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Comparison of the dynamics of population difference gratings created in a two-level and three-level medium by half-cycle light pulses

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Based on the numerical solution of the system of Maxwell-Bloch equations, a comparison is made of the dynamics of the population difference gratings and the polarization of the medium, modeled in the two- and three-level approximation. It is shown that gratings also appear in a three-level medium, but their dynamics are not qualitatively different from a two-level medium with the selected model parameters.

Keywords: population difference gratings, half-cycle pulses, attosecond pulses, coherent effects, two-level medium.

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Introduction

The progress in shortening of electromagnetic pulses enabled the formation of radiation structures containing only a few field oscillations of femto- and attosecond duration [1–3]. The generation of such pulses provided an opportunity to study and control the dynamics of electrons in matter [4–6]. Advances in this field have been recognized by the 2023 Nobel Prize in Physics [7].

The shortest duration in a fixed spectral interval is obtained if one removes all but one half-waves of the field from a normal multi-cycle pulse. A pulse obtained in this case is a unipolar half-cycle one [8–11]. One important parameter of half-cycle pulses of this kind is their electric area, which is defined at a given point in space as the integral of field strength $\mathbf{E}$ over time t [12–17]:

$$\mathbf{S}_E = \int \mathbf{E}(t)dt. \quad (1)$$

The interest in generation of such pulses has been on the rise lately. They have the capacity to act quickly and unidirectionally on charges, which makes them promising candidates for various applications. The possibility of application of such pulses for ultrafast control of the properties of various quantum systems (atoms, molecules, and nanostructures) was demonstrated in [18–24]. The latest research results in this field were summarized in reviews [12–17] and papers cited therein.

However, since half-cycle pulses are very short, standard theories of radiation–matter interaction and traditional approximations used for long multi-cycle pulses may be inapplicable in this case. Specifically, Keldysh's theory of photoionization becomes inapplicable when a pulse is shorter than the orbital period of an electron in an atom [25].

The two-level approximation is another traditional approximation used in optics and laser physics for the analysis of resonant coherent interaction with matter [26–29]. It is assumed in it that the medium has only two energy levels, and the remaining levels are disregarded. Half-cycle pulses do not have a carrier frequency and feature a wide spectrum that may cover several resonant transitions of the medium at once. The issue of application of the two-level approximation in studies of coherent propagation of ultrashort pulses (USPs) in resonant media becomes relevant in this case. The process of coherent propagation of such pulses has been studied most extensively in the two-level approximation [30–35].

In the case of coherent interaction between USPs and a medium with its polarization relaxation time T_2 being longer than the pulses and the delays between them, creation and ultrafast control of population difference gratings in this medium are possible [36–38]. Recent and earlier results of studies focused on this issue were reviewed in [39,40] and papers cited therein. When a short pulse emerges from a medium, it leaves behind traveling polarization waves that subsist within an interval corresponding to phase memory time of the medium T_2. If a second pulse enters the medium after the first, a population difference grating may emerge as a result of interaction with this polarization wave [36–38]. In the approximation of a small field amplitude within perturbation theory, the formation of gratings may also be analyzed in terms of „interference“ of pulse areas [41].

A number of early studies of the dynamics of such gratings under the influence of half-cycle pulses were carried out in the two-level approximation [36–38]. The dynamics of gratings in a three-level medium with its parameters corresponding to hydrogen and rubidium atoms was examined in [42,43]. In the present study, the dynamics

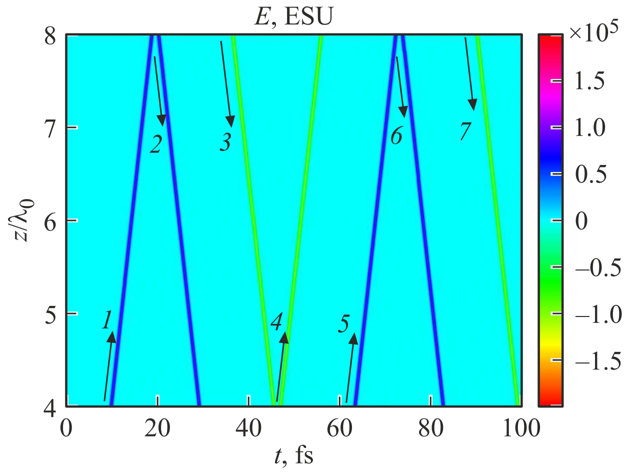

Figure 1. Diagram of propagation of pulses in the medium. Arrows indicate the directions of their propagation on entry into the medium. The pulses are numbered.

of population difference gratings is compared with the dynamics of medium polarization under the influence of half-cycle attosecond pulses in two- and three-level media based on a numerical solution of the Maxwell–Bloch system of equations. The case when pulses do not overlap in the medium is considered.

System under study and formulation of the problem

Analysis was performed in the following way. The dynamics of polarization and populations in two-level and three-level media was calculated numerically. The parameters of the medium and excitation are listed in the table. The relaxation times are on the order of nanoseconds (i.e., are orders of magnitude greater than the length of excitation pulses and intervals between them (femto- and attoseconds)). Therefore, their values are nonessential in the examined scenario and, for simplicity, are taken to be the same for all transitions.

The medium was excited by a sequence of half-cycle Gaussian pulses moving towards each other:

$$E(z=0,t)=E_{01}e^{-\frac{(t-\Delta_1)^2}{\tau^2}}, \tag{2}$$

$$E(z=L,t)=E_{02}e^{-\frac{(t-\Delta_2)^2}{\tau^2}}. \tag{3}$$

Here, $\Delta_1 = 2.5\tau$ and $\Delta_2 = 30.5\tau$ are delays chosen so that pulses do not overlap in the medium.

Zero boundary conditions were set at the boundaries of the integration region, which had length $L = 12\lambda_0$, to produce a pulse sequence. The medium was located in the center of the integration region between points $z_1 = 4\lambda_0$ and $z_2 = 8\lambda_0$. The propagation diagram of such pulses is shown in Fig. 1.

Dynamics of population difference gratings and polarization structures in a two-level medium

The present section is focused on studies into the dynamics of population difference and polarization of a two-level medium. The Maxwell–Bloch system of equations for a two-level medium takes the form [44]

$$\frac{\partial\rho_{12}(z,t)}{\partial t}=-\frac{\rho_{12}(z,t)}{T_2}+i\omega_0\rho_{12}(z,t)-\frac{i}{\hbar}d_{12}E(z,t)n(z,t), \tag{4}$$

$$\frac{\partial n(z,t)}{\partial t}=-\frac{n(z,t)-n_0(z)}{T_1}+\frac{4}{\hbar}d_{12}E(z,t)\,\mathrm{Im}\,\rho_{12}(z,t), \tag{5}$$

$$P(z,t)=2N_0d_{13}\,\mathrm{Re}\,\rho_{12}(z,t), \tag{6}$$

$$\frac{\partial^2E(z,t)}{\partial z^2}-\frac{1}{c^2}\frac{\partial^2E(z,t)}{\partial t^2}=\frac{4\pi}{c^2}\frac{\partial^2P(z,t)}{\partial t^2}. \tag{7}$$

System of equations (4)−(7) contains the following parameters: ρ_{12} is the off-diagonal element of the density matrix of a two-level medium, $n=\rho_{11}-\rho_{22}$ is the population difference (inversion) of the two-level medium, P is the polarization of the medium, t is time, z is the longitudinal coordinate, c is the speed of light in vacuum, d_{12} is the dipole moment of a transition, ω_0 is the transition frequency, N_0 is the concentration of two-level particles, $\hbar$ is the reduced Planck constant, and n_0 is the population difference of the medium in zero electric field ($n_0 = 1$ for an absorbing medium). Wave equation (7) characterizes the dynamics of the electric field strength. Equations (4)−(7) with initial conditions (2), (3) were solved numerically.

Figures 2 and 3 illustrate the typical spatiotemporal dynamics of the population difference and polarization of the medium, respectively. The formation of a harmonic population grating is observed after the passage of the second pulse. Subsequent pulses control the parameters of these gratings: alter their shape and multiply their spatial frequency. Both travelling and standing polarization waves form under the influence of pulses. A similar dynamics pattern has been observed earlier in a two-level medium [36–38].

Dynamics of population difference gratings and polarization structures in a three-level medium

Let us now introduce the third level of a medium. With all the parameters of incident pulses and the medium left as they were in the previous section, we consider the dynamics of polarization and population difference in a three-level medium. The system of equations for the density matrix of

Parameters of excitation pulses and the medium

Pulse amplitude	$E_{01} = E_{02} = 100000\,\text{ESU}$
Length of excitation pulses	$\tau = 390\,\text{as}$
Frequency of transition 12 (transition wavelength)	$\omega_{12} = 2.69 \cdot 10^{15}$ rad/s ($\lambda_{12} = \lambda_0 = 700\,\text{nm}$)
Dipole moment of transition 12	$d_{12} = 20\,\text{D}$
Frequency of transition 13	$\omega_{13} = 4\omega_{12}$
Dipole moment of transition 13	$d_{13} = d_{12}$
Frequency of transition 23 in a hydrogen atom (transition wavelength)	$\omega_{23} = \omega_{13} - \omega_{12}$
Dipole moment of transition 23	$d_{23} = 0$
Concentration of atoms	$N_0 = 10^{13}\,\text{cm}^{-3}$
Relaxation times T_{ik}	$T_{ik} = 1\,\text{ns}$

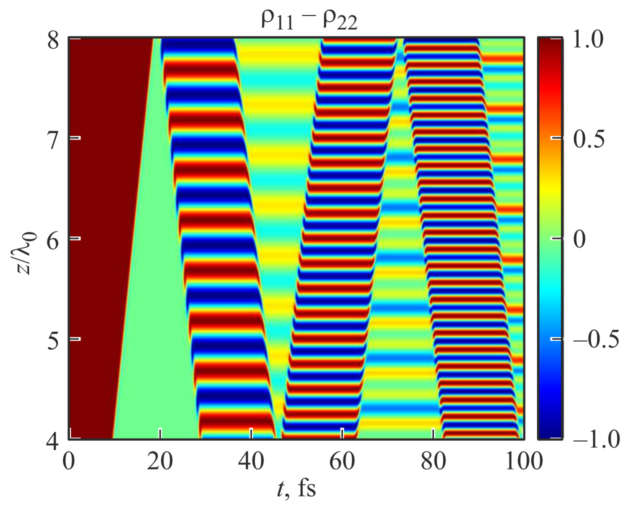

Figure 2. Spatiotemporal dynamics of population difference $n(z,t) = \rho_{11} - \rho_{22}$ of a two-level medium.

a three-level medium has a well-known form [44]

$$\frac{\partial}{\partial t}\rho_{21} = -\rho_{21}/T_{21} - i\omega_{12}\rho_{21} - i\,\frac{d_{12}}{\hbar}\,E(\rho_{22} - \rho_{11}) - i\,\frac{d_{13}}{\hbar}\,E\rho_{23} + i\,\frac{d_{23}}{\hbar}\,E\rho_{31}, \tag{8}$$

$$\frac{\partial}{\partial t}\rho_{32} = -\rho_{32}/T_{32} - i\omega_{23}\rho_{32} - i\,\frac{d_{23}}{\hbar}\,E(\rho_{33} - \rho_{22}) - i\,\frac{d_{12}}{\hbar}\,E\rho_{31} + i\,\frac{d_{13}}{\hbar}\,E\rho_{21}, \tag{9}$$

$$\frac{\partial}{\partial t}\rho_{31} = -\rho_{31}/T_{31} - i\omega_{13}\rho_{31} - i\,\frac{d_{13}}{\hbar}\,E(\rho_{33} - \rho_{11}) - i\,\frac{d_{12}}{\hbar}\,E\rho_{32} + i\,\frac{d_{23}}{\hbar}\,E\rho_{21}, \tag{10}$$

$$\frac{\partial}{\partial t}\rho_{11} = \frac{\rho_{22}}{T_{22}} + \frac{\rho_{33}}{T_{33}} + i\,\frac{d_{12}}{\hbar}\,E(\rho_{21} - \rho_{21}^*) - i\,\frac{d_{13}}{\hbar}\,E(\rho_{13} - \rho_{13}^*), \tag{11}$$

$$\frac{\partial}{\partial t}\rho_{22} = -\frac{\rho_{22}}{T_{22}} - i\,\frac{d_{12}}{\hbar}\,E(\rho_{21} - \rho_{21}^*) - i\,\frac{d_{23}}{\hbar}\,E(\rho_{23} - \rho_{23}^*), \tag{12}$$

$$\frac{\partial}{\partial t}\rho_{33} = -\frac{\rho_{33}}{T_{33}} - i\,\frac{d_{13}}{\hbar}\,E(\rho_{13} - \rho_{13}^*) + i\,\frac{d_{23}}{\hbar}\,E(\rho_{23} - \rho_{23}^*), \tag{13}$$

$$P(z,t) = 2N_0 d_{12}\,\mathrm{Re}\,\rho_{12}(z,t) + 2N_0 d_{13}\,\mathrm{Re}\,\rho_{13}(z,t) + 2N_0 d_{23}\,\mathrm{Re}\,\rho_{32}(z,t). \tag{14}$$

Here, ρ_{21}, ρ_{32}, and ρ_{31} are off-diagonal elements of the density matrix; ρ_{11}, ρ_{22}, and ρ_{33} are the populations of the 1st, 2nd, and 3rd states of the medium that specify the dynamics of polarization of the medium; ω_{12}, ω_{23}, and ω_{13} are the frequencies of resonant transitions; and d_{12}, d_{13}, and d_{23} are their dipole moments. The equations also contain relaxation terms T_{ik}.

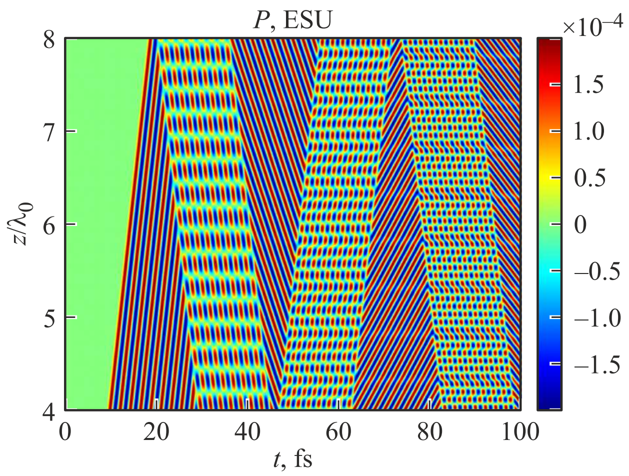

Figure 3. Spatiotemporal dynamics of polarization $P(z,t)$ of a two-level medium.

Figures 4–7 illustrate the spatiotemporal dynamics of polarization and population differences at different resonant transitions for a three-level medium. It is evident that this dynamics matches qualitatively the one observed in a two-level medium (see Figs. 2,3). Thus, despite popular belief, the introduction of an additional level does not suppress the effect of emergence of population difference gratings.

Therefore, the results obtained within the two-level model remain valid when additional levels are introduced. This result speaks in favor of the two-level approximation and agrees qualitatively with the data from other studies into coherent interaction of ultrashort pulses with resonant media [45,46], where the formation of dissipative solitons of self-induced transparency, which was initially predicted in two-level media, was demonstrated to be preserved in a three-level medium.

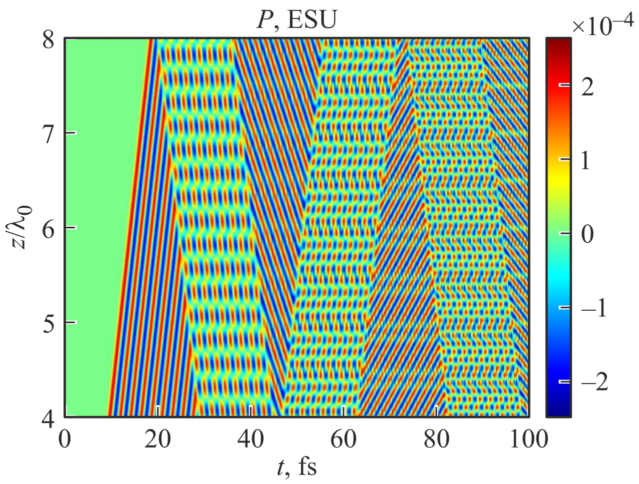

Figure 4. Spatiotemporal dynamics of polarization $P(z,t)$ of a three-level medium.

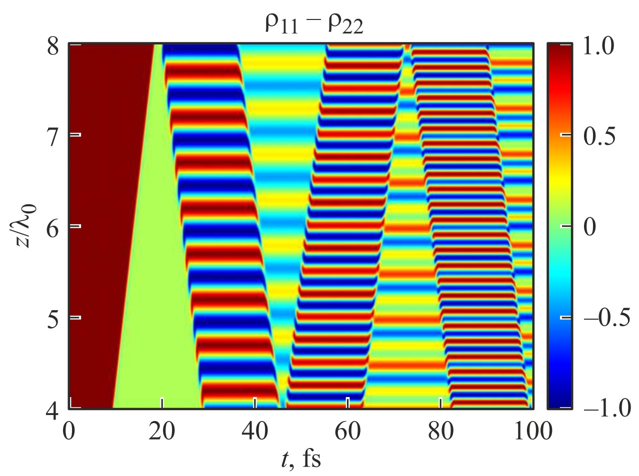

Figure 5. Spatiotemporal dynamics of population difference $\rho_{11} - \rho_{22}$ of a three-level medium.

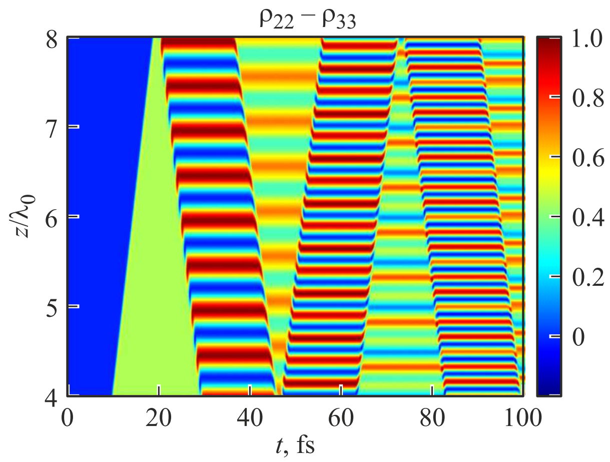

Figure 6. Spatiotemporal dynamics of population difference $\rho_{22} - \rho_{33}$ of a three-level medium.

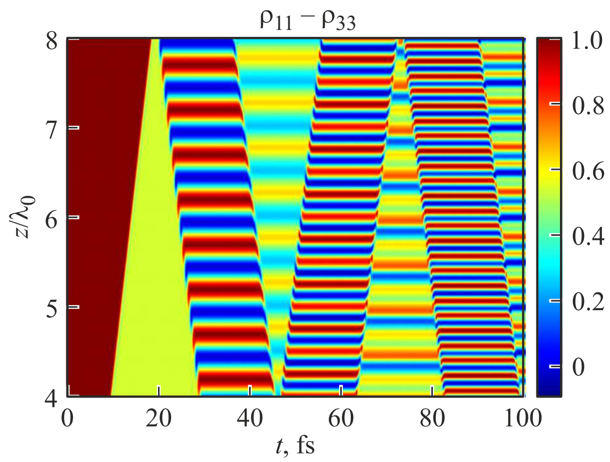

Figure 7. Spatiotemporal dynamics of population difference $\rho_{11} - \rho_{33}$ of a three-level medium.

Conclusion

The dynamics of population difference gratings was compared (by solving the Maxwell–Bloch system of equations numerically) with the behavior of polarization of two-level and three-level resonant media excited by a sequence of half-cycle pulses that do not overlap in the medium. It was demonstrated that the dynamics of population difference gratings and polarization of a three-level medium is qualitatively similar to that of a two-level medium (at the chosen calculation parameters).

The results presented above for a three-level medium verify the validity of earlier data obtained with the use of the two-level medium model, thus expanding the range of its applicability. Therefore, the formation of population difference gratings by USPs, which was first predicted in a two-level medium, is apparently a common feature of multi-level media. This statement is supported by the experimental observation of carrier-wave Rabi flopping, which was also predicted initially in a two-level medium [31], under the influence of a single-cycle pulse in semiconductors [47]. Note that the above example of analogy in system dynamics in two-level and multi-level approximations is by no means the only one; the results of other studies also support this conclusion [48–51].

The following simple physical considerations explain why gratings are preserved in a multi-level medium. A short half-cycle pulse passing through the medium leaves it in the so-called superposition state. Off-diagonal elements of the density matrix (coherence of the medium) then oscillate at each resonant transition of the medium at the transition frequency within polarization relaxation interval T_2. Regardless of the actual number of levels of a medium considered in a given problem, these oscillations will always be present at each resonant transition of the medium. Each subsequent pulse will interact with these coherence oscillations, producing population gratings at each resonant transition of the medium. This conclusion is consistent with the numerical solution of the Schrödinger equation obtained with account for ionization of the medium [52].

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Conflict of interest

The authors declare that they have no conflict of interest.

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