09

# Comparison of the effects of unipolar half-cycle and resonant multi-cycle electromagnetic pulses on quantum systems

© R.M. Arkhipov[1,2], M.V. Arkhipov[1], N.N. Rosanov[1,2]

[1] St. Petersburg State University,
199034 St. Petersburg, Russia
[2] Ioffe Institute,
194021 St. Petersburg, Russia

e-mail: arkhipovrostislav@gmail.com, m.arkhipov@spbu.ru, nnrosanov@mail.ru

*Received February 25, 2022*
*Revised February 25, 2022*
*Accepted March 11, 2022*

It is shown that not only multicycle resonant pulses with zero electric area, but also nonresonant unipolar pulses can be used to efficiently excite quantum transitions in a medium. Arguments are presented in favor of the advantages of unipolar pulses for excitation of quantum systems in comparison with resonant radiation. A simple relationship has been found showing that in order to compare the effects of unipolar half-cycle and resonant multicycle pulses on atomic systems. It shows that it is necessary to compare the electric area of ??a unipolar pulse and the area of ??the envelope of a bipolar pulse, rather than their energies.

**Keywords:** unipolar pulses, electric pulse area, pulse area, extremely short pulses, attosecond pulses.

DOI: 10.21883/EOS.2022.07.54728.3318-22

## Introduction

Ultrashort femtosecond and attosecond electromagnetic pulses produced in practice contain several field oscillation cycles and are bipolar [1–3]. The electric area of such pulses, which is defined at each point in space as [4]

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

($E$ is the electric field strength and $t$ is time), is always close to 0.

The limit of contraction of light pulses is the transition to unipolar pulses that contain a marked field half-wave of one polarity (see reviews [5–7]). The electric area of such pulses may differ from zero. The possibility of existence of such pulses and their propagation in space has long remained a debated topic in scientific literature.

However, the existence of unipolar pulses does not contradict Maxwell equations and may propagate in coaxial waveguides. These issues were discussed in detail in reviews [4,5,7] and papers cited therein. In fact, different practical arrangements for the production of femtosecond and attosecond electromagnetic pulses close in shape to unipolar ones (containing a marked field half-wave of one polarity and a trailing edge of the opposite polarity) have already been proposed in recent studies [8–13]. Soliton solutions of nonlinear optics equations in the form of unipolar pulses are also being examined [14,15].

The interest in unipolar pulses stems from their potential to exert prompt and unidirectional influence on charges [5]. This opens up the opportunities to use such pulses for ultrafast control over the dynamics of microobjects [16–18], acceleration of charges [19], holography with an ultrahigh temporal resolution [20], etc. [5].

Radiation in the form of long multicycle pulses resonant with the given transition in a medium are commonly used in optics to excite quantum transitions in a resonant medium [21]. The resonant interaction of optical radiation with two-level media has already been studied thoroughly [21,22].

Unipolar pulses feature a wide spectrum starting from zero frequency. Owing to their short duration and the nonresonant nature of interaction with quantum systems, the efficiency of application of such pulses for excitation of resonant transitions in a medium in comparison with long resonant multicycle pulses was questioned.

It follows from the results of theoretical analysis that if a half-cycle pulse is shorter than characteristic time $T_g$ associated with the ground state energy ($T_g = 2\pi\hbar/E_1$, where $E_1$ is the particle energy in the ground state), the effect of such a pulse on level populations and the ionization probability of a system are defined by the electric area of a pulse normalized to its atomic measure rather than by the pulse energy or its peak amplitude [23,24]. In this context, a unipolar pulse with a nonzero area may, in contrast to a bipolar pulse with a zero area, exert a considerable effect on quantum transitions of a medium. However, this assertion remains true only for pulses that are significantly shorter than the oscillation period of an electron in the ground state.

In view of this, it appears necessary to compare the impact of a unipolar nonresonant pulse, which is shorter than the characteristic oscillation period of an electron in a

quantum system, and a bipolar resonant multicycle pulse on quantum transitions in a medium.

In the present study, an approximate solution of the Schrödinger equation in the first-order perturbation theory is used to find a simple relation demonstrating that a unipolar pulse may, under certain conditions, excite populations of levels of a medium just as efficiently as a resonant multicycle pulse.

This relation also demonstrates that in order to estimate the relative effectiveness of unipolar and multicycle bipolar pulses, one should compare the electric area of ??a unipolar pulse and the envelope area of a bipolar pulse instead of the energies of these pulses.

## Comparison of the effect of a unipolar half-cycle pulse and a resonant multicycle pulse on a quantum system

The specifics of the impact of ultrashort pulses on populations of bound states in a medium were examined in [25]. It was demonstrated that when a pulse is shorter than the period of a transition in a medium, the populations of bound states are defined by electric area (1) of a pulse.

The effect of long multicycle pulses, which are longer than the periods of resonant transitions in a medium, is governed by the envelope area,

$$\Theta \equiv \frac{d_{1n}}{\hbar}\int\limits_{-\infty}^{\infty}\varepsilon(t)dt, \tag{2}$$

where $\varepsilon(t)$ is the slow envelope of a pulse, $d_{1n}$ is the transition dipole moment, and $\hbar$ is the reduced Planck constant.

This result is well known to researchers specializing in optics of coherent resonant interaction of short laser pulses with resonant media [21,22]. The envelope area of a pulse (pulse area) has first been introduced for long multicycle laser pulses in the theory of the self-induced transparency effect by McCall and Hahn [26].

**Let us pose the following question:** *which values of parameters (amplitude, duration, etc.) of a unipolar half-cycle pulse (shorter than the period of a resonant transition in a medium) and a resonant multicycle pulse (longer than the transition period in a medium) ensure that their impact on a bound state of a quantum system is the same?*

Let us assume that a system is excited by a pulse with a Gaussian envelope:

$$E_e(t) = E_0 e^{\frac{-t^2}{\tau^2}}\cos(\Omega t + \phi). \tag{3}$$

Here, $\Omega$ has the meaning of frequency, and $\phi$ is the phase (carrier envelope phase, CEP).

The following approximate expression for population of excited state $n$ after the interaction with a pulse may be obtained based on an approximate solution of the temporal Schrödinger equation in the first-order perturbation theory [25,27]:

$$w_n = 0.5\pi\frac{d_{1n}^2}{\hbar^2}E_0^2\tau^2\exp\left[-\frac{(\omega_{1n}^2+\Omega^2)\tau^2}{2}\right] \times\left[\cosh(\omega_{1n}\Omega\tau^2)+\cos 2\phi\right]. \tag{4}$$

Indices $u$ and $b$ are used hereinafter for unipolar and bipolar pulses, respectively.

*Case 1, unipolar pulse*: $\Omega = 0$, $\phi = 0$, the pulse is assumed to be shorter than the transition period, $\omega_{1n}\tau \ll 1$.

The following is then derived from (4):

$$w_{u,n} = \pi\frac{d_{1n}^2}{\hbar^2}E_0^2\tau^2. \tag{5}$$

*Case 2, bipolar multicycle resonant pulse*: $\Omega = \omega_{1n} \equiv \omega_0$, $\phi = 0$, the pulse is assumed to be longer than the transition period, $\omega_0\tau \gg 1$.

The following is then derived from (4):

$$w_{b,n} = \frac{d_{1n}^2}{\hbar^2}\pi E_0^2\tau^2\exp[-\omega_0^2\tau^2]\cosh(\omega_0^2\tau^2)$$

$$= 0.5\frac{d_{1n}^2}{\hbar^2}\pi E_0^2\tau^2\exp[-\omega_0^2\tau^2]\frac{\exp[\omega_0^2\tau^2]+\exp[-\omega_0^2\tau^2]}{2}.$$

As long as $\omega_0\tau \gg 1$, the second exponential function in brackets may be considered negligible in comparison to the first one. Then,

$$w_{b,n} \simeq 0.25\frac{d_{1n}^2}{\hbar^2}\pi E_0^2\tau^2 = 0.25\Theta^2. \tag{6}$$

Equating $w_{u,n} = w_{b,n}$, we obtain a condition for pulse parameters

$$\frac{d_{1n}^2}{\hbar^2}\pi S_E^2 = 0.5\Theta^2, \tag{7}$$

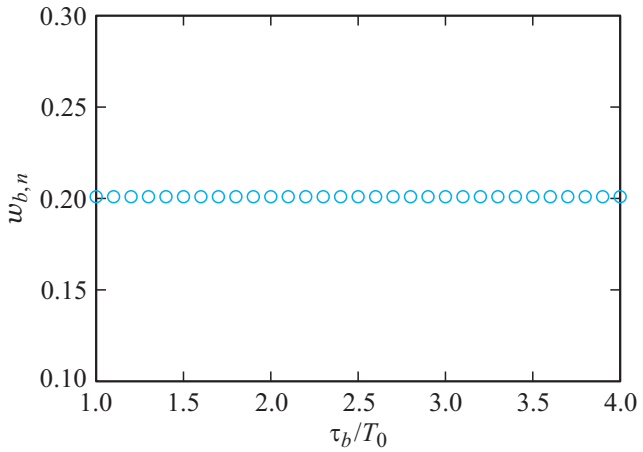
where $S_E = \int E(t)dt$ is the electric area of unipolar pulse (1) and $\Theta \equiv \frac{d_{1n}^2}{\hbar^2}\int_{-\infty}^{\infty}\varepsilon(t)dt = \frac{d_{1n}^2}{\hbar^2}E_{b,0}\tau\sqrt{\pi}$ — is the envelope area of resonant bipolar pulse (2).

In simplified form,

$$E_{u,0}^2\tau_u^2 = 0.25E_{b,0}^2\tau_b^2. \tag{8}$$

It follows from these relations that the electric area squared of a unipolar pulse, which is shorter than the transition period, should be equal (except for constant factor) to the envelope area squared of a resonant multicycle pulse if the impact of these pulses on a quantum transition in a medium is to be the same.

The results of numerical calculations performed using general formula (4) verify the above assertion. In calculations, we set specific amplitude and duration values for a unipolar pulse, $E_{0u} = 8\cdot 10^5$ V/cm, $\tau_u = 10$ fs. The frequency of the resonant transition in a medium is assumed to be $\omega_0/2\pi = 1$ THz (this corresponds to vibrational




Population $w_{b,n}$ as a function of duration $\tau_b$ of a bipolar pulse (in units of transition period $T_0 = 2\pi/\omega_0$).

transitions in a number of molecules), and the transition dipole moment is $d_{1n} = 10$ D. Population $w_{b,n}$ of a bound state is calculated as a function of duration $\tau_b$ of a bipolar pulse. Amplitude $E_{b,0}$ of the bipolar pulse is determined using condition (8) at each value of $\tau_b$.

The results of numerical calculations are shown in the figure. It can be seen that the level population is independent of the bipolar pulse duration if condition (8) is satisfied. This population value agrees with the one calculated for a unipolar pulse with the indicated parameters.

## Discussion. Conclusions

Thus, simple relation (8) for comparing the impact of unipolar half-cycle pulses and bipolar multicycle pulses on populations of levels of a medium was derived.

Relation (8) yields a very important result. It indicates that in order to compare the impact of unipolar half-cycle pulses (shorter than the transition period in a medium) and long resonant multicycle pulses on the populations of quantum transitions in a medium, one should compare the electric area of ??a unipolar pulse and the envelope area of a bipolar one instead of the energies of these pulses.

This relation also has an intuitive interpretation. A resonant multicycle pulse may be presented as a sequence of unipolar bumps (half-waves) with the distance between them being equal to the period of the considered quantum transition. Each half-wave then transfers a momentum, which is specified by the electric area of this half-wave, to the system. The envelope area squared is approximately equal to a square of the sum of electric areas of unipolar components of a multicycle pulse. Therefore, a trivial result follows from (8): one half-wave of the unipolar pulse field exerts the same influence on a system as a sequence of half-waves (components of a multicycle pulse) if the electric area of this half-wave is equal to the area of the sequence of half-waves of a bipolar pulse (i.e., the area of its envelope).

The integral of envelope area at the right-hand side of (8) has the meaning of Rabi frequency of a pulse [21]. It follows that a short unipolar pulse and a long bipolar pulse exert the same influence on a system if the Rabi frequency of the multicycle pulse matches its „counterpart“ for the half-cycle pulse.

We note in conclusion that, according to (7), a unipolar pulse carrier more energy than a bipolar pulse if their electric area and envelope area, respectively, are equal.

However, when used to excite transitions in quantum systems, unipolar pulses also have an advantage over resonant bipolar pulses. Unipolar pulses do not feature a carrier frequency and are „multi-purpose“in this respect. They do not require adjusting the carrier frequency to the object (atom, molecule, etc.) resonance. They may excite any quantum object efficiently within a much shorter time interval.

The latter feature is especially important in applications related to ultrafast control over the dynamics of wave packets. Specifically, this allows, e.g., for creation and ultrafast control of population density gratings in a resonant medium [6,27], holographic recording with an ultrahigh temporal resolution [20], etc.

### Funding

This study was supported by grant 21-72-10028 from the Russian Science Foundation.

### Conflict of interest

The authors declare that they have no conflict of interest.

## References

[1] F. Krausz, M. Ivanov. Rev. Mod. Phys., **81**, 163 (2009).
[2] F. Calegari, G. Sansone, S. Stagira, C. Vozzi, M. Nisoli. J. Physics B: Atomic, Molecular and Optical Physics, **49**, 062001 (2016).
[3] J. Biegert, F. Calegari, N. Dudovich, F. Quéré, M. Vrakking. J. Physics B: Atomic, Molecular and Optical Physics, **54**, 070201 (2021).
[4] N.N. Rosanov, R.M. Arkhipov, M.V. Arkhipov. Phys. Usp., **61**, 1227 (2018).
[5] R.M. Arkhipov, M.V. Arkhipov, N.N. Rosanov. Quantum Electronics, **50**, 801 (2020).
[6] R.M. Arkhipov, JETP Lett., **113**, 611 (2021).
[7] R.M. Arkhipov, M.V. Arkhipov, A. Pakhomov, I. Babushkin, N. Rosanov. Las. Phys. Lett., **19** (4), 043001 (2022).
[8] M.T. Hassan, T.T. Luu, A. Moulet, O. Raskazovskaya, P. Zhokhov, M. Garg, N. Karpowicz, A.M. Zheltikov, V. Pervak, F. Krausz, E. Goulielmakis. Nature, **530**, 66 (2016).
[9] H.-C. Wu, J. Meyer-ter-Vehn. Nat. Photonics, **6**, 304 (2012).
[10] J. Xu, B. Shen, X. Zhang, Y. Shi, L. Ji, L. Zhang, T. Xu, W. Wang, X. Zhao, Z. Xu. Sci. Rep., **8**, 2669 (2018).
[11] M.I. Bakunov, A.V. Maslov, M.V. Tsarev. Phys. Rev. A, **5**, 063817 (2017).
[12] A.V. Bogatskaya, E.A.Volkova, A.M. Popov. Phys. Rev. E, **104**, 025202 (2021).

[13] Y. Shou, R. Hu, Z. Gong, J. Yu, J. Chen, G. Mourou, X. Yan, W. Ma. New J. Phys., **23**, 053003 (2021).
[14] S.V. Sazonov, N.V. Ustinov. JETP Lett. **114**, 380 (2021).
[15] S.V. Sazonov. Laser Phys. Lett., **18** (10), 105401 (2021).
[16] R.M. Arkhipov, M.V. Arkhipov, I. Babushkin, A. Demircan, U. Morgner, N.N. Rosanov. Opt. Lett., **44**, 1202 (2019).
[17] R. Arkhipov, A. Pakhomov, M. Arkhipov, A. Demircan, U. Morgner, N. Rosanov. Opt. Express, **28**, 17020 (2020).
[18] I.A. Aleksandrov, D.A. Tumakov, A. Kudlis, V.M. Shabaev, N.N. Rosanov. Phys. Rev. A, **102**, 0231020 (2020).
[19] N.N. Rosanov N.V. Vysotina. JETP, **130**, 52 (2020).
[20] R.M. Arkhipov, M.V. Arkhipov, N.N. Rosanov. JETP. Lett., **111**, 484 (2020).
[21] L. Allen, J.H. Eberly. *Optical resonance and two-level atoms* (Wiley, N.Y., 1975).
[22] P.G. Kryukov, V.S. Letokhov. Sov. Phys. Usp., **12**, 641 (1970).
[23] R.M. Arkhipov, M.V. Arkhipov, A.V. Pakhomov, N.N. Rosanov. JETP Lett., **114** (3), 129 (2021).
[24] N. Rosanov, D. Tumakov, M. Arkhipov, R. Arkhipov. Phys. Rev. A, **104** (6), 063101 (2021).
[25] R.M. Arkhipov, M.V. Arkhipov, I. Babushkin, A.V. Pakhomov, N.N. Rosanov. JETP Lett., **14** (5), 250 (2021).
[26] S.L. McCall, E.L. Hahn. Phys. Rev., **183** (2), 457 (1969).
[27] R. Arkhipov, A. Pakhomov, M. Arkhipov, I. Babushkin, A. Demircan, U. Morgner, N. Rosanov. Sci. Rep., **11**, 1961 (2021).