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Two-dimensional bound states of ultrashort light pulses and polarisation of light in ferroelectric impurity crystals

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***Abstract*. The polarisation evolution of a ferroelectric crystal after its illumination by ultrashort laser pulses is described theoretically and numerically by using the microscopic pseudospin formalism. Parameters are found for which quasi-two-dimensional long-lived states can exist in ferroelectrics. For different initial conditions, the dynamics of bound states of the electromagnetic field and polarisation of the crystal is presented and properties of ferroelectrics with impurities and without them are compared.**

Keywords: *ultrashort pulses, long-lived states, polarisation of ferroelectrics.*

1. Introduction

Nonlinear structures, including nonperiodic structures, attract considerable interest because they can be used to control the transfer and reflection of waves, which opens up new possibilities for application of crystals used in nonlinear optics to process optical signals [1 – 4]. The latest theoretical and experimental studies have shown that a nonlinear localisation of light can exist in the optically-induced refractive medium (such long-lived localised states can be produced when the dispersion and diffraction effects are balanced by the self-induced change in the medium) [5, 6]. Usually the propagation of waves is controlled with the help of structural defects of the medium. However, the media, in which the internal structure and symmetry of the nonlinear state determine the choice of the wave-propagation direction periodic structures without defects, seem more promising. The symmetry of such states, which are often solitons, is determined by the physical mechanisms responsible for the light localisation. Optically-induced photon gratings are an important experimental tool for studying nonlinear effects during the light propagation in periodic structures. The existence of these effects suggests that the propagation of light pulses can be controlled with the help of light.

In this paper we consider the possibility of existence of long-lived states in a ferroelectric medium with an impurity. Because the self-induced transparency is caused by the interaction of a laser pulse with the impurity electric dipole momenta of a solid, we can expect a situation when the ferroelectric instability and optical bleaching of the resonance medium will be observed simultaneously.

The most promising from this point of view are compounds similar to $\mathrm{KCl:Li^+}$, $\mathrm{KTaO_3:Li^+}$, $\mathrm{PbTe:Ge^{++}}$ (i.e. compounds which include impurity noncentral ions). In addition, the intrinsic ferroelectric properties of KDP crystals (KDP, DKDP, ADP, etc.) widely used in lasers can substantially influence the formation of self-induced transparency pulses [7]. The study of these crystals can both open up new fields of their application and enrich our concepts about the behaviour of strongly nonlinear systems.

The structure of the majority of ferroelectrics of the KDP type is determined by hydrogen bonds, and, hence, their single crystals can be easily doped with impurity centres of different nature. In addition, due to the dissociation of hydrogen bonds in ferroelectrics and the capture of free charge carriers by the dissociated bonds, the impurity centres can appear in an initially undoped sample. Note that the nonlinear processes in ferroelectrics with hydrogen bonds have been studied theoretically by neglecting the fact that in real samples the impurity subsystem can play a substantial role in the excitation dynamics. Note also that consideration of this subsystem can considerably affect the character and peculiarities of the intrinsic ferroelectric phase transition [8].

2. Formulation of the problem and basic equations

In this paper we consider the properties of intrinsic ferroelectrics of the KDP type with impurity two-level atoms. The dynamics of the self-induced transparency pulses will be studied by using the pseudospin formalism. This approach describes rather well the perturbation dynamics in ferroelectric media, which was demonstrated in [9]. The main advantage of this formalism is the fact that elementary excitations (pseudospin waves) exist both in the ordered and disordered phases and, hence, the properties of a ferroelectric can be studied in both phases from the unified point of view. The Heisenberg equations of motion for the average values of pseudospin operators $\langle S^{\alpha} \rangle$ in the chaotic phase approximation and in the continuum limit have the form [8, 9]

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$$\frac{\mathrm{d}\langle S^x\rangle}{\mathrm{d}t}=(J\langle S^z\rangle+A\langle S^z\rangle_{\xi\xi}+B\langle S^z\rangle_{\eta\eta}+2\mu_0E)\langle S^y\rangle,$$

$$\frac{\mathrm{d}\langle S^y\rangle}{\mathrm{d}t}=\Omega\langle S^z\rangle-(J\langle S^z\rangle+A\langle S^z\rangle_{\xi\xi}+B\langle S^z\rangle_{\eta\eta}+2\mu_0E)\langle S^x\rangle, \qquad (1)$$

$$\frac{\mathrm{d}\langle S^z\rangle}{\mathrm{d}t}=-\Omega\langle S^y\rangle,$$

where

$$\langle S^z\rangle_{\xi\xi}=\frac{\mathrm{d}^2\langle S^z\rangle}{\mathrm{d}\xi^2};\quad \langle S^z\rangle_{\eta\eta}=\frac{\mathrm{d}^2\langle S^z\rangle}{\mathrm{d}\eta^2};$$

$$J=\sum_j J_{ij}=\sum_j J(j-i);\quad A=Ja^2;\quad B=Jb^2;$$

a is the distance between the neighbouring sells in the crystal in the direction of the ξ axis; b is the distance between the neighbouring sells in the crystal in the perpendicular direction; Ω is the tunnelling integral; J_{ij} is the exchange integral; E is the electric field; μ_0 is the dipole moment of a ferroelectric sell. In the problem under study, the coordinates x, y, z correspond to the axes in the pseudospin space. The axes in the real space are denoted as ξ, η. The $\xi\eta$ plane corresponds to the plane perpendicular to the optical axis of the ferroelectric, while the polarisation vector of the electric field is chosen parallel to this axis.

Note that because for typical ferroelectrics the constants Ω and J are approximately equal to 10^{13} and 10^{14} Hz, respectively, the pulses whose frequency falls in this interval will interact most efficiently with our medium. A more detailed consideration is required when the pulse frequency coincides with the frequency of the 'soft' mode [9], which depends on the sample temperature. We will consider an impurity atom in the model of a two-level system, assuming that higher energy levels are not excited in the interval of temperatures within which a ferroelectric crystal exists. This approximation is a standard one in self-induced transparency models. If the probability amplitudes of the upper (asymmetric) and lower (symmetric) states of an atom, whose difference in energies is $\hbar\Omega_{\mathrm{i}}$ (hereafter, $\hbar=k=c=1$) are denoted as Ψ_+ and Ψ_-, and if we assume that the transitions between the levels occur due to the interaction of a dipole moment of the impurity atom d_{i} with the electric field of the laser pulse, then we obtain the equations in partial derivatives in time t for the quantities

$$N=\Psi_+^*\Psi_+-\Psi_-^*\Psi_-,$$

$$P_+=\Psi_-^*\Psi_++\Psi_+^*\Psi_-,$$

$$P_-=\mathrm{i}(\Psi_-^*\Psi_+-\Psi_+^*\Psi_-)$$

in the form

$$N_t=-2d_{\mathrm{i}}EP_-,$$

$$(P_+)_t=-\Omega_{\mathrm{i}}P_-, \qquad (2)$$

$$(P_-)_t=\Omega_{\mathrm{i}}P_++2d_{\mathrm{i}}EN.$$

Let us supplement system of Eqns (1), (2) with the equation describing the dynamics of the electric field E in the case of the spatially homogeneous ferroelectric without free charges:

$$E_{tt}-c^2E_{\xi\xi}-c^2E_{\eta\eta}+\chi\langle S^z\rangle_{tt}+\chi_{\mathrm{i}}(P_+)_{tt}=0, \qquad (3)$$

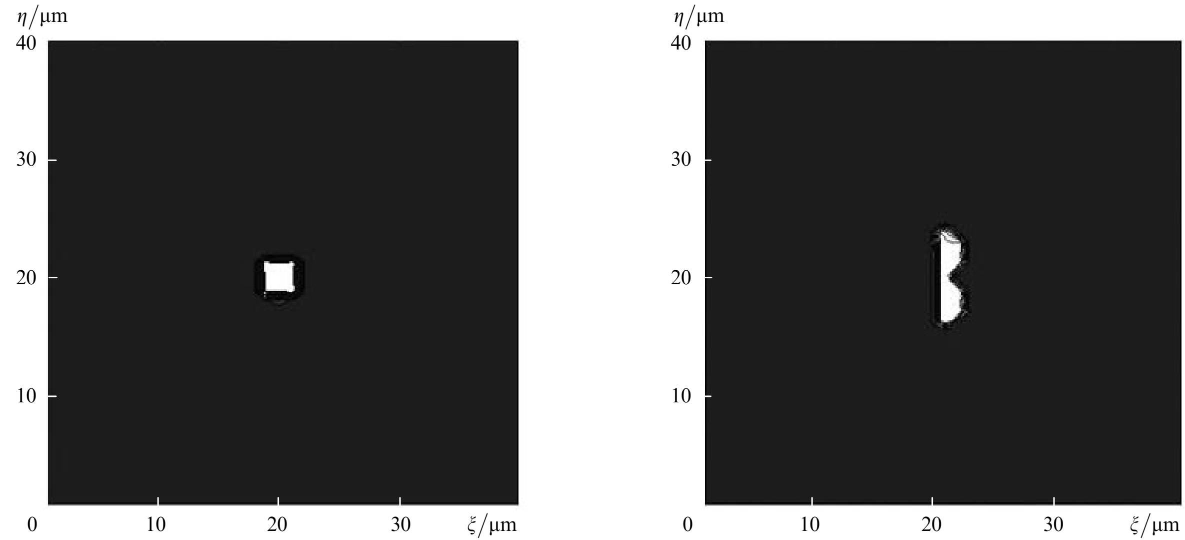
where $\chi=4\pi\mu_0n_{\mathrm{s}}$; $\chi_{\mathrm{i}}=4\pi d_{\mathrm{i}}n_{\mathrm{i}}$; n_{s} and n_{i} are the concentrations of ferroelectric sells and impurities, respectively; double subscripts below denote the corresponding partial derivative. The obtained complete system of equations (1)–(3) is a system of equations in partial derivatives, which will be analysed below. Note that the results of its analysis in the one-dimensional case can be found in paper [10].

3. Results and discussion

System of equations (1), (2) together with Eqn (3) for the electric field was solved numerically for different parameters of the problem and different initial conditions. The typical initial polarisation states are shown in Fig. 1. Figures 2 and 3 present a change in the initial state shown in Fig. 1a for a ferroelectric with impurities and without them. Note that we consider here a ferroelectric in the low-temperature phase near the phase transition point, but when the Ginzburg criterion is still valid and fluctuations can be neglected. Thus, this temperature for a ferroelectric of the KDP type will be ~120 K.

One can clearly see in Figs 2 and 3 the formation of a long-lived bound state of the electromagnetic field and ferroelectric polarisation. The light regions in Fig. 1 correspond to the crystal region initially illuminated by laser pulses. Then, this state evolves according to system of equations (1)–(3). One can also see from Figs 2 and 3 that although the bound localised state of polarisation and the electric field evolves differently for ferroelectrics with impurities and without them, for some values of the problem parameters there exist qualitatively similar stages (Fig. 2c, d and Fig. 3c, d). Therefore, in both cases we can speak about long-lived states. In particular, when the tunnelling integral Ω, which is related to the degree of the sample deuteration, increases, the polarisation achieves its quasi-equilibrium state faster. This can be explained by the fact that Ω increases with increasing the nonlinear efficiency in the pseudospin subsystem [11]. Note also that in the case of the impurity ferroelectric, less time is required to produce these states for large values of Ω_{i}. A similar situation is observed for the initial state shown in Fig. 1b, which is demonstrated in Figs 4 and 5.

This behaviour can be explained by the fact that when the energy Ω_{i} of the impurity subsystem increases, the frequencies of natural oscillations become closer to the frequencies of the natural oscillations of the pseudospin subsystem. In this case the energy exchange between these subsystems and between the electric field and the ferroelectric subsystem through the impurity susbsystem occurs



a b

Figure 1. Initial states, whose evolution is described in this paper.

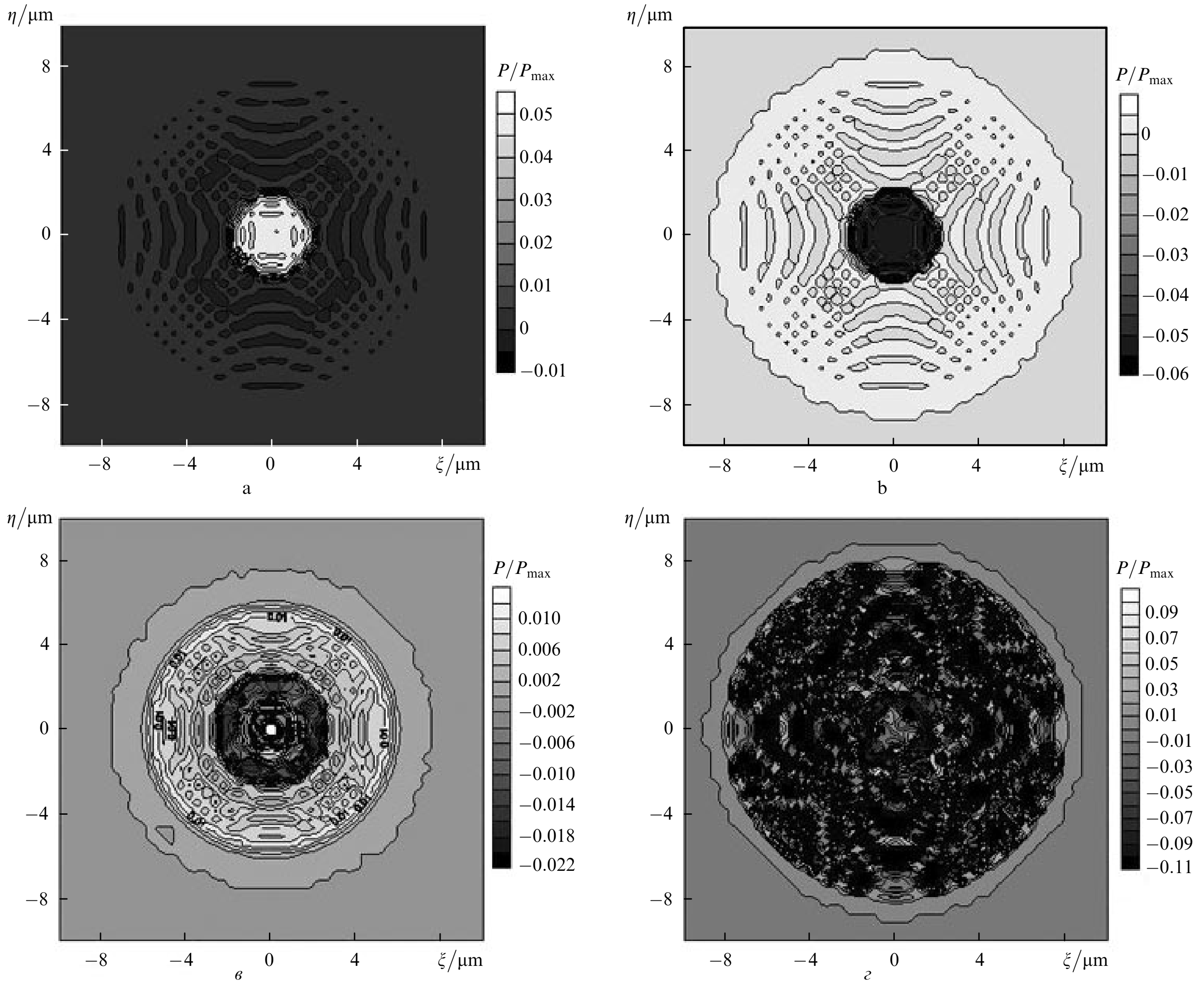
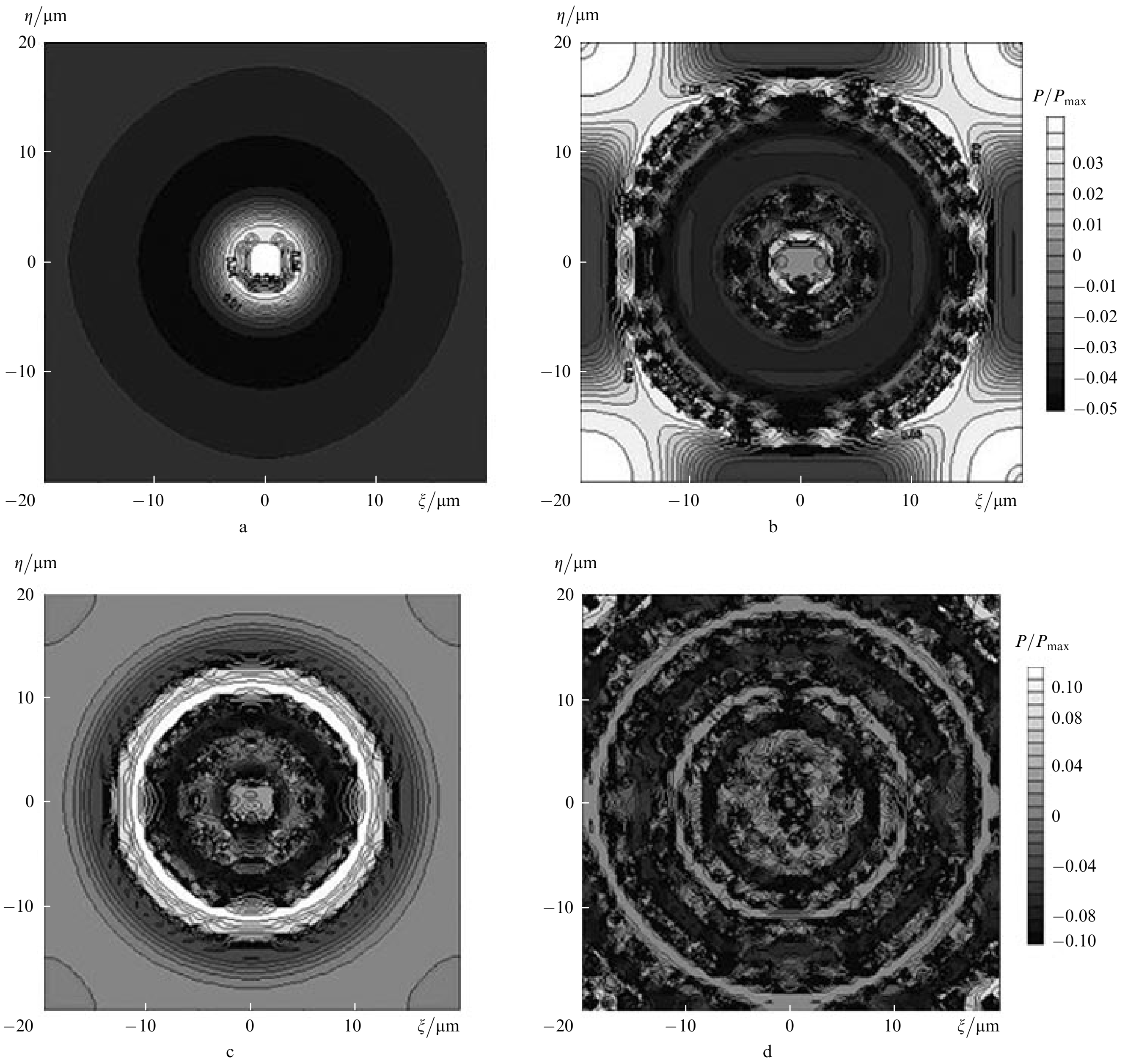


Figure 2. Change in the initial state shown in Fig. 1a for a ferroelectric crystal with impurities at the instant of time $t = 10^{-10}$ s for $\hbar J = 1.7 \times 10^{-21}$ J, $\mu_0 = 1.6 \times 10^{-30}$ C m, $\hbar\Omega = 3.4 \times 10^{-21}$ J, $\hbar\Omega_{\mathrm{i}} = 3.4 \times 10^{-25}$ J (c); $\hbar J = 1.7 \times 10^{-24}$ J, $\mu_0 = 1.6 \times 10^{-30}$ C m, $\hbar\Omega = 3.4 \times 10^{-21}$ J, $\hbar\Omega_{\mathrm{i}} = 3.4 \times 10^{-25}$ J (b); $\hbar J = 1.7 \times 10^{-24}$ J, $\mu_0 = 1.6 \times 10^{-28}$ C m, $\hbar\Omega = 3.4 \times 10^{-21}$ J, $\hbar\Omega_{\mathrm{i}} = 3.4 \times 10^{-25}$ J (c); and $\hbar J = 1.7 \times 10^{-24}$ J, $\mu_0 = 1.6 \times 10^{-30}$ C m, $\hbar\Omega = 3.4 \times 10^{-21}$ J, $\hbar\Omega_{\mathrm{i}} = 3.4 \times 10^{-21}$ J (d). Here and in Figs 3–5, the black colour gradation shows polarisation P expressed in fractions of saturation polarisation $P_{\max}$.



a b c d

Figure 3. Initial state evolution shown in Fig. 1a for a ferroelectric crystal without impurities at the instants of time $t = 10^{-10}$ (a, c) and 2×10^{-11} s (b, d) for $\hbar J = 1.7 \times 10^{-21}$ J, $\mu_0 = 1.6 \times 10^{-29}$ C m, $\hbar\Omega = 0.8 \times 10^{-22}$ J (a, b) and $\hbar J = 1.7 \times 10^{-21}$ J, $\mu_0 = 8 \times 10^{-29}$ C m, $\hbar\Omega = 1.7 \times 10^{-22}$ J (c, d).

more efficiently. As a result, the quasi-stationary state is established faster.

Note also that quasi-two-dimensional long-lived states described in this paper can be used to control the laser radiation field scattered by these states. These states can be also easily detected by the methods of neutron and light scattering [10]. Note that in the presence of these states, the central peak intensity increases. This makes the spectroscopic studies of ferroelectrics possible upon their simultaneous irradiation by laser pulses and neutrons.

4. Conclusions

Based on the performed numerical simulation we can draw the following conclusions:

(i) In a pseudospin system, which is widely used to describe real ferroelectrics, including ferroelectrics with hydrogen bonds, quasi-two-dimensional long-lived states of polarisation and the electric field related to it can exist in the ferroelectrics doped with impurities. These states weakly depend on the choice of initial conditions and have the axial symmetry at large times. Thus, in particular, long-lived states exist in the range of the change in their main parameters (in SI units): $\hbar J \approx 1.7 \times 10^{-24} - 1.7 \times 10^{-21}$ J, $\hbar\Omega \approx 3.4 \times 10^{-21} - 3.4 \times 10^{-23}$ J, $\hbar\Omega_{\rm i} \approx 3.4 \times 10^{-26} - 3.4 \times 10^{-24}$ J, $\mu_0 \approx 1.6 \times 10^{-28} - 1.6 \times 10^{-30}$ C m.

(ii) The parameters of the problem are found, which are responsible for the polarisation evolution regimes of the pseudospin subsystem. It is found that the greater the tunnelling integral Ω, the less time is required to produce long-lived states. In the range of the parameters under study, this time decreased linearly with increasing Ω. In the case of ferroelectrics with impurities, this time depends on the distance between the levels of the two-dimensional

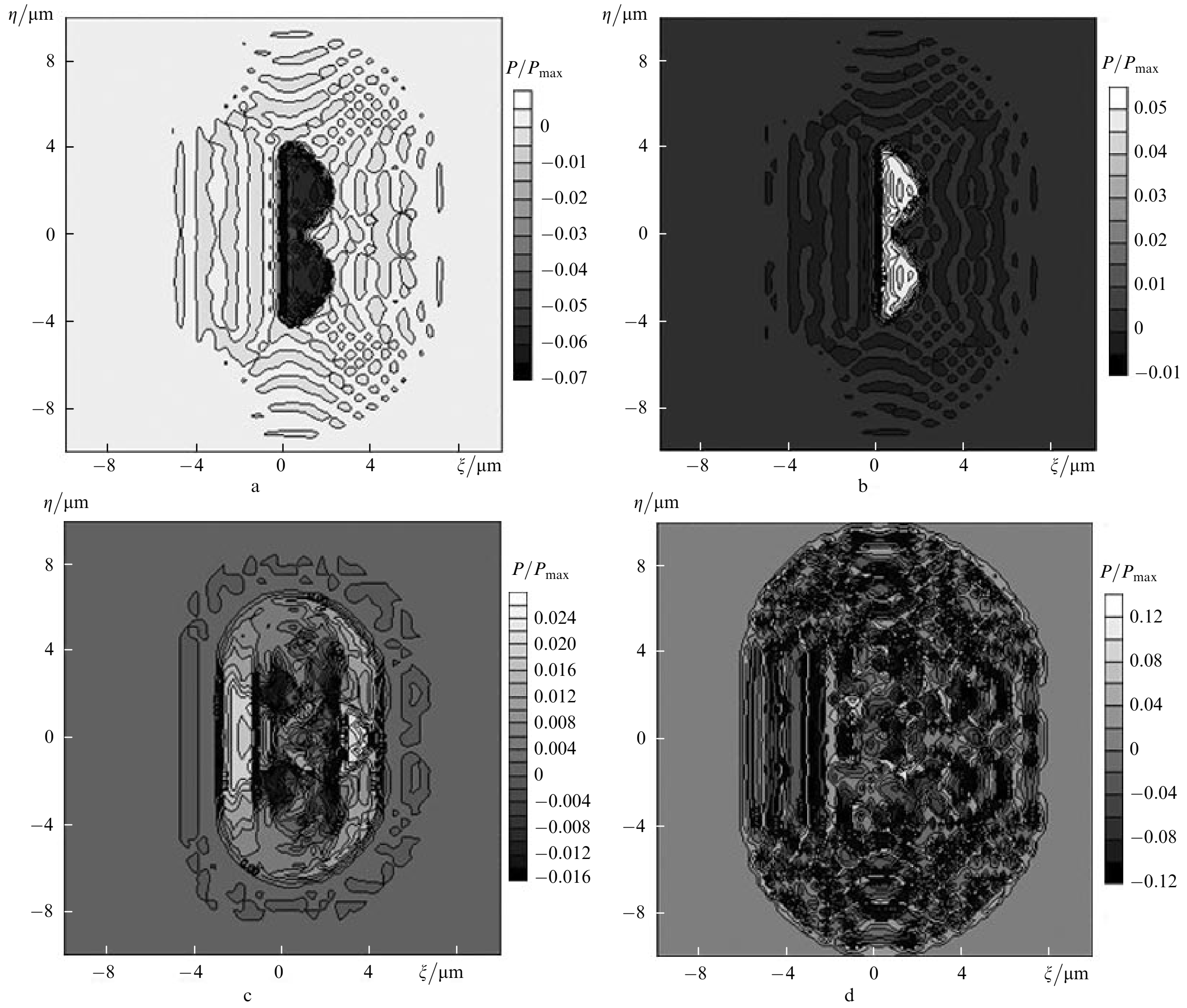


Figure 4. Change in the initial state shown in Fig. 1b for a ferroelectric crystal with impurities at the instant of time $t = 10^{-10}$ s for $\hbar J = 0.8 \times 10^{-22}$ J, $\mu_0 = 1.6 \times 10^{-30}$ C m, $\hbar\Omega = 3.4 \times 10^{-21}$ J, $\hbar\Omega_i = 3.4 \times 10^{-25}$ J (a); $\hbar J = 1.7 \times 10^{-21}$ J, $\mu_0 = 1.6 \times 10^{-30}$ C m, $\hbar\Omega = 3.4 \times 10^{-21}$ J, $\hbar\Omega_i = 3.4 \times 10^{-25}$ J (b); $\hbar J = 0.8 \times 10^{-22}$ J, $\mu_0 = 4.8 \times 10^{-28}$ C m, $\hbar\Omega = 3.4 \times 10^{-21}$ J, $\hbar\Omega_i = 3.4 \times 10^{-25}$ J (c) и $\hbar J = 0.8 \times 10^{-22}$ J, $\mu_0 = 1.6 \times 10^{-30}$ C m, $\hbar\Omega = 3.4 \times 10^{-21}$ J, $\hbar\Omega_i = 3.4 \times 10^{-21}$ J (d).

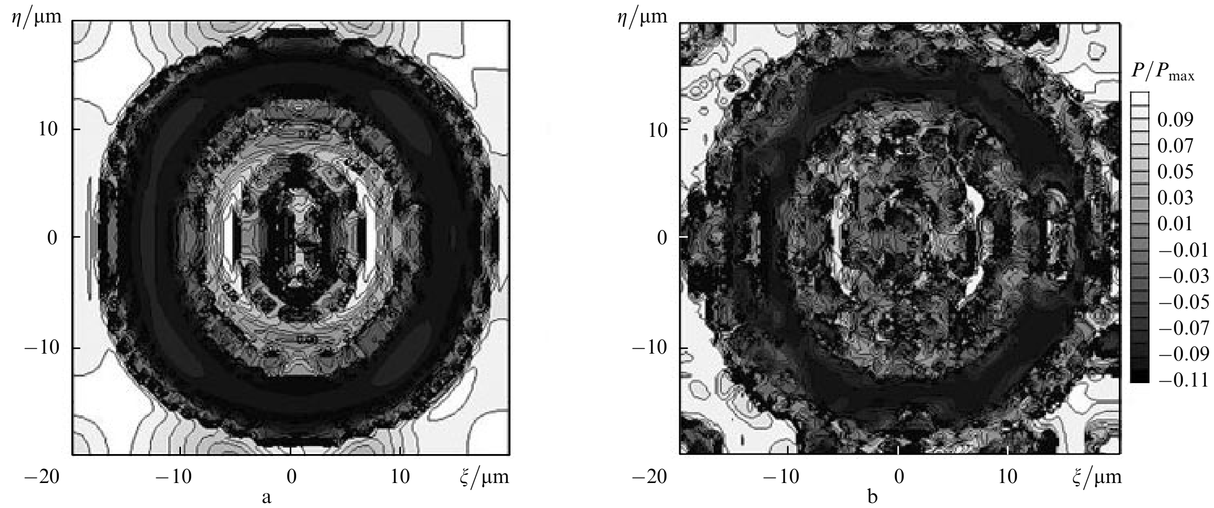


Figure 5. Initial state evolution shown in Fig. 1b for a ferroelectric crystal without impurities at the instants of time $t = 10^{-11}$ (a) и 2×10^{-11} s (b) for $\hbar J = 1.7 \times 10^{-21}$ J, $\mu_0 = 1.6 \times 10^{-29}$ C m, $\hbar\Omega = 1.7 \times 10^{-22}$ J.

impurity subsystem Ω_i: the lower is Ω_i, the more time is required to produce the long-lived polarisation state. In the range of the parameters under study, an inverse linear dependence of the formation time of long-lived states on Ω_i was observed.

Therefore, the use of impurities in ferroelectric crystals can change the evolution time of processes, which opens up new possibilities for the application of these crystals in holography and storage devices.

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