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Nonharmonic Spatial Population Difference Structures Created by Unipolar Rectangular Pulses in a Resonant Medium

© R.M. Arkhipov[1,2], M.V. Arkhipov[2], A.V. Pakhomov[1], O.O. Dyachkova[1], N.N. Rosanov[1,2]

[1] St. Petersburg State University, St. Petersburg, Russia
[2] Ioffe Institute, St. Petersburg, Russia

e-mail: arkhipovrostislav@gmail.com, m.arkhipov@spbu.ru, antpakhom@gmail.com, o.o.dyachkova@gmail.com, nnrosanov@mail.ru

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In the case of coherent interaction with a medium of extremely short light pulses (ESPs) having a carrier frequency and harmonic shape (when the pulse durations are shorter than the population relaxation times T_1 and polarization relaxation time T_2 of the medium), electromagnetically induced gratings (EMIGs) of the population difference, which have a pronounced harmonic dependence on the coordinates, may appear in it. These structures can occur when pulses do not overlap or overlap in the medium. Recently, the possibility of obtaining unipolar electromagnetic pulses in the optical and adjacent ranges of non-harmonic shape, for example, rectangular and triangular, with a duration less or comparable to the duration of the extremely-short pulse in this range, has attracted interest. In this work, using the numerical solution of the system of Maxwell-Bloch equations, we study EMIG formation by rectangular attosecond pulses in a two-level resonant medium. The possibility of inducing an EMIG of a non-harmonic shape in the form of light-induced channels microresonators, (microcavities) with a size of the order of the wavelength of the resonant transition of the medium, whose parameters can be controlled, for example, by the amplitude of the incident pulses, is shown. It has been suggested that it is possible to create an EMIG of a predetermined non-harmonic shape only in the general case of using unipolar pulses.

Keywords: attosecond pulses, unipolar pulses, rectangular pulses, electromagnetically induced gratings, polarization waves, light-induced microresonators.

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Introduction

The study of the possibility for generating electromagnetic extremely-short pulses (ESP) of femto- and attosecond duration and their interaction with matter is an important part of modern physics, since such pulses are actively used to control the dynamics of ultrafast processes in resonant media [1–7]. Typically, light pulses obtained in practice have a harmonic shape and contain several field oscillations. Further shortening of the pulse duration requires a transition to unipolar pulses (UPs) containing a burst of electric field of the same sign (see reviews [8,9], works [10–20] and cited literature). Unlike conventional pulses with a harmonic shape, UPs can have a non-zero electric area $\mathbf{S}_E(\mathbf{r},t) = \int \mathbf{E}(\mathbf{r},t)dt$ ($\mathbf{E}(\mathbf{r},t)$ is electric field strength, t is time). Interest in UPs is associated with the possibility of their unidirectional action on charges, and as a result, the ability to quickly and efficiently control the properties of atoms [21], molecules [22] and nanostructures [23].

With the coherent interaction of ultrashort harmonic pulses with resonant media it is possible to create spatial harmonic electromagnetically induced gratings (EMIGs) of a population difference when the pulses simultaneously overlap or do not overlap in the medium, see reviews [24,25] and the cited literature. The possibility of creating such EMIGs using non-overlapping long multicycle pulses has been studied for a long time [26–28] (the results of previous works are summarized in more detail in reviews [24,25]). This approach differs significantly from the traditional method of creating harmonic EMIGs based on the interference of two or more long monochromatic laser beams [29]. EMIGs created in this way have numerous applications [25,29]. In connection with the interest in the generation of femto- and atto-second pulses, the dynamics of EMIGs induced by a sequence of ESPs [25,30–37] has recently been actively studied. The physical mechanism of creating an EMIG is associated with the interference of the polarization waves of the medium, induced by the previous pulse, with the next pulse entering the medium after the previous pulse has left it [24–28,30–37].

The possibility of obtaining non-harmonic pulses of various shapes is also being actively discussed [10–20]. In particular, schemes are proposed in which the time dependence of the electric field strength has a rectangular and triangular shape [10,11]. Obtaining such UPs is possible, for example, by integrating and differentiating the time form of the field strength in thin metal films [38,39]. Experimentally, rectangular UPs in the THz frequency range in the form of precursors in an electro-optical crystal were obtained quite recently [19], and the possibility of obtaining rectangular terahertz unipolar pulses by excitation of a metal film was theoretically shown [20] using femtosecond laser pulse.

In all previous studies, the induced EMIGs had a harmonic shape or their rather simple superposition, and to create them, short pulses a harmonic shape or unipolar Gaussian pulses were also mainly used. Therefore, the question arises of the possibility of creating non-harmonic EMIGs using non-harmonic light pulses. In this work, one theoretically study the possibility of inducing and controlling the EMIGs of a population difference using a sequence of rectangular UPs, both overlapping and non-overlapping in a two-level medium. The possibility of creating in the medium of narrow channels of the population difference (microresonators) i.e. segments along which the population difference has a pronounced nonharmonic form, namely, a constant value of the population difference, is shown. There is a step in the population difference at the boundary of the plots. These structures arise when rectangular pulses collide in a resonant medium. In the absence of pulse overlap, such structures were not detected by us.

The simulating shows the existence of alternating sections in a resonant medium, where polarization waves arise, running in opposite directions. Previously, such structures were observed when harmonic pulses or unipolar Gaussian pulses overlapped at the center of the medium [33,34,37].

Theoretical model

Numerical calculations were carried out using the well-known system of Maxwell-Bloch equations, which describes the evolution of the off-diagonal element of the density matrix ρ_{12}, the difference in populations of the medium (inversion) $n = \rho_{11} - \rho_{22}$ of a two-level medium, its polarization P and electric field strength. E [40]

$$\frac{\partial \rho_{12}(z,t)}{\partial t} = -\frac{\rho_{12}(z,t)}{T_2} + i\omega_0\rho_{12}(z,t) - \frac{i}{\hbar} d_{12}E(z,t)n(z,t), \quad (1)$$

$$\frac{\partial n(z,t)}{\partial t} = -\frac{n(z,t) - n_0(z)}{T_1} + \frac{4}{\hbar} d_{12}E(z,t)\,\mathrm{Im}\,\rho_{12}(z,t), \quad (2)$$

$$P(z,t) = 2N_0 d_{12}\,\mathrm{Re}\,\rho_{12}(z,t), \quad (3)$$

$$\frac{\partial^2 E(z,t)}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E(z,t)}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 P(z,t)}{\partial t^2}. \quad (4)$$

In the (1)−(4) system: t — time, z — longitudinal coordinate, N_0 — concentration of active centers, c — speed of light in vacuum, $\hbar$ — reduced Planck constant, ω_0 — resonant transition frequency of medium ($\lambda_0 = 2\pi c/\omega_0$ is wavelength of the resonant transition), d_{12} is dipole moment of the working transition, n_0 is population difference of two working levels in the absence of electric fields ($n_0 = 1$ for an absorbing medium).

The system of equations (1)−(4) is written without the approximation of slowly varying envelopes and a rotating wave. For justification of the applicability of the two-level model in such problems, see the works [41,42] and comments therein. The use of a one-dimensional wave equation is justified, for example, in coaxial waveguides, in which UPs propagation is possible without significant loss of unipolarity [43].

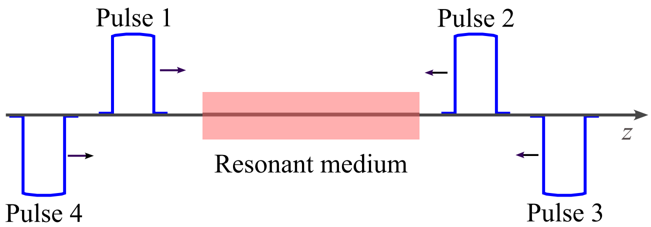

Figure 1. A sequence of rectangular pulses propagating towards each other in a resonant medium.

Fig. 1 illustrates the situation in which the theoretical model was applied. The medium is excited by a sequence of rectangular UPs propagating towards each other, as shown in Fig. 1. To create such a sequence of pulses in numerical calculations, a rectangular pulse 1 was launched from vacuum into the medium from left to right at the initial moment of time, described by the hyper-Gaussian function

$$E(0,t) = E_{01}e^{-\frac{(t-\tau_1)^{20}}{\tau^{20}}}. \quad (5)$$

And a similar impulse 2 from right to left

$$E(L,t) = E_{02}e^{-\frac{(t-\tau_2)^{20}}{\tau^{20}}}. \quad (6)$$

Here $\tau_{1,2}$ are delays that regulate the moment of meeting and impulses.

In order to create a sequence of pulses, zero boundary conditions were taken in the calculations. The system of Maxwell-Bloch equations (1)−(4) was numerically solved for various parameters indicated below; the spatial region of integration had length $L = 12\lambda_0$. The two-level resonant medium was located along the z axis in the center of the integration region between the points $z_1 = 4\lambda_0$ and $z_2 = 8\lambda_0$. The system of equations for the density matrix (1)−(3) was solved numerically by the 4th order Runge-Kutta method. The wave equation (4) was solved by the finite difference method. Delays $\tau_{1,2}$ were selected in such a way that the pulses either did not overlap at the same time, or met in the medium. Below are the results of numerical simulation of the EMIGs dynamics in these situations.

EMIG dynamics when the sequence of rectangular attosecond pulses does not overlap in the medium

The parameters used in the numerical calculation in this case are given in the table.

Such short relaxation times and dipole moments of tens of Debye are characteristic of various nanostructures [44–46], Parameters used in numerical calculations

Resonance transition wavelength	$\lambda_0 = 700$ nm
Transition dipole moment	$d_{12} = 20$ Debay
Inversion relaxation time	$T_1 = 1$ ps
Polarization relaxation time	$T_2 = 0.5$ ps
Atomic concentration	$N_0 = 5 \cdot 10^{14}$ cm^{-3}
Filed amplitude	$E_{02} = E_{01} = 74000$ ESU
Parameter τ	$\tau = 466$ as
Delay parameter	$\tau_1 = 2.5\tau$, $\tau_2 = 25\tau$

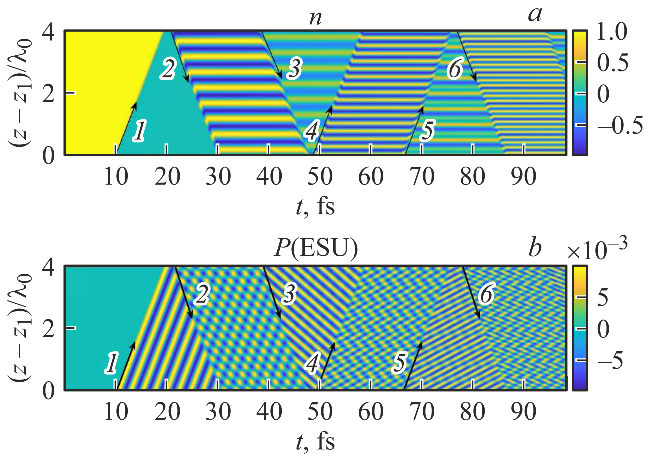

Figure 2. (a) Dynamics of the population difference $n(z, t)$, (b) dynamics of the polarization $P(z, t)$ under the action of single-cycle attosecond pulses 1 and 2 in the form (4) and (5). The calculation parameters are given in the table.

and unipolar pulses seem promising for ultrafast population control in quantum dots, nanoparticles etc. [23]. Note that the interaction of atto-second pulses with nanostructures has been the subject of active study lately [47–49].

Fig. 2 illustrates the dynamics of the population difference (a) and polarization (b) during coherent propagation in the medium of a pair of rectangular pulses (5) and (6), when the pulses do not overlap in the medium, at parameters given in the table. The amplitude of the pulses is chosen in such a way that they act like $\pi/2$-pulses, i.e. left after its propagation the medium in a state with zero population difference ($n = 0$). Numbers of pulses and directions of their propagation are shown by numbers and arrows, respectively.

The first rectangular pulse, propagating from left to right, as is in Fig. 1, brings the medium into a state with zero inversion, and leaves behind a harmonic polarization wave traveling from left to right in the medium (Fig. 2, a, b). Then, when the first pulse has left the medium, the same counter pulse 2 enters the medium. As a result of interaction with the polarization wave, a harmonic EMIG of populations

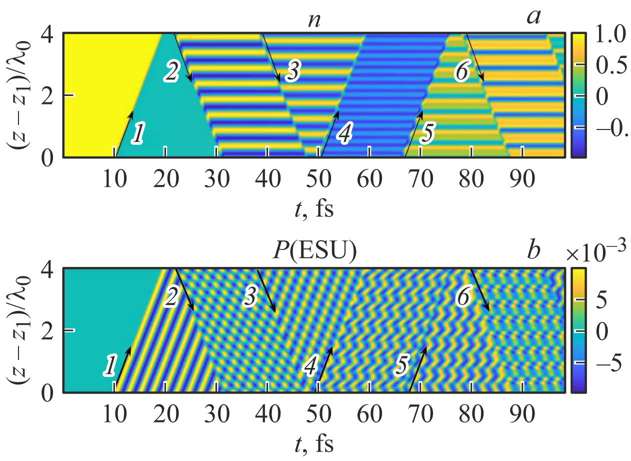

Figure 3. (a) Dynamics of the population difference $n(z, t)$, (b) dynamics of the polarization $P(z, t)$ under the action of single-cycle attosecond pulses 1 and 2 in the form (4) and (5). Parameter $\tau_2 = 27\tau$. Other calculation parameters are given in the table.

with a period of $\lambda_0/2$ appears in the medium. The propagation of subsequent pulses leads to multiplication of the EMIGs period and the appearance of standing waves of polarization (Fig. 2). This dynamics is similar to those considered earlier in the papers [30–32], in which harmonic-shaped ESPs or unipolar Gaussian pulses were used to create an EMIG. Thus, rectangular pulses can also be used to create and superfast control of EMIG in a resonant medium.

Fig. 3 illustrates how the EMIG dynamics of inversion and polarization changes with a change in the delay value ($\tau_2 = 27\tau$). It can be seen that the 2nd pulse also induces the EMIG of the population difference, but pulse 3 shifts the grating in space without changing its spatial period. Subsequent pulses create an inversion grating, which has a complex spiking structure. The polarization dynamics in this case is also interesting (Fig. 4, b). One can see alternating sections of small sizes in the form of zigzag structures at times 50−85 fs: polarization waves appear in these sections, running in opposite directions. Previously, such structures have already been observed in numerical calculations, when harmonic one-cycle and sub-cycle pulses collided in the center of the medium [25,33,34,37]. In the case of non-crossing ESPs, such structures were not observed.

EMIGs created by a sequence of rectangular pulses overlapping in the medium

The EMIG dynamics is considered when a pair of rectangular pulses (5) and (6) overlap inside the medium; the place of overlap was regulated by choosing τ_1 and τ_2.

Fig. 4 shows an example of the EMIG dynamics of the population and polarization difference when a pair of $\pi/2$ pulses collide near the right edge of the medium in the

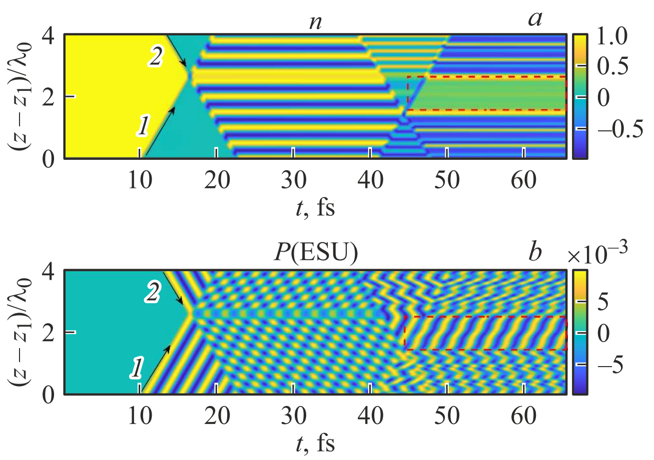

Figure 4. (*a*) Dynamics of the population difference $n(z,t)$, (*b*) dynamics of the $P(z,t)$ polarization: a pair of $\pi/2$-pulses collide near the right edge of the medium near the point $z - z_1 = 2.8\lambda_0$ at $\tau_1 = 2\tau$, $\tau_2 = 8\tau$. Other calculation parameters are given in the table.

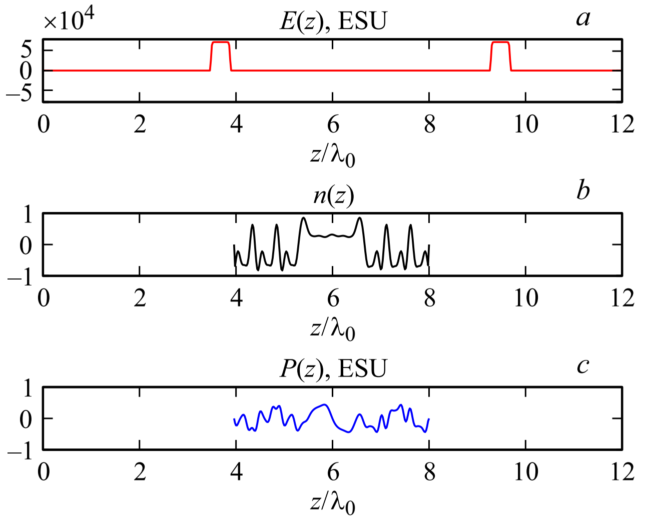

Figure 5. Instantaneous value of the field strength $E(z)$ (*a*), population difference $n(z)$ (*b*), and polarization $P(z)$ (*c*) as functions on the z coordinate at the time $t = 65.3$ fs for the situation shown in Fig. 4. The medium is located between the points $z_1 = 4\lambda_0$ and $z_2 = 8\lambda_0$.

region of the point $z - z_1 = 2.8\lambda_0$ at $\tau_1 = 2\tau$, $\tau_2 = 8\tau$ (other calculation parameters are the same as in the table). If, after the passage of pulses 1 and 2, ordinary EMIGs inversions with a period $\lambda_0/2$ are formed, then after the passage of pulses 3 and 4, an unusual situation arises in the medium (Fig. 4, *a*). Between the points $z - z_1 = 5.5\lambda_0$ and $z - z_1 = 6.5\lambda_0$, a light-induced „channel" or microresonator appears i.e. the region with a constant value of the population difference $n \cong 0.3$, at the ends which jumps in the population difference arise up to the value $n = 1$. This „channel" is marked with a red dashed rectangle in Fig. 4, *a*. For clarity, Fig. 5 presents the instantaneous value of the field strength $E(z)$ (*a*), the population difference $n(z)$ (*b*), and the polarization $P(z)$ (*c*) depending on the coordinate z at time $t = 65.3$ fs. The formation of a microresonator is clearly seen in Fig. 5, *b*. In Fig. 4, *b* at times 50–65 fs, zigzag polarization structures are again seen.

It is possible to increase the depth of this „microresonator" or light-induced channel by slightly decreasing the amplitude of the second pulse field: Fig. 6 illustrates the change in the dynamics of EMIG of population inversion and polarization at $E_{02} = 0.9E_{01}$. The formation of a light-induced microresonator with a constant inversion value $n = 0$ and a sharp jump in the inversion value at the boundary is also visible (the area is highlighted with dashes in Fig. 4, *a*). Snapshot of the channel is shown in Fig. 7, *b*, which shows the value of the inversion $n(z)$ at time $t = 65.3$ fs for the case in Fig. 6. The figures show the formation of a deep inversion channel, on both sides of which the spatial distribution of the inversion has the form of a sinusoid with two peaks.

It is interesting to note that, as shown by numerical calculations, this rectangular structure of the population difference in the form of a channel is preserved in the medium if the length of the medium is shortened, leaving

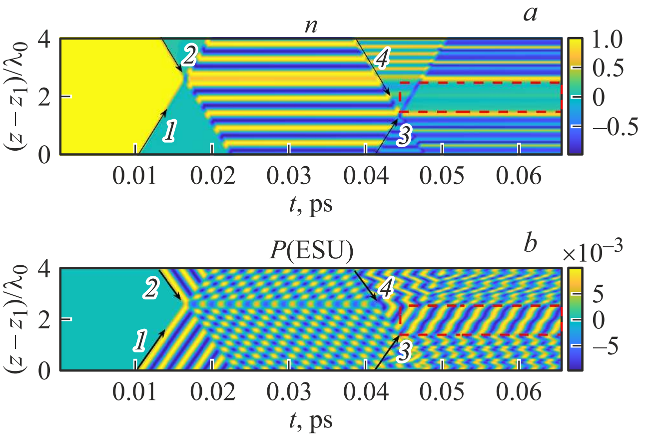

Figure 6. (*a*) Dynamics of the population difference $n(z,t)$, (*b*) dynamics of the $P(z,t)$ polarization: a pair of $\pi/2$-pulses collide near the right edge of the medium near the point $z_1 = 2.8\lambda_0$; $E_{02} = 0.9E_{01}$. Other calculation parameters are given in the table.

only the region in which the channel exists in it. In this case, the parts of the medium removed from consideration do not affect the formation of this channel (Fig. 8 and 9).

If the 2nd pulse amplitude is increased two times, the mentioned channel in the form of a well disappears. In this case, the appearance of a rectangular barrier is possible, in which the population difference is close to unity along this section, and then drops abruptly towards smaller values. Example of the appearance of such a structure in the form of a barrier is shown in Figs 10 and 11 at $E_{02} = 1.3E_{01}$. Other parameters are the same as in Fig. 9, 10. This structure is preserved with a smooth increase in the amplitude of the pulse field 2 E_{02}, and then disappears.

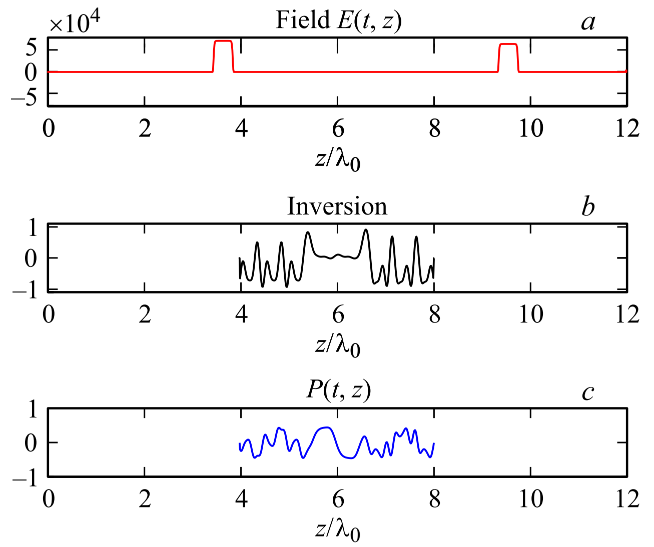

Figure 7. Instantaneous value of the field strength $E(z)$ (a), population difference $n(z)$ (b) and polarization $P(z)$ (c) depending on the z coordinate at the time $t = 65.3$ fs for the case in Fig. 6. The medium is located between the points $z_1 = 4\lambda_0$ and $z_2 = 8\lambda_0$.

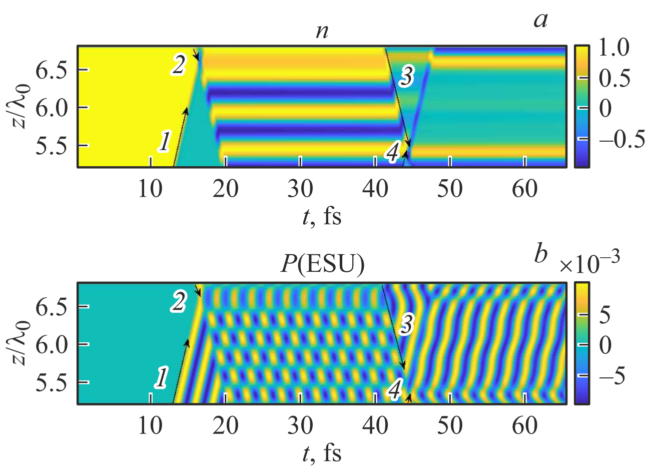

Figure 8. (a) Dynamics of the population difference $n(z, t)$, (b) dynamics of the polarization $P(z, t)$ for the parameters in Fig. 6, 7, for shortening of the medium length of the region in which the light-induced microresonator exists.

The possibility of inducing light-induced channels in photosensitive materials has been actively studied lately [50–52]. When a laser beam propagates in a photosensitive medium, a step in the refractive index is formed, which leads to the formation of self-written waveguides. Such structures remain in the medium after the passage of the laser beam; however, in our case, during the formation of EMIG during coherent propagation of ESPs in a resonant medium, they exist in the medium during the coherence time T_2. The induction of such microresonators is topical in connection with the problem of self-stopping of short light pulses in a resonant medium [42]. The above results show the possibility of ultrafast EMIG formation and switching on atto-second time scales and illustrate the rich dynamics of the system.

Conclusion

In this work, based on the numerical solution of the system of Maxwell-Bloch equations for a two-level medium, the dynamics population inversion EMIGs induced by atto-second rectangular pulses was studied. In the case when the pulses did not overlap in the medium, the EMIG dynamics is completely similar to that observed when the medium was excited by harmonic pulses [30,31]. In this case, not only induction is possible, but also the multiplication of the EMIG spatial frequency. The appearance of alternating

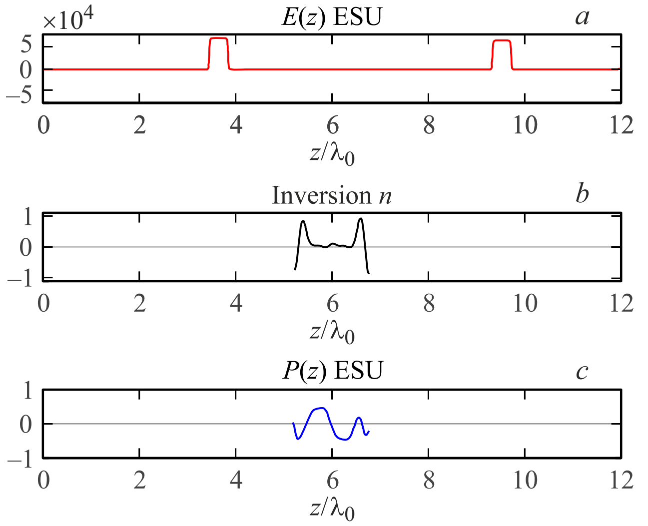

Figure 9. Instantaneous value of the field strength $E(z)$ (a), population difference $n(z)$ (b) and polarization $P(z)$ (c) depending on the coordinate z at the time $t = 65.3$ fs with the parameters in Fig. 8. The medium is located between the points $z_1 = 4\lambda_0$ and $z_2 = 8\lambda_0$.

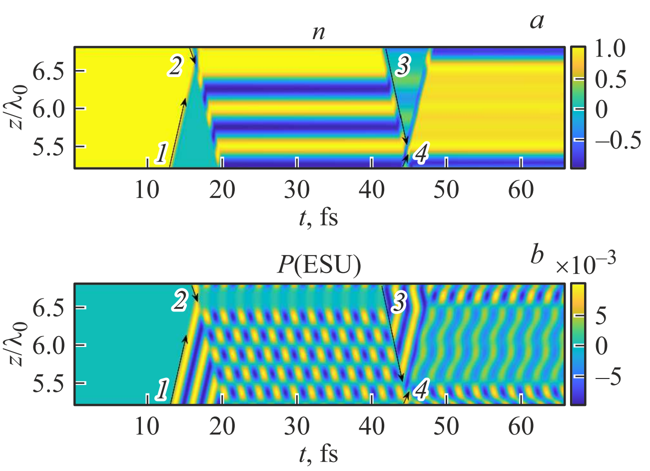

Figure 10. (a) Population difference dynamics $n(z, t)$, (b) polarization dynamics $P(z, t)$ at $E_{02} = 1.3E_{01}$. Other parameters are the same as in Figs. 8, 9.

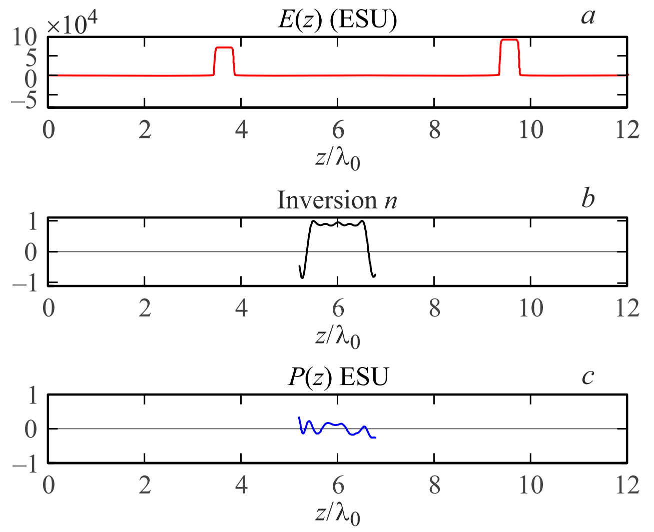

Figure 11. Instantaneous value of the field strength $E(z)$ (a), population difference $n(z)$ (b) and polarization $P(z)$ (c) depending on the z coordinate at the time $t = 65.3$ fs with the parameters in Fig. 10. The medium is located between the points $z_1 = 4\lambda_0$ and $z_2 = 8\lambda_0$.

sections is shown, in which polarization waves arise, running in opposite directions. Previously, similar situations were observed only when the pulses collided in the center of the medium [25,33,34,37].

Unusual form of population inversion EMIG was discovered in the case when rectangular pulses collided: the formation of light-induced microresonators („channels“) i.e. sections of the medium along which the inversion has a constant value, and at the ends an inversion step occurs. Note that the demonstrated structures can be considered as dynamic short-lived fibers for radiation propagating in the direction perpendicular to the radiation that forms them. Such channels can be induced at different distances from the edge of the medium.

Similar EMIG structures can be quickly created in various materials, and their lifetime is limited by the relaxation time T_1 of the medium. The presented results once again show the possibility of ultrafast control and switching of the state of the spatial parameters of the medium using unipolar nonharmonic pulses.

Finally, it is appropriate to make a general remark. When exposed to two or more multi- or low-cycle pulses of the usual harmonic form, a situation arises when the spatial parameters of the medium are modulated by interference between pulses (it does not matter whether interference occurs when pulses overlap or in the absence of simultaneous overlap due to interference with medium polarization waves), which has harmonic look. In the medium, depending on the nature of the nonlinear response, the harmonic form is transformed by some means or other. This seems obvious, but we do not know about the formulation of the problem of obtaining structures in a resonant medium with a pronounced desired non-harmonic shape. The example of numerical simulation given in the work shows that such a situation is most likely possible. Although the work gives the result in a one-dimensional model, it is interesting to carry out a theoretical analysis of systems of equations similar to the one used by us, where it will be possible to demonstrate in a general form the existence of fields capable of inducing EMIG structures of a predetermined shape. In our opinion, it will be essential to remove restrictions on the harmonic multicycle form of radiation pulses, i.e., the fields capable of solving such a problem will not be harmonic, but the impulses will be unipolar.

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Conflict of interest

The authors declare that they have no conflict of interest.

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