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Selective ultrafast control of multi-level quantum systems by subcycle and unipolar pulses

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Abstract: The most typical way to optically control population of atomic and molecular systems is to illuminate them with radiation, resonant to the relevant transitions. Here we consider a possibility to control populations with the subcycle and even unipolar pulses, containing less than one oscillation of electric field. Despite the spectrum of such pulses covers several levels at once, we show that it is possible to selectively excite the levels of our choice by varying the driving pulse shape, duration or time delay between consecutive pulses. The pulses which are not unipolar, but have a peak of electric field of one polarity much higher (and shorter) than of the opposite one, are also capable for such control.

1. Introduction

Generation of ultrashort pulses of electromagnetic radiation is one of the most important topics in modern optics [1–3]. Ultrashort pulses with a duration at the femtosecond and attosecond timescales, comparable to a period of field oscillations, are widely used in studying the dynamics of ultrafast processes in matter, medicine, data transfer etc. [3–7]. Although ordinary electromagnetic waves are bipolar, the possibility of obtaining short electromagnetic pulses with a high degree of unipolarity $\xi \sim 1$, with ξ defined as

$$\xi = \frac{|\int_{t=-\infty}^{+\infty} \vec{E}(t)dt|}{\int_{t=-\infty}^{+\infty} |\vec{E}(t)|dt}, \tag{1}$$

where $\vec{E}(t)$ is the electric field strength in dependence on time t, has been attracting attention in the last years, see e.g. [8–14] and references therein. In the numerator of Eq. (1) stands so called electric area of the electromagnetic pulse [15]:

$$\vec{S}_E = \int_{t=-\infty}^{+\infty} \vec{E}(t)dt. \tag{2}$$

Assuming in the following the linear polarization of the electric field, we can talk about scalar electric pulse area S_E.

Bipolar pulses, typically produced in optics, have nearly zero area $S_E \approx 0$ and degree of unipolarity $\xi \approx 0$. Experimentally available nowadays are strong subcycle pulses containing a

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high-amplitude burst of electromagnetic field and a long decaying weak tail of opposite polarity, which we may call quasi-unipolar. Such subcycle pulses have been obtained in the optical [7,16,17] and terahertz frequency range [18–20]. In addition, different methods of subcycle quasi-unipolar pulse formation in the form of solitons in nonlinear medium [9,21–23] and via reflection of bipolar pulse from thin film of metal or dielectric [10,24] have been recently proposed. For some of such mechanisms, the degree of unipolarity can deviate quite noticeably from zero [10,24].

The presence of a DC-component in a pulse allows to effectively accelerate charges and thus to excite and control the dynamics of wave packets, see [14,25] and references therein. For instance, half-cycle terahertz pulses (whether having a DC component or not) were used to effectively control and study the dynamics of Rydberg atoms [26] and semiconductor nanostructures [16]. Subcycle attosecond pulses in the optical range, as shown in [7], are capable to efficiently excite atomic systems. The use of unipolar pulses together with multi-cycle bipolar pulses during excitation of various media allows to efficiently generate isolated attosecond pulses via high-order harmonic generation [27–29].

A number of papers discusses a possibility of controlling the shape of unipolar pulses as they are being generated. In particular, in [30–34] a method of obtaining rectangular quasi-unipolar pulses based on coherent control and free induction decay of low-frequency oscillations in nonlinear media, excited by femtosecond laser pulses was proposed and theoretically considered in different medium geometries. In [35] solitonic rectangular unipolar pulses in a three-level resonant medium were found (rectangular waveshape corresponds not to the envelope, but to the fast optical field itself). The possibility of obtaining rectangular THz waveshapes in the form of precursors propagating ahead of high-power laser pump pulses in electro-optical crystals was theoretically predicted in [36–38]. It was also found, that unipolar pulses can be not only generated, but also transported for long distances [39].

Besides that, a few studies have been devoted so far to interaction of subcycle pulses with atomic systems. Ionization of a hydrogen atom using pulses with rectangular waveshapes has been considered in [40,41]. In [42–45] it was shown, that unipolar and subcycle pulses are especially efficient in excitation of atoms and molecules, when the pulse duration is smaller than the inverse transition frequency between the levels. The reason of such efficiency can be classically easily understood: subcycle unipolar waveshapes provide more efficient "push" to electrons than a usual multi-cycle optical pulse. In [46] a possibility of generation of population gratings in a multi-level quantum systems by two half-cycle pulses was demonstrated.

In this work we show that using unipolar and subcycle pulses, despite they have extremely delocalized spectra covering many transitions at once, allows nevertheless to control population of multi-level atomic systems in a very selective way. For instance, we can target one particular level by choosing a correct pulse waveshape, duration, or distance between pulses in a pair of pulses. Furthermore, we show, that quasi-unipolar pulses with $S_E = 0$, but consisting of a single strong spike and a long unipolar tail of the opposite sign, which are much more feasible in optical experiments, work almost as good as exactly unipolar ones.

2. Interaction of unipolar pulse with a hydrogen-like atom

For a theoretical description it is necessary to solve the time-dependent Schrödinger equation for the wave function ψ [47]:

$$i\hbar\frac{\partial\psi}{\partial t} = [\hat{H}_0 + V(t)]\psi. \tag{3}$$

Here $\hbar$ is the reduced Planck constant, $\hat{H}_0$ is the field-free Hamiltonian of the system, $V(t) = -\mathbf{dE}(t)$ is its interaction potential with the driving pulse in the dipole approximation, $\mathbf{d} = q\mathbf{r}$ is the operator of atomic dipole moment, q is the electron charge. We assume, that before the interaction the atom was in the ground state Ψ_1 of the field-free Hamiltonian $\hat{H}_0$. In the weak-field approximation, the

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Schrödinger Eq. (3) can be approximately solved using a perturbation theory. In the first order of the perturbation theory, the probability of transition of the system from the ground state to the m-th excited state of the discrete spectrum has the form [47]:

$$w_{1m} = \frac{1}{\hbar^2}\left|\int V_{1m}e^{i\omega_{1m}t}dt\right|^2. \tag{4}$$

Here $V_{1m} = -d_{1m}E$ is the matrix element of the perturbation operator, d_{1m} is the transition dipole moment, ω_{1m} is the resonant transition frequency. In the following subsections we consider the effect of unipolar pulses of various shapes, as well as pair of pulses on a hydrogen-like atom using this perturbation approach.

2.1. Rectangular pulse

Let us consider a linearly polarized unipolar pulse of rectangular waveshape, amplitude E_0 and duration τ_p, i.e., the one described with a scalar field $E(t)$ with $E(t) = E_0$ if $0<t<\tau_p$ and 0 elsewhere. Using Eq. (4), we obtain for the transition probability from the ground state w_{1k}:

$$w_{1m} = 2\frac{d_{1m}^2E_0^2}{\hbar^2\omega_{1m}^2}\left(1-\cos\omega_{1m}\tau_p\right). \tag{5}$$

From Eq. (5) it can be seen, that w_{1m} depends periodically on the pulse duration. It was shown in [44,45], that, if the pulse duration is shorter than the period of resonant transitions in the system ($\omega_{1m}\tau_p \ll 1$), the exponential term in Eq. (4) can be approximately calculated at $t = 0$. That is the transition probability is determined by the electric pulse area:

$$w_{1m} = \frac{d_{1m}^2}{\hbar^2}S_E^2.$$

This fact shows a possibility of effective control of quantum systems by extremely-short unipolar pulses having nonzero pulse area. In the case of a long enough pulse, one can see, that the transition probabilities w_{1m} for different levels m in Eq. (5) oscillate with different frequencies as we vary τ_p. This directly indicates a possibility of selective excitation of certain resonant transitions in the system.

To illustrate this idea, we consider a hydrogen-like atom. For hydrogen-like atoms with a nuclear charge qZ the frequencies of atomic transitions are defined as [47]:

$$\omega_{km} = \frac{m_e q^4 Z^2}{2\hbar^2}\left(\frac{1}{k^2}-\frac{1}{m^2}\right). \tag{6}$$

Here m and k are the pair of main quantum numbers, m_e is the mass of the electron. As it is known, this equation describes transition frequencies of various spectral series in the radiation spectrum of a hydrogen atom [47]. For example, the case $k = 2$ is known as Balmer series, whereas $k = 1$ defines the Lyman series. Here we are interested in the latter case. For instance, for the first four wavelengths of the Lyman series we have: $\lambda_{12} = 1216$ Å, $\lambda_{13} = 1026$ Å, $\lambda_{14} = 972$ Å. The corresponding transition dipole moments are $d_{12} = 3.27$ Debay, $d_{13} = 1.31$ Debay, $d_{14} = 0.77$ Debay [48].

It is seen from Eq. (5), that for those transition frequencies and pulse durations satisfying $\omega_{1m}\tau_p = 2\pi n$, $n = 1, 2, 3, \ldots$ the population of m-th level vanishes. In contrast, for $\omega_{1m}\tau_p = (2n-1)\pi$, $n = 1, 2, 3, \ldots$ the population attains the maximum. Since all of these frequencies are different (and mostly even incommensurate), we can use this fact for selective excitation of the quantum transitions. For instance, Fig. 1 shows periodic dependence of the probability w_{1m} on the pulse duration τ_p for three lowest excited levels. By correctly selecting the pulse duration,

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we can control positioning the excited electron on particular level (see vertical lines in Fig. 1). Note, that the pulse considered here has a rectangular shape of the electric field (and not the envelope). In particular, for $\tau_p \approx 1$ fs the most of the excited population is located on the second level $m = 2$, whereas at $\tau_p \approx 1.7$ fs only the third level is excited, whereas levels 2 and 4 are nearly empty. For $\tau_p \approx 2.4$ fs, the level 4 is populated, whereas levels 2 and 3 are mostly empty. We do not consider here further levels $5, 6, \ldots$ since the excitation probabilities quickly decay with the quantum number. Their populations might be nonzero, and our approximation here is to neglect those populations.

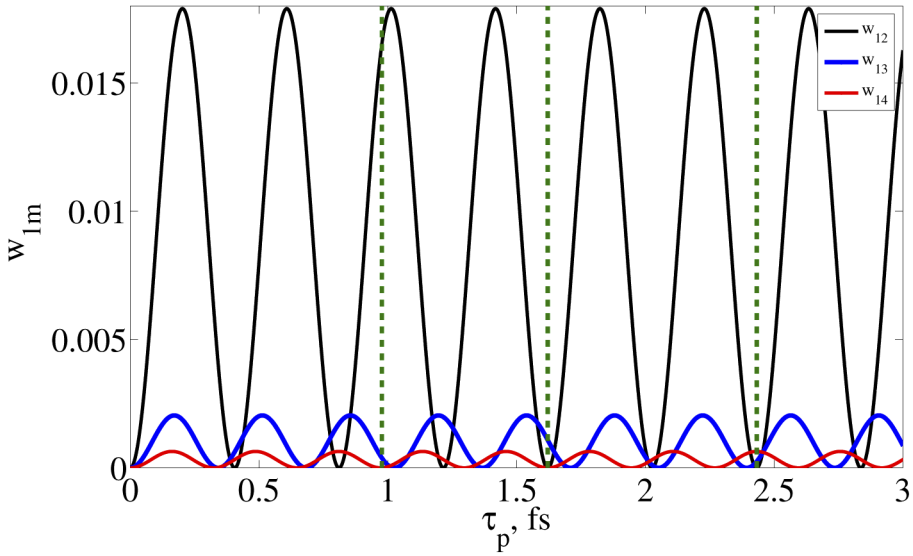

Fig. 1. Probability of the transitions w_{1m} of the Lyman series versus the duration τ_p of a unipolar pulse with rectangular waveshape for $m = 2, 3, 4$. The vertical lines indicate the positions, where selective population is achieved; $E_0 = 10^8$ V/cm.

The periodic dependence of the transition probability can be simply explained on the basis of the semi-classical theory of the Bohr atom [49]. If an electron rotates along the classical Bohr orbit with certain frequency, then, depending on the ratio between the pulse duration and that frequency, the pulse can either slow down the electron or accelerate it (that is, facilitate transfer to another orbit).

2.2. Gaussian and super-Gaussian pulses

Now, instead of a rectangular waveshape let us consider a super-Gaussian pulse:

$$E(t) = E_0 e^{\frac{-t^n}{\tau_p^n}},$$

where n is an integer number. For $n = 2$ this gives a Gaussian shape, whereas it approaches rectangle for large n. We note again, that the waveshape here is the fast optical field and not the envelope. To find the transition probabilities we need to calculate the integral:

$$w_{1m} = \frac{d_{1m}^2 E_0^2}{\hbar^2} \left| \int e^{\frac{-t^n}{\tau_p^n}} e^{i\omega_{1m} t} dt \right|^2,$$

which is possible only numerically. Figure 2 shows the dependencies of the probability of the transition from the ground state to the first excited state w_{12} of the hydrogen atom for a rectangular pulse and super-Gaussian pulses with $n = 4, 10, 20$. Electric field amplitude is $E_0 = 5 \cdot 10^7$ V/cm.

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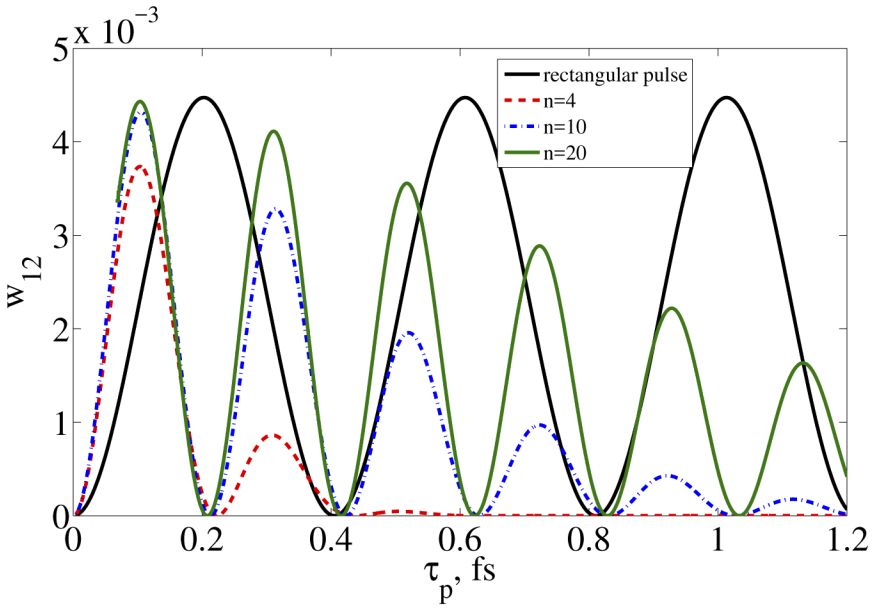

Fig. 2. Excitation probabilities w_{12} for the Lyman-alpha line of hydrogen versus pulse duration τ_p for a rectangular pulse (black) and a super-Gaussian pulse for different values of parameter $n = 4, 10, 20$; $E_0 = 5 \cdot 10^7$ V/cm.

Whereas in the case of a rectangular pulse (black line) the dependence is cosine-like, in the case of a super-Gaussian pulse, the maxima decay with increasing of the pulse duration and their decay rate decreases with increase of n. The larger is n, the steeper are the fronts of the super-Gaussian pulse. One can understand the result in the following way: the pulse edges can be approximated as a series of rectangular pulses with the increasing amplitudes and positioned at different times, that is, delayed to each other. For each rectangular pulse the probability w_{12} is proportional to $\sim \cos \omega_{12}\tau_p$. Thus, the total probability can be approximated as a sum of such elementary impacts with different amplitudes and phases. This interference between those impacts leads to the decay of the maxima in Fig. 2 corresponding to the different values of n. Thus we see, that super-Gaussian pulses with small n are less suitable for controllable population of the levels, since it needs rather large τ_p, for which super-Gaussian waveshapes are not effective.

2.3. *Lorentzian pulse*

As the last possibility, let us consider a pulse with a Lorentzian waveshape (again, we consider the waveshape of the fast field and not the envelope). The advantage of this waveshape is that, although it looks not so "sharp-edged" as a rectangular one, the calculation of the transition probabilities is still possible to do analytically. The Lorentzian shape is given by:

$$E(t) = \frac{E_0}{1 + \left(\frac{t}{\tau_p}\right)^2}.$$

For the transition probability, according to Eq. (4) it is possible to obtain using Cauchy's theorem:

$$w_{1m} = \frac{d_{1m}^2 E_0^2 \tau_p^2}{\hbar^2 \pi^2} e^{-2\omega_{1m}\tau_p}. \tag{7}$$

Unlike a rectangular pulse, the probability decreases monotonically with increasing pulse duration as long as $\omega_{1m}\tau_p > 1$; therefore, these pulses are not of significant interest.

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2.4. Pair of rectangular pulses

Next, we extend the range of possible control parameters by considering a pair of rectangular unipolar pulses. If both pulses have the same duration and one of the pulses is delayed in time by Δ to another, the following expression can be obtained for the transition probability [46]:

$$w_{1m} = 4\frac{d_{1m}^2 E_0^2}{\hbar^2 \omega_{1m}^2}\left(1-\cos\omega_{1m}\tau_p\right)\left(1+\cos\left[\omega_{1m}\left(\Delta+\tau_p\right)\right]\right). \tag{8}$$

Here time delay Δ between pulses is measured between the trailing edge of the first pulse and the leading edge of the second pulse. This equation contains the terms oscillating with changing of τ_p and Δ with different frequencies, and thus a selective effect on population dynamics is also to be expected. For instance, Fig. 3 illustrates the dependence of the transition probabilities w_{12}, w_{13} and w_{14} on the delay Δ and pulse duration τ_p for the case of hydrogen atom. Varying both Δ and τ_p, it is possible to control transitions more effectively.

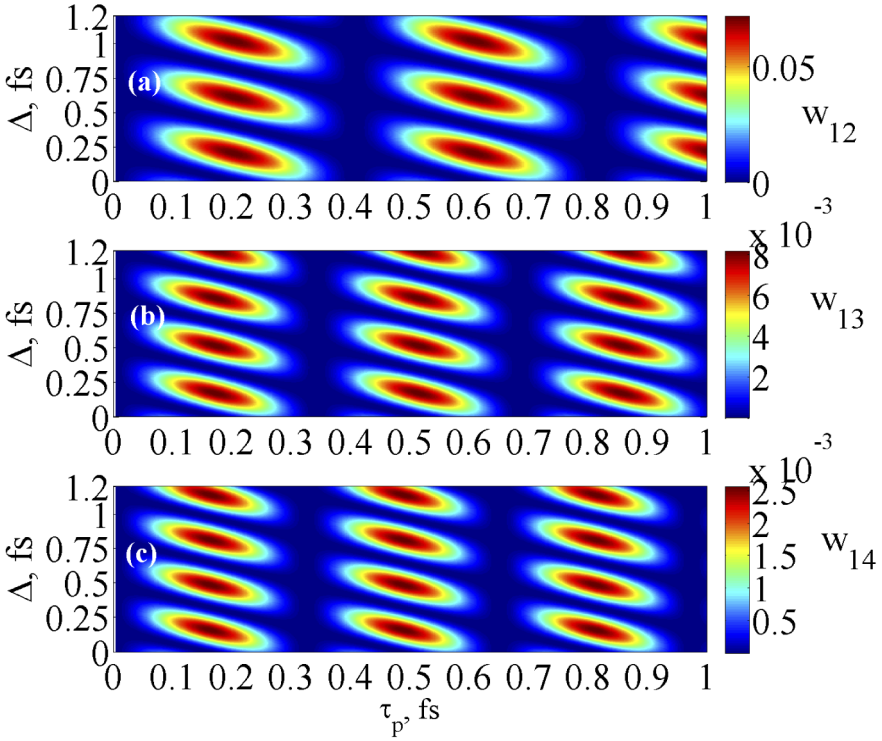

Fig. 3. Excitation probabilities w_{12} (a), w_{13} (b) and w_{14} (c) as a function of the delay Δ between the pulses and their duration τ_p for the Lyman series of the hydrogen atom, interacting with a pair of rectangular pulses for $E_0 = 10^8$ V/cm.

We remark, that the periodic dependence on the delay between pulses allows to create a periodic population gratings, if we illuminate a spatially-extended gas-filled cell with a pair of counterpropagating pulses [50,51]. In this case the counterpropagating pulses arrive to every point in space at different times. Thus, spatial grating is mapped into a periodic modulation of population with the delay between pulses. The result of Fig. 3 shows now, that we are able to selectively create a grating in population not only between the fundamental and the first excited levels, but also between the fundamental and the second or even higher levels, all this with an

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ultra-broadband subcycle pulses. Such gratings can find various applications in nonlinear optics and spectroscopy [52].

2.5. Pair of Gaussian or Lorentzian pulses

Here, we consider a pair of pulses with the waveshape different from a rectangular one, and with a tunable delay Δ. For the following consideration, we need the pulses with arbitrary durations and amplitudes, that is, we consider the waveshape in the form:

$$E(t) = E_1 e^{\frac{-t^2}{\tau_{p1}^2}} + E_2 e^{\frac{-(t-\Delta)^2}{\tau_{p2}^2}}.$$

For the transition probability we get:

$$w_{1m} = \frac{d_{1m}^2 S_{E1}^2}{\hbar^2} e^{-\frac{\omega_{1m}^2 \tau_{p1}^2}{2}} + \frac{d_{1m}^2 S_{E2}^2}{\hbar^2} e^{-\frac{\omega_{1m}^2 \tau_{p2}^2}{2}} + 2\frac{d_{1m}^2}{\hbar^2} S_{E1} S_{E2} e^{-\frac{\omega_{1m}^2 (\tau_{p1}^2 + \tau_{p2}^2)}{4}} \cos \omega_{1m} \Delta, \qquad (9)$$

where S_{E1} and S_{E2} are the electric pulse areas. This equation was obtained in [45]; here we apply Eq. (9) to demonstrate the control of population inversion.

Rather similar expression can be obtained in the case, when the medium is excited by a pair of Lorentzian pulses with delay Δ (here we consider for simplicity identical pulses):

$$w_{1m} = \frac{2 d_{1m}^2 E_0^2 \tau_p^2}{\hbar^2 \pi^2} e^{-2\omega_{1m}\tau_p} \left(1 + \cos \omega_{1m} \Delta\right). \qquad (10)$$

In both of these cases, similar features can be observed in dependence of the population on τ_p

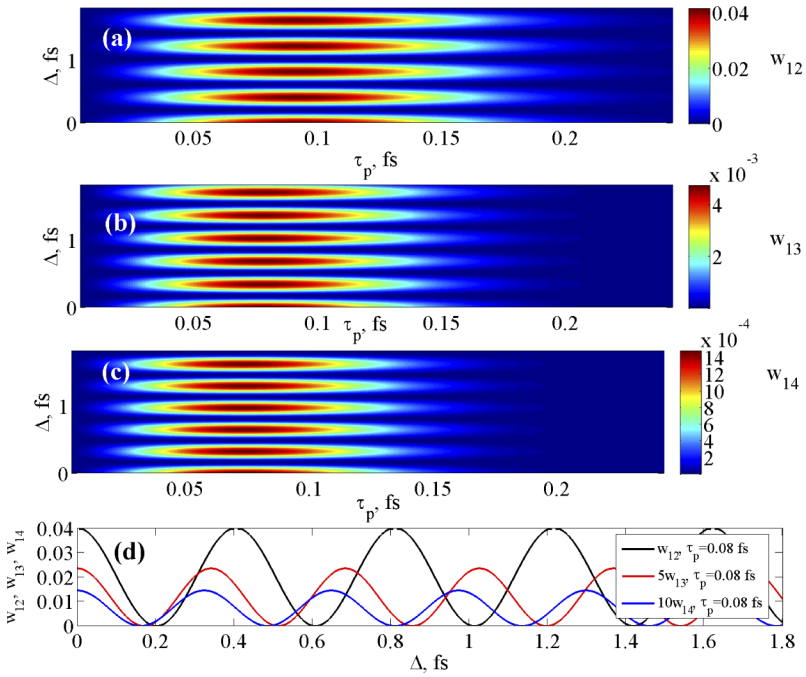

Fig. 4. Excitation probabilities w_{12} (a), w_{13} (b) and w_{14} (c) for the Lyman series of hydrogen versus pulse delay Δ and pulse duration τ_p; (d): cross-sections of (a-c) for fixed $\tau_p = 0.08$ fs. Field amplitude is $E_0 = 10^8$ V/cm.

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and Δ: the amplitude of the transition decays exponentially with τ_p, so very short pulses have significant advantage. On the other hand, periodic dependence on Δ is also present, pointing to a possibility to control the populations in the same way we did it for a single rectangular pulse, only using Δ as a control parameter. We note that, as it was shown in [50,51], this periodic dependence allows also to produce population-density gratings with counter-propagating pump pulses.

In Fig. 4 the probabilities of transitions w_{12}, w_{13}, w_{14} are shown versus the pulse delay Δ and durations for the Lyman series ($k = 1$) of the hydrogen atom according to Eq. (9) for a pair of identical Gaussian pulses with equal durations $\tau_{p1} = \tau_{p2} \equiv \tau_p$ and amplitudes $E_1 = E_2 \equiv E_0 = 10^8$ V/cm. From this figure one can see, that the selective excitation of the atomic levels is indeed possible by varying the pulse-to-pulse delay Δ. For example, for $\Delta \approx 1$ fs the population of the second level is around zero. At the same time the population of the third level and the fourth level are close to their maximum values (at τ_p around 0.08 fs), see Fig. 4(d).

Equation (9) can be also used to show, that our results are applicable to subcycle pulses of more general shape than, for instance, Gaussian unipolar ones. For example, subcycle pulses generated experimentally [7,16–20] contain a giant spike of nearly half-wave duration, followed by a weak decaying tail of the opposite polarity and much longer duration. With Eq. (9) we can describe the action of such pulses by modeling them using Eq. (9) as a pair of Gaussian pulses with opposite polarity and significantly different pulse durations. In this case, if $\omega_{1m}\tau_{p2} \gg 1$, the first term will dominate in Eq. (9), while the second and the third terms can be neglected. Hence, the action of such a single half-cycle pulse with strictly zero area coincides with the action of a single unipolar pulse having nonzero electric pulse area. Figure 5 illustrates this idea. Figure 5(a) shows an example of a quasi-unipolar half-cycle pulse for delay $\Delta = 0.8$ fs.

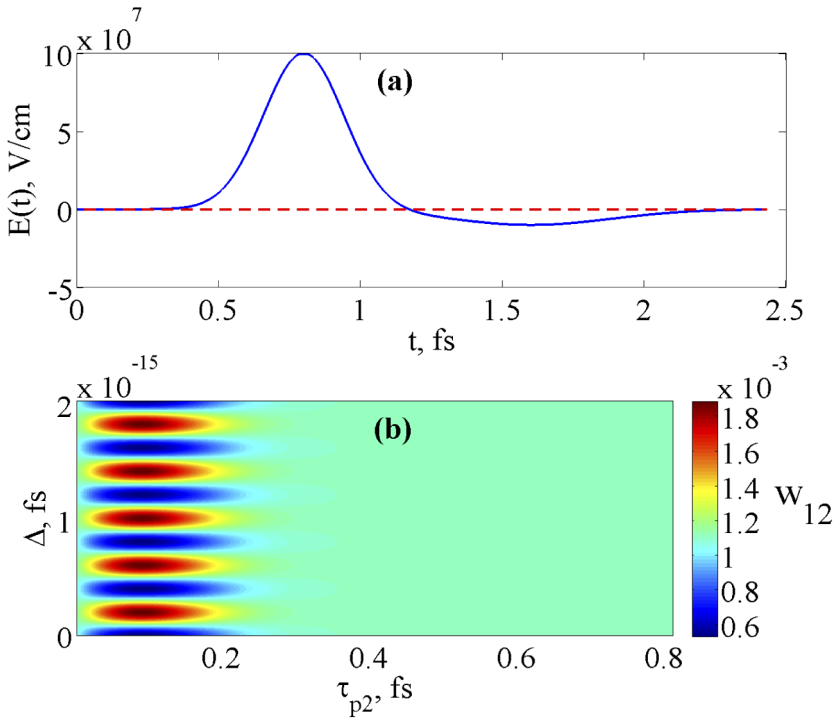

Fig. 5. (a): Example of the pump pulse shape for $\Delta = 0.8$ fs, $\tau_{p1} = 0.2$ fs, $E_{01} = 10^8$ V/cm, $E_{02} = -0.1E_{01}$; (b): Excitation probability w_{12} for the Lyman-alpha line of hydrogen versus delay between the pulses Δ and the duration of the second pulse τ_{p2}.

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In Fig. 5(b) probability transition w_{12} versus pulse delay and duration of the second pulse is plotted, when duration of the first pulse $\tau_{p1} = 0.2$ fs is fixed, $E_{01} = 10^8$ V/cm, $E_{02} = -0.1E_{01}$. It can be seen from Fig. 5, that for large values of the second pulse duration τ_{p2} the transition probability w_{12} approaches a constant value $\neq 0$, which does not depend on τ_{p2}. This result is in the qualitative agreement with the experimental data of Hassan *et al.* [7], which showed efficient action of a half-cycle pulse on an atomic gas with respect to a bipolar single-cycle pulse.

In the example above the area of a half-cycle pulses is in general nonzero. Nevertheless, similar results can be obtained for half-cycle pulses with exactly zero electric pulse area, as it is illustrated in Fig. 6. Figure 6(a) shows an example of such a pulse, whereas in Fig. 6(b) the transition probability w_{12} versus the delay Δ and duration τ_{p2} of the second pulse is plotted. For this figure, we select, for every τ_{p2}, the amplitude of the second half-pulse as $E_{02} = -\dfrac{E_{01}\tau_{p1}}{\tau_{p2}}$, which ensures that the total area of the pulse is zero for every τ_{p2}. One can see, that, as in Fig. 5, the excitation probability is peaked at low pulse durations (in this case the peak is even higher than in the case of Fig. 5), and approaches a constant value at large values of τ_{p2}. Such quasi-unipolar pulses, that is, having $S_E = 0$ but consisting of a single prevailing peak of a certain polarity, are much more accessible in experiments than the strictly unipolar ones. Our results show clearly that such pulses are also useful for the selective population excitation.

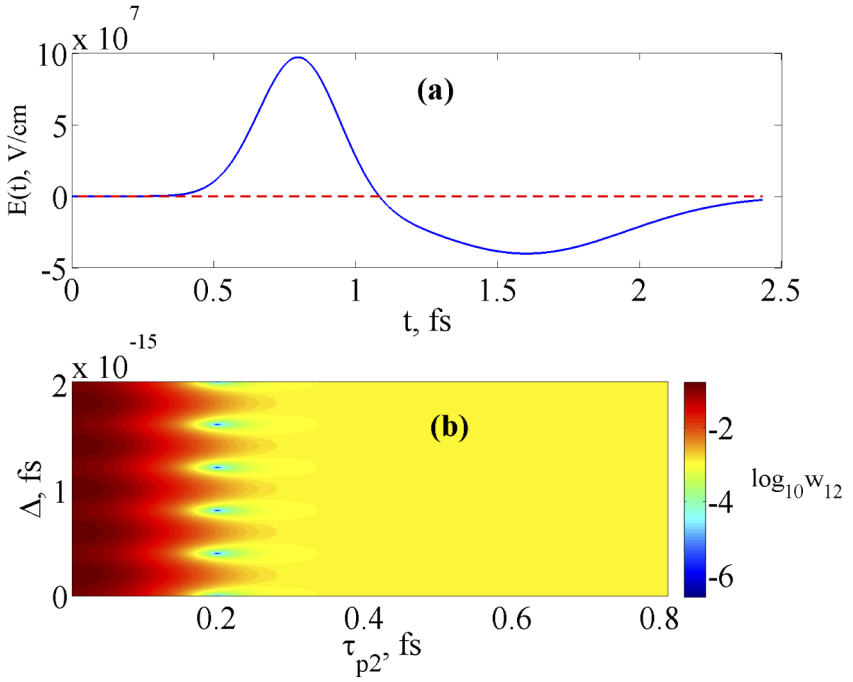

Fig. 6. (a): Example of the pump pulse shape for $\Delta = 0.8$ fs, $\tau_{p1} = 0.2$ fs, $\tau_{p1} = 0.5$ fs, $E_{01} = 10^8$ V/cm, $E_{02} = -\frac{\tau_{p2}}{\tau_{p1}}E_{01} = -4 \cdot 10^7$ V/cm; (b): Excitation probability w_{12} for the Lyman-alpha line of hydrogen versus delay between the pulses Δ and the duration of the second pulse τ_{p2} (note the logarithmic color scale); for each value of τ_{p2}, E_{02} is choosen as $E_{02} = -E_{01}\frac{\tau_{p1}}{\tau_{p2}}$, which ensures $S_E = 0$.

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3. Simulations with Schrödinger equation

Here we corroborate our findings, up to now based on the perturbation theory, by a direct solution of the Schrödinger equation written for the bound states in the form:

$$\psi(\vec{r},t) = \sum_{m=1}^{N} a_m(t)\psi_m(\vec{r})e^{-\frac{iE_m t}{\hbar}}, \qquad \omega_{ml} = \frac{E_m - E_l}{\hbar},$$
$$\dot{a}_m(t) = \frac{i}{\hbar}\sum_{l=1}^{N} d_{lm}a_l(t)E(t)e^{i\omega_{ml}t}, \tag{11}$$

where d_{lm} is the transition dipole moment between levels m and l, ω_{ml} is the frequency of the corresponding transition.

Let us suppose, that the quantum system is in the ground state before the excitation, i.e. $a_1 = 1$ and $a_m = 0$ for $m \neq 1$. Then, if the following condition holds for each m:

$$\frac{d_{1m}E_0\tau_p}{\hbar} \ll 1, \tag{12}$$

in Eq. (11) one can neglect in the first approximation all terms on the right-hand side except for the ones, proportional to a_1, and obtain:

$$\dot{a}_m(t) \approx \frac{id_{1m}E(t)}{\hbar}a_1(t)e^{i\omega_{m1}t}, \qquad m = 2,3,4,\ldots$$

Eq. (4) can be recovered from this equation by assuming $a_1(t) \approx 1$ during the whole duration of an excitation pulse.

Beyond the small signal approximation, one can obtain an analytical solution for the action of a rectangular unipolar pulse on a two-level medium. As it was shown in [53], polarization P and population inversion n of a two-level medium under the action of a rectangular unipolar pulse behave as:

$$n(t) = \frac{4\Omega_R^2\cos\Omega t + \omega_0^2}{4\Omega_R^2 + \omega_0^2},$$
$$P(t) = \frac{2E_0 d_{12}^2\omega_0}{\hbar(4\Omega_R^2 + \omega_0^2)} \cdot \Big[1 - \cos\Omega t\Big]. \tag{13}$$

Here Ω_R is the Rabi frequency and $\Omega = \sqrt{4\Omega_R^2 + \omega_0^2}$. It is seen, that polarization and population inversion oscillates in time with the frequency different from the Rabi frequency, as it takes place in the case of resonant interaction with longer pulses [54]. Similar behavior could be expected to take place for strong fields in the case of a multi-level system as well.

As the next step we solve the Schrödinger equation Eq. (11) numerically, taking into account first 4 levels of the hydrogen atom. The transition frequencies are calculated from Eq. (7) and the dipole moments are taken as before from Ref. [48]. Figure 7 shows the temporal dynamics of the populations of this 4-level system under the action of a rectangular unipolar pulse for two different values of the electric field strength. One can see, that for $E_0 = 10^6$ V/cm the condition Eq. (12) gives for the largest dipole moment d_{12}:

$$\frac{d_{12}E_0\tau_p}{\hbar} \approx 0.0414\cdots,$$

so that Eq. (5) should be well applicable. Indeed, from Fig. 7(a) one can see that all excitation probabilities w_{1m} change periodically with frequencies ω_{1m}, as predicted by Eq. (5). Thus, also

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the possibility of the selective control of level populations does not disappears on this level of description. However, when the electric field strength is increased to $E_0 = 10^8$V/cm (see Fig. 7(b)), the dynamics becomes more complex and all excitation probabilities start to collectively oscillate in a complicated way. In this case the selective control of the level populations becomes quite challenging.

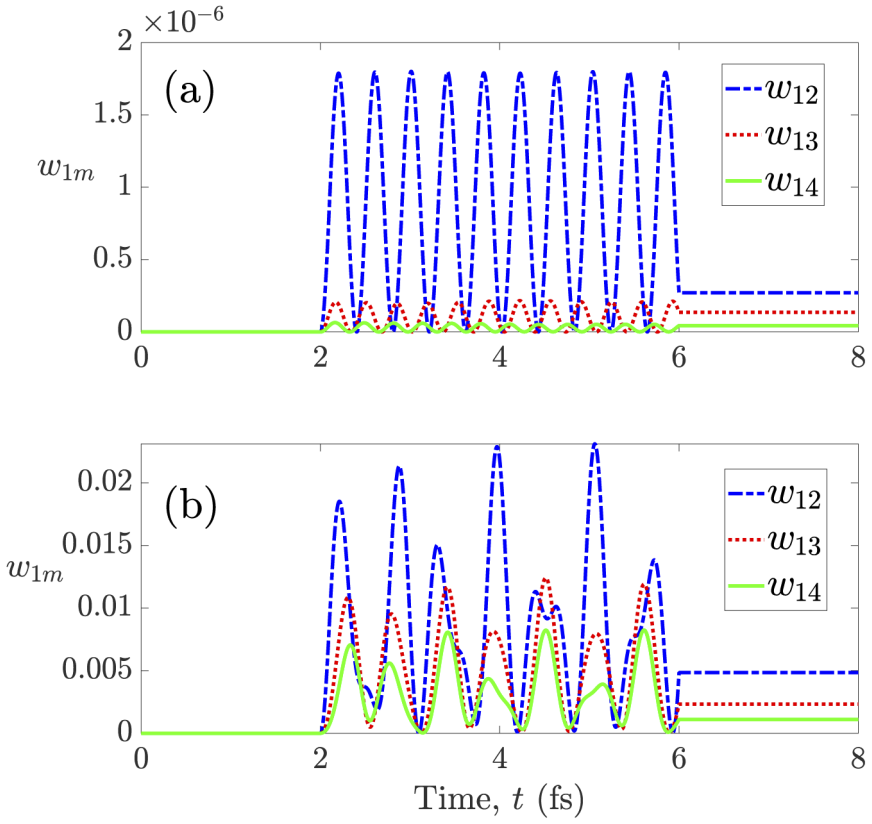

Fig. 7. Temporal dynamics of the excitation probabilities w_{1m} for a 4-level medium upon excitation by a rectangular pulse of duration $\tau_p = 4$ fs and electric field strength: (a) $E_0 = 10^6$ V/cm; (a) $E_0 = 10^8$ V/cm.

Following the above reasoning, we examine now excitation of our 4-level system by a pair of unipolar pump pulses delayed by Δ with respect to each other. Figure 8 shows the excitation probabilities vs. the time delay Δ for a pair of rectangular pulses of duration $\tau_p = 1$ fs and electric field strength $E_0 = 10^8$ V/cm. One can see, that when the driving pulses do not overlap, i.e. delay between pulses is larger than pulse duration, the excitation probabilities w_{1m} for different levels change periodically with time, but with different periods. This means, that the selective control of the population appears to be possible even in strong fields, when a pair of unipolar excitation pulses is used instead of a single pulse. This result can be easily explained. Even if all populations oscillate in time with the same collective frequencies in the field, in between the pulses, when the driving electric field equals zero, all phase factors in Eq. (11) start to oscillate at the frequencies ω_{1m}. Therefore all expansion amplitudes a_m in Eq. (11) acquire different phase shifts to the arrival of the second excitation unipolar pulse, resulting in final level populations w_{1m} oscillating with different periods as delay Δ is varied. Figure 9 demonstrates the dependence of the excitation probabilities w_{1m} on the time delay Δ for a pair of Gaussian pulses. The main

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findings here match the ones for a pair of rectangular pulses, in particular, selective excitation and control of higher levels becomes feasible by changing the time delay Δ.

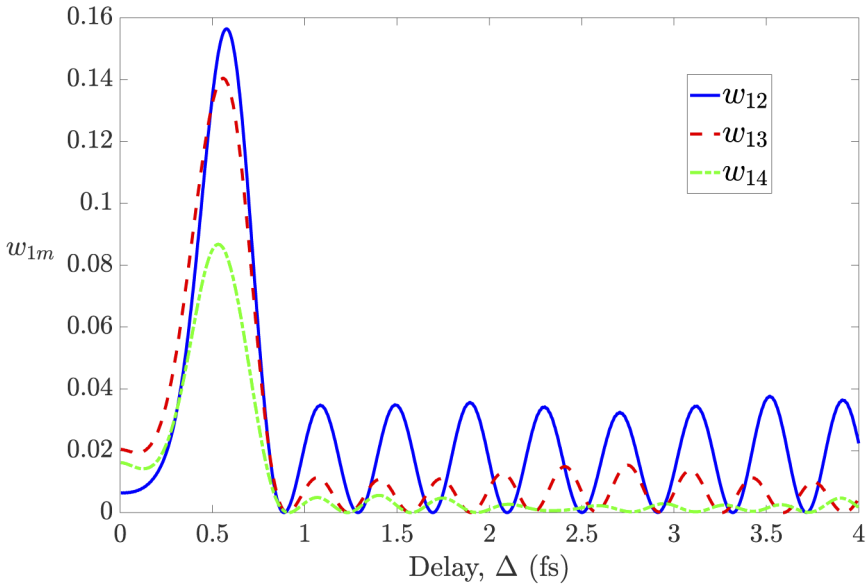

Fig. 8. Excitation probabilities w_{1m} for a 4-level medium, excited by a pair of rectangular pulses of duration $\tau_p = 1$ fs and electric field strength $E_0 = 10^8$ V/cm, propagating with time delay Δ.

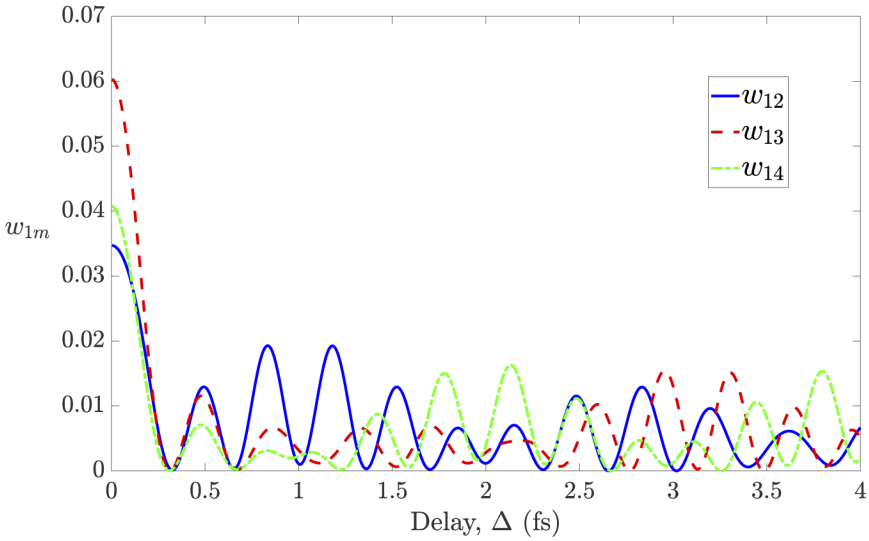

Fig. 9. Excitation probabilities w_{1m} for a 4-level medium, excited by a pair of Gaussian pulses of duration $\tau_p = 0.2$ fs and electric field strength $E_0 = 10^8$ V/cm, propagating with time delay Δ.

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4. Conclusions

In conclusion, here we considered interaction of unipolar and subcycle pulses of various shapes with a hydrogen-like atom. We showed that, despite of ultrashort and nonresonant character of the optical excitation, the transition probability depends periodically on the pulse duration for certain waveshapes. The period depends on the transition energy, which allows for selective excitation of the level of our choice.

In a hydrogen and hydrogen-like atoms, we were able to obtain analytical expressions for the probability of the transitions if the driving waveshapes are rectangular or Gaussian. A pair of pulses with a certain delay between them was also considered. Beyond the analytical perturbative theory, we provided simulations using the Schrödinger equation for amplitudes of quantum states in a 4-level medium.

We remark that, despite our unipolar pulses contain, by definition, nonzero component at zero frequency, the situation considered here is very different from the case of static electric field which does not change in time. Ultrashort unipolar pulses act on the atom/molecule only during a very short time, whereas the static electric field acts permanently, from $t = -\infty$ to $t = +\infty$. This latter situation is well studied in literature and leads, for instance, to Stark shift of energies [55], or, if the field is strong enough, to tunneling of an electron from the bound state to continuum [56].

In spite of the fact that unipolar pulses seem to be exotic today, the results presented above illustrate the possibility to use them in ultrafast non-resonant control of an electronic wavepacket dynamics in the medium. Beside, as we have shown here, the pulses must not necessarily be unipolar. Subcycle pulses which have one strong spike of one polarity, accomplished by a longer and weaker tail of the opposite polarity, works exactly as good as the unipolar pulses. This is easy to understand, since the whole process is nonlinear, that is, intensity dependent, and therefore the weak tail of the pulse produces much lower effect.

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Disclosures

The authors declare no conflicts of interest.

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