

Unipolar half-cycle pulse propagation in an amplifying three-level medium

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Received March 07, 2024

Revised March 07, 2024

Accepted March 21, 2024

The propagation of a unipolar half-cycle pulse through a layer of a non-equilibrium three-level resonant medium with different energy-level schemes is studied theoretically. It is shown that in such a case initial unipolar pulse gets gradually transformed into a bipolar one due to the formation of oscillating tails at the trailing edge of the pulse. Depending on the specific energy-level scheme both amplification and absorption of a leading half-cycle field burst can take place with the propagation distance. At the same time in all cases the conservation rule of the electric area is obeyed, although the unipolarity degree of the pulse always decreases over its propagation in the medium layer.

Keywords: subcycle pulses, electric pulse area, unipolar pulses, light-matter interaction.

DOI: 10.61011/EOS.2024.04.58886.6127-24

Introduction

The optics of attosecond light pulses, including both the search for methods of generation of attosecond pulses and the study of specific features of their interaction with different media [1–5], is progressing rapidly at present. The main advantage of such pulses is that they provide an opportunity to monitor (and even control) various ultrafast processes in nanostructures, molecules, and even individual atoms [1,5,6].

Subcycle pulses (primarily half-cycle ones) are of particular interest [7–9]. Although half-cycle attosecond pulses are rather hard to obtain, a considerable number of methods for their generation have been proposed in recent years [10–16]; half-cycle pulses generated in certain studies were as short as several attoseconds.

Half-cycle pulses have the important property of unipolarity, since they contain only one half-wave with a constant sign of the electric field. The electric area of a pulse [17], which is written as

$$\bar{S}_E(\bar{r}) = \int_{-\infty}^{+\infty} \bar{E}(\bar{r}, t') dt', \quad (1)$$

is the time integral in infinite limits of the electric field strength at a given point in space. This quantity allows one to formulate a more precise definition of unipolar pulses: these are the pulses with a non-zero value of electric area (1).

Electric area of a pulse (1) is not only convenient for introducing a general definition of unipolar pulses, but also has a number of important features. The most important of them is the fundamental rule of conservation of electric area

in space

$$\frac{d}{dx} S_E(x) = 0,$$

which follows directly from Maxwell's equations and is satisfied in the propagation of ultrashort pulses in an arbitrary medium with dissipation in one-dimensional geometry [17,18] (in certain scenarios, such as propagation in ferromagnetic media [19], deviations from this rule may be observed). In addition, it was found that when a quantum system is excited by pulses shorter than the characteristic time of the internal system dynamics, their effect on the system is no longer determined by the energy of a pulse; instead, it is specified completely by its electric area (1) [20–24].

A very challenging accompanying problem arising in the application of attosecond pulses is the control of their time profile. Several methods for solving this problem, such as attosecond synthesis [25–27] or coherent control of induced polarization in a resonant medium layer [28,29], have been proposed in recent years. Thus, the development of techniques for controlling the parameters of unipolar pulses (specifically, their shape and amplitude) becomes relevant.

The fundamental rule of conservation of the electric area of ultrashort pulses imposes certain restrictions on possible transformations of half-cycle pulses in the process of their propagation in different media. That said, the possibility of amplification of pulses (subcycle ones included) in non-equilibrium media has been demonstrated in several studies. Specifically, amplification of subcycle pulses in a non-equilibrium two-level medium during coherent propagation without relaxation was reported in [30–32]. This amplification was coupled with an intensification of alternating-sign oscillations of the field. The authors of [33,34] also used

the example of non-equilibrium plasma to demonstrate the possibility of both a reduction and an increase in electric area of subcycle pulses within the unidirectional propagation approximation. However, the approximate models used in [30–34] do not satisfy the rule of conservation of electric area; therefore, they may yield physically incorrect results (see [18]). The existence of unipolar half-cycle soliton-like objects in a non-equilibrium medium was demonstrated in [35,36].

One intriguing research direction in this context is the study of feasibility of amplification of half-cycle pulses propagating in a non-equilibrium medium. The propagation of a half-cycle unipolar pulse in a layer of an amplifying two-level medium was examined in [37,38]. It was found that this pulse is inevitably transformed in this case into a bipolar one with an extended oscillating tail, and the shape of this tail depends strongly on the phase relaxation time in the resonant medium. At the same time, the applicability of the two-level approximation itself to a resonant medium interacting with a subcycle attosecond pulse is questionable due to the extreme width of the spectrum of such excitation pulses. Therefore, a greater number of energy levels of a resonant medium need to be taken into account in order to obtain reliable data on the transformation of half-cycle pulses in propagation within this medium.

In the present study, the propagation of a half-cycle unipolar pulse in a layer of an amplifying three-level medium is investigated. Specifically, the variation of pulse parameters (its profile, energy, and electric area) in the process of propagation through a medium is examined. It is demonstrated that, despite the exact fulfillment of the fundamental rule of conservation of electric area, a unipolar half-cycle pulse is transformed into a bipolar one due to the excitation of oscillations of polarization of the medium at the frequencies of allowed transitions. Owing to the extreme width of the spectrum of an incident pulse, its stimulated amplification in a nonequilibrium medium is not observed in the general case. However, depending on the energy level diagram of the medium, the formation of oscillating tails at the trailing edge of a pulse may be accompanied by an increase in the maximum amplitude of the leading half-cycle field burst.

Model

Let us consider a planar layer of a three-level resonant medium with thickness L and axis x being orthogonal to this layer. Layer thickness L is thought to be much greater than the wavelengths of all resonant transitions in the medium. Let us also assume that a linearly polarized half-cycle pulse of the form

$$E(t) = E_0 e^{-t^2/\tau^2} \quad (2)$$

with duration τ and amplitude E_0 is incident normally on a multi-level resonant medium layer. If we assume for simplicity that the medium layer has very large transverse

Parameters of the considered three-level resonant medium, the initial incident pulse, and the medium layer

Incident pulse duration	$\tau = 500$ as
Incident pulse amplitude	$E_0 = 10^5$ CGSU
Volume concentration of atoms in the layer	$N_0 = 10^{20}$ cm ⁻³
Dephasing time	$T_2 = 5$ fs
Upper level lifetime	$T_1 = 50$ fs
Thickness of the medium layer	$L = 3.5$ μ m
Frequency of transition 1 \rightarrow 2	$\omega_{12} = 2.7 \cdot 10^{15}$ rad/s
Dipole moment of transition 1 \rightarrow 2	$d_{12} = 20$ D
Frequency of transition 1 \rightarrow 3	$\omega_{13} = 4 \cdot 10^{15}$ rad/s
Dipole moment of transition 1 \rightarrow 3	$d_{13} = 40$ D
Frequency of transition 2 \rightarrow 3	$\omega_{23} = 1.3 \cdot 10^{15}$ rad/s
Dipole moment of transition 2 \rightarrow 3	$d_{23} = 0$ D

dimensions, the problem for this linearly polarized excitation pulse is effectively reduced to a one-dimensional one.

The response of the resonant medium to the field of an excitation pulse is characterized using the equations for evolution of the density matrix of a three-level medium [39]:

$$\begin{aligned} \frac{d}{dt} \rho_{12} &= -i\omega_{12}\rho_{12} - \frac{\rho_{12}}{T_2} - \frac{id_{12}}{\hbar} E(t)(\rho_{22} - \rho_{11}) \\ &\quad + \frac{id_{23}}{\hbar} E(t)\rho_{13} - \frac{id_{13}}{\hbar} E(t)\rho_{32}, \\ \frac{d}{dt} \rho_{13} &= -i\omega_{13}\rho_{13} - \frac{\rho_{13}}{T_2} - \frac{id_{13}}{\hbar} E(t)(\rho_{33} - \rho_{11}) \\ &\quad - \frac{id_{12}}{\hbar} E(t)\rho_{33} + \frac{id_{23}}{\hbar} E(t)\rho_{12}, \\ \frac{d}{dt} \rho_{23} &= -i\omega_{23}\rho_{23} - \frac{\rho_{23}}{T_2} - \frac{id_{23}}{\hbar} E(t)(\rho_{33} - \rho_{22}) \\ &\quad + \frac{id_{13}}{\hbar} E(t)\rho_{12} - \frac{id_{12}}{\hbar} E(t)\rho_{13}, \\ \frac{d}{dt} \rho_{11} &= \frac{1 - \rho_{11}}{T_1} - \frac{2d_{12}}{\hbar} E(t)\text{Im}\rho_{12} + \frac{2d_{12}}{\hbar} E(t)\text{Im}\rho_{13}, \\ \frac{d}{dt} \rho_{22} &= -\frac{\rho_{22}}{T_1} + \frac{2d_{12}}{\hbar} E(t)\text{Im}\rho_{12} + \frac{2d_{23}}{\hbar} E(t)\text{Im}\rho_{23}, \\ \frac{d}{dt} \rho_{33} &= -\frac{\rho_{33}}{T_1} - \frac{2d_{13}}{\hbar} E(t)\text{Im}\rho_{13} - \frac{2d_{23}}{\hbar} E(t)\text{Im}\rho_{23}, \end{aligned} \quad (3)$$

where $\rho_{12}, \rho_{13}, \rho_{23}$ are off-diagonal elements of the density matrix that specify the polarization of the medium; $\rho_{11}, \rho_{22}, \rho_{33}$ are the populations of the 1st, 2nd, and 3rd levels of the medium, respectively; $\omega_{12}, \omega_{13}, \omega_{23}$ are the frequencies of resonant transitions; d_{12}, d_{13}, d_{23} are the dipole moments of these transitions; and T_1 and T_2 are the energy and phase relaxation times (for simplicity, these times are assumed to be equal for all transitions in the medium). The values of parameters of equation system (3) used in the modeling are listed in the table.

The evolution of the electric field in the considered system may be characterized by a one-dimensional scalar wave equation:

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

where $E(x, t)$ is the strength of the linearly polarized electric field, $P(x, t)$ is the induced polarization of the three-level resonant medium, and c is the speed of light in vacuum. It is also worth noting that one-dimensional wave equation (4) may characterize the propagation of ultrashort pulses (half-cycle ones included) in coaxial waveguides [40]. In turn, induced macroscopic polarization of the medium $P(x, t)$ on the right-hand side of (4) is expressed in terms of the off-diagonal elements of the density matrix and volume concentration N_0 of resonant centers in the layer in the following way:

$$P(x, t) = 2N_0(d_{12}\text{Re}\rho_{12} + d_{13}\text{Re}\rho_{13} + d_{23}\text{Re}\rho_{23}). \quad (5)$$

Equations (3)–(5) form a closed system that characterizes completely the spatiotemporal dynamics of the field and the medium within the model under consideration.

Amplifying medium layer

Let us examine the passage of a unipolar half-cycle pulse (2) through an optically thick layer of a three-level medium. The parameters of this layer and excitation pulse (2) are listed in the table. As a first step, we assume that the medium initially was in an excited nonequilibrium state. For definiteness, we also assume that all resonant centers of the medium were brought to the lower excited level (i.e., level 2). If a sufficiently long multi-cycle pulse

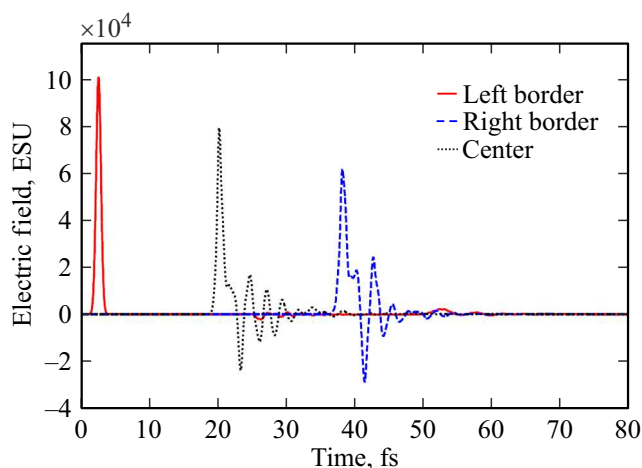


Figure 1. Time dependence of the electric field at three fixed points in space located to the left of the medium layer (red solid line), in the center of the medium layer (black dotted line), and to the right of it (blue dashed line). Incident half-cycle pulse (2) has duration $\tau = 500$ as and amplitude $E_0 = 10^5$ CGSU; the parameters of the medium are given in the table.

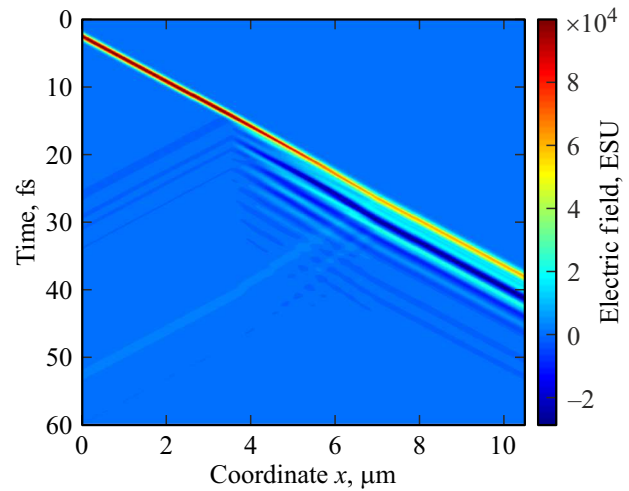


Figure 2. Spatiotemporal dynamics of the electric field in the process of passage of a unipolar half-cycle pulse (2) with duration $\tau = 500$ as and amplitude $E_0 = 10^5$ CGSU through an amplifying three-level medium layer with a thickness of $3.5\mu\text{m}$ and other parameters listed in the table.

with its central frequency close to the $1 \rightarrow 2$ transition frequency were to propagate through such a medium, it would undergo stimulated amplification [41]. A similar amplification effect was also demonstrated in a two-level amplifying medium for ultrashort pulses with a length of several optical cycles (e.g., by modulating the spatial distribution of the dipole moment of the resonant transition in the medium [42]). However, in the case of half-cycle unipolar pulse (2), the interaction with the medium becomes significantly non-resonant due to the extreme width of its frequency spectrum. Therefore, the dynamics of variation of pulse parameters is not a priori clear in this case and has to be studied through appropriate numerical modeling.

The system of equations (3)–(5) was solved numerically to determine the field. Wave equation (4) was solved using the FDTD (finite-difference time-domain) method, and equations (3) for evolution of the density matrix of a three-level medium were solved by the fourth-order Runge–Kutta method. Numerical modeling was carried out for a computational domain $10.5\mu\text{m}$ in extent. A resonant medium layer with a thickness of $3.5\mu\text{m}$ was positioned symmetrically in its center.

Figure 1 shows the time dependences of the electric field calculated at three fixed points within the computational domain. The first point (red solid line) was located on the left border of the calculation domain (i.e., at a distance of $3.5\mu\text{m}$ from the left boundary of the medium layer). The second point was positioned in the center of the medium layer (black dotted line). The third point (blue dashed line) was located on the right border of the calculation domain (i.e., at a distance of $3.5\mu\text{m}$ from the right boundary of the medium layer).

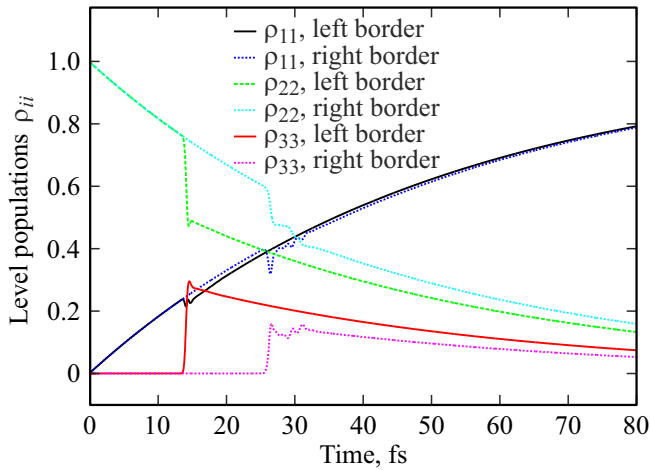


Figure 3. Temporal variation of the populations of levels of a three-level medium at points on the left and right borders of the medium layer for the example from Fig. 2.

It can be seen from Fig. 1 that oscillations form at the trailing edge of an initially unipolar pulse as it propagates in the medium. The intensity of these oscillations, which are caused by macroscopic polarization induced in the medium under the influence of the incident pulse itself, increases with distance traveled in the medium. Thus, the initial unipolar pulse transforms gradually into a bipolar one. The dynamics of this process is seen even clearer in Fig. 2, where both spatial and temporal variations of the electric field during the passage of half-cycle pulse (2) through the medium layer are presented in the form of a two-dimensional diagram.

Figure 3 shows the time dependences of the populations of levels of a three-level medium at two spatial points located at the boundaries of the resonant medium layer. All resonant centers were initially excited to level 2. In zero external field, all resonance centers relax gradually to ground level 1 at rate T_1 . At the same time, more complex dynamics of coherent interaction of a pulse with the medium and stimulated transitions between all levels of the medium is observed under the influence of an incident pulse and its emerging tails.

Specifically, the population of the second level drops sharply under the influence of an incident pulse; since this pulse needs a certain time to pass through the medium layer, the indicated reduction occurs at different moments at points on the left and right boundaries of the layer. In contrast, the population of the third level increases abruptly at the moment of arrival of half-cycle pulse (2). After the passage of a pulse, the populations of both excited levels decrease gradually to zero with characteristic time T_1 ; i.e., all resonance centers relax to the ground state. These results are presented in more detail in Fig. 4 that illustrates the spatiotemporal dynamics of the population of the first (ground) and second (initially populated) levels of the medium.

Figure 5 shows the time dependence of intensity (absolute value of the Poynting vector) at two given points in space located to the left and right of the medium layer. Thus, the dashed line in Fig. 5 represents the time dependence of intensity of the transmitted pulse, while the solid line corresponds to the intensity of both the initial incident pulse (2) and the reflected field trailing after with a certain time delay. It can be seen from Fig. 5 that, despite the presence of a non-equilibrium population of the resonant medium, the energy of the transmitted pulse is significantly lower than the energy of the initial half-cycle pulse. This is attributable to the presence of an initially unpopulated 3rd energy level of the medium: a significant part of resonant centers transition to it. As a result, the pulse energy is eventually dissipated as spontaneous emission. Thus, stimulated amplification of a half-cycle pulse is not observed, which is consistent with the results of calculations for a two-level amplifying medium [37,38].

Let us now examine the behavior of electric area (1) of a pulse in the considered example. For greater clarity, we use, in addition to electric area (1), the following convenient parameter introduced in [43]:

$$\xi(\vec{r}) = \frac{\left| \int_{-\infty}^{+\infty} \vec{E}(\vec{r}, t') dt' \right|}{\int_{-\infty}^{+\infty} |\vec{E}(\vec{r}, t')| dt'}, \quad (6)$$

which is hereinafter referred to as the degree of unipolarity of a pulse. It follows directly from definition (6) that this parameter assumes a value of 1 for a pulse with a constant electric field sign, while the degree of unipolarity in the contrary limiting case of an alternating bipolar field is zero. Thus, quantity ξ serves as a convenient indicator of the extent in which a pulse remains unipolar as it is transformed in the medium.

Figure 6 shows the spatial dependence of electric area (1) and degree of unipolarity $\xi(x)$ (6). It is evident that the fundamental rule of conservation of the electric pulse area is fulfilled exactly in this case. At the same time, degree of unipolarity $\xi(x)$ has a strong dependence on coordinate within the medium layer. Specifically, at the chosen values of parameters of the medium, the degree of unipolarity first drops sharply in the left part of the layer, but then reaches a certain saturation level and remains virtually unchanged in the right part of the layer. Thus, despite the emergence of field oscillations at the trailing edge of a pulse, the degree of unipolarity does not decrease to zero, but tends to a certain stationary non-zero value.

Influence of the energy level diagram

Resonant transitions $1 \rightarrow 2$ and $1 \rightarrow 3$ are allowed in the dipole approximation in the example three-level medium from the table (i.e., this medium has a so-called V level diagram). At the same time, owing to population inversion

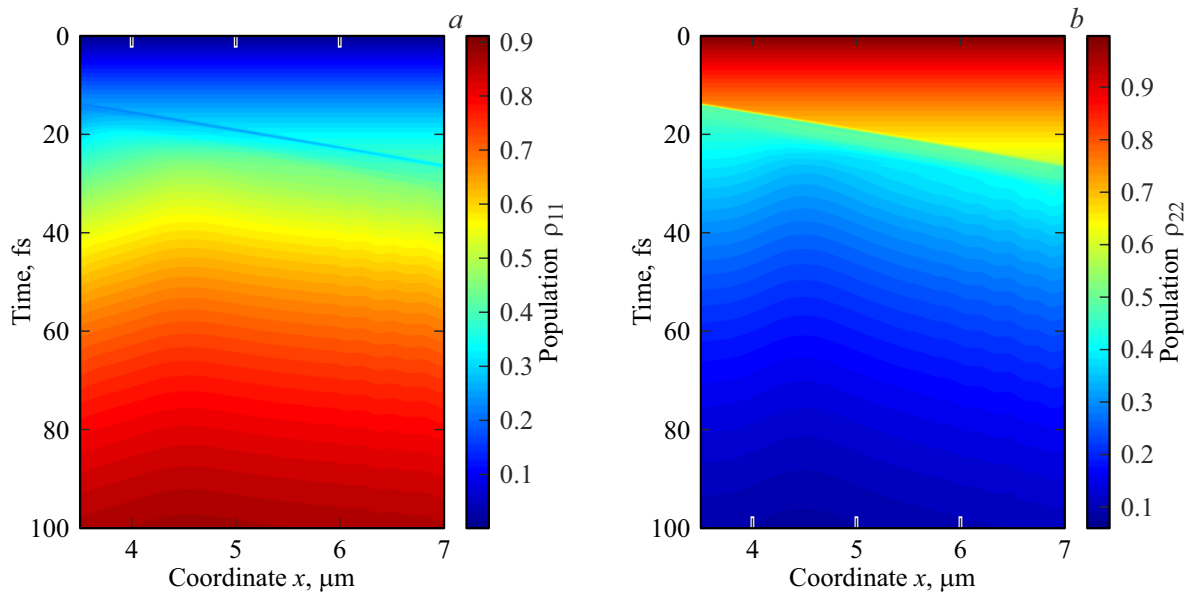


Figure 4. Spatiotemporal dynamics of the populations of the 1st and 2nd levels of a three-level medium in the process of passage of unipolar half-cycle pulse (2) through the amplifying three-level medium layer with a thickness of $3.5\ \mu\text{m}$ for the example from Fig. 2.

in the medium, the specific type of an energy level diagram is expected to exert a potentially significant influence on the nature of amplification of propagating half-cycle pulses in such a medium.

To clarify the influence of the energy level diagram, we turn to another case: the so-called cascade level diagram. In this case, resonant transitions $1 \rightarrow 2$ and $2 \rightarrow 3$ are dipole-allowed in a three-level medium, while transition $1 \rightarrow 3$ is forbidden. Thus, atoms of the medium may migrate from the initially populated 2nd level directly both to the ground level and to the upper 3rd level. For ease of comparison, we use the same transition frequencies and other system parameters as the ones given in the table (with the exception of dipole moments of transitions $2 \rightarrow 3$ and $1 \rightarrow 3$, which are now taken to be equal to $d_{23} = 30\text{ D}$ and $d_{13} = 0$).

Figure 7 shows the time dependences of the electric field calculated at the same three fixed points as in Fig. 1. Several significant differences between Figs. 1 and 7 may be noted. First of all, Fig. 7 demonstrates amplification of the leading half-cycle field burst; i.e., its amplitude increases gradually with the distance traveled, while the amplitude of this half-cycle pulse in Fig. 1 decreases. In both cases, an extended oscillating tail is formed at the trailing edge of a pulse due to oscillations of induced polarization of the medium.

To uncover the reason for the observed differences in variation of the maximum amplitude of a half-cycle pulse, we examine the dynamics of populations of levels of the medium. The corresponding time dependences at points located at the boundaries of the resonant medium layer (similar to those presented in Fig. 3 for the medium with a V level diagram) are shown in Fig. 8. It follows from a comparison of Figs. 3 and 8 that the population of the ground level in a cascade diagram increases much more

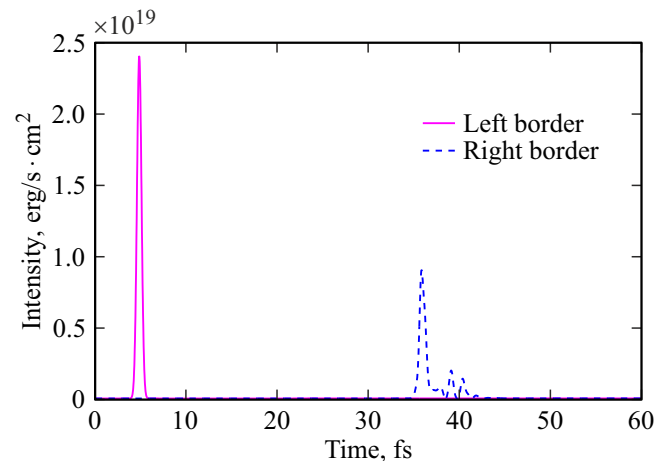


Figure 5. Time dependence of the radiation intensity at fixed points on the left (solid line) and on the right (dashed line) of the medium layer for the example from Fig. 2.

profoundly under the influence of an incident pulse than in a medium with a V level diagram. This implies that a non-equilibrium medium transfers a part of the stored energy to a half-cycle pulse, raising its maximum amplitude. After the passage of a pulse, a significant residual population of excited levels remains in both cases, which leads to the formation of alternating oscillating pulse tails.

A spatial dependence of electric area (1) and degree of unipolarity (6) for a medium with a cascade level diagram was plotted (see Fig. 9) to determine the contribution of tails forming at the trailing edge of a pulse. While electric area (1) again remains constant in space (as required by the fundamental rule of its conservation), degree of

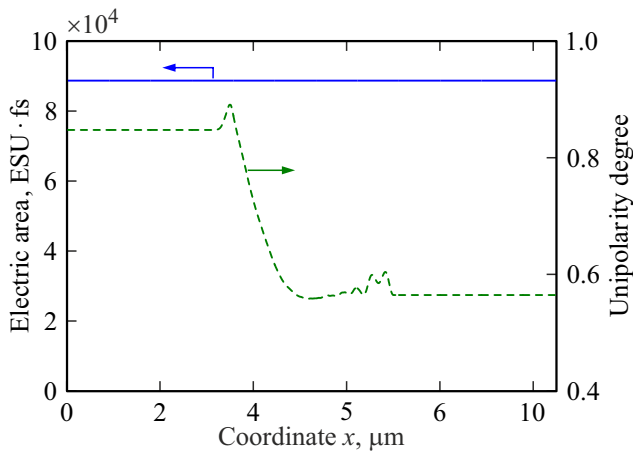


Figure 6. Dependence of electric area (1) and degree of unipolarity of the field (6) on the spatial coordinate for the example from Fig. 2.

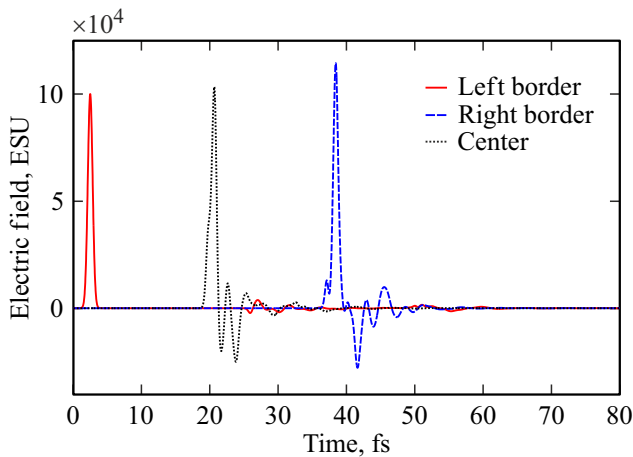


Figure 7. Time dependence of the electric field at three fixed points in space located to the left of the medium layer (red solid line), in the center of the medium layer (black dotted line), and to the right of it (blue dashed line). Incident half-cycle pulse (2) has duration $\tau = 500$ as and amplitude $E_0 = 10^5$ CGSU; the medium has a cascade level diagram.

unipolarity (6) decreases rapidly with distance from the left boundary of the medium layer. As in Fig. 6, the degree of unipolarity reaches saturation and assumes a stationary value near the right boundary of the layer, which is indicative of stabilization of the contribution of forming extended tails to the total electric area of the field.

Conclusion

The issue of feasibility of amplification of a half-cycle pulse passing through a non-equilibrium multilevel resonant medium layer was examined theoretically. Numerical modeling of propagation of such an attosecond pulse through a layer of a three-level medium, which was brought to the

lower excited level prior to the arrival of this pulse, was performed for this purpose.

It was demonstrated that a propagating pulse causes oscillations of induced polarization at resonant transitions of the medium, which produce an alternating field. As a result, an extended oscillating tail forms at the trailing edge of a half-cycle pulse. This leads to gradual transformation of a unipolar pulse into a bipolar one. The rule of conservation of electric area (1) is fulfilled exactly due to the emergence of backward radiation, but the degree of unipolarity of the field, which is given by expression (6), decreases with distance from the left boundary of the medium layer.

The possibility of stimulated amplification of half-cycle pulses in a medium with a nonequilibrium population of levels is an important issue. The obtained calculated data revealed that, owing to the extreme width of their spectrum, the amplification of half-cycle pulses with preservation of the pulse shape in an amplifying medium is not observed in

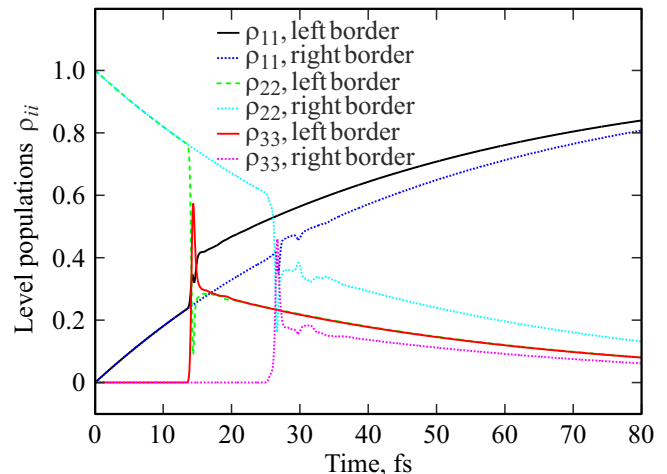


Figure 8. Temporal variation of the populations of levels of the three-level medium at points on the left and right borders of the layer of the medium with a cascade level diagram.

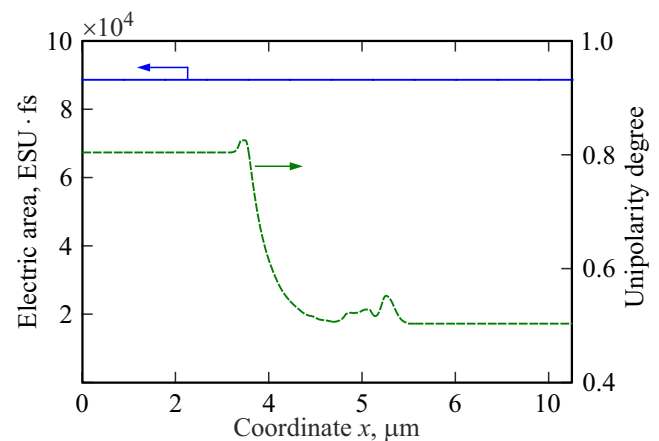


Figure 9. Dependence of electric area (1) and degree of unipolarity of the field (6) on the spatial coordinate for the medium with a cascade level diagram.

the general case. A redistribution of populations of excited levels of the medium occurs instead of a simple directed transfer of energy from the excited medium to a pulse. However, amplification of the leading half-cycle field burst is possible in certain kinds of level diagrams of the medium. Specifically, in a V level diagram, the energy in the leading half-cycle field burst decreases with distance traveled. The energy lost by a pulse is then converted into spontaneous radiation of the medium and into oscillating tails that form at the trailing edge of the initial pulse. At the same time, the amplitude of the leading half-cycle field burst in a medium with a cascade level diagram increases due to stimulated transitions of the medium from the initially excited level to the ground level.

It is important to note that half-cycle attosecond pulses used in the above modeling may now be generated in experiments, which was demonstrated in a number of theoretical and experimental studies that utilized a variety of different approaches [10–16]. Thus, the presented modeling data may be relevant to the development of methods for controlling the amplitude, duration, and shape of generated half-cycle sub-femtosecond pulses.

Funding

This study was supported by grant No. 21-72-10028 from the Russian Science Foundation.

Conflict of interest

The authors declare that they have no conflict of interest.

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Translated by D.Safin