the sample with a speed equal to that of the acoustic wave; when a standing wave is present in the sample, the periodic domain structure has a stable spatial distribution determined by the period of the standing wave.

Thus, a stripe-domain structure with 90° domain walls can form in magnetic substances. It has been found that the magnetization is able to adjust itself to the elastic deformations generated by the acoustic wave if the relaxation time is shorter than the vibration period. Acoustic vibrations with frequencies up to several megahertz satisfy these conditions for real magnetic substances.

Qualitatively, the kinetics of formation of acoustically induced domains differ from processes of ordinary domain formation in magnetic substances. Since the relationship between the domain width d and the width of the domain wall δ depends on the deformation amplitude u in the acoustic

wave as

$$d = \frac{\lambda}{2} - \delta\,, \tag{12}$$

where $\delta = (\alpha\lambda/\gamma u)^{1/3}$, and λ is the inhomogeneous exchange constant, the width of the domain wall decreases with an increase in u while the boundary width increases to $\lambda/2$. The stripe-domain structure also emerges at $\delta \approx d$, but the change in the direction of magnetization is less than 90° in this case.

The theoretical calculations were verified by subsequent experiments. For instance, Chetkin and Lykov [39] detected a traveling domain structure generated by a longitudinal acoustic wave propagating in the easy-axis antiferromagnet $FeBO_3$. Standing and traveling periodic domain structures were observed by Avakyan et al. [85] in hematite. Here, the traveling periodic domain structures moved with a speed equal to that of the acoustic wave. Resonant amplification of acoustic vibrations was observed [86] on an induced domain structure in iron borate under conditions where an integral number of domains was distributed within the sample's bulk.

4. Propagation and generation of light and acoustic waves in periodic domain structures

A periodic domain structure can be considered as a system of anisotropic media separated by walls at which, depending on the medium's symmetry class, the electric, electrooptical, elastic, piezoelectric, or magnetoelastic properties change. Specifically, for noncentrosymmetric oxide ferroelectrics, the sign of the elements of the third-rank tensors describing the quadratic dielectric susceptibility and the piezoelectric effect changes at the walls of 180° domains. Thus, the domain walls provide an additional contribution to the optical or acoustic nonlinearity of the crystal and in this way broaden the possibilities of using such linear and nonlinear optical and acoustic effects as reflection, refraction, and generation of the fundamental and higher harmonics and parametric frequency conversion.

4.1 Propagation of light

Since the dielectric susceptibility and permittivity tensors χ and ε that describe the optical properties of dielectric media do not change from domain to domain in a periodic domain structure, the propagation of a light wave is independent of the domain structure. However, when a constant electric field is applied to the periodic domain structure, the value and sign of the refractive index and the dielectric constant vary periodically due to the electrooptical effect.

The change in the refractive index, Δn, leads to a phase shift φ in the wave propagating through the domain:

$$\varphi = \frac{\pi r n_e^3 E L}{\lambda}(\mathbf{E}_0\mathbf{P}_0)\,, \tag{13}$$

where r is the electrooptical coefficient, n_e is the extraordinary refractive index of the medium, $\mathbf{P}_0$ and $\mathbf{E}_0$ are the unit polarization vector and the unit vector of the applied field, and L is the sample length in the direction of propagation of the ray.

Due to the difference in signs of the coefficient r, the phase angles also have opposite signs in the adjacent domains. The maximum effect occurs when the polarization vector in the domain and the electric field vector are collinear, which makes it possible to consider the periodic domain structure as a phase grating with a period equal to that of the periodic domain structure.

When an electric field is applied to the periodic domain structure perpendicular to the domain wall, periodic variations of the values and sign of the elements of the dielectric tensors set in. Due to the smallness of the effect [10], the change in the tensor ε can be represented by a periodic perturbation:

$$\Delta\varepsilon = \varepsilon r_{ijk} E n_o^2 n_e^2 l\,, \tag{14}$$

where r_{ijk} is the corresponding element of the electrooptical tensor, n_o is the medium's ordinary refractive index, and $l = \pm 1$ for domains with positive and negative polarizations, respectively.

The electrically induced variation $\Delta\varepsilon$ can be interpreted as a rotation of the optical axes in each domain through the angle

$$\varphi = l \arctan\left[2rE\left(\frac{1}{n_o^2} - \frac{1}{n_e^2}\right)^{-1}\right]. \tag{15}$$

A coherent light beam propagating along a periodic domain structure under the given conditions is reflected and refracted at each interface. As a result, there emerges a complex structure of waves propagating in the forward and backward directions in relation to the primary waves and interacting with each other. In the coupled-modes theory [10] describing the propagation of electromagnetic radiation in a periodic layered medium, all interacting waves are reduced to two waves (wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$) that propagate either in the same direction or in the opposite direction. Two conditions are needed for the waves to strongly interact:

(1) longitudinal phase matching:

$$\Delta k = k_1 - k_2 - m\frac{2\pi}{d} = 0\,, \qquad m = 1, 2, 3\ldots, \tag{16}$$

for waves propagating in the same direction, and

$$\Delta k = k_1 + k_2 - m\frac{2\pi}{d} = 0 \tag{17}$$

for waves propagating in opposite directions;

(2) a dynamic relationship between the wave vector and the length L of the periodic domain structure: maximum transmission (Fig. 7) of the light beam through the periodic domain structure corresponds to

$$|k_m|L = (2m+1)\frac{\pi}{2}\,, \qquad m = 0, 1, 2, 3\ldots, \tag{18}$$

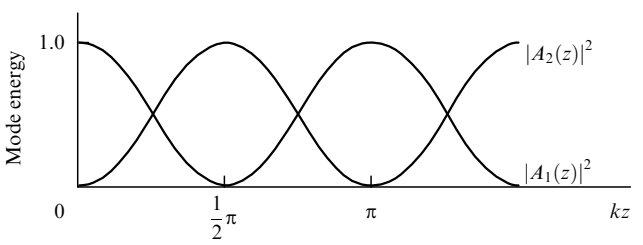

Figure 7. Spatial distributions of the energies of two beams propagating in the same direction.

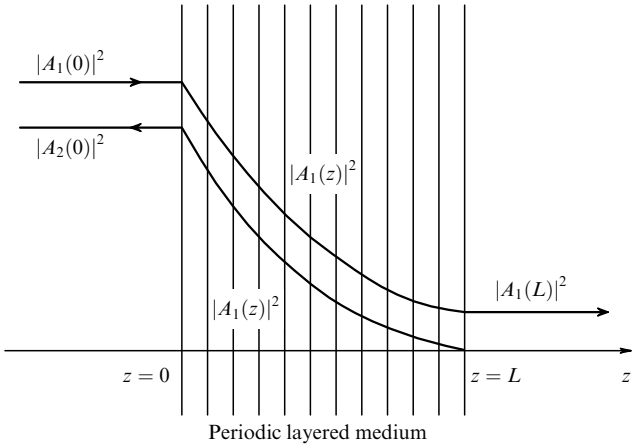

Figure 8. Spatial distributions of the energies of two beams propagating in opposite directions.

and maximum reflection (Fig. 8) corresponds to

$$|k_m|L = m\pi\,. \tag{19}$$

Experimental studies of the propagation of coherent light beams through periodic domain structures with electric fields applied to the sample began at the Moscow State University in the 1970s. Aleksandrovskiĭ et al. [87] found that there is diffraction of the light beam on a periodic domain structure formed in crystals of barium sodium niobate and developed the first design for a modulator. After that, light beam diffraction was observed by Blistanov et al. [26] in crystals of lithium niobate in which a more perfect structure with a period of 40 μm had been formed. The researchers found that when a plane-polarized beam of light propagates along domain walls with an electric field applied along them, Raman–Nath diffraction is observed and a conversion efficiency of 98% can be reached.

The use of periodic domain structures made it possible to considerably raise the speed with which the parameters of the light beam could be controlled and to build modulators with control in the gigahertz range [88]. Cheng [89] used a periodic domain structure in lithium niobate to build a highly effective deflector of light beams.

4.2 Propagation of acoustic waves

Acoustic waves propagating through periodic domain structures in piezoelectric ferroelectrics undergo reflection and refraction because of the difference in the signs of the piezoelectric coefficients in adjacent domains. In the general case of an arbitrary angle of incidence of the acoustic wave on a domain wall in a piezoelectric crystal, there emerge four reflected and four refracted waves (two transverse and one longitudinal acoustic waves and one electrostatic wave). Under certain conditions, there is also conversion of bulk waves into surface waves. However, the number of converted waves decreases when the wave propagates in a direction in which there are only pure modes. Analyses of special cases of the passage of waves through a periodic domain structure and their conversion for crystals with different symmetries can be found in Refs [9, 90–93]. As shown by Kessenikh and Shuvalov [91] and Gorkunova and Shuvalov [92], the coefficients of reflection and refraction of acoustic waves propagating along a periodic domain structure can be determined using the model of coupled modes, which in many respects is similar to the model used earlier for light waves. For transverse acoustic waves in ferroelectric antiphase gratings, the reflection and refraction coefficients are determined by the angle θ of incidence of the wave on the domain structure and the electromechanical coupling coefficients K. For instance, the square of the modulus of the reflectance has the form

$$|R|^2 = Q^2\left[Q^2 + \frac{\sin^2(k_1 D)}{\sin^2(Nk_1 D)}\right]^{-1}, \tag{20}$$

where $Q^2 = 4K^4 \tan^2\theta\left[\cos(k_2 d_A) - K^2\sin(k_2 d_A)\right]^2$, k_1 is the wave vector of the periodic domain structure, k_2 is the acoustic wave vector, and $D = d_A + d_B$ is the structure period.

A characteristic feature of the reflection spectrum is its high selectivity, similar to the case of propagation of light waves through a periodic domain structure. A quasi-periodic structure ($d_A \neq d_B$) exhibits modulation of resonance peaks in the intensity and the disappearance of some of these peaks, which is determined by the relationship between d_A and d_B.

Experimentally reflection of bulk acoustic waves was observed practically at the same time on a periodic domain structure in lead germanate [94] and on a holographic grating generated by a light beam in lithium niobate [95]. Later came the detection of reflection [32] and refraction [33] of surface acoustic waves on induced periodic domain structures. The results of these experiments, which are in full agreement with the results of theoretical calculations by Zhu et al. [31] and Tourlog et al. [96], demonstrated the possibility of using periodic domain structures to convert bulk and surface acoustic waves in the entire megahertz range.

4.3 Generation of acoustic waves

A periodic domain structure generated in a crystal that is placed in an alternating electric or magnetic field can be considered as a system of periodically arranged sources of acoustic waves (Fig. 9). These sources are formed by the domain walls whose piezoelectric or magnetoelastic coefficients have opposite signs. Effective generation occurs when the phases of the sources coincide, which means that the structure's period either coincides with the wavelength of the acoustic wave or is an integral multiple of that wavelength. Resonance of any type of acoustic wave on a periodic domain structure appears at the frequencies

$$f = \frac{nV}{D}\,, \qquad n = 1, 2, 3\ldots, \tag{21}$$

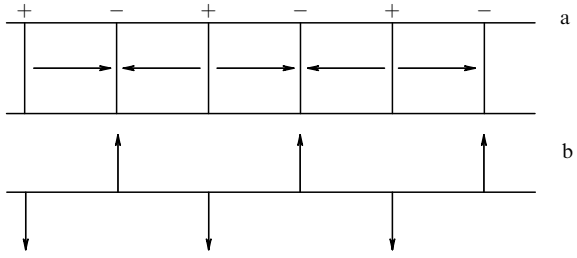

Figure 9. Periodic domain structure (a) and the corresponding sources of ultrasonic vibrations (b).

while for a quasi-periodic domain structure this happens at

$$f = \frac{(m + n\tau)V}{D}\,, \tag{22}$$

where V is the velocity of the bulk or surface waves.

The total length L of the periodic domain structure has no effect on the parameters of the resonance frequency but determines the value of the factor M characterizing the conversion of electrical energy into acoustical:

$$M \sim K^2 N_{\rm d}\,, \tag{23}$$

where K is the electromechanical or magnetoelastic coupling coefficient, and $N_{\rm d}$ is the number of domain walls.

The first experiments in the generation of surface and bulk acoustic waves by an alternating electric field were done on a light-induced holographic grating [98] and on an induced domain structure in lead germanate [97]. These experiments showed a relative increase (in comparison with single-domain samples) in the coefficients of conversion of the energy of the electric field into acoustic energy. A substantial broadening of the generation frequency range (up to 1000 MHz) was achieved by Zhu et al. [29, 31]; this became possible after developing methods for forming periodic domain structures with domain sizes of several microns. Later, Zhu et al. [79] obtained generation of acoustic waves in lithium niobate by exciting a quasi-periodic domain structure with an alternating field. The experimentally studied spectra of generated frequencies fully correspond to the theoretical calculations for periodic and quasi-periodic domain structures done by Cheng [99] and Chen et al. [100] (Fig. 10).

The theoretical aspects of generation of acoustic waves by domain walls in magnetic substances have been studied in Refs [101 – 104], where the various features of excitation of domain walls in different classes of magnetic crystals were investigated. In their experiments, Avakyan et al. [85] and Bogdanova et al. [104] studied the amplification of magnetoelastic vibrations by an acoustically induced periodic domain structure in iron borate.

In view of the realization of photorefractive gratings and periodic domain structures in ferroelectrics with a strong piezoelectric effect, it became possible to generate acoustic waves by acting with a laser beam on periodic structures. Note that this method, based on converting the pulses of a light-induced electric field into acoustic vibrations, differs from the well-known methods of laser generation of ultrasound [105, 106]. The latter use one of several macroscopic generation mechanisms: thermal expansion, surface evaporation, optical breakdown, or striction.

Since the writing and erasing of a light-induced holographic grating in a piezoelectric crystal are accompanied by a substantial change in the electric-field gradients [55 – 59], it was assumed (and this was later corroborated in experiments) that acoustic waves are generated in the interval between writing and erasing [107 – 109]. In the most perfect method of Deev and Pyatakov [109], pulse generation of acoustic waves was carried out by simultaneously irradiating a bismuth germanate crystal with (a) a spatially periodic, time-continuous laser beam that formed a holographic grating and (b) a pulsed laser beam from another source. The temporal periodicity of the acoustic pulses generated in this process was determined by the periodicity of the laser pulses, while the wavelength of the acoustic wave was determined by the period of the holographic grating. Ten years later, Petrov et al. [110] used light to excite surface acoustic waves.

Such a method of generating acoustic vibrations has proved to be useful in studies of photogeneration and relaxation of free carriers but could not be used as an effective method of generating acoustic waves. More convenient from this viewpoint is a method of generating acoustic waves based on laser pulsed irradiation of a periodic domain structure. This method was realized only recently when a periodic domain structure formed in lithium niobate was irradiated with short laser pulses with $\lambda = 0.53$ μm [111]. Each laser pulse irradiating the periodic domain structure was registered by an acoustic pickup detecting pulses (Fig. 11) that were related, according to the time of passage, to the signals of the surface acoustic wave (*1*), the transverse component of the near-surface acoustic wave (*2*), and the second harmonic of the surface acoustic wave (*3*). The centers

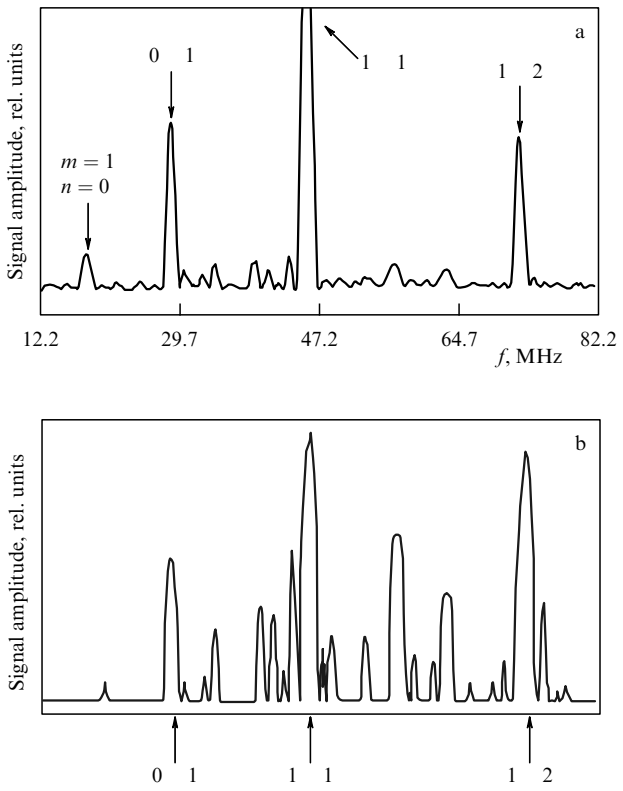

Figure 10. Spectra of ultrasonic vibrations for a quasi-periodic domain structure: (a) theoretical and (b) experimental.

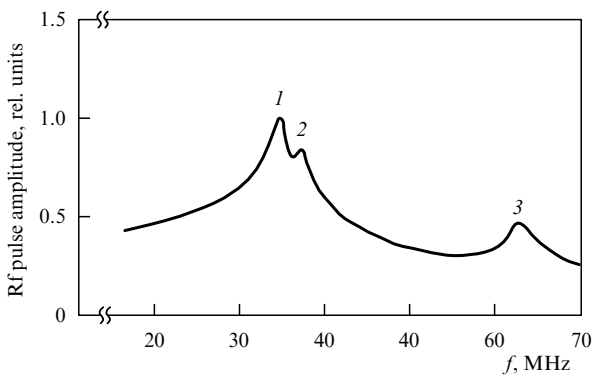

Figure 11. Spectra of laser generation of acoustic waves on a periodic domain structure in lithium niobate.

of the frequency spectrum of these types of wave correspond to the condition that the wavelength be equal to the period of the structure.

Qualitatively, the process of laser generation can be interpreted [112] as the variation of the electric field in each domain due to the photogeneration of electrons in the course of a pulse. The resulting jump in the electric field strength in each domain leads, due to the inverse piezoelectric effect, to generation of acoustic vibrations. Since the field induced by the laser is spatially homogeneous, the changes of the field in the adjacent domains have opposite signs, similar to the case of generation by an alternating electric field.

After a laser pulse has passed, the induced field relaxes. If the relaxation time is shorter than the interval between pulses, by the time the next pulse arrives a quasi-equilibrium state has set in. Since for most oxide ferroelectrics the recombination time does not exceed 10^{-8} s, the laser pulse period-to-pulse duration ratio will be sufficiently high [113].

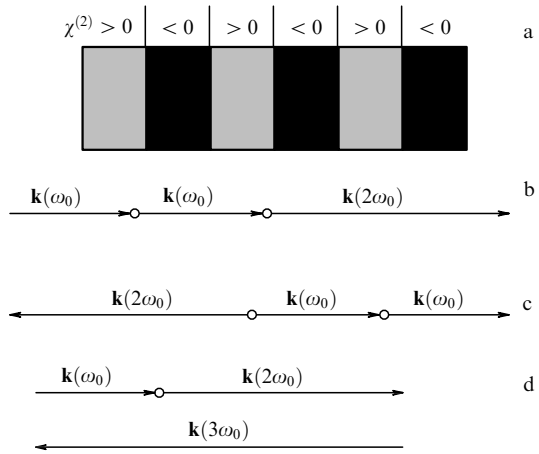

Figure 12. Configuration of light beams in generation of harmonics.

5. Nonlinear optical and acoustic effects in periodic domain structures

5.1 Generation of optical harmonics

The generation of the second and higher harmonics of laser radiation and the design of tunable parametric oscillators have for a long time attracted the attention of scientists and engineers since the laser frequency range can be broadened in this way. In recent years, the problem of converting the infrared radiation of semiconductor lasers into visible light (often called 'blue light') has emerged. In all these cases, the generation of tunable radiation and harmonics is related to the use of nonlinear optical media. Below, we examine the nonlinear features of the dielectric susceptibility and their application in periodic domain structures.

As is known, in any atomic system the polarization P_i induced by an electric field is a nonlinear function of the field strength:

$$P_i = \chi_{ij}E_j + \chi_{ijk}E_jE_k + \chi_{ijkl}E_{jkl} + \ldots , \tag{24}$$

where χ_{ij} is the linear susceptibility, χ_{ijk} is the second-order nonlinear susceptibility responsible for second-harmonic generation and parametric amplification and generation, and χ_{ijkl} is the third-order nonlinear susceptibility responsible for third-harmonic generation and Raman scattering.

Earlier, it was shown [10] that the resonant interaction of light waves in a homogeneously nonlinear medium occurs most effectively when there is phase matching of the fundamental-frequency and higher-harmonic waves:

$$\Delta k = k_2 - 2k_1 = 0 , \tag{25}$$

where $k_2 = 2\pi n_2/\lambda_2$ and $k_1 = 2\pi n_1/\lambda_1$ are the wave vectors of the second-harmonic and the fundamental-frequency waves, and n_2 and n_1 are the effective refractive indices at frequencies 2ω and ω, respectively.

Earlier, natural birefringence was the common property used in most nonlinear optical materials, but the efficiency of optical conversion was limited by the smallness of the nonlinear optical coefficients d and by the fact that the temperatures and angles of incidence of beams onto the medium needed for phase matching were not the same. As early as in the 1960s, a new method was proposed for the phase matching of coherent light beams, which employed one-dimensional spatially periodic modulation of the nonlinear dielectric susceptibility. This method could be used for birefringent crystals in which the nonlinear optical coefficients cannot be phase matched. Since the nonlinear dielectric susceptibility χ_{ijk} is described by a third-rank tensor, it is obvious that at the walls of domains in media without a symmetry center such nonlinear susceptibilities change sign (Fig. 12a) and are represented by a simple space distribution of the type

$$\chi(r) = \frac{4}{\pi}\,\chi_0\,\frac{1}{m}\cos(k_m r) , \tag{26}$$

where $k_m = 2\pi m/D$ is the wave vector of the periodic domain structure, and m is the quasi-matching period.

The linear increase in the second-harmonic intensity is the result of a choice of the domain size such that at each wall separating two domains the generalized phase shifts by π. The quasi-matching condition for the second harmonic in this case acquires the form

$$\Delta k = k_2 - 2k_1 - k_m = 0 . \tag{27}$$

The use of periodic and quasi-periodic domain structures made it possible to realize the three main advantages of such structures over homogeneously nonlinear structures.

First, because of the increase in the nonlinear quality factor $\eta = d_{\text{eff}}^2/n_1^2 n_2$ and the use of the most effective nonlinear optical coefficients. For instance, for lithium niobate and lithium tantalate, which are usually used as nonlinear elements, the largest nonlinear element d_{33} cannot be phase matched, so that for conversion in homogeneous elements the coefficients d_{13} and d_{15} are used. However, when domain structures are used, the coefficient d_{33} becomes suitable for phase matching, since in this case $d_{ij\,\text{eff}} = k_m d_{ij}$ and the optical conversion coefficient increases $(2d_{33}/\pi d_{15})^2$-fold. The effectiveness of using a periodic domain structure is illustrated by Table 1.

Table 1. Nonlinear quality factors in materials with a homogeneous nonlinearity and with periodic domain structures.

Homogeneously nonlinear materials			Periodically nonlinear materials		
Material	d_{ij}	$d^2/(n_1^2 n_2)$	Material	d_{ij}	$d^2/(n_1^2 n_2)$
$LiNbO_3$	$d_{15} = 6$	3,4	$LiNbO_3$	$d_{33} = 30$	34
$KTiOPO_4$	$d_{24} = 4$	2,7	$KTiOPO_4$	$d_{33} = 17$	18
$LiTaO_3$	$d_{31} = 3$	2,6	$LiTaO_3$	$d_{33} = 19$	13

Subsequently, the theoretical ideas were fully corroborated by experiments. For instance, waveguide-type structures formed on the surfaces of $LiMnO_3$ and $KTiOPO_4$ crystals and containing periodic domain structures with a period of 4 – 7 μm were used to generate blue light from tunable laser diodes (770 – 1040 nm) [114]. Here, the diode laser and the waveguide transducer constitute a single structure. With 120 – 150-mW laser diodes, the second-harmonic radiation power amounted to 25 – 30 mW.

Second, the use of domain structures made it possible to lift most limitations on the frequency and temperature ranges of conversion to higher harmonics. Due to the use of various types of nonlinear interactions, especially for quasi-periodic domain structures [115 – 117], it became possible to realize multiwave conversion to the second harmonic. Since in the event of phase matching the positions of the peak values of the wavelengths of the second harmonic are determined by the wave vector of the structure, for a quasi-periodic domain structure the generation spectrum, given by the expression

$$\left(\frac{I}{\lambda}\right)_{mn} = \frac{k_{mn}}{4\pi\left[n_2(\lambda) - n_1(\lambda)\right]}\,, \tag{28}$$

is, apparently, limited only by the optical transparency window of the material with the periodic structure.

Indeed, Zhu et al. [117] used a quasi-periodic domain structure in lithium tantalate to generate the second harmonic from a parametrically tunable laser in the blue, green, red, and IR ranges with an energy conversion efficiency of roughly 5%. Here, the spectrum of the generated waves fully corresponded to the results of calculations (Fig. 13).

Regular domain structures formed in oxide ferroelectrics have made it possible to use new configurations of propagation of light beams (see Fig. 12). While earlier the main configurations used in homogeneously nonlinear media were those in which all beams participating in the conversion were made to propagate in the same direction (see Fig. 12b), devices using periodic domain structures can generate the second harmonic propagating in the opposite direction to that of the incoming beam (see Fig. 12 c); they are even able to generate the third harmonic due to the three-wave interaction $\omega + 2\omega \rightarrow 3\omega$ (see Fig. 12d) [118, 119].

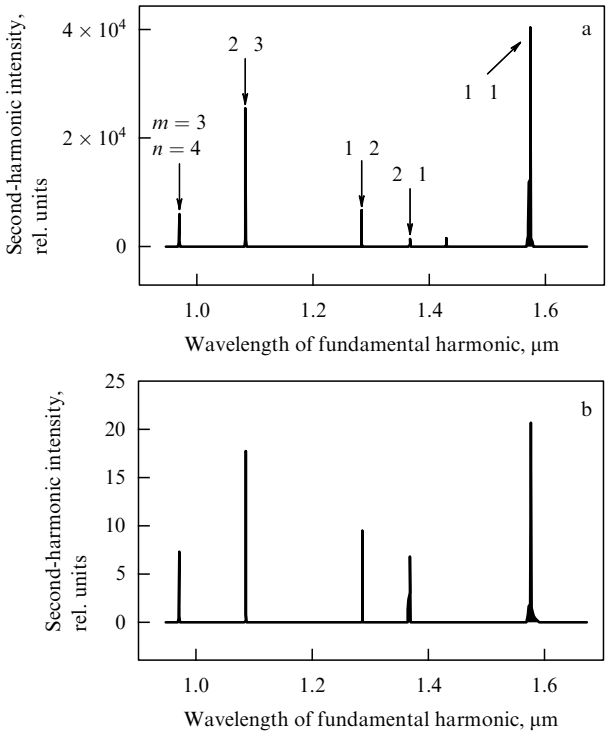

Figure 13. Spectra of the second optical harmonic converted on a quasi-periodic domain structure in lithium tantalate: (a) theoretical and (b) experimental.

Third, the use of periodic domain structures results in a radical increase in operation speed due to the decrease in the interaction lengths of the waves, which contributes to the possibility of frequency conversion up to the femtosecond range. Zhu et al. [117] recently achieved a conversion to the second harmonic for a pulse whose length was several femtoseconds.

In the same way as the dielectric nonlinear susceptibility is the cause of generation of the second and higher optical harmonics, the nonlinear magnetic susceptibility described by the third-rank tensor $\chi_{ijk}(\mu)$ can lead to nonlinear optical effects [120]. In particular, the conversion of a light beam of intensity $I(\omega)$ into the second harmonic can be expressed by the following formula:

$$I(2\omega) = \chi_{ijk}(\mu) I^2(\omega)\,. \tag{29}$$

Second-harmonic generation was detected by Wierenga et al. [121] in an experiment in which a laser beam with $\lambda = 800$ nm impinged on the layered structure Co : Cu : Co. The researchers found that the $I(\omega)$ dependence followed a hysteresis loop as the applied magnetic field changed sign with the field varying from −15 Oe to +15 Oe and the film thickness varying from 1 to 25 monolayers. Such behavior can be explained by the transformation of the domain structure of the magnetic layers.

The theoretical study of the effect of the domain structures in magnetic materials and ferroelectrics on the nonlinear characteristics of acoustic waves began in the late 1970s [103], and the focus was on the movement of domain walls in a magnetic or electric field. The first experiments involved ferroelastic (Seignette salt and gadolinium molybdate) [122]. The experiments revealed the presence of second-harmonic generation when a longitudinal acoustic wave traveled through an electrically induced periodic domain structure. As predicted by theory, the change in sign of the spontaneous deformation at the domain walls produced an additional contribution to the acoustic nonlinearity of the ferroelastic, which manifested itself in the proportionality of the second-harmonic amplitude to the number of domain walls. Theoretical and experimental studies [111, 112] of second-harmonic generation on periodic domain structures in oxide ferroelectrics with a large photorefractive effect came later [111, 112]. The biggest contribution to the acoustic nonlinearity is provided by the concentration mechanism due to photogeneration of mobile charge carriers followed by the charge exchange of the donor – acceptor centers. A detailed description of this process can be found in Ref. [114]. Second-harmonic generation was observed in experiments involving a lithium niobate crystal in which a periodic domain structure was irradiated with a laser pulse [111] (see Fig. 11).

5.2 Parametric conversion

Today, optical parametric oscillators, the idea of which was put forward by Akhmanov and Khokhlov [123] in 1962, have found many applications as sources of coherent radiation in

frequency ranges for which lasers have yet to be built or where a broad tuning interval is required. Earlier, when homogeneously nonlinear media were used in optical parametric oscillators, the phase-matching condition

$$\Delta k = k_p - k_1 - k_2 = 0 \quad \text{when} \quad \omega_p = \omega_1 + \omega_2 , \tag{30}$$

where k_p and ω_p are the wave vector and frequency of the pumpfield, and k_1, k_2, ω_1, and ω_2 are the wave vectors and frequencies of the parametrically generated oscillations, was met by selecting the proper matching angles between $\mathbf{k}_p$ and the optical axes of the crystal, the temperature interval, or the strength of the applied electric field. The use of periodic domain structures made it possible to substantially broaden the frequency range of frequency conversion employing angle matching or temperature tuning [124].

Assuming that all the wave vectors are collinear, the wave matching condition for periodic domain structures with first-order interaction has the form

$$\Delta k = k_p - k_1 - k_2 - k_m = 0 . \tag{31}$$

Calculations have shown that by varying the period of the periodic domain structure one can vary the wavelength of parametrically generated oscillations within substantial limits (Fig. 14). Myers et al. [125] used a set of 25 periodic domain structures formed on a single plate of a lithium niobate crystal with the size of the discrete periods varying from 26 to 32 μm to achieve parametric generation in the 1.3–4.8 μm range with a pump wavelength of 1.06 μm. The use of periodic domain structures with domain sizes in the 2–4 μm range has made it possible to build parametric oscillators pumped by a second-harmonic wave from a Nd : YAG laser ($\lambda = 0.53$ μm) [126].

When the phase matching condition is met, the amplitudes of the generated waves gradually increase, at the expense of the energy of the pumpfield, as the waves propagate along the crystal, with the wave excitation having a threshold. Since for this case the threshold pumping intensity is defined as

$$I_{th} = \frac{n_p n_1 n_2 \lambda_1 \lambda_2}{8\pi^2 d_{eff}^2 L^2} , \tag{32}$$

where L is the length of the periodic domain structure, the use of the largest nonlinear coefficients d_{ij} makes it possible to appreciably lower the threshold value of the pumpfield (just as it does for the case of second-harmonic generation).

In modern practice, parametric generation is achieved by exciting periodic domain structures through the use of cw laser beams [127, 128] and femtosecond laser pulses [129] with a conversion efficiency of tens of percent.

Although the first attempts to build optical parametric oscillators with periodic domain structures began only in the first half of the 1990s and the first papers appeared in 1994–1995, at the time of publication of the present review the total number of papers on this topic is probably several dozen. The reader will find an exhaustive bibliography on the subject in Ref. [130].

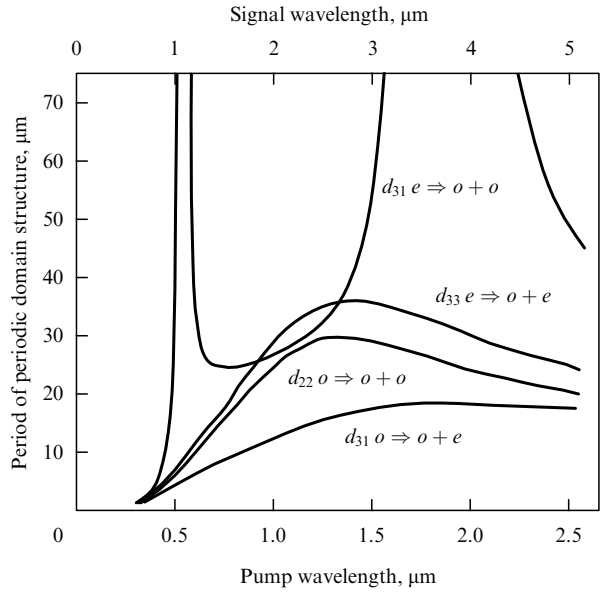

Figure 14. Relationship between the wavelengths of parametrically generated signals and the period of the periodic domain structure: *e*, the extraordinary ray; and *o*, the ordinary ray.

6. Conclusion

The state of the art of studies in the physics of domain states and the applicational aspects of using periodic domain structures reviewed in the present paper can be expected to be substantially augmented by new theoretical and experimental results. Since today attention is focused on the use of periodic domain structures in nonlinear-optics devices, further achievements in this field could be related to the decrease in the period of the periodic domain structures to 0.5–1 μm and to building easily tunable periodic domain structures with a varying period (quasi-periodic structures). Possible instruments to do this are pencil light beams [131, 132], electron beams, or local injection of electric charges from a needle-shaped electrode [133]. The recent results in third-harmonic generation involving the use of quasi-periodic domain structures suggest that the coherent optical beams will successfully 'occupy' the UV range [134]. We can probably expect the emergence of new optical multiplex and demultiplex systems for injecting signals of different wavelengths into or extracting from an optical waveguide, since the first samples of such devices based on holographic gratings in lithium niobate have already been developed [135]. There are successful examples of the generation of higher harmonics via quasi-periodic domain structures [99, 100]. The use of a single periodic or quasi-periodic domain structure to amplify the acoustooptical interaction in modulators with simultaneous phase matching of the acoustic and light beams also appears to be very promising.

Possibly, in the near future we will discover new ways of controlling stripe-domain structures with sizes of several nanometers formed by oriented charges in partially substituted lanthanum manganite [136]. Szymczak [137] studied magnetoelastic interactions in materials with a large magnetoresistive effect and pointed out the possibility of nonlinear conversion and generation not only of light waves but also of acoustic waves.

One should expect the emergence of generators of higher harmonics and other transducers of acoustic waves based on dynamically induced domain structures in magnetic materials with a strong magnetoelastic interaction, e.g., in iron borate and hematite.

However, the development of methods for forming new periodic domain structures is not only a problem of technology. New physical effects may emerge in regular periodic structures of micrometer and especially submicrometer sizes, e.g., related to the tunneling of free charges through domain walls and to the formation of mesoscopic states. These problems have yet to be studied more thoroughly by theoreticians [138].

In addition to the widening of the contribution of domain structures to nonlinear optics and acoustics, one should expect further development of studies on using domains and periodic domain structures in devices for recording, storing, and processing signals in optical and digital form. In the simplest case, individual microdomains with oppositely directed polarizations can be considered as elements of the binary code. Any collection of microdomains in a material can represent a system for recording data in holographic form.

Before actually designing an information system based on periodic domain structures, one must answer the following questions: What is the minimum size of a stable state? Are such states to be microdomains or nanodomains? In what way will local polarization switching be created in such domains? These questions can be answered only by further studies in the physics of domain formation. At present, such studies are in their infancy. First, we must thoroughly investigate the role of the spatial redistribution of the charges of the impurity ions in an external or light-induced electric field, since, as assumed in Refs [62, 71, 112, 139], this process plays the leading role in the formation of domains in lithium niobate and barium titanate.

Preliminary experiments have shown that domain polarization switching can be achieved using fairly low-powered light beams ($I \sim 1$ W cm^{-2}) or acoustic beams in crystals with a low coercive field, e.g., in barium titanate [140]. Applying this method to crystals with a strong piezoelectric effect, we can use the piezoelectric field to stimulate the formation of an additional number of domains, whose spatial density will be proportional to the wave's intensity.

The possibility of recording acoustic signals on individual grains of magnetic or ferroelectric ceramics has been demonstrated by Bondarenko et al. [141] and Chaban [142], respectively. In both cases the reading of the recorded signals can be done by acoustic or optical devices.

Thus, the results that have been obtained so far irrevocably support the prediction that further studies will lead to the development of new media and physical principles for recording optical and acoustic information in analog, digital, and holographic form on domain structures.

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