

09

Coherent control and creation of population gratings for a pair of attosecond pulses in a resonant medium based on one-dimensional rectangular quantum wells

© R.M. Arkhipov^{1,2}, P.A. Belov³, M.V. Arkhipov¹, A.V. Pakhomov¹, N.N. Rosanov^{1,2}

¹ St. Petersburg State University,
199034 St. Petersburg, Russia

² Ioffe Institute,
194021 St. Petersburg, Russia

³ I.N. Ural'tsev Laboratory of Spin Optics, SPSU,
198504 St. Petersburg, Russia

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

Received December 29, 2021

Revised January 21, 2022

Accepted January 21, 2022

Attosecond pulses can be used to create and control coherence in resonant media, since their duration is shorter than the population relaxation times T_1 and medium polarization T_2 . Previously, the possibility of creating and ultrafast control of electromagnetically induced gratings (EMIG) of atomic populations in a resonant medium was shown using a sequence of extremely short light pulses, when the pulses coherently interact with the medium and do not simultaneously overlap in the medium. These studies were carried out in various approximations, when a finite number of energy levels of the medium is taken into account, or when the pulse amplitude is small. In this paper, based on a direct numerical solution of the time dependent Schrödinger equation without the indicated approximations, we study the possibility of ultrafast coherent control of populations and the creation of an EMIG by a pair of attosecond pulses in a multilevel resonant medium with a low density of particles. The medium is modeled using a one-dimensional rectangular potential well with infinitely high walls. The studies performed show the possibility of ultrafast coherent control of the properties of resonant media based on quantum wells using attosecond pulses.

Keywords: electromagnetically induced gratings, coherent interaction, extremely short pulses, unipolar pulses, attosecond pulses, medium coherence.

DOI: 10.21883/EOS.2022.06.54715.3098-21

Introduction

In recent years attosecond electromagnetic pulses are obtained experimentally [1]. These pulses are actively used to study the dynamics of wave packets in atoms, molecules and solid bodies [2–5]. Recently the interaction of ultra short pulses with different nanostructures [6,7], in particular, with metal nanostructures [8,9], is actively investigated. The interest in recent studies is related to the possibility to convert ultrafast optical signals into low-frequency signals due to creation of the electron current under the interaction of the initial pulse with a nanoparticle.

The limit for reduction of light pulse length is obtaining of already unipolar half-cycle pulse containing a field half-wave of single polarity and having a nonzero electric pulse area (see review in [10] and cited literature). These pulses can faster and in a more effective manner control the properties of wave packets as compared with normal bipolar multi-cycle pulses having a nonzero electric pulse area [11–13].

Length of femtosecond and attosecond pulses can be shorter than relaxation times T_1 and T_2 , therefore coherent interaction of these pulses with the medium is possible. Results of recent experiments show the possibility to

induce coherence and to control it in molecules using such pulses [14,15]. In case of coherent excitation of a resonant medium by a short pulse, it excites coherence of the medium and a free induction decay arises [16]. XUV-attosecond pulses make it possible to create and control the free induction decay [17,18].

The coherence in resonant medium can also be used to create periodical electromagnetically induced gratings (EMIG) of atom populations using ultrashort [19] and extremely short pulses [20], not occurring one-time in the medium. Usually, EMIGs are resulted from interference of a pair of long monochromatic laser beams overlapping in the medium [21–25]. EMIGs created in this manner are actively studied in recent years and have different applications in optics and spectroscopy.

However, creating an interference pattern and, as a consequence, inducing EMIGs using extremely short pulses seems impossible due to their short length, which can be of an order of field cycle or less. However, this is not true. As shown before, creating EMIG without one-time overlapping of pulses in the medium is possible at coherent interaction of pulses with the medium [26–31].

In this case EMIGs are created due to interference of the medium polarization wave (induced coherence), that has been created by the first pulse, with the second pulse that propagates in the opposite direction to the first pulse and does not meet it one-time in the medium. The possibility of EMIG creation in these conditions using a train of one-cycle and half-cycle attosecond and femtosecond pulses is actively studied in recent years.

In the case of coherent excitation in a resonant medium by a pair of extremely short pulses, the first pulse induces coherence in the medium, while the second pulse, acting with a certain delay in relation to the first pulse, allows controlling it, which makes it possible, for example, to affect selectively the populations of states, create an inverse population, etc. [20,28–31]. The latest results of investigations in the field of EMIG creation and coherence control using extremely short pulses are described in detail in the review [20] and cited literature.

However, in previous studies [20,26–31] the possibility was investigated to create and control EMIG and coherent excitation by a train of extremely short pulses, when the medium was modelled in a low-level approximation. The possibility to create EMIGs in multilevel media was also investigated using an approximated solution to the Schrödinger equation in the weak field approximation [30,31].

For a more accurate analysis of the possibility of ultrafast control of populations and EMIG creation in a resonant medium, it is necessary to solve the time dependent Schrödinger equation for wave function without the above-mentioned approximations.

In this study, on the basis of numerical solution to the time dependent Schrödinger equation, the possibility is theoretically investigated to create population gratings in a multilevel medium with low density of particles, as well as the possibility of ultrafast coherent control of bonded states populations of the medium using half-cycle attosecond pulses. We consider the simplest case when the medium is modelled using one-dimensional model of rectangular potential well with infinitely high walls.

This medium model and its different variants are extensively used for analysis of interaction of metal nanoparticles with short laser pulses [9], modelling excitons in semiconductor structures, etc. (see [32–34] and cited literature).

Relaxation times in these systems can lie in the femtosecond range. Therefore, superfast coherent control of these systems requires the use of shorter pulses with durations and interpulse delays lying in the attosecond range.

Creating EMIG and control of bound states population in a resonant medium on the basis of one-dimensional rectangular quantum wells

Just as in [20,26–31], we consider a rarefied medium (it allows neglecting the absorption of the excitation pulse

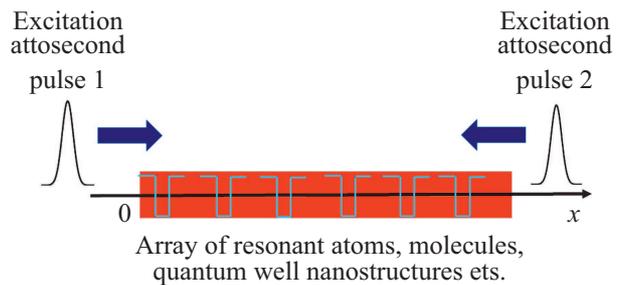


Figure 1. Diagram of population gratings creation in a resonant medium using a pair of attosecond pulses propagating towards each other but without one-time overlapping in the medium. Particles of the medium are shown in blue as one-dimensional quantum wells.

radiation propagating in the medium), which is excited by a pair of attosecond pulses propagating towards each other, see Fig. 1. The first pulse moves from left to right, the second pulse moves from right to left.

In this case, as shown in these articles, the problem of resonant medium excitation by a pair of pulses is reduced to the problem of excitation of an isolated quantum system by a pair of short pulses with variable delay between them, see [20,26–31].

Indeed, in the diagram shown in Fig. 1 the propagating pulse 1 leaves behind a medium coherence oscillating at frequencies of resonant transitions (polarization and non-diagonal elements of the density matrix). And phase of this oscillation will be dependent on the position of resonant dipole, i.e. on the x coordinate.

Then pulse 2, moving in the opposite direction, interacts with the oscillating coherence (polarization) of the medium induced by pulse 1. Since phase of this oscillation depends on the x coordinate, the second pulse can coherently control this oscillation. For example, in some point the polarization oscillation at some resonant transition will be extinguished, while at another point they will be amplified. The situation is similar to the control of a classical pendulum (swing) oscillation by successive pushes.

Thus, the result of the medium interaction with the pair of pulses is dependent to a significant extent on the moment of time of the second pulse arrival to the given point of the medium, i.e. it is defined by the delay between pulses $\Delta \sim \frac{x}{c}$ (c — speed of light). It means, that a sine (or a more complicated, other than harmonic waveform) population grating arises in the medium at each resonant transition of this medium.

And thus, the problem of interaction between an extended rarefied medium and a pair of pulses can be reduced to the problem of interaction between a single quantum system and a pair of extremely short pulses with variable delay [20,26–31].

Let us consider a medium model, which we describe as a one-dimensional potential well with absolutely rigid walls. Such a model, although being the simplest, however is used in physics, for example, in description of metal nanoparticles and semiconductor nanostructures [9,32–35].

Potential energy of the particle in this case can be written as follows:

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

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

where a — width of the well.

Eigen energies of the particle in this case are as follows [36]

$$E_n = \frac{\hbar^2}{2m} \left(\frac{\pi n}{a} \right)^2, \quad n = 1, 2, 3, \dots,$$

while eigen functions are a set of standing waves

$$\varphi_n = \sqrt{\frac{2}{a}} \cos \frac{\pi n}{a} x, \quad n = 1, 3, 5, \dots;$$

$$\varphi_n = \sqrt{\frac{2}{a}} \sin \frac{\pi n}{a} x, \quad n = 2, 4, 6, \dots$$

The system is excited by a pair of attosecond pulses of the following form:

$$E_e(t) = E_0 e^{-\frac{t^2}{\tau^2}} \cos(\omega t + \phi) + E_0 e^{-\frac{(t-\Delta)^2}{\tau^2}} \cos(\omega[t-\Delta] + \phi). \quad (1)$$

Here ω — frequency of pulses, ϕ — carrier envelope phase (CEP), Δ — delay between pulses, t — time, τ — pulse duration.

We consider a coherent interaction between pulses and the medium, i.e. time intervals shorter than relaxation times of the medium — duration of pulses and delay between them should be shorter than these times for the interaction to be coherent. Therefore we neglect the relaxation in the following.

The interaction between a quantum system and a field of external pulses is described by the Schrödinger equation for wave function $\Psi(x, t)$ [36]

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

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

In the assumption of weak amplitude of the external field, population of the k -th bound state w_n of the quantum system after the end of pulses can be calculated in the first order of the perturbation theory and defined by the following relationship [20,30]

$$w_n = \frac{d_{1n}^2}{\hbar^2} E_0^2 \tau^2 \exp \left[-\frac{(\omega_{1n}^2 + \omega^2)}{2} \tau^2 \right] \times [\cosh(\omega_{1n}\omega\tau^2) + \cos 2\phi][1 + \cos(\omega_{1n}\Delta)], \quad (3)$$

where d_{1n} — dipole moment of transition, ω_{1n} — frequency of resonant transition.

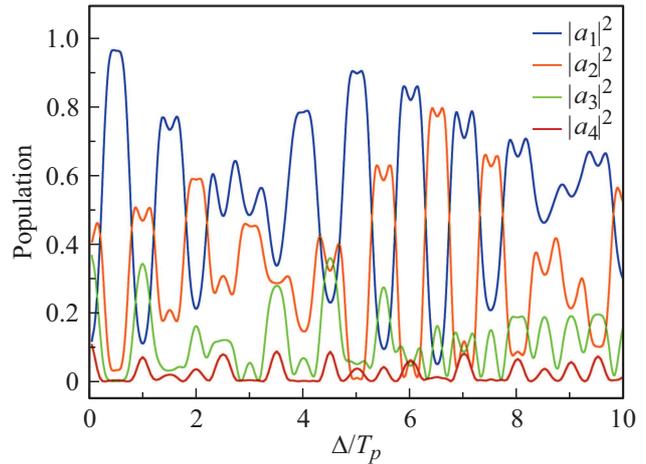


Figure 2. Populations of the first four bound states in a rectangular well with infinitely rigid walls $|a_i|^2$ as a function of delay between attosecond pulses Δ .

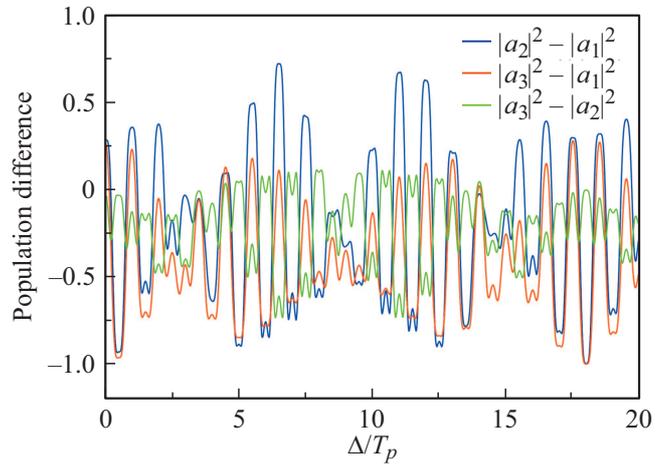


Figure 3. Difference of populations in a rectangular well with infinitely rigid walls for parameters shown in Fig. 1 as a function of delay between attosecond pulses Δ .

This approximated expression predicts the emergence of harmonic population gratings in the medium and the possibility of selective coherent control of bound state populations of isolated nanostructure [20,30,31]. However, in strong fields the form of induced gratings may be other than the simple harmonic form [30].

For a more precise analysis, we solved numerically the time dependent Schrödinger equation (TSE) (2) for a particle in a one-dimensional potential well with absolutely rigid walls, which is exposed to the action of a pair of attosecond pulses of form (1). We used the Crank-Nicolson method [37] to solve the TSE numerically.

Figures 2 and 3 illustrate the population of the first four bound states and the difference of populations, respectively, after the end of pulses as a function of the delay between pulses Δ . Parameters of the calculation were as follows: incident field amplitude $E_0 = 2 \cdot 10^8$ V/cm, well

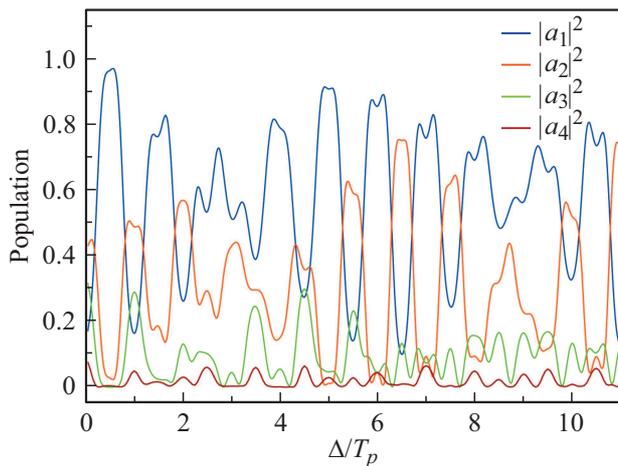


Figure 4. Populations of the first four bound states in a rectangular well with infinitely rigid walls of levels $|a_i|^2$ as a function of delay between attosecond pulses Δ . Excitation pulse length $\tau = 0.3T_p = 134.6$ as. Other parameters are the same as in Fig. 2.

width $a = 1.2$ nm, frequency of pulses $\omega = 14 \cdot 10^{15}$ rad/s (wavelength $\lambda_a = 134.6$ nm, period $T_p = \frac{2\pi}{\omega} = 448.8$ as), phase $\phi = 0$, excitation pulse length $\tau = 0.2T_p = 89.76$ as. Pulses with a length of tens of attoseconds and less can be obtained experimentally [38,39].

It should be noted that with these parameters pulses have a half-cycle unipolar form, their length is less than the field period, therefore for such short pulses values of the parameter ω and wavelength λ_a have conventional meaning.

Figure 4 illustrates the same as Fig. 2 at longer excitation pulses $\tau = 0.3T_p = 134.6$ as.

It can be seen from Figs 2 and 4, that there is a complex spike dependence of populations on the delay between pulses. The form of induced gratings at given parameters differs from the simple harmonic form predicted by formula (3). The behavior of populations, as can be seen from the analysis of Figs 2 and 4, has similar type.

The behavior of population dependence on delay has a complex spike form as well (Fig. 3). These dependencies show the possibility of superfast control of bound states in quantum wells and creation of inverse populations in wells using a pair of attosecond pulses with variable delay.

Previously this possibility was shown in atom-molecular systems [28–31]. The degree of population inversion, as can be seen from Fig. 3, can be controlled by varying the delay between pulses.

Conclusion

In this study, on the basis of numerical solution to the time dependent Schrödinger equation the possibility is shown to create EMIGs and ultrafast control of state populations in rectangular quantum wells with infinitely high walls at coherent excitation of the system by a pair

of half-cycle attosecond pulses. These results show the possibility to induce EMIGs by extremely short pulses in multilevel media taking into account all medium levels, which extends the previous results [20,26–31] obtained, as noted above, in the low-level approximation or in the weak field approximation.

A similar simple model of potential well with infinitely high walls is used to describe different nanostructures. The obtained results indicate the prospects to use the half-cycle attosecond pulses with varied delay for ultrafast control of populations in nanostructures based on quantum wells.

The possibility to create an inverse population can be used to create laser generation in such systems. Also, the inversion value can be controlled by changing the delay between the pulses. Induced EMIGs can be used for holographic recording with ultra-high time resolution using sub-cycle attosecond pulses [40].

Funding

The study was funded by the Russian Science Foundation within the framework of scientific project 21-72-10028.

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] D.M. Villeneuve, P. Hockett, M.J.J. Vrakking, H. Niikura. *Science*, **356**, 1150–1153 (2017).
- [3] E. Goulielmakis, Z. Loh, A. Wirth, R. Santra, N. Rohringer, V.S. Yakovlev, S. Zherebtsov, T. Pfeifer, A.M. Azzeer, M.F. Kling, St.R. Leone, F. Kraus. *Nature*, **466**, 739–743 (2010).
- [4] F. Calegari, G. Sansone, S. Stagira, C. Vozzi, M. Nisoli. *J. Physics B: Atomic, Molecular and Optical Physics*, **49**, 062001 (2016).
- [5] 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).
- [6] M.F. Ciappina, J.A. Pérez-Hernández, A.S. Landsman, W.A. Koell, S. Zherebtsov, B. Förg, J. Schöötz, L. Seiffert, T. Fennel, T. Shaaram, et al. *Report on Progress in Physics*, **80** (5), 054401 (2017).
- [7] L. Seiffert, S. Zherebtsov, M.F. Kling, T. Fennel. *arXiv preprint*, arXiv:2109.02367 (2021).
- [8] C. Karnetzky, P. Zimmermann, C. Trummer, C.D. Sierra, M. Wörle, R. Kienberger, A. Holleitner. *Nat. Commun.*, **9**, 2471 (2018).
- [9] L. Shi, I. Babushkin, A. Husakou, O. Melchert, B. Frank, J. Yi, G. Wetzel, A. Demircan, C. Lienau, H. Giessen, M. Ivanov, U. Morgner, M. Kovacev. *Laser & Photonics Reviews*, **15** (8), 2000475 (2021).
- [10] R.M. Arkipov, M.V. Arkipov, N.N. Rosanov. *Quant. Electron.* **50**(9), 801 (2020).
- [11] R.M. Arkipov, M.V. Arkipov, I. Babushkin, A. Demircan, U. Morgner, N.N. Rosanov. *Opt. Lett.*, **44**, 1202 (2019).

- [12] R.M. Arkhipov, M.V. Arkhipov, I. Babushkin, A.V. Pakhomov, N.N. Rosanov. *JETP Lett.* **114** (5), 250 (2021).
- [13] N. Rosanov, D. Tumakov, M. Arkhipov, R. Arkhipov. *Phys. Rev. A*, **104** (6), 063101 (2021).
- [14] P. Peng, Y. Mi, M. Lytova, M. Britton, X. Ding, A.Yu. Naumov, P.B. Corkum, D.M. Villeneuve. *Nat. Photon.*, (2021). DOI: 10.1038/s41566-021-00907-7
- [15] M. Garg, A. Martin-Jimenez, M. Pizarra, Y. Luo, F. Martin, K. Kern. *Nat. Photon.*, (2021). DOI: 10.1038/s41566-021-00929-1
- [16] L. Allen, J.H. Eberly, *Optical resonance and two-level atoms*. NY: Wiley, 1975.
- [17] S. Bengtsson, E.W. Larsen, D. Kroon, S. Camp, M. Miranda, C.L. Arnold, A. L'Huillier, K.J. Schafer, M.B. Gaarde, L. Rippe, J. Mauritsson, *Nature Photonics*, **11** (4), 252–258 (2017).
- [18] R.M. Arkhipov, M.V. Arkhipov, A.V. Pakhomov, M.O. Zhukova, A.N. Tsyppkin, N.N. Rosanov. *JETP Letters*, **113**, 242 (2021). 10
- [19] E.I. Shtyrkov, V.S. Lobkov, N.G. Yarmukhametov. *JETP Lett.*, **27**, 648 (1978).
- [20] R.M. Arkhipov. *JETP Lett.*, **113** (10), (2021).
- [21] H.J. Eichler, P. Günter, D.W. Pohl. *Laser-Induced Dynamic Gratings*. (Springer-Verlag, Berlin, Heidelberg, NY, Tokyo), (1981).
- [22] H. Zhang, J. Yuan, S. Dong, C. Wu, L. Wang. *Appl. Sci.*, **10**, 5740 (2020).
- [23] Z. Zhang, S. Liang, F. Li, S. Ning, Y. Li, G. Malpuech, Y. Zhang, M. Xiao, D. Solnyshkov. *Optica*, **7**, 455 (2020).
- [24] J. Yuan, S. Dong, H. Zhang, C. Wu, L. Wang, L. Xiao, S. Jia. *Opt. Express*, **29** (2), 2712 (2021).
- [25] T. Jones, W.K. Peters, A. Efimov, D. Yarotski, R. Trebino, P. Bowlan. *Opt. Express*, **29** (8), 11394 (2021).
- [26] R.M. Arkhipov, M.V. Arkhipov, I. Babushkin, A. Demircan, U. Morgner, N.N. Rosanov. *Opt. Lett.*, **41**, 4983 (2016).
- [27] R.M. Arkhipov, M.V. Arkhipov, I. Babushkin, A. Demircan, U. Morgner. *N.N. Rosanov. Scientific Reports*, **7**, 12467 (2017).
- [28] R. Arkhipov, A. Pakhomov, M. Arkhipov, A. Demircan, U. Morgner, N. Rosanov, I. Babushkin. *Optics Express*, **28**, 17020 (2020).
- [29] R.M. Arkhipov. *Opt. Spectrosc.*, **128**, 1865 (2020).
- [30] R. Arkhipov, A. Pakhomov, M. Arkhipov, I. Babushkin, A. Demircan, U. Morgner, N.N. Rosanov. *Scientific Reports*, **11** (1961) (2021).
- [31] R.M. Arkhipov, M.V. Arkhipov, A.V. Pakhomov, Yu.M. Artem'ev, N.N. Rosanov. *Opt. Spectrosc.*, (2021).
- [32] Zh.I. Alferov. *Semiconductors*, **32**, 1 (1998).
- [33] E.L. Ivchenko. *Optical Spectroscopy of Semiconductor Nanostructures*, Alpha Science, 2005.
- [34] P.A. Belov. *Phys. E*, **112**, 96 (2019).
- [35] M. Belloni, R.W. Robinett. *Phys. Rep.*, **540** (2), 25-122 (2014).
- [36] L.D. Landau, E.M. Lifshitz. *Kvantovaya mekhanika. Nerelativistskaya teoriya*. M.: Nauka, 1989. 768 p. (in Russian). (L.D. Landau, E.M. Lifshitz. *Quantum mechanics* (Pergamon, 1974)).
- [37] J. Crank, P. Nicolson. *Mathematical Proceedings of the Cambridge Philosophical Society*, **43** (1), 50–67 (1947). DOI: 10.1017/S0305004100023197
- [38] T. Gaumnitz, A. Jain, Y. Pertot, M. Huppert, I. Jordan, F. Ardana-Lamas, H.J. Wörner. *Opt. Express*, **25**, 27506 (2017).
- [39] Y. Shou, R. Hu, Z. Gong, J. Yu, Jia erh Chen, G. Mourou, X. Yan, W. Ma. *New J. Phys.*, **23**, 053003 (2021).
- [40] R.M. Arkhipov, M.V. Arkhipov, N.N. Rosanov. *JETP. Lett.*, **111**, 484 (2020).