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Generation of isolated attosecond pulses with large electric area in a dense resonant medium

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Obtaining unipolar half-cycle optical pulses of femto- and attosecond duration with a large electrical area is an urgent but difficult task. The reason for the emerging difficulties lies in the existence of the rule of conservation of the electrical area of the pulse, which does not allow converting a bipolar pulse into a unipolar one. In this work, it is shown that in a resonant medium a few-cycle pulse can be converted into two unipolar pulses separated in time by a distance that is an order of magnitude or more longer than the duration of the initial pulse. This allows in a number of problems to consider such pulses separately as unipolar. The estimation of the electric area value relative to its „atomic scale"is carried out.

Keywords: attosecond pulses, unipolar pulses, subcycle pulses, electrical area of pulses.

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Introduction

Pulses in the attosecond range of durations contain several oscillation cycles, that allows to use them to study the ultrafast dynamics of wave packets in atoms, molecules and solids [1–6]. The field strength vector components in them reverse direction several times over the pulse duration. Therefore, the electric area of such pulses, calculated as the integral of the electric field strength over time $S_E = \int E(t)dt$, is 0, and they are not unipolar [7].

It is possible to reduce the duration of the pulse containing one cycle of oscillations, if „cut off" one of the field half-waves and turn it into a unipolar half-cycle pulse containing a half-wave of the field of one polarity.

Unipolar pulses, due to non-resonant action on micro-objects, are able to quickly and more effectively change the state of quantum systems compared to long multicycle pulses [3,8–14], accelerate charged particles [15], rotate the electron spin [16,17], perform holographic recording with ultra-high temporal resolution [18].

For unipolar pulses one of the most important characteristics is the pulse electric area S_E. The impact of such pulses on micro-objects is determined precisely by the pulse electric area, and not by its energy [9–17].

In the experiments and theoretical studies it is most often possible to obtain quasi-unipolar pulses containing a unipolar field burst with a large amplitude and a long trailing edge of opposite polarity [3,5,19–23]. As theoretical and experimental studies show, it is the unipolar pulse component, that has a significant effect, and the long reverse polarity front practically does not affect the system [3,11,12].

One can try to obtain unipolar pulses of a large electric area using the phenomenon of self-induced transparency (SIT) [24,25]. In the case of SIT the leading edge of a short pulse transfers the medium from the ground state to an excited one, and at the trailing front the medium returns the absorbed energy to the pulse and passes into the ground unexcited state. In this case, the radiation propagates in the medium practically without loss. Besides a strong change in the pulse shape can occur.

A two-stage single-cycle pulse compressor based on the SIT phenomenon was considered in the study [26]. In it the original bipolar one-cycle pulse, consisting of two half-waves of opposite polarity, experienced the following transformation. Unipolar half-waves shortened their duration and were attracted to each other. As a result of compression and attraction of half-waves of a single-cycle pulse, the duration of a single-cycle pulse was reduced by several times. Using several media with multiple transition frequencies, the initial pulse duration can be reduced from a few femtoseconds to a few attoseconds [26].

However, the impulse thus obtained remains bipolar. In this study we will show, that there is another scenario for changing the shape of a single-cycle pulse during propagation in the SIT mode. The unipolar pulse half-waves will not approach each other, but, on the contrary, will lag behind each other. Such a possibility is indicated by an example of a numerical solution of the problem of propagation of a low-cycle pulse in a dense resonant medium [27]. The pulse incident on the medium was divided into several subpulses. The first and main pulse contained several cycles. It was followed by a pair of spaced apart unipolar pulses with opposite polarity. Two pulses with opposite polarities were obtained by calculating the reflection of a low-cycle pulse from a medium with a quadratic or cubic nonlinearity [22]. In the case of dissipative SIT solitons, a similar solution was observed in studies [28,29].

The issue of possible applications of a resonant medium for the separation of two unipolar pulses in time has not been studied and will be considered in this study. A comparison of the electric area of individual unipolar components with the values of the electric area required to excite the simplest quantum systems will be made.

Be reminded, that in one-dimensional problems of light propagation in media with dissipation, the rule of preservation of the pulse electric area [7,30,31] applies. Therefore, if there is no significant reflection from the boundaries of the radiation media, which can have a unipolar character, then it is impossible to obtain unipolar radiation from an initially bipolar pulse. The area of the radiation that has passed through the medium should be equal to zero.

The scale of the quantum systems pulse electric area

In the recent study [32] the value „of the electric area atomic measure"is introduced, it specifies the unipolar pulse electric area required to empty the ground state of the quantum system. The expression has a universal form:

$$S_0 = \frac{2\hbar}{aq}. \qquad (1)$$

It includes the charge q, $\hbar$ — the reduced Planck constant and a — the typical size of the system.

Expression (1) is applicable to various systems. By virtue of the Heisenberg uncertainty relation, in a system with size $\sim a$, the momentum has a magnitude of the order of $\hbar/a$ [33]. On the other hand, the pulse electric area coincides with the change under the action of the average quantum-mechanical pulse value, referred to the unit electric charge of the system [34]. Thus, the atomic measure of the pulse electric area is equal to the typical quantum-mechanical pulse „of a free" system.

For a hydrogen atom (1) $a = a_0$, where $a_0 = 0.5 \cdot 10^{-8}$ cm –radius of the first Bohr orbit of the hydrogen atom. Then $S_0 = \frac{2\hbar}{a_0 q} = 8.78 \cdot 10^{-10}$ erg·s/cm·ESU. This value of the electric area serves „as an atomic scale" of the electric area and sets the scale of the electric area during the interaction of the unipolar pulse with the atomic system. If the pulse electric area is greater, than this value, such an impulse is able to change the system state and has a large electric area on this scale.

For hydrogen-like formations in solids, vibrations and rotations of molecules, electrons in conducting nanoparticles, the typical dimensions a are orders of magnitude larger and, accordingly, the values „of the scale" are smaller.

Bipolar to unipolar conversion factor

Be noted once again, that in one-dimensional problems of light propagation in media with dissipation, the electric area conservation rule does not allow to obtain unipolar radiation from an initially bipolar pulse, if there is no significant reflection from the boundaries.

However, in the considered problem of converting a single-cycle pulse into two „isolated" (remote at a distance much greater than their length) unipolar pulses with opposite polarities, it is appropriate to raise the question of how the value of the area of the positive/negative bipolar pulse component will change, when converted to spaced impulses. To do this, it is proposed to introduce a conversion factor U equal to the ratio of the electric area E_{out} of one of the unipolar pulses to half of the electric area of the field strength modulus E_{in} in the bipolar pulse:

$$U = \frac{|\int E_{out} dt|}{1/2 \int |E_{in}| dt}.$$

This coefficient can be useful in assessing the efficiency of converting low-cycle pulses into „solitary" unipolar ones.

Modeling the production of „solitary" attosecond pulses of a large electric area

To theoretically describe the propagation of short pulses in a resonant medium, the system of Maxwell-Bloch equations was used, which describes the evolution of the off-diagonal elements of the density matrix of a two-level medium, the level population difference, and the electric field. This system of equations is as follows [26]:

$$\frac{\partial \rho_{12}(z,t)}{\partial t} = -\frac{\rho_{12}(z,t)}{T_2} + i\omega_0 \rho_{12}(z,t) - \frac{i}{\hbar} d_{12} E(z,t) n(z,t), \qquad (2)$$

$$\frac{\partial n(z,t)}{\partial t} = -\frac{n(z,t) - n_0(z)}{T_1} + \frac{4}{\hbar} d_{12} E(z,t) \mathrm{Im}\rho_{12}(z,t), \qquad (3)$$

$$P(z,t) = 2N_0 d_{12} \mathrm{Re}\rho_{12}(z,t), \qquad (4)$$

$$\frac{\partial^2 E(z,t)}{\partial z^2} - \frac{1}{c^2}\frac{E(z,t)}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 P(z,t)}{\partial t^2}. \qquad (5)$$

Here ρ_{12} — off-diagonal element of the density matrix, $n \equiv \rho_{11} - \rho_{22}$ — population difference between the ground and excited states of two-level system, P — medium polarization, N_0 — concentration of active centers, E — electric field strength with fixed linear polarization, c — speed light in vacuum, ω_0 — resonant transition frequency of the medium ($\lambda_0 = 2\pi c/\omega_0$ — resonant transition wavelength), n_0 — equilibrium population difference ($n_0 = 1$ for an absorbing medium). See [26] for more details on the applicability of this model for this problem.

In this paper, the equations for the density matrix (2) and (3) were solved numerically using the Runge-Kutta

Parameters used in the given example

Parameter	Value
Medium thickness, μm	4.5
Concentration N_0, cm^{-3}	$8.5 \cdot 10^{22}$
Dip. moment d_{12}, D	5
Relaxation time T_1, ps	10
Relaxation time T_2, ps	10
Central wavelength of the transition λ_0, nm	700
Transition frequency ω_0, s^{-1}	$2.69 \cdot 10^{15}$
Input pulse amplitude E_0, ESU	$1.8 \cdot 10^6$
Input pulse duration, τ_p, fs	2.61

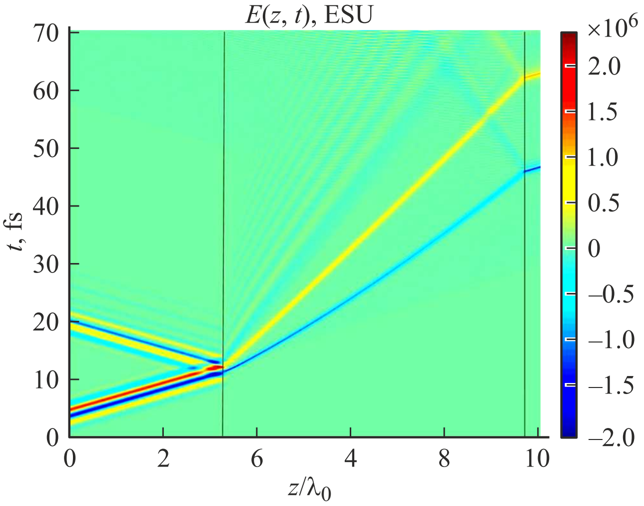

Figure 1. Evolution of a single-cycle pulse during propagation in a two-level resonant medium, the boundaries of which are indicated by vertical lines. Vacuum is located on both sides of the medium.

method, the wave equation (5) — by the finite difference method. The incident pulse is as follows

$$E(t) = E_0 e^{-\frac{(t-z/c)^2}{\tau_p^2}} \sin\left(\omega\left(t - \frac{z}{c}\right)\right), \qquad (6)$$

where E_0 – pulse amplitude, τ_p – pulse duration, and ω – center frequency. In further calculations, the pulse center frequency is equal to the medium transition frequency, $\omega = \omega_0$. The parameters used in the given example of calculations are given in the table. Similar values of medium parameters can be implemented in various nanostructures and semiconductor materials [26,35–37]. Pulses with the parameters used in the calculations can now be obtained experimentally [38].

The results of numerical simulation are shown in Fig. 1. A single-cycle pulse in the form (6), containing two unipolar half-waves of opposite polarity, enters the resonant medium, the boundaries of which are shown by vertical lines in Fig. 1. These half-waves propagate in the medium at different speeds, which leads to their spreading. As a result, a pair of unipolar pulses of opposite polarity spaced apart in time is formed at medium output. The time dependence of the pulse electric field strength at the medium input and output is shown in Fig. 2.

Calculations have shown, that the distance between pulses at the outlet of the absorbing medium will be affected by the concentration of particles. The concentration dependence of the distance between pulses is given in Fig. 3. It has three characteristic areas (highlighted in color). In the first section, when the concentrations are less than $3 \cdot 10^{22}$ cm^{-3}, the distance between pulses is equal to the distance between half-waves in the initial pulse. Then comes the concentration range up to $5.5 \cdot 10^{22}$ cm^{-3}, where the pulse compression is observed, that was described in the study [26]. After the section with compression, there is a region where „repulsion“ of unipolar pulses occurs.

At a concentration value of $9 \cdot 10^{22}$ cm^{-3} and more, the second pulse begins to change its shape, its duration increases, and oscillations appear (Fig. 4).

The dependence of the delay between pulses on time T_2 is interesting, it is shown in Fig. 5. The figure demonstrates, that a decrease in time T_2 at first does not affect the delay

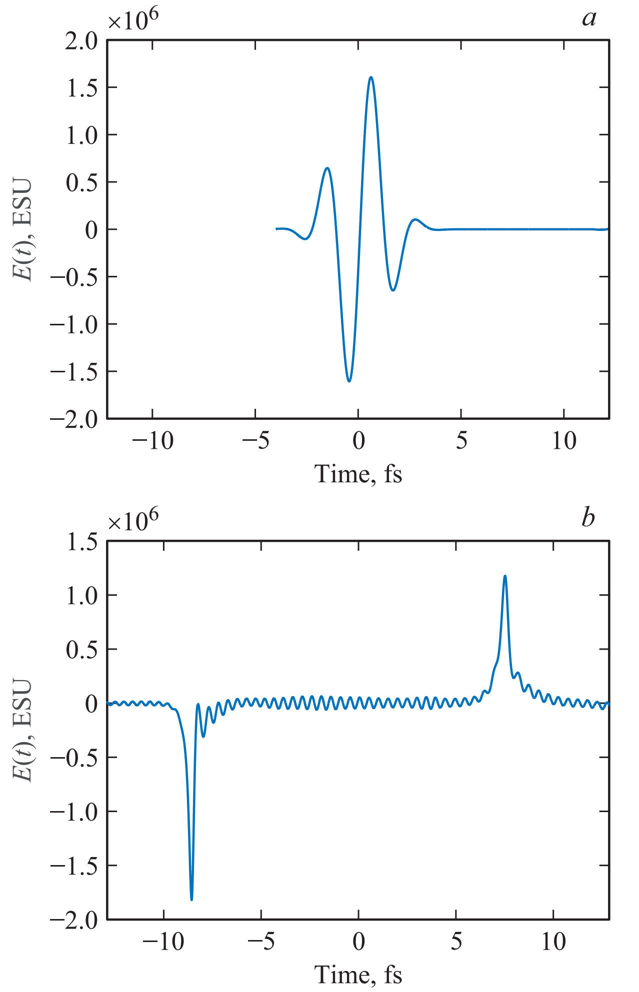

Figure 2. Dependences of the electric field strength $E(t)$ on time at the medium input (a) and output (b).

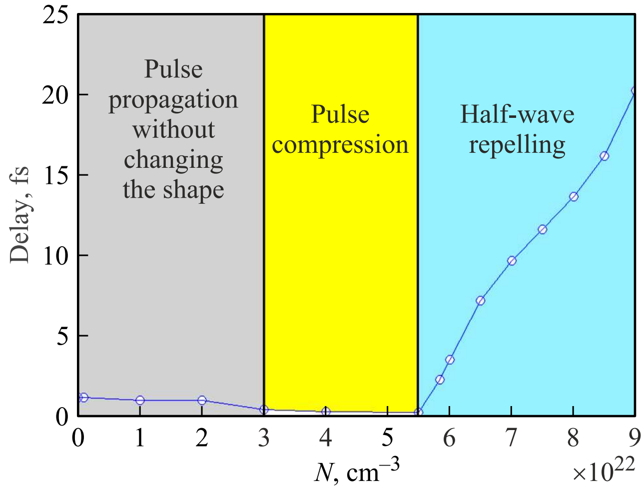

Figure 3. Dependence of the delay between pulses on the concentration N_0. Other parameters are the same, as in Fig. 1. The figure shows the areas of parameters, at which the pulse propagates without changing its shape, the input pulse compression zone, and the area, where the distance between unipolar waves, increases.

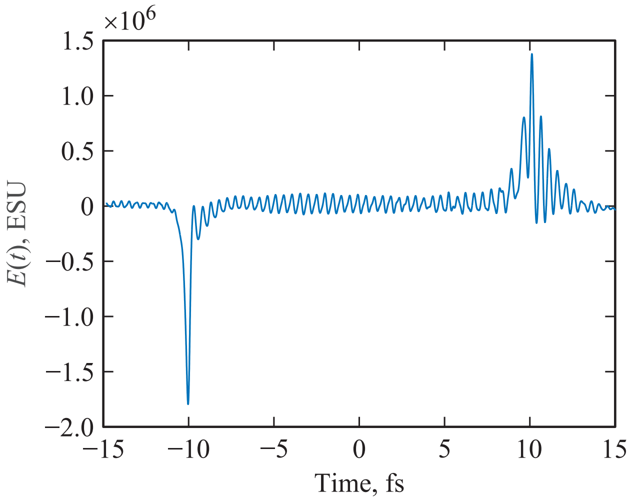

Figure 4. The field strength $E(t)$ at medium output at $N_0 = 9 \cdot 10^{22}$ cm^{-3}.

between pulses, and then results in its rapid decrease. This behavior is understandable, if we assume, that the first pulse leaves behind the medium in a state, that slows down the movement of the second pulse. For an extremely small value of time T_2, the influence of the first pulse on the second one disappears, and therefore the pulses do not „push apart".

The above example demonstrates, that for a medium length of $6.4\,\mu$m the time interval between two pulses is 20 fs. Increasing the medium length will increase the distance between the pulses, however, the destruction of such a propagation regime can also occur simultaneously.

As it is already noted, two unipolar pulses separated in time have the ability to selectively excite quantum systems. The impact selectivity is determined by the delay between them [11,12]. With the proper choice of delay, the second pulse can amplify the effect of the first. This is relevant, for example, in problems of obtaining superradiance „of stopped polarization" [39]. The amount of delay can be

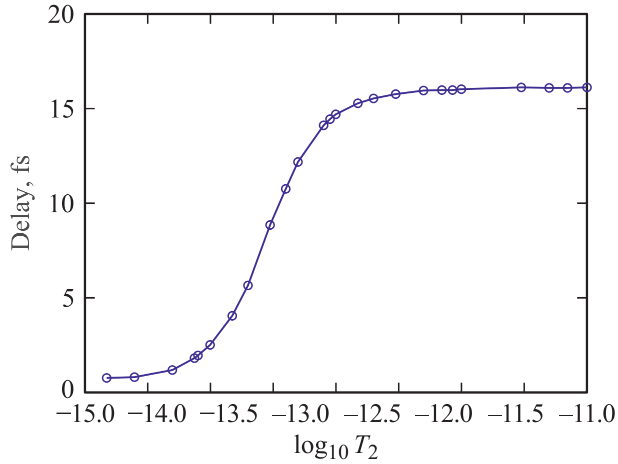

Figure 5. Dependence of the delay between pulses on the relaxation time T_2. The abscissa shows the decimal logarithm of T_2, expressed in seconds. Other parameters are the same, as in Fig. 1.

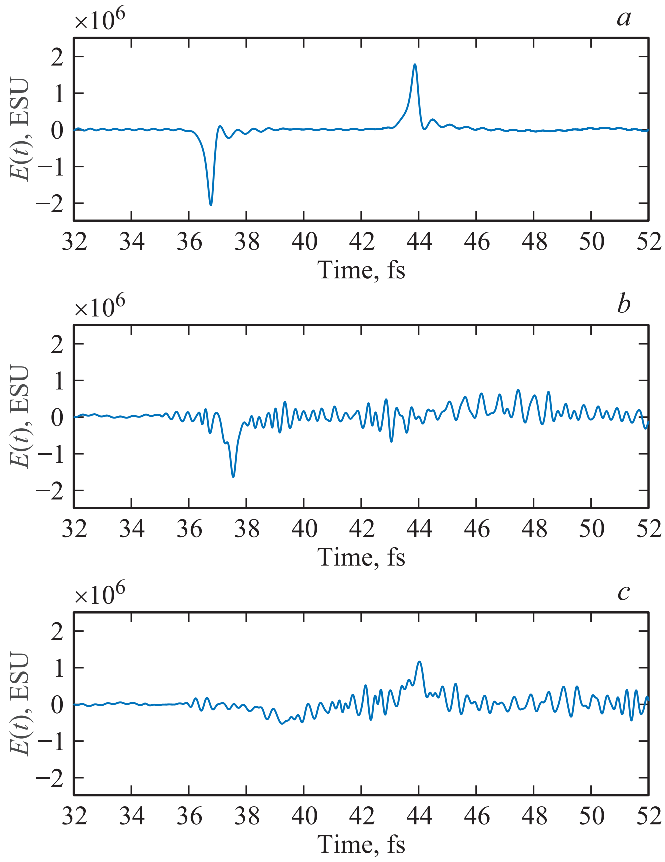

Figure 6. Radiation at the medium output: a — in the absence of a counter pulse; b — in the presence of a counter pulse with envelope amplitude $E_0 = -1.7 \cdot 10^6$ ESU; c — for the amplitude of the counter-pulse envelope $E_0 = 1.7 \cdot 10^6$ ESU.

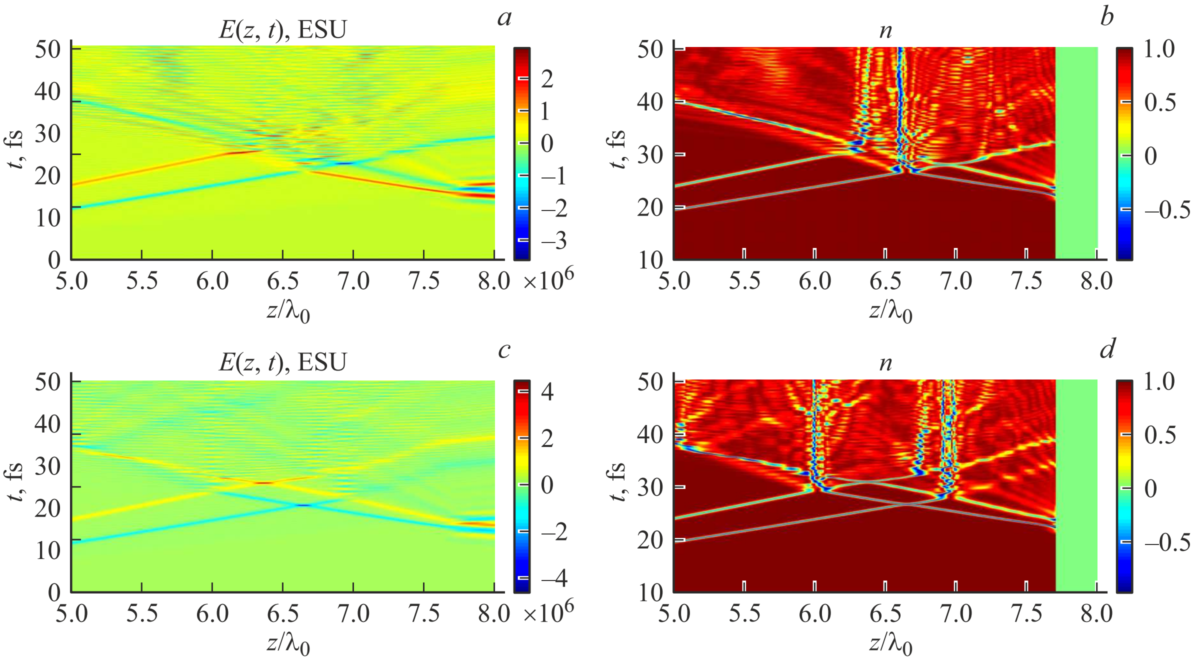

Figure 7. Areas of the medium, where pulses collide. Dependence of the field strength E (a, c) and the population difference n (b, d) under the conditions of Fig. 6, ,b (a, b) and 6, c (c, d).

adjusted by the medium length. A change in thickness can be created by tilting a thin plate relative to the beam propagation direction.

Note, that the pulse area in Fig. 2, b is $8.7 \cdot 10^{-10}$ erg·s/cm·ESU. Half the area of the modulus of the incident pulse field strength in Fig. 2, a is equal to $1.7 \cdot 10^{-9}$ erg·s/cm·ESU. In the above example the conversion factor is $U = 0.5$.

Suppression of one „solitary" component in a pulse collision

If radiation is formed in the medium in the form of two pulses of opposite polarity, as in Fig. 2, b, then one can try to isolate one of them by directing a short bipolar pulse towards it. Below is an example, which demonstrates this possibility. The results of the corresponding calculation are given in Fig. 6. The calculation is performed for a medium of length $3.8\,\mu$m, concentration $N_0 = 7 \cdot 10^{22}\,\text{cm}^{-3}$ and relaxation time $T_2 = 10^{-13}$ s. Other parameters are the same as in the calculation in Fig. 1.

In the example given in Fig. 6, b, the counter pulse had a lower amplitude and its sign was opposite to the initial pulse amplitude. When the amplitude sign changed (Fig. 6, c), both short unipolar bursts disappeared.

The collision scenario, leading to a decrease in the amplitude of the second unipolar burst and an increase in its duration, is shown in the graphs of the field behavior and population difference in the collision zone (Fig. 7).

Fig. 7 shows, that the counterpropagating pulse also splits into two unipolar pulses. In a situation, when it is possible to isolate one pulse, the first component of the initial pulse and the counter pulse have opposite polarities. The amplitude of the counter pulse is smaller. Developing in the area of collision, the counter pulse field slightly attenuates the first unipolar pulse and then significantly attenuates the second unipolar wave. This is shown in Fig. 6, b. In case, when their amplitudes have the same sign, the first wave is correspondingly attenuated as well (Fig. 6, c). Let us note, that after the collision of the first parts of counterpropagating pulses, a small-scale structure of the population difference is formed (Fig. 7, b, c), through which the delayed wave should pass. This structure is the source of radiation. Therefore, we observe fast field modulation in the situations in Fig. 6, b, c.

Discussion and conclusion

The study theoretically demonstrates the possibility of obtaining subcycle unipolar pulses with a large electric area based on the SIT phenomenon during the propagation of a single-cycle bipolar pulse in a two-level resonantly absorbing medium. It is shown, that during the propagation of the initial pulse, its unipolar components with opposite polarity move in the medium at different speeds, which leads to the formation of two „solitary " unipolar pulses with opposite polarity separated in time at the medium output. The duration of the obtained subcycle pulses is on the order of 390 as. The pulses electric area coincides with the value „of the atomic measure of electric area" for the hydrogen atom, and such a pulse has a large electric area.

It is shown, that it is possible to form a subcycle pulse with a short burst of high polarity of one polarity and a long front of opposite polarity upon collision of two bipolar pulses.

The considered situations of transformation of a bipolar pulse can be considered as options for obtaining unipolar pulses with a large electric area, which are necessary for fast and efficient control of quantum systems and acceleration of charged particles.

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Information on authors contribution

The calculations were performed using programs modified by S.V. Fedorov.

Conflict of interest

The authors declare, that they have no conflict of interest.

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