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Population gratings produced in a quantum system by a pair of sub-cycle pulses

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Abstract. **Possible generation of light-induced harmonic population gratings in a quantum system by two half-cycle pulses is studied using the approximate solution of the Schrödinger equation. The case is considered where the duration of pulses is shorter than the resonance transition periods in the system and the pulses do not overlap in the medium. The grating modulation depth is determined by the pulse electric area.**

Keywords: *light-induced gratings, attosecond pulses, sub-cycle pulses, ultrafast optics.*

1. Introduction

Presently, electromagnetic ultrashort (few-cycle) pulses (USPs) have been experimentally obtained in the attosecond range (1 as equals 10^{-18} s) [1–6]. Such pulses are actively used to control the dynamics of wave packet in various substances at times on the order of an oscillation period of a light wave [2–4]. Recently, half-cycle attosecond pulses became available in the optical range [4–6]. Conventionally, half-cycle pulses obtained experimentally comprise a unipolar electromagnetic field 'spike' (half-wave) and a long damping small-intensity tail of opposite polarity [4–6]. Such pulses may have a 'close-to-unity degree of unipolarity', which is defined as [7–9]:

$$\xi = \frac{\left|\int \boldsymbol{E}\,\mathrm{d}t\right|}{\int |\boldsymbol{E}|\,\mathrm{d}t}. \tag{1}$$

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The numerator in (1) is the electric area of the pulse determined by the relationship [10]

$$S_E = \int_{t=-\infty}^{+\infty} \boldsymbol{E}(t)\,\mathrm{d}t.$$

It is an important characteristic in problems of interaction of half-cycle USPs with resonance media. The pulse area is retained during pulse propagation through the macroscopic media with dissipation in the 1D case [10, 11], and substantially affects the interaction of ultrahigh-power USPs with quantum systems [8, 9, 12–16]. Interest in quasi-unipolar pulses is related to the possibility of unidirected action on charged particles, which may be used to efficiently control the dynamics of wave packets in matter and for accelerating charged particles (see, for example, reviews [7–9] and papers [11–15]. Various methods of generating quasi-unipolar half-cycle pulses are described in [7–9, 11] and references therein.

The duration of attosecond pulses may be well below the polarisation relaxation time T_2 and inversion time T_1 in a resonance medium. This determines a possibility of coherent interaction of pulses with matter [17], in which Rabi oscillations may occur – a fast change of atomic inversion at times on the order of a light wave oscillation period [18, 19]. This fact confirms the possibility of superfast control over medium state and poses the task of producing inversion gratings in a resonance medium by a sequence of successive sub-cycle pulses.

Presently, monochromatic laser radiation is used for prepare electromagnetic-induced gratings in a medium due to interference of two or more light beams, which overlap in the medium [20–22]. A periodical distribution of brightness in the beam overlapping domain changes populations of atomic levels, and a population grating arises in the medium. Diffraction of a probe beam on the electromagnetically induced gratings has numerous applications in optics and spectroscopy (see, for example, [20–22]).

However, in interference of the ultrashort pulses with a duration on the order of a light wave oscillation period, the overlapping domain will be rather small, which prevents generating even several interference fringes in the resonance medium. Nevertheless, if short pulses interact with medium coherently, light-induced population gratings may be produced in the medium when the pulses do not meet [23–29]. This fact was discovered already in first experiments on photon echo in [23, 24], in which long-duration (of nanosecond-range) pulses were used (see also review [25] and references therein). Producing gratings by a sequence of pulses not overlapping in the medium is related to the interaction of the incident pulses with travelling waves of medium macroscopic polarisation [23–29]. Indeed, if a short pulse (of duration $\tau_p < T_2$) passes into a medium then a travelling polarisation wave arises, which remains in the medium after the pulse leaves it and persists for the time T_2 in the medium. If another USP passes into the medium in a time interval shorter than T_2 then a harmonic polarisation gratings may arise as a result of interaction with this polarisation wave. By using diffraction of probe radiation on such gratings, the polarisation relaxation time T_2 of such a medium was measured [24, 25]. However, this method for generating gratings by using short pulses non-overlapping in a medium has not been widely applied in optics.

Recently, interest in obtaining attosecond and quasi-unipolar pulses initiated theoretical studies of the possibility of inducing and superfast controlling population difference gratings [26–29] under excitation of a resonance medium by a series of attosecond pulses (both bipolar [26, 27] and unipolar [28, 29]) non-overlapping in the medium. It was shown that gratings can not only be created but also erased and the spatial frequency of the gratings can be multiplicated. This circumstance opens new prospects in superfast optics of attosecond pulses because it allows one to change a state of matter at the attosecond scale. In all previous studies, theoretical analysis was based on numerical and analytical solutions of the Maxwell–Bloch system of equations, and the resonance medium was described in the two-level approximation. The problem of grating generation in multilevel media has not been considered thoroughly.

In the case of sub-cycle attosecond pulses, the pulse duration can be less than periods of resonance transitions in an atom. In this case, the employment of a two-level or low-number-level approximation is not sufficient for correct description of USP interaction with an atomic system. Thus, the question arises as to the possibility of inducing population difference gratings by sub-cycle pulses in real media possessing a large number of energy levels. One of the best descriptions of USP interaction with an atomic system is related to solving the Schrödinger equation for an atomic wavefunction.

The present article is aimed at analysis of the possibilities of a superfast control over a multilevel medium state by a pair of sub-cycle unipolar pulses. The duration of pulses is assumed less than the inverse frequencies of atomic transitions. The possibility of generating light-induced harmonic population difference gratings by a pair of such pulses in the medium is demonstrated.

2. Theoretical consideration

Consider a resonance medium of length L arranged along the z axis (Fig. 1). The concentration of particles we will assume small in order to neglect the profile variation of the pulse as it propagates through the medium. Let pulse 1 pass into the medium at the initial instant at the point $z = 0$ and propagate from left to right in the medium. We assume that as soon as it leaves the medium, pulse 2 enters the medium and propagates from right to left. Obviously, in this statement of the problem, the pulses never meet in the medium. Pulse 1, having left the medium, leaves behind oscillations of medium macroscopic polarisation, which persist for time T_2 [26–29]. We assume that pulse 2 enters the medium in a time interval much shorter than T_2. A result of its interaction with the polarisation wave will depend on a phase of medium polarisation oscillations at each spatial point and on the moment of pulse arrival at the point prescribed. Indeed, depending on the delay between the pulses, pulse 2 may either stop medium polarisation oscillation at the spatial point or, on the contrary, enhance it. Hence, in the case of a medium with a low particle concentration (Fig. 1) where a change in the pulse profile can be neglected, the problem concerning the action of two USPs on an extended medium can be reduced to the problem of unity response of the atom to the pair of USPs acting with a certain relative delay Δ. We are interested in the possibility of a superfast control over the medium state and generating gratings by using ultrashort sub-cycle pulses. We assume that the pulse duration is less than the inverse frequencies of transitions in a system. In this case, the pulse action on the system can be approximately reduced to the action of a delta-shaped pulse. Thus, for simplicity, let us first consider the medium response to the action of two delta-shaped pulses:

$$E(t) = S_{E1}\delta(t) + S_{E2}\delta(t - \Delta), \tag{2}$$

where S_{E1}, S_{E2} are the electric areas for pulses 1 and 2; and Δ is the delay between the pulses. Below, the calculation result is generalised to the case of a pulse with a finite attosecond duration.

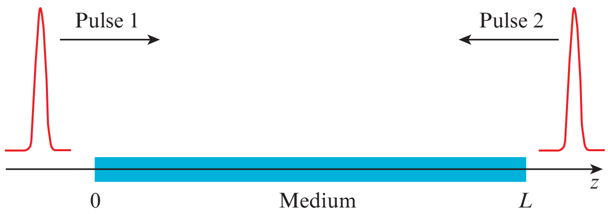

Figure 1. Resonance medium and a pair of sub-cycle pulses propagating towards each other from the opposite directions with a certain delay and not meeting in the medium.

Interaction of a USP with a quantum system is described by the temporal Schrödinger equation for a wavefunction ψ [30]:

$$i\hbar\frac{\partial\psi}{\partial t} = (\hat{H}_0 + V(t))\psi. \tag{3}$$

Here, $\hbar$ is the reduced Plank constant; $\hat{H}_0$ is the eigen Hamiltonian of the system; $V(t) = -dE(t)$ is the potential of system interaction with a radiation pulse, which in the case of not strong field can be considered as a weak perturbation; and d is the atomic dipole momentum. We restrict ourselves to the case of radiation with a fixed linear polarisation when the dipole moment is directed along the incident field. We assume that the perturbation acts for a finite very short time comparable to the period of resonance transitions in the atom or shorter. Let prior to the action the system be in the ground state ψ_0. Due to perturbation, the system may transfer to any other excited state. The incident field is assumed weak. Then, a standard perturbation theory can be applied for solving the wave equation (3) [30]. The probability of system transfer from the ground state of a discrete spectrum to the kth state can be found in the first-order perturbation theory [30]:

$$w_{0k} = \frac{1}{\hbar^2}\left|\int V_{0k}\exp(i\omega_{0k}t)\,dt\right|^2. \tag{4}$$

Here, $V_{0k} = -d_{0k}E(t)$ is the matrix element of the perturbation operator; and d_{0k} and ω_{0k} are the dipole moment and frequency of the resonance transition.

Now we assume that the pulses have equal areas $S_{E1} = S_{E2} = S_E$. By using (2) and (4) and performing integration, one can easily obtain the expression for the transition probability under the action of two USPs:

$$w_{0k} = 2\frac{d_{0k}^2 S_E^2}{\hbar^2}(1 + \cos\omega_{0k}\Delta). \quad (5)$$

From (5), one can see a periodical dependence of the transition probability (population of the kth excited state) on the delay between pulses Δ. Formula (5) explicitly demonstrates the possibility of a superfast control over the medium state by using half-cycle pulses and adjusting the delay between those. One can see that the result of pulse action on the medium is determined by the pulse electric area. This fact once more confirms the possibility of affecting a quantum system by unipolar pulses (in contrast to bipolar pulses with a zero electric area). Note that the possibility of efficiently affecting simplest quantum systems by a single high-amplitude quasi-unipolar USP (with a duration shorter than the inverse transition frequencies in the system) was also studied in the sudden-perturbation approximation in [13, 14].

We now consider generation of population gratings by a pair of sub-cycle pulses. Since in the case of the extended medium shown in Fig. 1 the delay $\Delta(z) \sim z/c$ determines the difference between the instants of pulse 1 and 2 arriving to the medium point with the coordinate z we may assume that formula (5) describes the harmonic population difference (inversion) gratings generated by two USPs in the extended medium. The modulation depth of the gratings is proportional to the pulse electric area and to dipole moment of the corresponding resonance transition. This conclusion was drawn from a solution of the Schrödinger equation, and hence the result obtained illustrates the possibility of generating gratings in multilevel media and generalises previous studies in the two-level approximation.

3. Inducing light-induced gratings by a pair of attosecond pulses

Previously, in view of a short duration of incident pulses we considered delta-shaped pulses to make calculations simpler. However, a similar result can be obtained for pulses with arbitrary profiles. To illustrate the suggested idea, we will consider the action of a pair of pulses of finite attosecond duration on the medium. For clearness, we use incident pulses of rectangular shape with amplitude E_0, duration τ_p, and delay between them Δ. Some methods for obtaining quasi-unipolar pulses of rectangular and triangle shape are described in [31–35] and reviews [7–9]. Using formula (4) one can easily obtain the expression for the transition probability:

$$w_{0k} = 4\frac{d_{0k}^2 E_0^2}{\hbar^2\omega_{0k}^2}(1 - \cos\omega_{0k}\tau_p)\{1 + \cos[\omega_{0k}(\Delta + \tau_p)]\}. \quad (6)$$

This expression shows that the grating modulation depth depends on the ratio of squares of the Rabi frequency of incident pulses $\Omega_R^2 = d_{0k}^2 E_0^2/\hbar^2$ and frequency of considered resonance transition ω_{0k}^2. Note that at $\tau_p \to 0$, by applying the Taylor formula to cosine in the first brackets we obtain $1 - \cos\omega_{0k}\tau_p \approx \omega_{0k}^2\tau_p^2/2$. In view of the expression for a rectangular pulse area $S_E = E_0\tau_p$, formula (6) transfers to formula (5).

We now present numerical estimates. Let the medium be excited by rectangular pulses with τ_p =500 as. We take the transition frequency $\omega_{0k} = 2.4 \times 10^{15}$ rad s^{-1}, which corres-

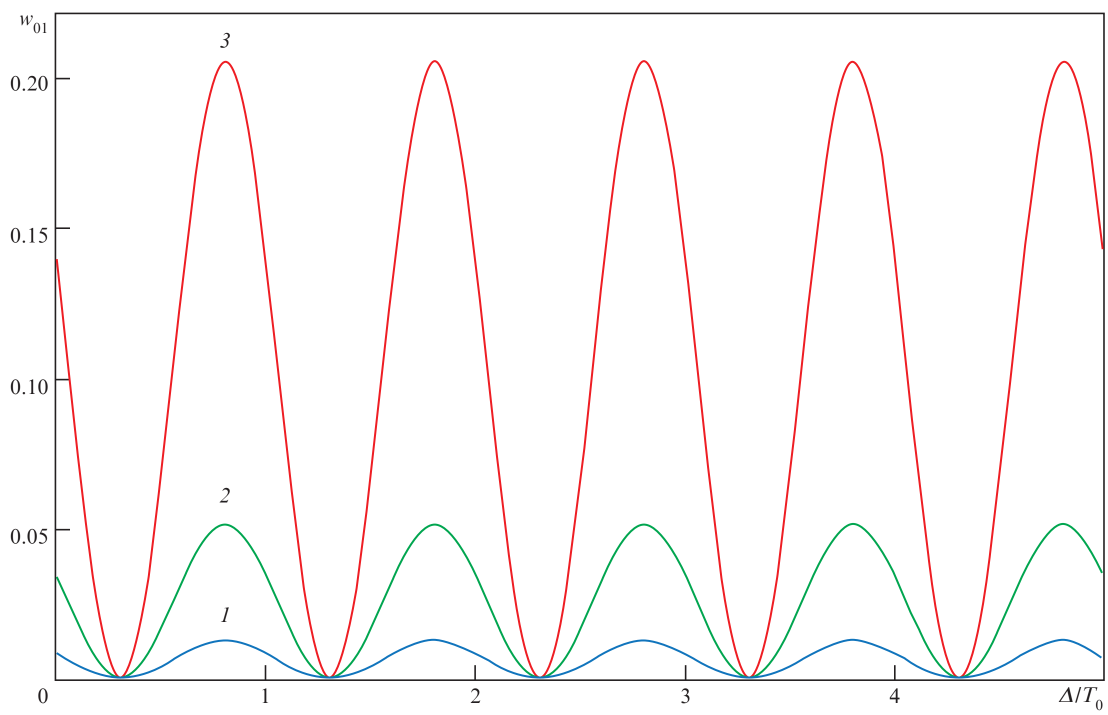

Figure 2. Dependences of the transition probability w_{01} on the delay between pulses Δ for field amplitudes E_0 = (*1*) 0.5×10^7, (*2*) 10^7, and (*3*) 2×10^7 V cm^{-1}; τ_p = 500 as, ω_{0k} =2.4×10^{15} rad s^{-1}, d_{0k} = 7.6 D, $T_0 = 2\pi/\omega_{0k}$ = 2.65 fs is the resonance transition period.

ponds to the resonance transition D_1 in rubidium vapours Rb^{87} with a wavelength of 794.767 nm ($5^2S_{1/2} \rightarrow 5^2P_{1/2}$). The dipole moment of the transition is d_{0k} = 7.6 D [36]. Figure 2 illustrates the dependence of the transition probability on the delay between pulses obtained from formula (6) at three values of the incident field amplitude E_0. The pulse amplitudes and durations are comparable to experimental results given in [4]. One can see that at $E_0 = 10^7$ V cm^{-1} the transition probability is ~ 0.05.

It also follows from (6) that the grating modulation depth depends periodically on the pulse duration τ_p. This makes us assume that in a multilevel medium, contributions of various transitions may be either great or small (depending on the ratio between the pulse duration and transition frequency). This is illustrated in Fig. 3, which presents the dependence of the transition probability on the delay between pulses and pulse duration. The modulation depth periodically depends on the pulse duration. A large modulation depth at a prescribed resonance transition requires pulses of duration $\sim$1 fs. Thus, in a multilevel medium, the contribution from various resonance transitions is determined by the ratio between the pulse duration and frequency of the considered transition.

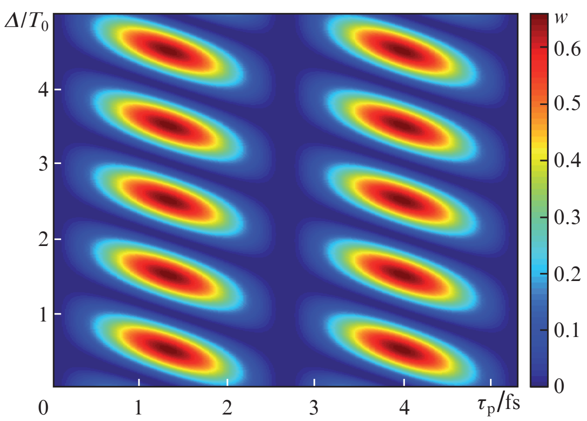

Figure 3. (Colour online) Dependences of the transition probability w on the pulse duration τ_p and delay between the pulses Δ at $E_0 = 2 \times 10^7$ V cm^{-1}, T_0 = 2.65 fs, d_{0k} = 7.6 D.

Concluding, note that formula (6) has been obtained according to the perturbation theory in the case of weak fields; hence, it is valid under the condition on a transition probability $w \ll 1$. This condition is generally satisfied when the pulse Rabi frequency is substantially less than the medium transition frequency ($\Omega_R << \omega_0$); however, it is valid for a greater Rabi frequency if the pulse duration is sufficiently short ($\omega_{0k}\tau_p << 1$).

4. Conclusions

Thus, basing on an approximate solution of the Schrödinger equation we have shown the possibility of a superfast control over parameters of a multilevel resonance medium by using a pair of sub-cycle unipolar pulses. In the case, where the pulse duration is shorter than the inverse frequencies of atomic transitions, the result of their action is determined by the pulse electric area, which again illustrates prospects of using quasi-unipolar pulses for efficient control of the dynamics of wave packets in matter.

It is shown that, similarly to a two-level medium, light-induced harmonic population gratings can be generated when the pulses do not overlap in the medium. The lattice modulation depth depends on the relationship between the pulse Rabi frequency, frequency of the considered transition, and pulse duration. Earlier studies of generating gratings by USPs considered a resonance medium in the two-level approximation only, which is not always applicable in the case of attosecond sub-cycle pulses. The result obtained above generalises the results obtained earlier in a two-level medium to the case of multilevel systems.

A possibility of inducing gratings by using a pair of attosecond pulses in rubidium vapours is studied theoretically, which confirms the possibility of controlling the quantum system state by a sequence of such pulses. Diffraction of probe radiation on such light-induced gratings can be used for measuring a polarisation relaxation time T_2 in various media, designing superfast switches and deflectors for laser radiation [23–29].

In experiments, it is necessary to employ media with a large relaxation time T_2. These are, for example, gases, atomic vapours, and quantum dots at low temperatures [37].

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References

1. Krausz F., Ivanov M. *Rev. Mod. Phys.*, **81**, 163 (2009).
2. Calegari F. et al. *J. Phys. B: Atom., Molec. Opt. Phys.*, **49**, 062001 (2016).
3. Ramasesha K., Leone S.R., Neumark D.M. *Ann. Rev. Phys. Chem.*, **67**, 41 (2016).
4. Hassan M.T., Luu T.T., Moulet A., Raskazovskaya O., et al. *Nature*, **530**, 66 (2016).
5. Wu H.-C., Meyer-ter-Vehn J. *Nat. Photonics*, **6**, 304 (2012).
6. Xu J., Shen B., Zhang X., et al. *Sci. Rep.*, **8**, 2669 (2018).
7. Arkhipov R.M., Pakhomov A.V., Arkhipov M.V., Babushkin I., Tolmachev Yu.A., Rosanov N.N. *JETP Lett.*, **105** (6), 408 (2017) [*Pis'ma Zh. Teor. Eksp. Fiz.*, **105** (6), 388 (2017)].
8. Rosanov N.N., Arkhipov M.V., Arkhipov R.M., Veretenov N.A., Pakhomov A.V., Fedorov S.V. *Opt. Spectrosc.*, **127** (1), 77 (2019) [*Opt. Spektrosk.*, **127** (1), 82 (2019)].
9. Arkhipov R.M., Arkhipov M.V., Shimko A.A., Pakhomov A.V., Rosanov N.N. *JETP Lett.*, **110** (1), 15 (2019) [*Pis'ma Zh. Teor. Eksp. Fiz.*, **110** (1), 9 (2019)].
10. Rosanov N.N. *Opt. Spectrosc.*, **107** (5), 721 (2009) [*Opt. Spektrosk.*, **107** (5), 761 (2009)].
11. Rosanov N.N., Arkhipov R.M., Arkhipov M.V. *Phys. Usp.*, **61** (12), 1227 (2018) [*Usp. Fiz. Nauk*, **188** (12), 1347 (2018)].
12. Dimitrovski D., Solov'ev E.A., Briggs J.S. *Phys. Rev. A*, **72**, 043411 (2005).
13. Rosanov N.N. *Opt. Spectrosc.*, **124** (1), 72 (2018) [*Opt. Spektrosk.*, **124** (1), 75 (2018)].
14. Arkhipov R.M., Pakhomov A.V., Arkhipov M.V., Babushkin I., Demircan A., Morgner U., Rosanov N.N. *Opt. Lett.*, **44** (5), 1202 (2019).
15. Rosanov N.N. *Opt. Spectrosc.*, **125** (6), 1012 (2018) [*Opt. Spektrosk.*, **125** (6), 818 (2018)].
16. Rosanov N.N. *Opt. Spectrosc.*, **126** (2), 140 (2019) [*Opt. Spektrosk.*, **126** (2), 211 (2019)].
17. Allen L., Eberly J. *Optical Resonance and Two-Level Atoms* (New York: Wiley, 1975; Moscow: Mir, 1978).
18. Hughes S. *Phys. Rev. Lett.*, **81**, 3363 (1998).
19. Mücke O.D., Tritschler T., Wegener M., Morgner U., Kärtner F.X. *Phys. Rev. Lett.*, **87**, 057401 (2001).
20. Eichler H.J., Günter E., Pohl D.W. *Laser-Induced Dynamic Gratings* (Berlin–Heidelberg–New York–Tokyo: Springer-Verlag, 1981).
21. Mitsunaga M., Imoto N. *Phys. Rev. A*, **59**, 4773 (1999).

22. Scholes G.D., Kim J., Wong C.Y. *Phys. Rev. B*, **73**, 195325 (2006).
23. Abella I.D., Kurnit N.A., Hartmann S.R. *Phys. Rev.*, **141**, 391 (1966).
24. Shtyrkov E.I., Lobkov V.S., Yarmukhametov N.G. *JETP Lett.*, **27** (12), 648 (1978) [*Pis'ma Zh. Eksp. Teor. Fiz.*, **27** (12), 685 (1978)].
25. Shtyrkov E.I. *Opt. Spectrosc.*, **114** (1), 96 (2013) [*Opt. Spektrosk.*, **114** (1), 105 (2013)].
26. Arkhipov R.M., Arkhipov M.V., Babushkin I.V., Demircan A., Morgner U., Rosanov N.N. *Opt. Lett.*, **41**, 4983 (2016).
27. Arkhipov R.M., Pakhomov A.V., Arkhipov M.V., Babushkin I., Demircan A., Morgner U., Rosanov N.N. *Sci. Rep.*, **7** (1), 12467 (2017).
28. Arkhipov R.M., Arkhipov M.V., Pakhomov A.V., Babushkin I., Rosanov N.N. *Quantum Electron.*, **47** (7), 589 (2017) [*Kvantovaya Electron.*, **47** (7), 589 (2017)].
29. Arkhipov R.M., Arkhipov M.V., Pakhomov A.V., Babushkin I., Rosanov N.N. *Las. Phys. Lett.*, **14** (9), 1 (2017).
30. Landau L.D., Lifshits E.M. *Quantum Mechanics. Non-Relativistic Theory* (London: Pergamon Press, 1958; Moscow: Nauka, 1989).
31. Arkhipov R.M., Arkhipov M.V., Belov P.A., Tolmachev Yu.A., Babushkin I. *Las. Phys. Lett.*, **13**, 046001 (2016).
32. Arkhipov R.M., Pakhomov A.V., Babushkin I.V., Arkhipov M.V., Tolmachev Yu.A., Rosanov N.N. *J. Opt. Soc. Am. B*, **33**, 2518 (2016).
33. Pakhomov A.V., Arkhipov R.M., Babushkin I.V., Arkhipov M.V., Tolmachev Yu.A., Rosanov N.N. *Phys. Rev. A*, **95**, 013804 (2017).
34. Pakhomov A.V., Arkhipov R.M., Arkhipov M.V., Demircan A., Morgner U., Rosanov N.N. *Sci. Rep.*, **9** (1), 7444 (2019).
35. Sazonov S.V., Ustinov N.V. *Phys. Rev. A*, **98**, 063803 (2018).
36. Steck D.A. http://steck.us/alkalidata (revision 2.1.1) (2009).
37. Bayer M., Forchel A. *Phys. Rev. B*, **65**, 041308 (2002).