

# COHERENT CONTROL OF POPULATION IN BOUND STATES IN QUANTUM WELLS BY A PAIR OF HALF-CYCLE ATTOSECOND PULSES

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**Abstract.** The study of interaction features between unipolar subcycle pulses and matter has shown the necessity of both revising standard theories of light-matter interaction and introducing new concepts in optics, such as pulse area interference. In this work, based on numerical solution of the time-dependent Schrödinger equation, we study the features of nonlinear interference of pulse areas during particle excitation in a rectangular potential well driven by a pair of half-cycle attosecond pulses. It is shown that when changing the delay between pulses, the population dependence of bound states on delay exhibits characteristic beating pattern, unlike the simple harmonic dependence obtained in the case of small field amplitude. The conducted research directly demonstrates the possibility of controlling quantum systems using sequences of half-cycle pulses, particularly the possibility of increasing ionization probability or its complete suppression and the possibility of inducing population difference gratings in multilevel media.

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## 1. INTRODUCTION

The problem of reducing electromagnetic pulse duration has been relevant since the invention of first lasers [1] and remains important to this day [2–10]. Currently, using practically obtained femto- and attosecond electromagnetic pulses, it has become possible to study the dynamics of wave packets in atoms, molecules, and nanostructures [6–10]; thus, the recent Nobel Prize in Physics was awarded specifically for experimental methods of generating attosecond light pulses for studying electron dynamics in matter [11].

The attosecond pulses obtained in practice using the high-order harmonic generation method are bipolar and contain several half-waves of electric field strength [6–10]. For a fixed spectral interval of radiation, the shortest duration is achieved with a half-cycle (unipolar) pulse, which is obtained by keeping only one half-wave of the field from a multi-cycle pulse [12]. An important characteristic of unipolar pulses is their electrical area, defined as the integral of the field strength over time at a given point in space [12]:

$$S_E(\mathbf{r}) = \int_{-\infty}^{+\infty} E(\mathbf{r}, t) dt, \quad (1)$$

Pulses with non-zero electrical area can quickly transfer mechanical momentum to a charged particle in one direction, which opens up various prospects for their application in ultrafast control of quantum systems, charge acceleration, etc., see reviews [12–14], monograph [15], and cited literature. The combination of many works in this direction [16–26] has led to the emergence of a new, but still not very well-known and sometimes difficult to comprehend research direction – “optics of unipolar and subcycle pulses”.

If the duration of such pulses is shorter than characteristic intra-atomic times (the orbital period of an electron in the Bohr orbit in the ground state), then the nature of their interaction with quantum systems differs significantly from the case of conventional bipolar multi-cycle pulses (see reviews [12–15] for more details). Many familiar optical phenomena in this case lose their meaning or occur according to different scenarios. For example, light interference becomes impossible in its usual understanding [14, 15] or Keldysh's photoionization

theory [27] becomes inapplicable, which is valid for long multi-cycle pulses and demonstrates good accuracy even for extremely short pulses with duration of several periods of optical oscillations with near-atomic electric field strength [28, 29].

It should be noted that the impact of both single unipolar ultrashort pulses (USP) and sequences of such pulses on quantum objects is insufficiently studied to date. To describe the interaction of such USPs with quantum systems, the introduction of new concepts was required. As shown by the results of numerous studies, the effect of a single USP, if its duration is  $\tau$  shorter than the orbital period of an electron in an atom (or the characteristic time  $T_g$  associated with the particle energy in the ground state  $E_g$ ,  $\tau < T_g = 2\pi\hbar/E_g$ ), on a quantum system is determined by the electric area of the pulse and its atomic scale, rather than the pulse energy [30–34].

The situation becomes more complex when a sequence of USPs affects the system. For example, when resonant media are excited by several USPs consisting of a small number of oscillations, it is possible to form a complex pattern of multi-frequency photon and combination echo [35–37].

In cases where pulse durations and delays between them become comparable to intra-atomic times, a non-trivial pattern of superposition of individual pulse contributions may arise. Without resorting to complex quantum mechanical calculations, it is possible to obtain a number of clear relationships in the case of low-amplitude pulse exposure when conventional perturbation theory is applicable. As we have shown earlier, in this case, the interaction of a USP sequence with quantum systems can be described based on the recently introduced concept of "interference of pulse electric areas" [38] (see also review [14]). In this case, the populations of bound states are determined by the sum of squares of pulse electric areas and an interference term, which resembles the expression for light intensity during interference of a pair of monochromatic light waves [39]. Note that when electromagnetic waves interact with quantum systems, one can also speak about the interference of bound states. It can be shown that the expression for the probability of system transition from one state to another is, again, formally similar to the classical one: the sum of squares of the system's basis state amplitudes and an interference term oscillating at the transition frequency [40].

Another possible application of USPs is the creation of population difference gratings using a sequence of pulses in a resonant medium during their coherent interaction with the medium, i.e., when the

duration of pulses and delays between them are shorter than the medium's polarization relaxation time  $T_2$ . Such population difference gratings were previously studied for quasi-monochromatic pulses, particularly for applications in echo holography [41–44]. At the same time, such population gratings can also be induced by extremely short pulses [45–48]. In the approximation when low-amplitude pulses are used and the medium under consideration is sufficiently rarefied (which allows neglecting not only the influence of neighboring atoms on each other but also the change in the shape of incident pulses during propagation), the emergence of these gratings can be easily understood based on the introduced concept of pulse area interference [35].

All previous studies of population grating dynamics were conducted using various approximations, such as the few-level approximation of the resonant medium or the approximate solution of the Schrödinger equation in the first order of perturbation theory [48]. In work [49], coherent excitation of the medium was studied based on the model of one-dimensional infinite potential wells, which is also a rather crude idealization.

Despite the large number of recent works in the field of unipolar subcycle light optics, several questions regarding the interaction of subcycle USPs with quantum systems remain unexplored. These questions include the features of already nonlinear "interference of pulse areas" when a sequence of powerful USPs acts on quantum systems, where perturbation theory is not applicable and strong ionization of the system is possible. The question of the possibility of creating population gratings in multilevel media remains unexplored, taking into account ionization, which can lead to significant depletion of the well and bound states.

Therefore, in this work, based on the direct numerical solution of the time-dependent Schrödinger equation, we study the dynamics of bound state population excitation and particle ionization probability in a one-dimensional rectangular quantum well of finite depth, excited by a pair of attosecond pulses, depending on the delay between pulses. In this case, unlike previous studies, we consider the effect of powerful pulses when the delays between them are comparable to intra-atomic times, making both conventional perturbation theory and sudden perturbation approximation inapplicable.

The simplest model of a one-dimensional quantum well is used. This model, despite its simplicity, finds various applications in the physics of interaction between ultrashort light pulses and quantum systems – atoms [50, 51], nanostructures [52–54], and other systems [55].

The features of nonlinear interference of pulse areas are studied. Based on the obtained results, the possibility of creating population gratings in an extended multilevel rarefied medium is discussed for the first time, taking into account its ionization when excited by a pair of non-overlapping USP.

## 2. THEORETICAL MODEL AND SYSTEM UNDER CONSIDERATION

The interaction of quantum systems with the field of external light USP is described by the time-dependent Schrödinger equation for the electron wave function  $\Psi(x, t)$  [56]:

$$i\hbar \frac{\partial \Psi}{\partial t} = [\hat{H}_0 + V(t)]\Psi. \quad (2)$$

Here  $\hat{H}_0$  is the system's own Hamiltonian and  $V(t) = -dE(t)$  — energy of system interaction with the field of external pulses in dipole approximation,  $d = qx$  is the dipole moment,  $q$  is the electron charge,  $\hbar$  is the reduced Planck constant.

The system is excited by a pair of attosecond USP, having the form

$$E_e(t) = E_1 \exp\left\{\frac{-t^2}{\tau_1^2}\right\} \cos(\omega_1 t + \phi_1) + E_2 \exp\left\{\frac{-t^2}{\tau_2^2}\right\} \cos(\omega_2[t - \Delta] + \phi_2), \quad (3)$$

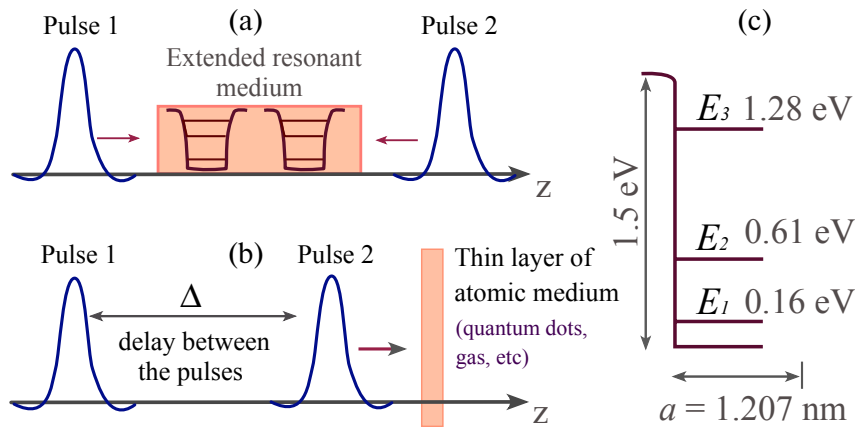
where  $t$  — time,  $E_{1,2}$  — amplitudes,  $\tau_{1,2}$  — durations of both pulses respectively,  $\omega_{1,2}$  is the central frequency of pulses,  $\phi_{1,2}$  — phase (carrier envelope phase, CEP) (for USP these parameters have

conditional meaning),  $\Delta$  — is the delay between acting pulses.

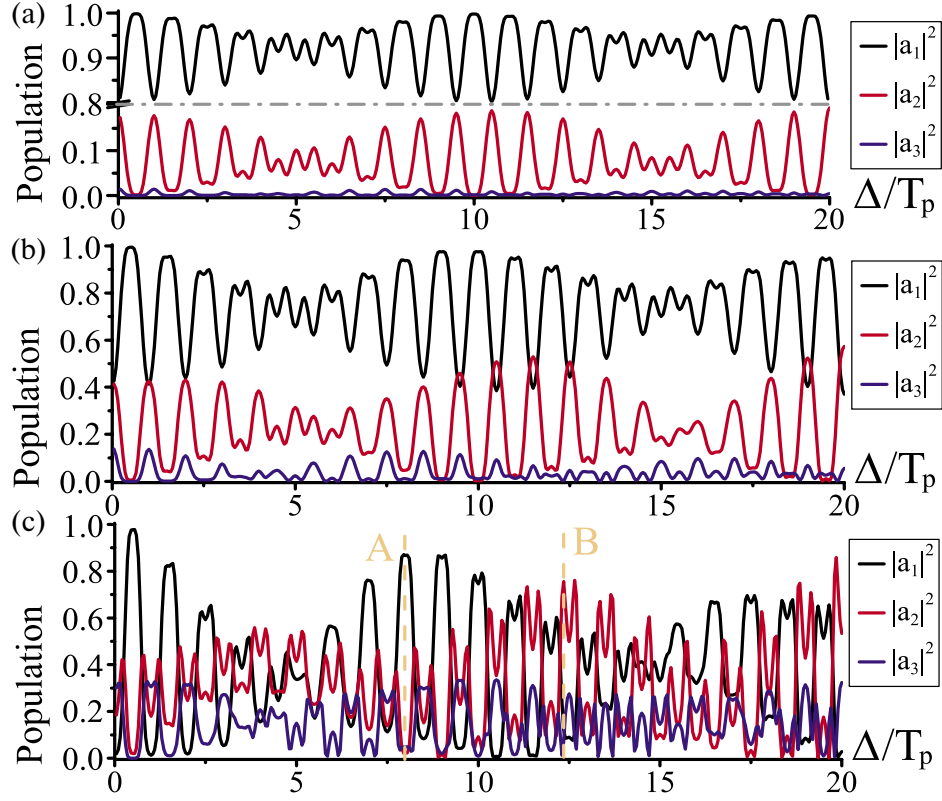
Simple relations for the population of bound states can only be obtained in the weak-field approximation in the first order of perturbation theory. Then the expression for the population can be written as

$$w_n = \frac{d_{1n}^2}{2\hbar^2} S_1^2 \exp\left\{-\frac{\omega_{1n}^2 \tau_1^2}{2}\right\} \frac{\cos 2\phi_1 + \text{ch}(\omega_1 \omega_{1n} \tau_1^2)}{\cos^2 \phi_1} + \frac{d_{1n}^2}{2\hbar^2} S_2^2 \exp\left\{-\frac{\omega_{1n}^2 \tau_2^2}{2}\right\} \frac{\cos 2\phi_2 + (\omega_2 \omega_{1n} \tau_2^2)}{\cos^2 \phi_2} + \frac{d_{1n}^2}{\hbar^2} S_1 S_2 e^{-\omega_{1n}^2 (\tau_1^2 + \tau_2^2)/4} \frac{1}{\cos \phi_1 \cos \phi_2} \times \left[ \cos \omega_{1n} \Delta \left( \cos(\phi_1 + \phi_2) \text{ch}\left(\frac{\omega_{1n}(\omega_1 \tau_1^2 - \omega_2 \tau_2^2)}{2}\right) + \cos(\phi_1 - \phi_2) \text{ch}\left(\frac{\omega_{1n}(\omega_1 \tau_1^2 + \omega_2 \tau_2^2)}{2}\right) \right) + \sin \omega_{1n} \Delta \left( \sin(\phi_1 + \phi_2) \text{sh}\left(\frac{\omega_{1n}(\omega_2 \tau_2^2 - \omega_1 \tau_1^2)}{2}\right) - \sin(\phi_1 - \phi_2) \text{sh}\left(\frac{\omega_{1n}(\omega_1 \tau_1^2 + \omega_2 \tau_2^2)}{2}\right) \right) \right], \quad (4)$$

where  $d_{1n}$  is the dipole moment of transition,  $\omega_{1n}$  is the resonant transition frequency, and the electric areas of both pulses are introduced



**Fig. 1.** a — Excitation of an extended medium by a pair of USPs propagating towards each other; medium particles are shown schematically as one-dimensional quantum wells. b — Coherent excitation of a thin layer of medium by a pair of USPs propagating with some delay  $\Delta$  relative to each other. c — Schematic representation of the considered quantum well



**Fig. 2.** Dependencies of bound state populations  $|a_i|^2$  in a rectangular potential well of finite depth on the delay between pulses  $\Delta/T_p$  at pulse amplitudes  $E_{02} = 5 \cdot 10^7$  B/cm (a),  $E_{03} = 1 \cdot 10^8$  B/cm (b),  $E_{04} = 2 \cdot 10^8$  B/cm (c)

**Table 1.** Parameters of pulses and quantum well model used in calculations

Varying Pulse Parameters			Quantum Well Parameters			
$E_{0i}$ , B/cm	$S_0$ , B · c/cm	$T_e$ , fs	$E_d = 1.5$ eV		$a = 1.207$ nm	
$1 \cdot 10^7$	$1.07 \cdot 10^{-9}$	128.04	$S_a = 1.09 \cdot 10^{-8}$ , B·c/cm			
$5 \cdot 10^7$	$5.36 \cdot 10^{-9}$	25.61	$E$ , eV		$T_i$ , fs	$\omega_i$ , 10 <sup>14</sup> rad/s
$1 \cdot 10^8$	$1.07 \cdot 10^{-8}$	12.80	$E_1$	0.1547	26.73	2.35
$2 \cdot 10^8$	$2.14 \cdot 10^{-8}$	6.40	$E_2$	0.6128	6.75	9.31
$3 \cdot 10^8$	$3.22 \cdot 10^{-8}$	4.27	$E_3$	1.2784	3.24	19.42

$$S_{1,2} = \sqrt{\pi} E_{1,2} \tau_{1,2} \exp \left\{ -\frac{\omega_{1,2}^2 \tau_{1,2}^2}{4} \right\} \cos \phi_{1,2}.$$

It is easy to see that the population of the bound state of the medium periodically depends on the delay between pulses  $\Delta$  and is determined by the

sum of squares of electric areas  $S_{1,2}$ . In this sense, we can talk about the interference of pulse areas, as discussed previously in work [38]. In the simplest case, with identical pulse parameters, i.e.

$$\begin{aligned} E_1 &\neq E_2, & \tau_1 &= \tau_2 = \tau, \\ \omega_1 &= \omega_2 = \omega, & \phi_1 &= \phi_2 = \phi, \end{aligned}$$

expression (4) reduces to

$$w_n = \frac{d_{1n}^2}{2\hbar^2} \exp \left\{ -\frac{\omega^2 \tau^2}{4} \right\} \frac{\cos 2\phi + \omega \omega_{1n} \tau_1^2}{\cos^2 \phi} \times \\ \times (S_1^2 + S_2^2 + 2S_1 S_2 \cos \omega_{1n} \Delta). \quad (5)$$

As shown previously [45–48], the case of an extended and rarefied medium (Fig. 1a) allows us to neglect both the influence of neighboring atoms on each other and the change in pulse shape during propagation. Expression (5) most clearly describes the interference of electric areas of a pair of exciting USPs acting on a quantum system.

Expression (5) also indicates the possibility of creating a population grating using a pair of pulses propagating towards each other without simultaneous overlap in the medium. In the case of a single structure (thin layer) (Fig. 1b), expression (5) shows the possibility of controlling the excitation of quantum systems using a pair of USP by changing the delay between them. Obviously, this consideration is approximate and does not allow taking into account the influence of medium ionization on the population of bound states.

Let's consider a model in the form of a one-dimensional potential well of finite depth. Despite its simplicity, this model is used in describing metal nanoparticles and semiconductor nanostructures [52–54]. The potential energy of a particle in this case is written as

$$U(x) = 0, \quad |x| > \frac{a}{2},$$

$$U(x) = -E_d, \quad |x| \leq \frac{a}{2}.$$

The calculation parameters had the following values: well width  $a = 1.2$  nm, depth  $E_d = 1.5$  eV; there are three bound states of the particle in the

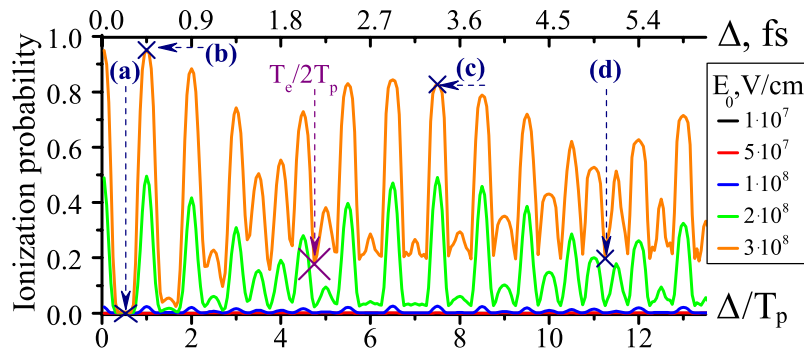
well with energies  $E_1 = 0.1574$  eV,  $E_2 = 0.6128$  eV,  $E_3 = 1.278$  eV (the associated characteristic frequencies ( $\omega_i = E_i/\hbar$ ) and times  $T_i$  are given in Table 1), which corresponds to transition wavelengths  $\lambda_{12} = 2706.49$  nm ( $T_{12} = 9.03$  fs) and  $\lambda_{13} = 1103.36$  nm ( $T_{13} = 3.68$  fs) (see Fig. 1c). The well parameters were chosen so that the wavelength of the transition from the ground to the first excited state was on the order of several micrometers, which is typical, for example, for quantum dots.

The value  $\omega$  was  $10 \cdot 14^{15}$  rad/s (wavelength  $\lambda_a = 134.6$  nm), period  $T_p = 2\pi/\omega = 448.8$ , phase  $\phi = 0$ , excitation pulse duration  $\tau = 0.2T_p \approx 89.7$  ac. Pulses of such duration, with amplitudes on the order of  $\sim 10^8$  B/cm can be obtained in various nonlinear processes [7, 20–26]. The amplitude was varied, see Table 1. The time-dependent Schrödinger equation was solved numerically using the Crank-Nicolson method [57].

### 3. INTERACTION OF A PAIR OF FCP WITH A SINGLE QUANTUM WELL

In this section, we will consider the interaction of a pair of USP with a single quantum well (optically thin layer). This situation is schematically shown in Fig. 1a. Fig. 2 illustrates the dependence of bound state populations after the pulse action on the amplitude of the field  $E_0$  and delay between pulses  $\Delta$ , normalized to  $T_p$ . A complex form of this dependence is visible; and with increasing amplitude  $E_0$  it becomes more "peaked", differing from the simple harmonic form predicted by formula (4) above. It is evident that the population behavior has a similar form up to a certain pulse intensity threshold.

The dependencies above show the possibility of ultrafast control of bound states in quantum wells and creation of population inversion in them using a pair of FCP. For example, by



**Fig. 3.** Dependencies of particle ionization probability in the considered quantum well on the delay between pulses  $\Delta$  at different pulse amplitudes  $E_0$  [B/cm]

selecting parameters, it is possible to maximize the population of one state without affecting the population of others, see indicators A and B in Fig. 2c. Thus, for A at  $\Delta/T_p = 8$  it is possible to obtain values  $|a_1|^2 = 0.87$ ,  $|a_2|^2 = 0.02$ , and  $|a_3|^2 = 0.05$ ; and at point B at  $\Delta/T_p = 12.35$  the bound level populations are and  $|a_3|^2 = 0.07$ . Such possibility was previously demonstrated in atomic-molecular systems [46, 48].

#### 4. CONTROL OF PARTICLE IONIZATION IN QUANTUM WELLS USING A PAIR OF FCP

Fig. 3 illustrates the dependence of ionization probability

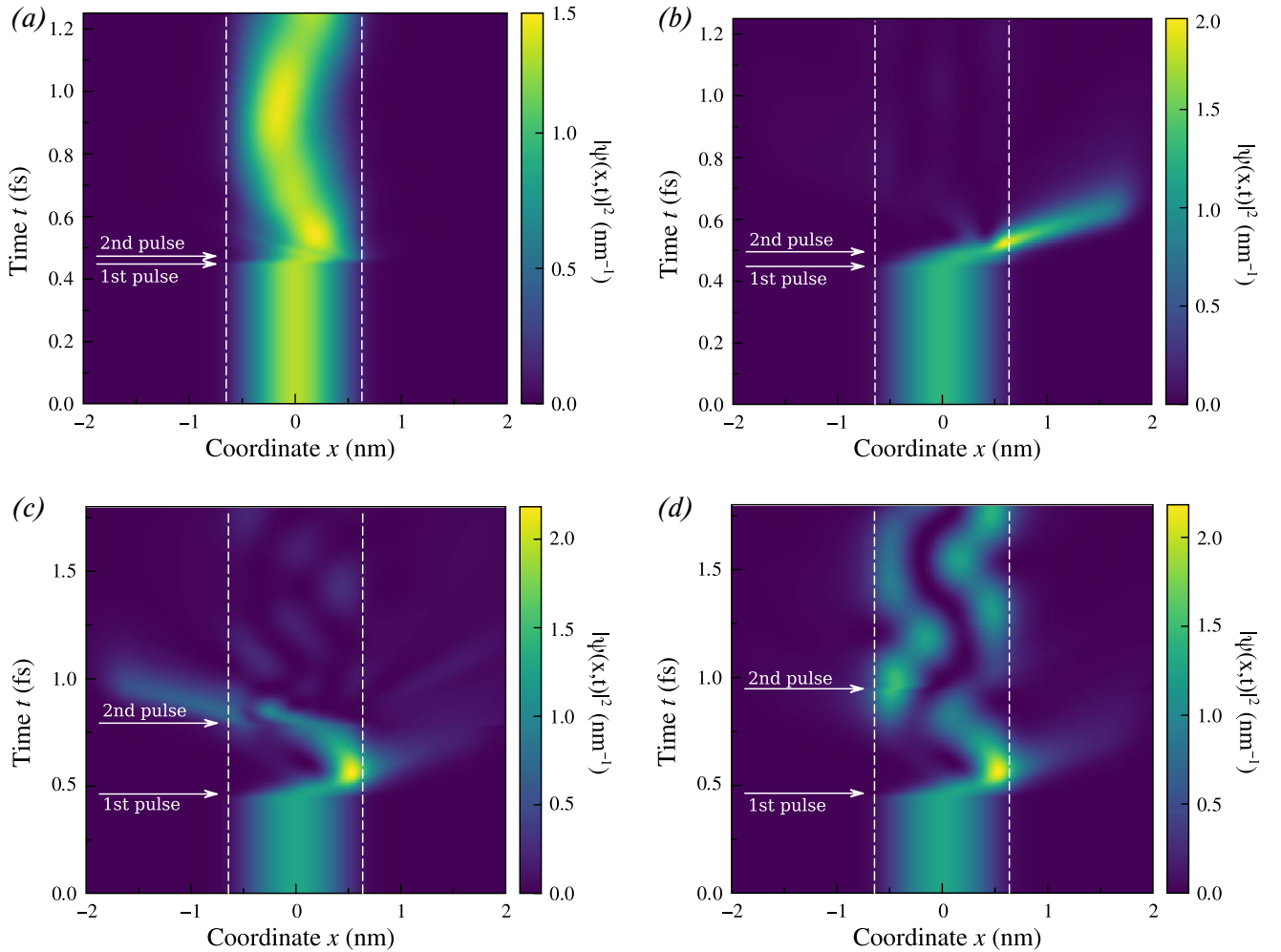
$$w_i = 1 - (|a_1|^2 + |a_2|^2 + |a_3|^2)$$

on pulse intensity  $E_0$  and delay between them  $\Delta$ . In the case of

$$E_0 = 1 \cdot 10^7 - 1 \cdot 10^8 \text{ B/cm}$$

the ionization value does not exceed 0.025; and for larger amplitude values, it is possible to select a delay value that minimizes ionization. It is interesting to note that for  $E_{04} = 2 \cdot 10^8 \text{ B/cm}$  and  $E_{05} = 3 \cdot 10^8 \text{ B/cm}$ , ionization reaches its minimum value in the section  $0.35 < \Delta/T_p < 0.75$ , after which the minimum inversion value reaches a "plateau"; the maximum value is achieved at  $\Delta/T_p = 1$ , point (b) in Fig. 3. Thus, by changing the ratio  $\Delta/T_p$  in a small range from 0.75 to 1, we can either exclude system ionization or completely remove particles from the well.

Let's examine in detail the ionization of particles in a quantum well for the most illustrative case  $E_{05} = 3 \cdot 10^8 \text{ B/cm}$ . For this, we will select four points marked in Fig. 3 and track the temporal evolution of the wave packet for cases of maximum and minimum



**Fig. 4.** Wave packet propagation in the considered quantum well at the amplitude of interacting pulses  $E_{05} = 3 \cdot 10^8 \text{ B/cm}$  and different delay values  $\Delta$ :  $\Delta/T_p = 0.5$  (a), 1 (b), 7.5 (c), 11.25 (d); these points are also marked in Fig. 3. Arrows indicate the moments of pulse action



ionization at different values of the ratio  $\Delta/T_p$ . In Figs. 4 *a, b* the absolute minimum and maximum for particle ionization in the quantum well are presented; while  $\Delta \sim T_p$ , i.e., the pulses are still close and can act as one, interfering in the process. Fig. 4*c* shows the local minimum of ionization; Fig. 4*d* shows the local maximum; for these cases  $\Delta > T_p$ , the pulses do not overlap.

Let's look at Fig. 4*b* – the case of absolute maximum ionization, and Fig. 4*c* – the case of local maximum. It can be seen that some time after the action of the second pulse in Fig. 4 with we still observe some wave packet distribution inside the well, while in Fig. 4*b* there is practically none. It is visible that in the case of Fig. 4*a* the wave packet begins slow oscillations while remaining completely within the well, and in Fig. 4*d* complex behavior of the wave function is observed, although most of it is confined within the well boundaries.

We can talk about periodic motion of the wave packet as long as it hasn't significantly distorted (spread). The characteristic spreading time (analog of diffraction length) can be obtained from the Schrödinger equation for a free electron:

$$i\hbar \frac{\partial \Psi}{\partial t} = -\frac{\hbar^2}{2m} \nabla^2 \Psi.$$

We assume by order of magnitude

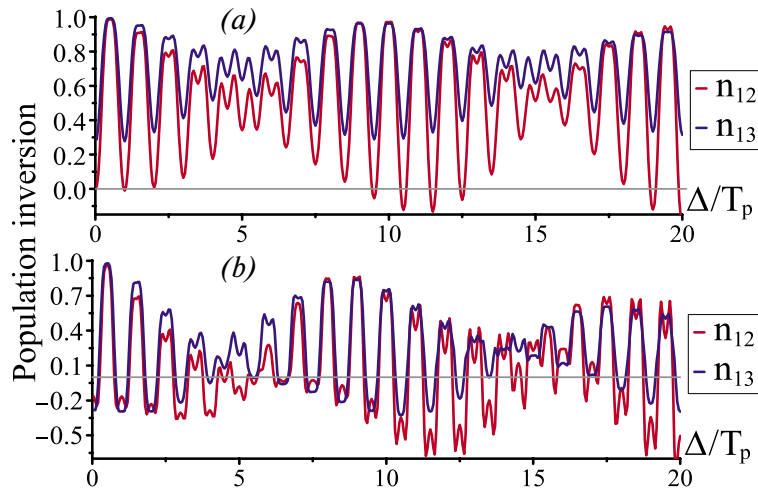
$$\frac{\partial \Psi}{\partial t} \sim \frac{\Psi}{T_d}, \quad \nabla^2 \Psi \sim \frac{\Psi}{a^2}.$$

Then  $T_d \sim 2ma^2/\hbar$  or  $T_d \approx 25.8$  fs, which is much longer than the considered times.

The relationship between the populations of bound states of the system and the parameters of the acting pulses can be described by comparing the area of the electric pulse  $S_E$  with the characteristic atomic scale of the system  $S_a$  [33, 34].  $S_E$  is a quantitative measure of the incident pulse's effect on the system, namely: depletion of the ground state and excitation of upper levels; the area values  $S_E$  for each amplitude value  $E_{0i}$  are given in Table 1. Using the definition of atomic measure of electric area introduced in work [33], for the quantum well considered in this work we get  $S_a = 2\hbar/ea = 1.09 \cdot 10^{-8}$  B · s/cm.

For  $E_{01}$  and  $E_{02}$  the atomic measure of electric area is larger than the electric area of the pulses interacting with it,  $S_a > S_E$ . Indeed, in Fig. 2*a* it can be seen that the pulse has almost no effect on the population of excited states, and only slightly populates the ground state,  $|a_1|^2$ ; no ionization is observed either (see Fig. 3). In Fig. 2*b*, where  $S_a > S_{E_3}$ , a significant population of states is visible, and in Fig. 5*a* there is population inversion. Ionization is present but does not have a noticeable effect on the system. For pulses with amplitude  $E_{04}$  and  $E_{05}$ , the relation  $S_a < S_{E_i}$  holds (see Table 1), and, as a result, significant ionization (complete for  $E_{05}$ , Fig. 3) and the difference in populations of bound states (see Fig. 5*b*).

Let's try to explain the dependence of ionization on the delay between pulses from another perspective. Let the first pulse transfer momentum of magnitude  $p_i = eS_E$  to the electron in the well; the corresponding wave packet will move between the well walls with velocity  $V_i = p_i/m_e$ . In the simplest model considered, the electron motion in the well after the first "push" will be periodic, with period  $\tau_i = 2a/V_i$ , or



**Fig. 5.** Dependencies of particle population inversion  $n$  in the final well on the delay between pulses  $\Delta/T_p$  at pulse amplitude  $E_{03} = 1 \cdot 10^8$  B/cm (a),  $E_{04} = 2 \cdot 10^8$  B/cm (b)

$T_e[c] = 1.3727 \cdot 10^{-22}/S_{E_i}$  [B · s/cm]; the dependence on the delay of the second pulse is characterized by the same period. The obtained period values  $T_e$  are listed in Table 1. It can be seen that they are 1–3 orders of magnitude larger than  $T_p$ . For  $E_{04} = 2 \cdot 10^8$  B/cm, the ratio is  $T_e/2T_p = 7.13$ ; for  $E_{05} = 3 \cdot 10^8$  B/cm, the ratio is  $T_e/2T_p = 4.75$ ; the latter is marked with a dot in Fig. 3. These are local minimums, not absolute ones.

## 5. CREATION OF POPULATION GRATINGS IN AN EXTENDED MEDIUM OF QUANTUM WELLS

In this section, we will consider the possibility of creating population gratings in an extended medium based on quantum wells using a pair of pulses propagating towards each other and not overlapping in the medium, see Fig. 1a. It was previously shown that the problem of the effect of such pulse pairs on an extended medium can be reduced to the problem of the effect of a pair of extremely short pulses on a single quantum object with variable delay between pulses, as in Fig. 1, in the case when particle density is low, which allows neglecting changes in pulse shape during propagation in the medium and the influence of neighboring atoms on each other, see [45, 46, 48]. By delay  $\Delta \sim z/c$  we should understand the moment of arrival of the second pulse into the medium at the point with coordinate  $z$ ,  $c$  — speed of light, therefore below we also use the results from the previous section. The

results of such simplified consideration are consistent with the results obtained based on numerical solution of Maxwell-Bloch equations taking into account pulse propagation effects in an extended medium [45, 46, 48].

For detailed analysis, let's focus on the data for pulse amplitudes  $E_{03} = 1 \cdot 10^8$  B/cm and  $E_{04} = 2 \cdot 10^8$  B/cm, (see Fig. 2); in this case, significant population values are visible and there is no large ionization (see Fig. 3). The resulting dependence of population inversion  $n_{li} = |a_1|^2 - |a_i|^2$  on ratio of delay between pulses  $\Delta$  to pulse length  $T_p$  is shown in Fig. 5.

In Fig. 5a, harmonic beats are clearly visible; let's try to describe this dependence in the form

$$n = C - \sum_{i=1}^n a_i \cos\left(2\pi \frac{\Delta}{T_p} v_i\right)$$

using numerical Fourier transformation of the data. The coefficients  $C$  we obtained, dimensionless frequencies  $v_i$  (equivalent to the value of ratio  $T_p/\Delta$ ) and their amplitudes  $a_i$  are presented in Table 2.

On the graph Fig. 5b beats with a more complex dependence, therefore when describing we will consider the phase  $\theta_i$ :

$$n = C - \sum_{i=1}^n a_i \cos\left(2\pi \frac{\Delta}{T_p} v_i + \theta_i\right),$$

coefficients  $C, a_i, \theta_i$  and frequencies  $v_i$  are also presented in Table 2.

**Table 2.** Parameters of population inversion approximation  $n$ , obtained using Fourier transformation of dependencies in Fig. 5

$E_0$	$E_0 = 1 \cdot 10^8$ B/cm		$E_0 = 2 \cdot 10^8$ B/cm			
	$n_{12}$	$n_{13}$	$n_{12}$		$n_{13}$	
$C$	0.53	0.7	0.09		0.24	
Frequency $v_i, T_p/\Delta$	$a_i$		$a_i$	Phase $\phi$ , rad	$a_i$	Phase $\phi$ , rad
	0.0242	0.1182		4.5113	0.0475	4.482
0.15	0.0212	0.1043		1.4241	0.0388	1.78
0.90	0.0134		—		0.0559	1.92
0.95	0.3061	0.1826	0.3629	0.4018	0.3217	0.1443
1.05	0.2655	0.1606	0.2909	0.3223	0.2745	0.424
1.10	0.0119		—		0.0425	1.2315
1.90	—		0.0632	−3.9111	—	
2.00	0.0980	0.0817	0.0746	−2.6365	0.0313	0.5828
2.95	—		0.1218	−1.8758	0.0447	−2.7384
3.05	—		0.1043	3.7552	0.0375	−1.817



Although formula (4) predicts a simple harmonic dependence at frequency  $\nu_i = T_p/\Delta = 1$ , calculations in both cases show that it is absent in the spectrum. Instead, the main contribution comes from two components near it with a small detuning:  $T_p/\Delta = 0.95$  and  $T_p/\Delta = 1.05$ , or  $T_p/\Delta = 1 \pm \delta$ , where  $\delta \sim 0.05$ , which corresponds to  $T_p - \delta = 21 - 24$  au, of the order of one atomic unit (24.189 au). The addition of these two oscillations with close frequencies generates the beats.

It can also be assumed that the low frequencies ( $\nu_i = 0.1$  and  $\nu_i = 0.15$ ) appear due to connection with characteristic transition times, the corresponding ratios have the form

$$\frac{T_p}{T_{13}} \approx 0.12, \quad \frac{T_p}{T_{12}} \approx 0.05, \quad \frac{T_p}{T_{23}} \approx 0.08.$$

The presence of additional frequencies, detuning, and, in the case of high field amplitude, the appearance of a significant phase shift  $\theta_i$  is difficult to explain; multi-frequency response is characteristic of quantum systems in strong fields; the complex picture of nonlinear interference cannot be underestimated.

## 6. CONCLUSIONS

Based on a one-dimensional model of a particle in a rectangular well, the system's response to a pair of half-cycle attosecond pulses was studied. Using numerical solution of the time-dependent Schrödinger equation, the dependencies of bound state populations and ionization probability on the delay between pulses were studied. It is shown that this dependence has the form of beats and with increasing field amplitude, it takes on a more complex peak structure. This demonstrates the manifestation of nonlinear interference of pulse areas in strong fields and its difference from the linear case, where the dependence of populations on the delay between pulses is harmonic and is determined by expressions (4) and (5).

The calculation results showed the possibility of both enhancement and suppression of particle ionization in the well when changing the delay between pulses, which in the case of a rarefied extended multilevel medium qualitatively predicts the possibility of inducing a spatial population difference grating. The results obtained in this work show the possibility of using a sequence of half-cycle pulses for ultrafast control of wave packet dynamics in matter and thereby open new directions in unipolar light optics.

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