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Generation of unipolar pulses of terahertz radiation with a large electric area

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A physical situation is proposed and theoretically analyzed, in which, in our opinion, it is possible to generate unipolar terahertz pulses with a large electric area. In this case, the gas in the tube is excited by a femtosecond IR laser pulse. In this case, the tube with gas is placed in a constant external electric field. The generation of a unipolar pulse is based on „three-step scheme" — ionization of gas atoms by a femtosecond pulse, subsequent acceleration of a free electron in a dc external field and subsequent annihilation of an electron upon collision with a tube wall or another atom (ion).

Keywords: unipolar pulses, ultrafast optics, electric pulse area, terahertz radiation, three-step model.

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Introduction

The problem of shortening the duration of light pulses is actual since the invention of lasers and at the present time [1]. Today, the possibilities of obtaining ultrashort pulses of terahertz radiation [2,3], as well as subfemtosecond pulses in the optical and extreme ultraviolet frequency range [4–6] are being actively studied. Such pulses find numerous applications in ultrafast optics and spectroscopy of various substances [7,8].

The limit for reducing the width of electromagnetic pulses is obtaining unipolar pulses containing one half-period of the electric field. Unlike ordinary multicycle pulses, they have a non-zero electric area, which is determined at each point in space as an integral of the field strength over time [9]:

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

where E is electric field strength and t is time. For conventional bipolar multicycle pulses, the electric area is always close to zero.

To excite quantum transitions in a medium, long resonant multicycle laser pulses are used. Interest in unipolar pulses is due to the fact that they are able to very quickly transfer a mechanical pulse to a charged particle in one direction. And therefore, they can be used for ultra-fast and efficient control of the dynamics of wave packets in matter, as compared to bipolar pulses, acceleration of charges, and other applications. See details of the latest results in this area in reviews [9–11] and cited literature.

If the width of such half-period pulse is much less than the characteristic time associated with the intraatomic motions of an electron in an atom (for example, the orbital period of an electron in an atom or the characteristic time associated with the energy of the ground state, $T_g = 2\pi\hbar/E_1$, where E_1 is the energy of the particle in the ground state), then the impact of such pulse on a microobject is determined by the electric area of the pulse, but not by its energy [10–15].

Therefore, when the populations of atomic systems are excited, unipolar pulses are „universal", not requiring frequency tuning to the frequency of the quantum transition under consideration, as during resonant excitation by bipolar multicycle pulses [15]. In this connection, the problem of obtaining unipolar pulses with large electric area, rather than energy, is actual. To specify the degree of ultrashort pulses effect on quantum systems, a new physical quantity has recently been introduced — an atomic scale of electric pulse area inversely proportional to the size of the microobject [13–15]. Despite the great interest in unipolar pulses, their experimental generation is still a difficult task.

For the first time, the presence of unipolarity was experimentally demonstrated for terahertz pulses generated at optical rectification and filamentation in liquid jets excited by femtosecond laser pulses [16]. In this paper estimates of the electric area of pulses are also given. The paper [17] theoretically shows the possibility of obtaining half-cycle isolated subfemtosecond pulses with large electric area due to the phenomenon of self-induced transparency.

In [18] terahertz pulses were experimentally obtained in the form of precursors arising from the propagation of the femtosecond pulse in the electro-optical crystal. In theoretical papers [19–21] the possibility of obtaining THz pulses by amplifying single-cycle pulse in a gas plasma is discussed. The soliton-like mode of generation of terahertz radiation in a quadratic nonlinear medium was theoretically studied in paper [22]. However, estimates of the electric area were not carried out in these papers.

In the present paper, we consider an experiment scheme in which, in our opinion, generation of unipolar terahertz pulse with large electric area is possible. This situation can be realized when single-cycle femtosecond laser pulse propagates in a gas-filled tube placed in a constant external electric field. The generation of terahertz pulse is based on the three-step scheme proposed below, which consists of three main steps (ionization, acceleration, annihilation) — ionization of gas atoms by a passing external pulse, subsequent acceleration of extracted electrons by DC external field and after a distance equal to about the free path length the annihilation of electron upon collision with another atom, ion, or tube wall. As a result of ionization in each transverse layer of the medium a short-lived dipole is formed, which is the source of unipolar radiation pulse.

The proposed three-step mechanism for generating terahertz pulses is somewhat similar to the well-known three-step model for generating high-order harmonics (HHG) proposed by P. Corkum, due to which subfemtosecond pulses are generated [23,24]. In this case, at the first step the atom is ionized by a laser pulse, then the free electron is accelerated and the subsequent recombination of the electron with the parent ion under the action of the same laser pulse occurs.

In our case, the generation of terahertz pulse also occurs in three steps, and the generation model is based on the processes of ionization, acceleration, and recombination of the extracted electron. Therefore, the model being basis of the process considered will also be called the „three-step model“. However, our situation differs from P. Corkum's three-step HHG mechanism. In our case, the incident pulse is considered to be very short, i.e., it only has time to ionize the electron, then it is accelerated by DC external field and annihilates when it collides with the tube wall or another atom.

Numerical estimates of the parameters of the generated pulse were carried out. It is shown that the electric area of the generated pulse can be comparable with the atomic measure for vibrational transitions in a molecule.

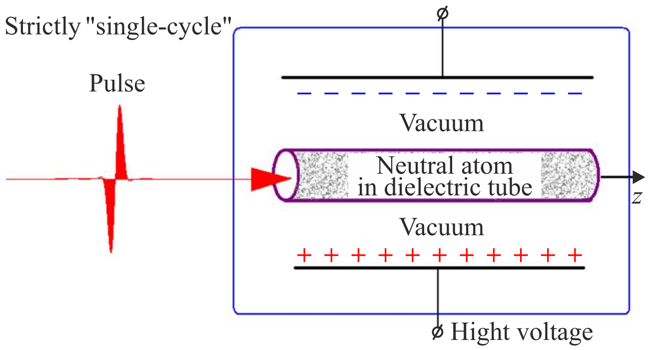

Figure 1. Scheme of experiment and main idea. The gas tube is placed in DC external electric field. A single-cycle laser pulse runs through the tube ionizing the electrons.

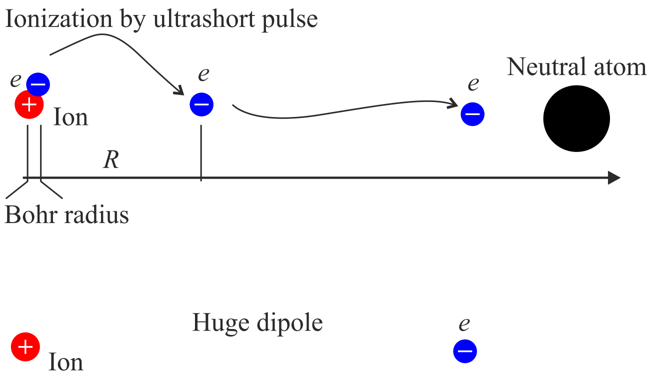

Figure 2. Schematic description of the three-step generation mechanism of unipolar terahertz pulse — the appearance of the free electron due to ionization, leading to the occurrence of a giant dipole. The subsequent electron acceleration by the DC external field and its annihilation upon collision with a wall or another atom.

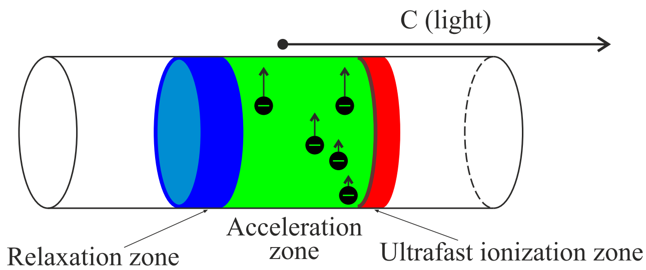

Figure 3. The movement of the ionization zone (red) along the waveguide, creating the field of generated pulses.

Description of the experiment and three-step model for obtaining unipolar terahertz pulse

The scheme of the proposed experiment and the underlying idea of obtaining the unipolar pulse are described in Figs. 1–3 (three-step generation mechanism).

Let us imagine a waveguide (thin capillary) filled with gas and placed in a DC external electric field between the plates of a capacitor. Note that a similar scheme was used in various experiments on the generation of terahertz radiation in filaments in air in the presence of DC external field, see e.g. [25–27]. However, the possibility of obtaining unipolar pulses was not discussed in these papers.

The terahertz pulse is generated in three steps. We will call the mechanism underlying these steps as the „three-step“ model for generating unipolar terahertz pulses by analogy with the three-step HHG mechanism [23,24]. The system is irradiated with a single-cycle femtosecond pulse shown schematically in Fig. 1 and propagating along the z axis. Such a pulse ionizes electrons as it propagates in the gas, creating a moving ionization front on its way (Fig. 3). The extracted electrons start to accelerate in the DC external field. In this case, in pair with the ion, formed as a result of ionization, the electron forms a giant dipole, which is the source of the unipolar pulse (Fig. 2).

Then, after traveling some distance, approximate the free path length, the free electron collides with another atom or the tube wall and annihilates. At the second step of acceleration by the external field, the electron does not have time to experience external effects in the form of collisions. All electrons will only be accelerated in exactly one direction, which creates the pulse of the terahertz field. But during collisions their accelerations will be in different directions. And collisions will occur at different time moments. This contribution of electrons deceleration can cause the appearance of a long tail of opposite polarity and a small amplitude in the main pulse. When acting on quantum objects, the contribution of this tail can be ignored [28]. Therefore, its contribution to the main radiation pulse is not taken into account in future.

Similar three processes occur in each transverse layer of the medium i.e. ionization, leading to the appearance of a free electron, its short-term acceleration in an external field and subsequent annihilation of the electron. A moving ionization front arises in the medium, which is the source of radiation of a half-period terahertz pulse. This situation is shown schematically in Fig. 3. In the situation under consideration, the total radiation field at the exit from the medium can be represented as the sum of the fields radiated by the short-lived dipole in each layer of the medium, see the next Section.

Note that the similar situation, in which the radiation of quasi-unipolar terahertz pulses is also possible, was considered earlier [29–31]. But in this case, the generation of unipolar pulses occurs due to the collective spontaneous emission of stopped polarization pulse created and switched off by a pair of pumping pulses, see details in review [32] and the cited literature.

Note the importance of using precisely single-cycle pulses in the scheme under consideration. When excited by a long multicycle pulse containing many cycles of field oscillations and with a duration greater than the orbital period of the electron, the extracted electrons can scatter in all directions. When excited by a single-cycle pulse in the form shown in Fig. 1, the emitted electrons will fly in one preferred direction, set by the DC external field, if the pulse width is comparable to or less than the orbital period of the electron.

In the next Section a theoretical description is given, and numerical estimates are made for the parameters of the generated pulses.

Theoretical model, numerical estimates and discussion of results obtained

For a theoretical description of the idea considered above, in the simplest case the following system of equations can be used, which contains the rate equation (2) for the concentration of extracted electrons n_e in gas under the action of the external field. The system also contains the wave equation (3), which describes the evolution of the electric field (polarized along the vertical axis perpendicular to the direction of excitation propagation in Fig. 1). This system of equations has the form

$$\frac{\partial}{\partial t} n_e = -\gamma n_e - [N_0 - n_e] W(t), \tag{2}$$

$$\Delta E(z,t) - \frac{1}{c^2}\frac{\partial^2}{\partial t^2} E(z,t) = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2} P(z,t). \tag{3}$$

In this system of equations, γ is the electrons recombination rate, N_0 is the concentration of gas atoms in the tube, $W(t)$ is the ionization rate, which is calculated using expression obtained in the refined Keldysh ionization theory [33–35],

$$W(t) = \alpha \frac{E_a}{|E_p(t)|} e^{-\beta \frac{E_a}{|E_p(t)|}}, \tag{4}$$

where α, β are some constants in the refined ionization model, $E_a = 5 \cdot 10^{11}$ V/m is intraatomic field, $E_p(t) = E_0 e^{-\frac{t^2}{\tau_p^2}} \sin \omega t$ is the excitation pulse that propagates along the z axis from left to right, as shown in Fig. 1.

In the approximation under consideration, the field source in equation (2) is a short-term polarization pulse P, which begins at the moment of the excitation arrival at a given point of the medium $t_1 = z/c$ and ends at time $t_2 = z/c + \tau$. This polarization is written as

$$P(z,t) = q n_e \frac{a[t - z/c]^2}{2}, \ \frac{z}{c} < t < \frac{z}{c} + \tau, \tag{5}$$

$$0, t < \frac{z}{c}, \ t > \frac{z}{c} + \tau,$$

where q is electron charge, a is electron acceleration in the DC external field. The time τ is determined by the acceleration that the electron acquires in the external field and by its free path length, see below.

We are interested in the electric field of the generated pulse at a point located on the z axis and at a large distance ($r \gg L$) from the right end of the tube, where L is its length. In this approximation, the radiated field is the sum of the fields of elementary dipoles located in each layer of the tube. The field amplitude of the generated pulse can be estimated

Model parameters

Model parameter	Its value
Excitation pulse intensity	$I = 10^{14}$ W/cm^2
Amplitude of excitation pulse field	$E_{op} = 2.7 \cdot 10^8$ V/cm
Center frequency (wavelength)	$\omega = 2.35 \cdot 10^{15}$ rad/s ($\lambda = 800$ nm)
Excitation pulse width	$\tau_p = 1; 5$ fs
Concentration of gas atoms at room temperature $T = 273$ K and pressure $P = 10^5$ Pa	$N_0 \sim 2.7 \cdot 10^{19}$ cm^{-3}
Cross-section of atom at its radius equal to two Bohr radiuses ($r_a = 10^{-8}$ cm)	$\sigma = \pi r_a^2 = 3.1 \cdot 10^{-16}$ cm^{-2}
Free path length of electron	$l \sim \frac{1}{\sigma N_0} \sim 3.1 \cdot 10^{-4}$ cm
Amplitude of DC external field	$E_{dc} = 260000$ V/cm
Achieved acceleration of electron	$a = \frac{eE_{dc}}{m_e} = 4.5 \cdot 10^{20}$ cm/s^2
Cross-section radius of gas tube	$R = 100\,\mu$m
Length of gas tube	$L = 1$ cm
Rate of electrons recombination	$\gamma = 10^{12}$ s^{-1}

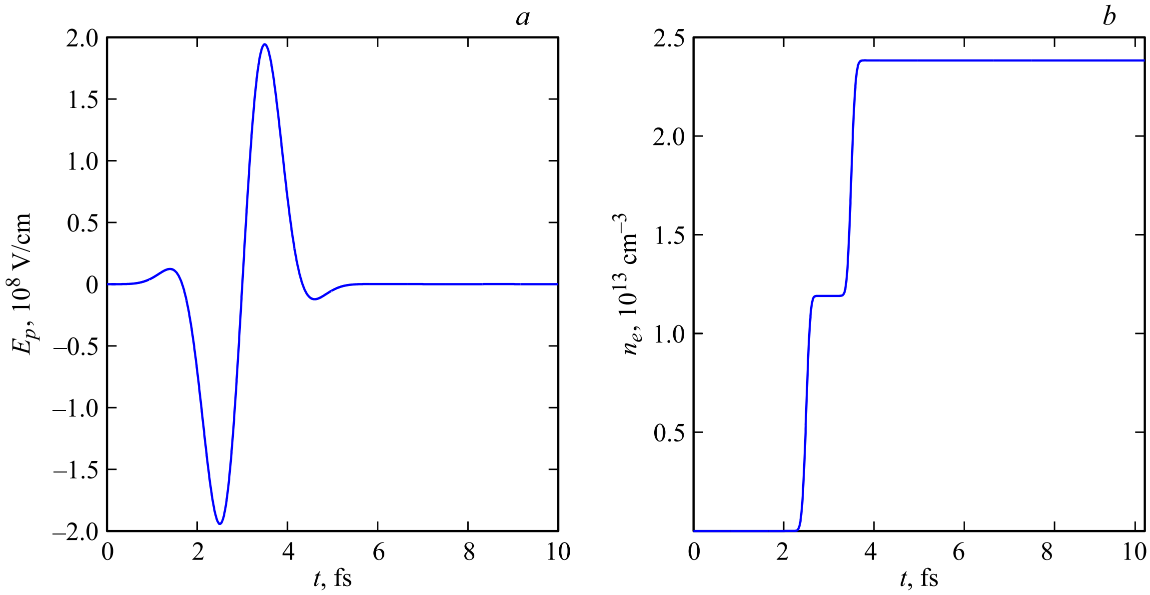

Figure 4. (a) Time shape of the excitation pulse $E_p(t)$, (b) concentration of extracted electrons $n_e(t)$ under action of this impulse vs. time. Excitation pulse width $\tau_p = 1$ fs. Other parameters are shown in Table.

using the well-known expression for the field amplitude of the emitting dipole in the far zone [36]:

$$E_0 = \frac{qa}{c^2 r} n_e \pi R^2 L, \tag{6}$$

where R is capillary radius.

Let us carry out numerical estimates of the amplitude of the generated field pulse, its duration and electric area, using the parameters given in Table. Then for the width of the generated pulse we obtain $\tau \sim \sqrt{2l/a} \approx 1$ ps.

The shape of the excitation pulse and the temporal behavior of the concentration of extracted electrons, calculated using the numerically solving of equation (3) for excitation durations $\tau_p = 1$ and 5 fs, are shown in Figs. 4 and 5, respectively.

For these parameters, as can be seen from these Figures, the concentration of extracted electrons can be about $n_e \sim 10^{13}{-}10^{16}$ cm^{-3}. The generated pulse is radiated over the free path length of the electron while it is accelerated by the external field.

At electrons concentration of $n_e \sim 10^{16}$ cm^{-3}, the generation field amplitude at the distance $r = 1$ cm from the tube can be estimated using formula (6). The estimate gives $E_0 = 2.3 \cdot 10^5$ V/cm.

Let us estimate the electric area of the pulse (for $\tau = 1$ ps): $S_E = E_0 \tau = 2.3 \cdot 10^{-5}$ V/m·s. This value must be compared with the value of the atomic scale of the area entered in [13]. The value of the atomic measure for the quantum harmonic oscillator can be estimated using the ratio [13] $S_{0,HO} = \frac{\sqrt{2\hbar\omega_0 m}}{q} \cong 10^{-5}$ V/m·s (parameters:

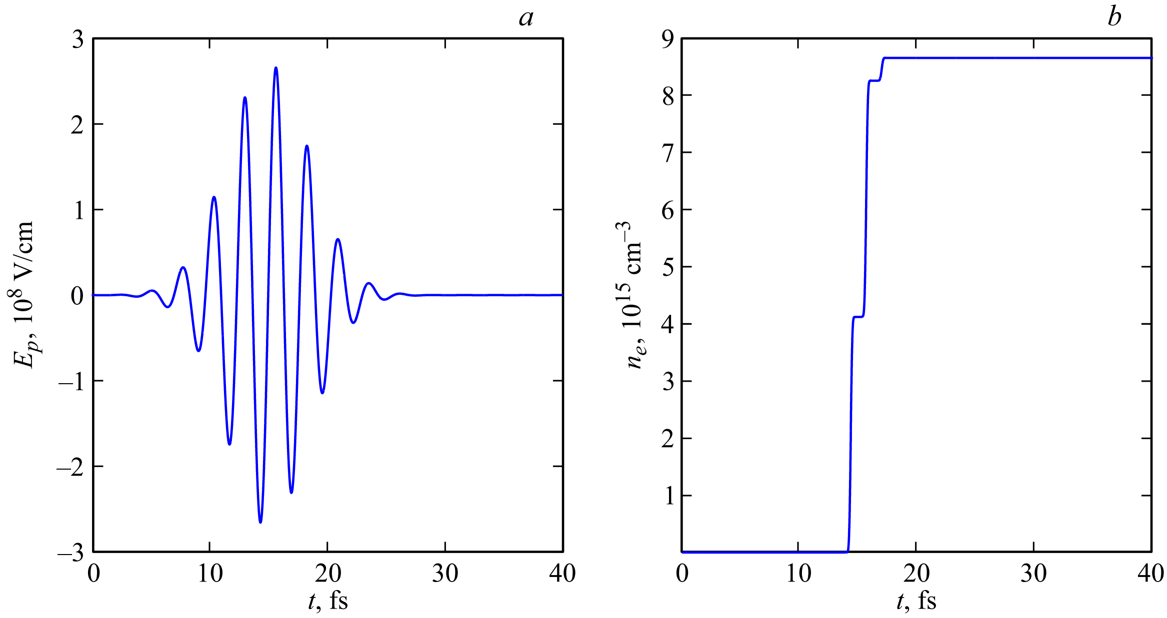

Figure 5. (a) Time shape of the excitation pulse $E_p(t)$, (b) concentration of extracted electrons $n_e(t)$ under the action of this impulse vs. time. Excitation pulse width $\tau_p = 5$ fs. Other parameters are shown in Table.

oscillator natural frequency $\frac{\omega_0}{2\pi} = 1$ THz, oscillator weight $m = 1.67 \cdot 10^{-27}$ kg was equal to the proton weight, the charge of the oscillator q was equal to the charge of the electron).

It can be seen that the value of the electric area obtained in our case is comparable in order of magnitude with the value of its area measure estimated for quantum harmonic oscillator with transition frequency in the terahertz range. Therefore, the generated pulses can be used for efficient excitation of vibrational transitions in molecules [15].

The above conclusions about the possibility of generating the unipolar pulse are confirmed by a direct numerical solution of wave equation (3) in the one-dimensional case. At the same time, in its right part there is a given field source i.e. unipolar polarization pulse moving at the speed of light. The calculation shows that when such given source moves, a moving single-cycle electric field pulse occurs, the amplitude of which increases linearly with distance. It is obvious that in this case the shape of the generation pulse in the far zone will be determined by the first time derivative of the given single-cycle pulse, i.e., will represent the half-cycle terahertz pulse of interest to us.

Note that the unusual situation considered above lies in the fact that the radiation source is an artificial short-term pulse of medium polarization moving at the speed of light. And real charges cannot move at the speed of light or higher.

However, there are well-known examples of various artificial sources of coherent radiation i.e., effective charges, light spots, etc., which can move at an arbitrary speed (including superlight). Various examples of such sources and their radiation are discussed in reviews [37–40] and in the cited literature.

Discussion of results. Conclusion

The scheme of the experiment is considered and analyzed, in which, in our opinion, it is possible to obtain unipolar terahertz pulses with large electric area. Generation occurs in three steps during ionization of gas excited by an external single-cycle femtosecond laser pulse, subsequent electron acceleration in an external static electric field, and subsequent annihilation of the free electron upon collision with the tube wall.

By analogy with the three-step HHG mechanism, the considered mechanism is also called as the three-step model for generating terahertz pulses. However, in the case of HHG, all three steps i.e. ionization, acceleration, and recombination of the extracted electron occur under the action of the initial excitation pulse. In the case considered the incident pulse only ionizes the atoms.

The proposed method makes it possible, in our opinion, to obtain unipolar pulses with electric area comparable to its atomic scale. Consequently, the resulting pulses can be used to effectively control the population of transitions in molecules [15].

In view of the small radius of the capillary with gas, one should expect a large divergence of the terahertz radiation and loss of unipolarity during pulse propagation. This may require the use of an additional light guide. The problem of transporting the unipolar pulses can be solved by using coaxial waveguides [41].

Note that the proposed model and the underlying system of equations (2)-(4) do not take into account the contribution of various types of nonlinearity, which can play a significant role in the formation of terahertz pulses in plasma [42,43]. However, the results of recent experiments [16] demonstrated the presence of unipolarity in terahertz pulses generated by water filamentation excited by femtosecond laser pulses. Therefore, the above three-step scheme for obtaining unipolar terahertz pulses, in our opinion, can serve as a good approximation for describing the actual experiment, the idea of which is considered in this paper.

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Conflict of interest

The authors declare that they have no conflict of interest.

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