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Superradiance of a Stopped Polarization Pulse in a Thin Layer of a Five-Level Medium Excited by Subcycle Attosecond Pulses

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Using the numerical solution of the system of equations for the amplitudes of bound states, together with the wave equation, we theoretically consider stopped polarization pulse superradiance upon excitation by a pair of half-period attosecond pulses in a thin layer of a five-level resonant medium, the parameters of which are the same as in a hydrogen atom. It is shown that in the case of a multilevel medium, at certain parameters of the exciting field, the superradiance pulse near the medium is also a single-cycle pulse, the shape of which is determined by the first time derivative of the stopped polarization pulse, as in the case when approximate low-level and classical models were used to describe the response of the medium.

Keywords: superradiance, attosecond pulses, stopped polarization pulse, unipolar pulses, subcycle pulses.

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Introduction

Obtaining ultrashort electromagnetic pulses of femto- and attosecond durations has occupied an important place in modern optics in recent decades [1]. A large number of reviews published recently [2–7] are devoted to this topic. The interest in obtaining ultrashort pulses is due to the fact that pulses of such short duration are actively used to study and control the dynamics of wave packets in atoms, molecules, solids, etc. [7–9].

For example, recently, the possibility of using attosecond pulses to study the dynamics of populations of bound states in a solid [10], studying the migration of charges in molecules [11] has been actively studied. With the help of attosecond pulses, the possibility of controlling the shape of the absorption line in molecules in the extreme ultraviolet (XUV) region of the spectrum [12] was revealed, the relative delay in the response of electrons in a dielectric system caused by a strong field of pulses lasting several periods of the field [13,14] was measured. The use of a sequence of XUV-attosecond pulses makes it possible to study the dynamics of multiphoton ionization of atomic systems [15].

To date, numerous schemes for obtaining attosecond pulses have been proposed. All of them are based on the high-order harmonic generation (HOHG) method that occur when various targets are irradiated with a femtosecond laser pulse [1–9]. Various gating schemes for isolating a single attosecond pulse from the general sequence [16] are discussed. In addition to obtaining attosecond pulses on account of HOHG, it is worth mentioning various methods for obtaining such pulses based on acceleration or deceleration of electrons in plasma [17–19]. There are also known methods for isolating single bursts of a single-cycle and subcycle-shaped field due to the Fourier synthesis of broadband pumping [13,14]. The possibility of obtaining already unipolar half-period pulses with a nonzero electric area, which can provide faster and more efficient excitation of quantum systems compared to conventional multi-cycle pulses [20–22], is discussed.

In all the proposed methods for obtaining femto- and attosecond pulses, their source is a short-term pulse of polarization of the medium or a pulse of free electron current created by an external ultrashort pumping pulse [1–9]. Recently, alternative methods have been proposed for obtaining ultrashort pulses in a resonant medium due to the superradiance of a pulse of stopped polarization in a resonant medium excited by a pair of ultrashort pumping pulses [22–29]. In this case, the first pulse creates oscillations of nonlinear polarization in the medium at the frequency of the resonant transition, and the second pulse, acting after a time interval equal to half of the oscillation period of the resonant transition $T_0/2$, stops these oscillations. In this case, the polarization pulse has the form of a half-wave and is conventionally called the stopped polarization pulse (SPP). This SPP can be a source of superradiance of the system of excited centers [29].

We note that the superradiance — is the collective spontaneous emission of a concentrated system of phased oscillators (dipoles). It was first predicted by Dicke in 1954 [30]. At present, this term „superradiance“ is used to describe a diverse class of problems in various systems [31].

The only thing that remains unchanged in all contexts is the meaning of the term „superradiance“, which characterizes the presence of phasing of radiation emitting centers — atoms, molecules, spins, quantum dots, etc. [31]. Such superradiance phenomena in the above sense, in which the collective nature of the radiation of a phased ensemble of active particles manifests itself, have been studied for a wide class of systems to date, see, for example, [31–36] and the cited literature.

In our case, the duration of the excited pulses and the interval between them are assumed to be shorter than the relaxation time of the polarization of the medium T_2, therefore, the emitting centers will emit in a phased manner at the time intervals under consideration. Also, the radiation of the field due to the SPP is formed between a pair of external radiation pulses, i.e., in fact, in the absence of an external field. In this sense, the radiation of the medium can be considered spontaneous [37]. Thus, below, for the process of SPP of the medium emission we are considering, we will use the term „superradiance“ in the broad sense indicated above — collective spontaneous emission of a phased system of dipoles.

In an extended medium, this SPP can be used to obtain unipolar pulses of unusual inharmonic shape — rectangular and triangular — in the THz range [23,24]. Also, when a thin layer of the medium is excited by a pair of pumping pulses in a reflected field, it is possible to obtain a single-cycle pulse of THz radiation [25] or a single-cycle pulse of attosecond duration in the XUV range due to the superradiance of the SPP [26,27]. The results of these studies are summarized in the review [29].

In [38], the features of the SPP superradiance propagating in an extended resonant medium and generated by a pair of half-cycle attosecond pulses are studied. This can lead to unipolar pulses of THz radiation of rectangular or triangular shape in an inhomogeneous medium [39].

However, the mentioned studies used the simplest models of the medium in the form of a classical nonlinear oscillator, or the medium was modeled in a low-level approximation, see the review [29]. But the real media are multilevel — when such a medium is excited by an ultrashort pulse with a wide spectrum, a large number of levels are excited. This may lead to the fact that the SPP, which can easily be created in an medium of classical oscillators (a two-level medium) having a single transition frequency, may not occur in a real multi-level environment. Therefore, the issue of SPP superradiance in a real environment with a large number of levels requires a separate study.

In this paper, based on the numerical solution of the equations for the amplitudes of bound states (arising from the Schrodinger time equation), the SPP superradiance is studied for a five-level medium excited by a pair of attosecond pulses following with a period equal to half the period of the resonant transition from the ground to the first excited state. The parameters of the medium are selected (transition frequencies and dipole moments), as in a hydrogen atom. It is shown that at certain parameters of external pulses, a single-cycle pulse of superradiance of the SPP medium also occurs in the reflected field, as in the case when approximate low-level or classical models of the medium were used.

The system and model under consideration

The considered scheme of excitation of the superradiance pulse is shown in Fig. 1. An optically thin layer of medium of thickness L is located perpendicular to the axis z, between the points z_1 and z_2. The medium is modeled in a five-level approximation with a converging scheme of levels, as in atomic systems (Fig. 1, *b*). Atomic hydrogen was taken as a model medium, the corresponding values of frequencies and dipole moments of transitions are given in the table below. It is worth noting that the values for the first 5 levels of the hydrogen atom with the values of the main quantum number from 1 to 5 were used, i.e. we did not take into account the presence of S-, P- and D-sublevels, but implied them all as part of the corresponding degenerate levels [40].

The layer is excited by a pair of unipolar attosecond pulses, which are considered in a one-dimensional approximation in the form of plane waves propagating along the z axis. One-dimensional propagation of unipolar pulses can be realized in coaxial waveguides [41].

The pulse repetition interval is equal to half of the period $T_0/2$ of the resonant transition in the medium from the ground state to the first excited one. The shape of the pulses is taken as

$$E(t) = E_0 e^{\frac{-t^2}{\tau^2}} + E_0 e^{\frac{-(t-T_d)^2}{\tau^2}}, \qquad (1)$$

$T_d = T_0/2$ — delay between pulses, E_0 — pulse amplitude, τ — their duration.

The response of a five-level medium to an external field is described using equations for the amplitudes of bound states, which have the form [42]:

$$\psi(\mathbf{r}, t) = \sum_{n=1}^{5} \psi_n(\mathbf{r}) a_n(t) e^{-iE_n t/\hbar},$$

$$\omega_{nl} = \frac{E_n - E_l}{\hbar},$$

$$a_n(t) = \frac{i}{\hbar} \sum_{n=1}^{5} d_{nm} a_m(t) E(t) e^{i\omega_{nm} t}, \qquad (2)$$

where $a_n(t)$ — the amplitudes of the decomposition of the atomic wave function by eigenfunctions $\psi(\mathbf{r})$, d_{nm} — the dipole moments of transitions between the levels n and m, and ω_{nm} — corresponding transition frequencies. We note that we neglect the relaxation terms in the equations (2), since the lifetimes of excited levels lie, as a rule, in the nanosecond range, while the periods of resonant transitions

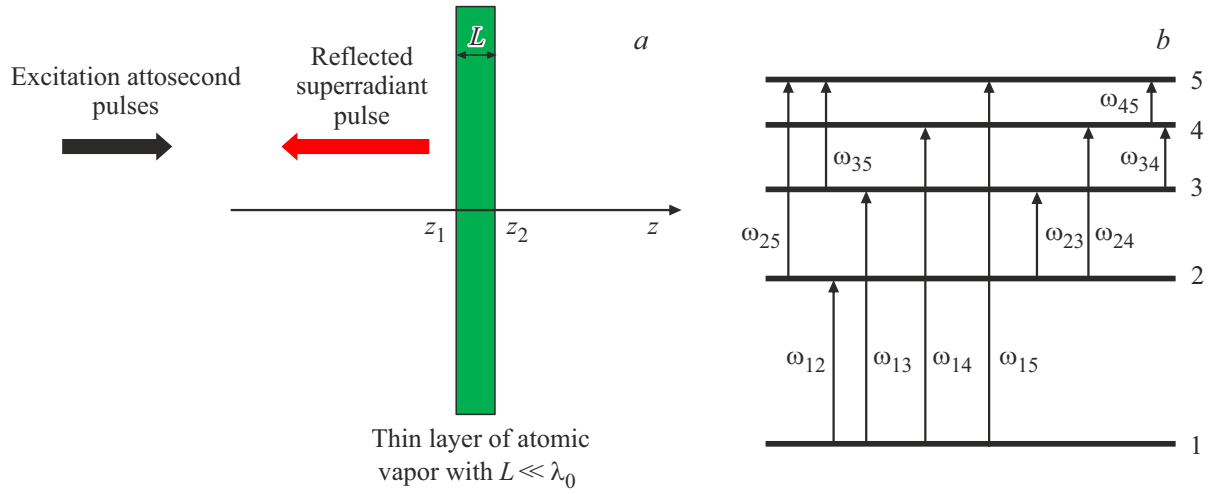

Figure 1. A scheme in which SPP superradiance occurs in a five-level resonant medium (a) excited by a pair of subcycle attosecond pulses; (b) diagram of the working levels of a five-level environment. The numbers of levels and frequencies of resonant transitions are indicated.

in the system under consideration, as can be seen from the table, are only a few femtoseconds.

Based on the numerical solution of a system of equations (2) the polarization of the medium was calculated, which is given by the expression [42]:

$$P(t) = N_0 \sum_{n=1}^{5} d_{nm} a_n(t) a_m^*(t) e^{i\omega_{nm}t} + c.c., \qquad (3)$$

where N_0 — volume concentration of atoms. The evolution of the electric field is described by a one - dimensional wave equation:

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

in which E — the electric field strength with fixed linear polarization, c — the speed of light in vacuum.

In the case under consideration, Fig. 1 analytical solution of the wave equation (4) for the field reflected from the medium layer (superradiance pulse) generated by the SPP $P(z,t)$ has the form [43]:

$$E_s(z,t) = -\frac{2\pi}{c}\int_{z_1}^{z_2} \frac{\partial}{\partial t} P\left(z', t - \frac{|z - z'|}{c}\right) dz'. \qquad (5)$$

In our case, the medium is an optically thin layer of atomic vapors, shown in Fig. 1, the thickness of which is assumed to be much less than the wavelength, $L \ll \lambda_0$, then the expression (5) for the superradiance pulse field is significantly simplified and has the form [26–28]:

$$E_s(t) = -\frac{2\pi}{c} L N_0 \frac{\partial}{\partial t} p(t), \qquad (6)$$

that is, the response of such a thin layer is determined by the time dependence of the first time derivative on the polarization of the medium. Here p — polarization of a single atom, $P = N_0 p$, N_0 — concentration of atoms in the layer.

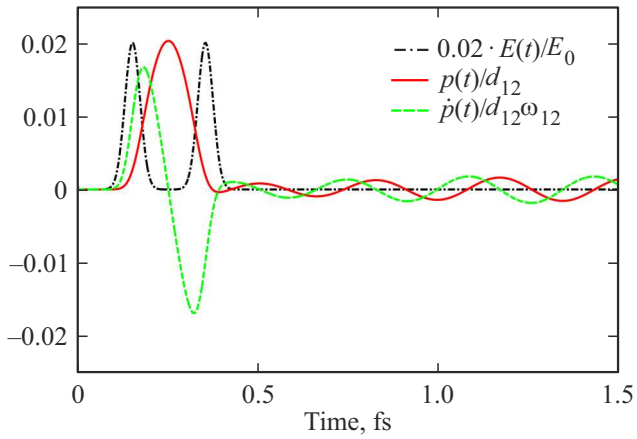

Figure 2. Optical response of a single hydrogen atom in a 5-level approximation when excited by a pair of subcycle unipolar pulses of duration $\tau = 30$ as and amplitude $E_0 = 10^5$ ESU (shown by a black dotted line); all values are displayed in dimensionless units.

Numerical simulation results and discussion of results

Let us first consider the excitation of a single hydrogen atom by a sequence of two attosecond unipolar pulses (1), and the delay between the pulses is equal to half the period for the transition between levels 1 and 2. Fig. 2 shows the corresponding values of the polarization of a single atom and its time derivative for the duration of the exciting pulses $\tau = 30$ as and the amplitude $E_0 = 10^5$ ESU. It can be seen that the second of the pulses (1) almost

Transition frequencies and dipole moments for the first 5 levels of atomic hydrogen, as well as parameters of exciting pulses and the medium layer

The duration of the excitation pulses	$\tau = 30\,\text{as}$
The transition frequency is 12 in the hydrogen atom (transition wavelength)	$\omega_{12} = 1.55 \cdot 10^{16}\,\text{rad/s}$ ($\lambda_{12} = 121.6\,\text{nm}$)
Dipole transition moment 12	$d_{12} = 3.27\,\text{D}$
The transition frequency is 13 in the hydrogen atom (transition wavelength)	$\omega_{13} = 1.84 \cdot 10^{16}\,\text{rad/s}$ ($\lambda_{13} = 102.6\,\text{nm}$)
Dipole transition moment 13	$d_{13} = 1.31\,\text{D}$
The transition frequency is 14 in the hydrogen atom (transition wavelength)	$\omega_{14} = 1.94 \cdot 10^{16}\,\text{rad/s}$ ($\lambda_{14} = 97.3\,\text{nm}$)
Dipole transition moment 14	$d_{14} = 0.77\,\text{D}$
The transition frequency is 15 in the hydrogen atom (transition wavelength)	$\omega_{15} = 1.98 \cdot 10^{16}\,\text{rad/s}$ ($\lambda_{15} = 95.0\,\text{nm}$)
Dipole transition moment 15	$d_{15} = 0.53\,\text{D}$
The transition frequency is 23 in the hydrogen atom (transition wavelength)	$\omega_{23} = 2.87 \cdot 10^{15}\,\text{rad/s}$ ($\lambda_{23} = 656.6\,\text{nm}$)
Dipole transition moment 23	$d_{23} = 12.63\,\text{D}$
The transition frequency is 24 in the hydrogen atom (transition wavelength)	$\omega_{24} = 3.88 \cdot 10^{15}\,\text{rad/s}$ ($\lambda_{24} = 486.1\,\text{nm}$)
Dipole transition moment 24	$d_{24} = 4.85$
The transition frequency is 25 in the hydrogen atom (transition wavelength)	$\omega_{25} = 4.34 \cdot 10^{15}\,\text{rad/s}$ ($\lambda_{25} = 434.1\,\text{nm}$)
Dipole transition moment 25	$d_{25} = 2.83\,\text{D}$
The transition frequency is 34 in the hydrogen atom (transition wavelength)	$\omega_{34} = 1.01 \cdot 10^{15}\,\text{rad/s}$ ($\lambda_{34} = 1875.1\,\text{nm}$)
Dipole transition moment 34	$d_{34} = 29.33\,\text{D}$
The transition frequency is 35 in the hydrogen atom (transition wavelength)	$\omega_{35} = 1.47 \cdot 10^{15}\,\text{rad/s}$ ($\lambda_{35} = 1281.8\,\text{nm}$)
Dipole transition moment 35	$d_{35} = 10.76\,\text{D}$
The transition frequency is 45 in the hydrogen atom (transition wavelength)	$\omega_{45} = 4.63 \cdot 10^{14}\,\text{rad/s}$ ($\lambda_{45} = 4067.2\,\text{nm}$)
Dipole transition moment 45	$d_{45} = 29.93\,\text{D}$
Particle concentration in the layer	$N_0 = 2.7 \cdot 10^{19}\,\text{cm}^{-3}$
The thickness of the medium layer	$L = 20\,\text{nm}$

completely stops the polarization oscillations caused by the action of the first pulse. As a result, the time dependence of the polarization is well approximated by a half-wave at the transition frequency of $1-2$. The contribution of the overlying levels, however, leads to the presence of noticeable tails, since the polarization oscillations at the frequencies of all other transitions will not be completely stopped by the second pulse, but may even be amplified on the contrary. Consequently, the relative contribution of the residual polarization tails depends on how strongly the levels above the second level will be populated compared to the population of the second level. The calculations show that the contribution of higher levels can be neglected, and, accordingly, describe the polarization of the medium half-wave at a transition frequency of $1-2$ for the amplitudes of exciting pulses up to values of the order 10^5 ESU.

Fig. 3 shows the calculated response of the medium when the pulse amplitude increases to the value $E_0 = 5 \cdot 10^5$ ESU.

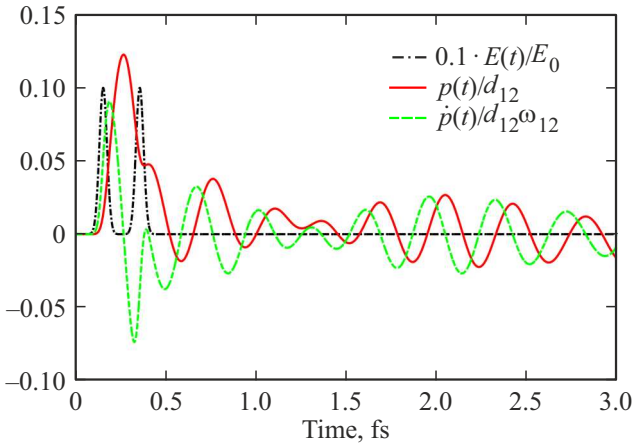

Figure 3. Optical response of a single hydrogen atom in a 5-level approximation when excited by a pair of subcycle unipolar pulses of duration $\tau = 30$ as and amplitude $E_0 = 5 \cdot 10^5$ ESU (shown by a black dashed line); all values are displayed in dimensionless units.

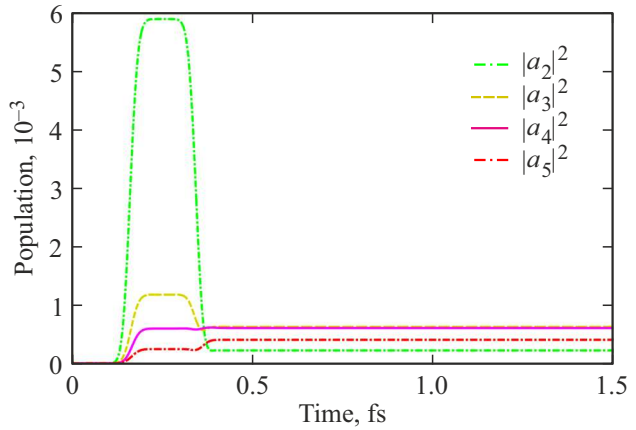

Figure 4. Population levels of the hydrogen atom in the 5-level approximation when excited by a pair of subcycle unipolar pulses for parameters from Fig. 3.

It is clearly seen that the second exciting pulse can no longer effectively stop the oscillations of the polarization of the medium due to the significant contribution of the upper levels. As a result, the residual polarization tails have an amplitude comparable to the amplitude of the main half-wave of polarization.

Let us now consider the form of the time derivative of the polarization of the medium, which, in accordance with equation (6), will determine the type of the radiated field. This time derivative for a single atom is shown by green dashed lines in Fig. 2–3. It can be seen that with the amplitude of the exciting pulses up to values of the order 10^5 ESU this time derivative can be described fairly accurately as one cycle of harmonic oscillations at a transition frequency of 1–2, i.e. one period of the sine wave. We note that the exact shape of the received pulse significantly depends on the duration of the exciting pulses τ, or rather on the ratio between it and the time delay between the pulses $T_d = T_0/2$. In the case of a decrease in the value of τ compared to the value from Fig. 2, instead of one period of the sine wave, we will get a sharper increase in the field during the action of the first pulse and a sharper decrease during the action of the second pulse, between which, however, there will be a harmonic half-wave of duration $T_d = T_0/2$ at a transition frequency of 1–2.

Residual tails in the radiated field arise due to the contribution to polarization from other possible transitions, as can be seen from equation (3). At large pulse amplitudes, as in Fig. 3, the approximation of the time derivative of polarization by the period of the sinusoid is no longer applicable, since the resulting tails have almost the same amplitude as the initial single-cycle sinusoid. The reason for this is the excitation of higher levels, as shown in Fig. 4. As can be clearly seen, the populations of levels 3, 4 and 5 after the passage of the second exciting pulse remain significantly higher than the population of level 2, which means the presence of residual polarization oscillations at the corresponding transitions.

Superradiance of an optically thin layer of hydrogen atoms

Finally, we turn to radiation not from a single atom, but from a whole thin layer, the parameters of which are given in the table. To do this, the wave equation (1) was solved numerically by the finite-difference time-domain (FDTD) method, while the equations were solved numerically for the response of the medium(2) by the Runge–Kutta method of high order. The time dependence of the electric field calculated in reflection from the layer is shown in Fig. 5.

It can be seen that the field in Fig. 5 agrees well with the time derivative of the polarization of a single atom in Fig. 2, which is provided by the choice of a small layer thickness. As a result, the radiated field is a single cycle of a sine wave

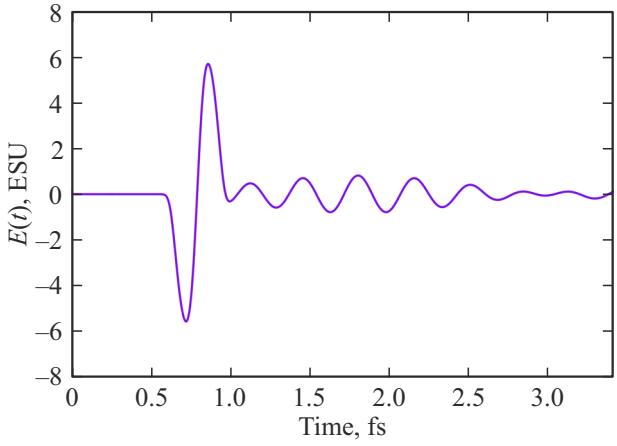

Figure 5. The reflected field from a layer of atomic hydrogen with a thickness of 20 nm and a pressure of 1 atm when excited by a pair of subcycle unipolar pulses of duration $\tau = 30$ as and amplitude $E_0 = 10^5$ ESU.

at a transition frequency of 1−2, followed by a residual tail from the contribution of other transitions in the multilevel medium under consideration.

Conclusion

Thus, in this work, the superradiance of a thin layer of a five-level medium with parameters like in a hydrogen atom was theoretically studied. The medium was excited by a pair of unipolar subcycle attosecond pulses following with a period equal to half the period of the resonant transition from the ground state of the atom to the first excited state. Based on the numerical solution of the equations for the amplitudes of bound states, it is shown that with the duration of excitation pulses of several tens of attoseconds, a half-wave of stopped polarization can be obtained in a multilevel medium. In this case, a medium superradiance pulse occurs in the reflected field, the shape of which near the medium layer is determined by the first time derivative of induced polarization. It should be noted, that this result is consistent with those obtained earlier, when approximate models of the medium were used, for the amplitudes of exciting pulses up to values of the order 10^5 ESU. At a higher excitation field strength, the upper levels of the medium begin to populate, as a result of which the form of polarization may differ markedly from the ideal half-wave.

The studied superradiance effect, in our opinion, can be observed in thin films (jets) of atomic and molecular gas media. Perhaps it should be expected when using a thin film of nanoparticles (quantum wells) as a medium. In quantum wells (for example, one-dimensional ones), the structure of the levels is no longer the same as in atoms, shown in Fig. 1. The distance between the energy levels increases when approaching the well ceiling, while in atomic systems the distance between the levels decreases. Therefore, the problem of the occurrence of SPP in quantum wells requires a separate study based on the numerical solution of the Schrodinger time equation and is beyond the scope of this article.

The conducted research shows the possibility of using half-cycle unipolar attosecond pulses for ultrafast control of radiation and properties of various materials and opens up new directions in the study of the interaction of unipolar subcycle pulses of light with matter in the framework of a new direction in modern optics — „Optics of unipolar and subcycle light “, which arose recently and has been actively developing recently, see the review [21].

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Conflict of interest

The authors declare that they have no conflicts of interest.

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