



The generalized kinetics model for the description of the photoprocesses induced by ultrashort laser pulses

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Received: 2 March 2023 / Accepted: 24 May 2023 / Published online: 9 June 2023
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Abstract

To explain the dynamics of the population of a quantum system excited by an ultrashort laser pulse, the generalized kinetic model is proposed and tested. It is shown that this model is accurate within the constraints of the applicability of the perturbation theory for any values of the laser pulse's duration and carrier frequency. By comparing the outcomes of our model to the two conventional methods and by precisely resolving the Bloch equations, we can assess the effectiveness of them. It is demonstrated that the usual techniques considerably overestimate the population values of the higher state of the excited quantum system outside the boundaries of their applicability. When using the Bloch equations is problematic or impossible, the suggested model can be utilized to characterize the population kinetics in the general situation.

1 Introduction

There are unique characteristics that distinguish the interaction of ultrashort laser pulses (USLP) with matter from those of lengthy pulses [1]. It was demonstrated, for instance, in [2–4] that the dependency of the probability of a photoprocess on the pulse duration might be non-linear even when the first order of the perturbation theory is appropriate. Additionally, the pulse characteristics (length, carrier frequency, envelope shape) largely dictate how this probability depends on the current time and might take on a linear or oscillatory form [5, 6].

It is required to create theoretical approaches that accurately take into account the features indicated above to characterize photoprocesses in the realm of such short laser pulses, which is related to the development of technology for producing laser pulses with particular parameters [7–9]. It is helpful to create straightforward methods that are universal, physically transparent, and predictive in addition to complex computation techniques like the numerical solution of the temporal Schrödinger equation [10, 11]. Such a strategy

was first presented in [12] and subsequently generalized in [13] to account for the likelihood of photoprocesses caused by USLP in the context of perturbation theory. To calculate the rate of photoprocesses, which is a factor in the kinetic equations for the populations of a quantum system controlled by USLP, one might extend the strategy presented in the mentioned publications.

The purpose of this paper is to propose and verify a simple model for the population kinetics of a quantum system excited by a short laser pulse based on the previously derived expression for time-dependent probability [13]. This model accounts for specifics of ultrashort electromagnetic interaction and can be applied particularly for description of the photoexcitation of spectral line with different types of broadening taking into account the impact of various perturbations of the excited system.

2 Models for photoprocesses rate

Let us consider the interaction of laser pulse with dipole-allowed transition between stationary states of quantum system $|1\rangle \leftrightarrow |2\rangle$ with energies $E_1 < E_2$, matrix element of dipole momentum operator d_0 , and eigenfrequency $\omega_0 = (E_2 - E_1)/\hbar$. For simplicity, we assume that these energy levels are non-degenerate. The kinetic equations for populations $N_{1,2}$ of these levels can be written as follows

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$$\frac{dN_1}{dt} = w(t) [N_2 - N_1] - \frac{N_1 - N_{1l}^e}{T_{1l}} \tag{1a}$$

$$\frac{dN_2}{dt} = -w(t) [N_2 - N_1] - \frac{N_2 - N_{2u}^e}{T_{1u}} \tag{1b}$$

where $w(t)$ is the rate of photoprocesses (absorption and stimulated emission) induced by laser pulse, $T_{1l,u}$ are population relaxation times of lower and upper levels, $N_{1l,u}^e$ are equilibrium populations (without the action of laser pulse). Note that lower level can be both ground or excited.

It is usually assumed that the rate of the photoprocess $w(t)$ is proportional to the square of the electric field strength at the given time moment, namely [14]:

$$w_{st}(t) = \sigma(\omega_c) j_{\text{phot}}(t, \tau) = \sigma(\omega_c) \frac{c E^2(t, \tau)}{4 \pi \hbar \omega_c}, \tag{2}$$

where ω_c is carrier frequency of the pulse, τ is pulse duration, $\sigma(\omega)$ is photoexcitation cross-section of dipole-allowed transition, j_{phot} is photon flux density, $E(t, \tau)$ is the electric field strength in laser pulse, c is light velocity.

We will call the equality (2) the *first standard approach* to describing the population kinetics. Equation (2) may be rewrite as follows

$$w_{1st}(t) = \pi G(\omega_c) \Omega_0^2 \tilde{E}^2(t, \tau), \tag{3}$$

where $\Omega_0 = d_0 E_0 / \hbar$ is Rabi frequency, E_0 and $\tilde{E}(t) = E(t) / E_0$ are amplitude and dimensionless electric field strength of the pulse, $G(\omega)$ is spectral profile of the photoexcitation cross-section.

In the case of monochromatic radiation, photoexcitation rate (3) is constant in time and after averaging over oscillation period: $T = 2 \pi / \omega$ equal to

$$w_{\text{mon}} = \frac{\pi}{2} G(\omega) \Omega_0^2 = \text{const.} \tag{4}$$

Sometimes, instead of (2), another photoprocess rate is used [15], which is determined by Einstein equality

$$w_E(t) = B \rho(t, \omega), \tag{5}$$

where $B = \frac{4 \pi^2}{\hbar^2} d_0^2$ is Einstein coefficient for induced process, $\rho(t, \omega)$ is spectral density of laser pulse energy at given time moment. In terms of the Rabi frequency, formula (5) can be rewritten as

$$w_{2st}(t) = B \frac{\rho(t)}{\Delta \omega_L} = \pi \Omega_0^2 \frac{\tau}{\pi} \tilde{E}^2(t). \tag{6}$$

Here and below, it is assumed that the laser pulse is spectrally limited, so that the width of its spectrum is completely determined by its duration. We will call the use

of the photoprocess rate (6) in the kinetic equations the *second standard approach*.

In our *generalized kinetics model*, the rate of the photoprocess is expressed through dimensionless probability of the process at given time $W(t)$ according to the relation

$$w(t) = \frac{d}{dt} W(t). \tag{7}$$

We have previously derived the expression for the photoexcitation probability [13] which in terms of Rabi frequency has the form

$$W(t, \tau) = \omega_0 \Omega_0^2 \int_0^\infty G(\omega) \frac{\tilde{D}(t, \tau, \omega)}{\omega} d\omega, \tag{8}$$

where

$$\tilde{D}(t, \tau, \omega) = \left| \int_{-\infty}^t dt' \exp(i \omega t') \tilde{E}(t', \tau) \right|^2 \tag{9}$$

is squared modulus of the incomplete Fourier transform of the dimensionless electric field strength in a laser pulse (D-function for short). Taking into account (7) and (8), we have, for the photoprocess rate in our model, the following expression

$$w(t) = \omega_0 \Omega_0^2 \int_0^\infty \frac{G(\omega)}{\omega} \frac{d}{dt} \tilde{D}(t, \tau, \omega) d\omega. \tag{10}$$

Thus, the time dependence of the photoprocess rate is determined by the time derivative of the D-function.

It is simple to numerically solve the system of Eqs. (1) and determine how populations vary on time within the context of various approaches using the expressions (4)–(6) and (10). It is helpful to derive an integral representation of the solution to system (1) in the case of a two-level quantum system. In particular, this representation makes it possible to find analytical limiting cases.

3 Two-level system

In the case of two-level quantum system, the normalization condition $N_1(t) + N_2(t) = 1$ is satisfied, and instead of the system of Eqs. (1), we have the following equation for the population of the upper level under the assumption that $N_2^e = 0$ and $T_{1l} = T_{1u} = T_1$

$$\frac{dN_2}{dt} + \frac{N_2}{T_1} = w(t) [1 - 2N_2]. \tag{11}$$

The solution of Eq. (11) is equal to

$$N_2(t, \tau) = \frac{1}{2} \left\{ 1 - \frac{1}{T_1} \exp \left[-\frac{t}{T_1} - 2W(t, \tau) \right] \int_{-\infty}^t \exp \left[\frac{t'}{T_1} + 2W(t', \tau) \right] dt' \right\}. \tag{12}$$

where $W(t) = \int_{-\infty}^t w(t') dt'$. It should be noted that (12) includes only the dimensionless probability W , and not the photoexcitation rate w .

It is interesting to consider a couple of various limiting cases of the expression (12). Neglecting the upper level relaxation, from equality (12) we have

$$N_2(t, \tau, T_1 \rightarrow \infty) = \frac{1}{2} \{ 1 - \exp[-2W(t, \tau)] \}. \tag{13}$$

Note that within the framework of the validity of perturbation theory (when $W(t, \tau) \ll 1$), it follows from (13) that $N_2(t, \tau) \cong W(t, \tau)$.

In monochromatic case (4), it follows from formula (12) for laser pulse which turns on at zero time the expression

$$N_2^{\text{mon}}(t) = \theta(t) \frac{w_{\text{mon}} T_1}{1 + 2w_{\text{mon}} T_1} \left(1 - \exp \left(-\frac{t}{T_1} - 2w_{\text{mon}} t \right) \right), \tag{14}$$

where $\theta(t)$ is Heaviside step-function. Thus, for $t \gg T_1 / (1 + w_{\text{mon}} T_1)$ one has from (14) well-known relation [14]

$$N_2^{\text{mon}}(t \gg T_1) = \frac{w_{\text{mon}} T_1}{1 + 2w_{\text{mon}} T_1} \tag{15}$$

which describes the saturation effect if $w_{\text{mon}} T_1 \gg 1$.

The system of Eqs. (1) also provides a simple analytical solution if both levels are excited with the same population relaxation times $T_{1l} = T_{1u} = T_1$ and zero equilibrium populations $N_{1l,u}^e = 0$. Then we have

$$N_2(t, \tau) = \frac{1}{2} \exp(-t/T_1) \{ 1 - \exp[-2W(t, \tau)] \}. \tag{16}$$

4 Verification of the model

Let us verify our model by comparing it with the exact solution of the Bloch equations for a two-level system. Then we can use expression (12) for the population of the upper level and (9) for excitation probability. We also use expressions (3) and (6) for the photoprocess rate in the framework of standard approaches for comparison with the exact solution and the result of our generalized kinetics model.

The findings of the integral representation (12) and the solution of the system of kinetic Eqs. (1) for a two-level system are identical, according to numerical analysis.

The equations for the optical Bloch vector \mathbf{R} in terms of Rabi frequency have the form [16]

$$\frac{dR_1}{dt} = \omega_0 R_2 - \frac{R_1}{T_2}, \tag{17a}$$

$$\frac{dR_2}{dt} = -\omega_0 R_1 - \frac{R_2}{T_2} + 2\Omega_0 \tilde{E}(t) R_3, \tag{17b}$$

$$\frac{dR_3}{dt} = -2\Omega_0 \tilde{E}(t) R_2 + \frac{R_3^e - R_3}{T_1}. \tag{17c}$$

Here, T_2 is phase relaxation time, R_3^e is equilibrium value of the third component of the Bloch vector. The third component of the optical Bloch vector is related to the population inversion and is equal by definition to

$$R_3 = -N \equiv N_1 - N_2. \tag{18}$$

Taking into account the normalization condition, we have for the population of the upper level

$$N_2 = \frac{1 - R_3}{2}. \tag{19}$$

Substituting the solution of the system of Eqs. (17) for the third component of the optical Bloch vector into formula (19), we find the exact value for the population of the upper level of a two-level system excited by laser pulse.

5 Comparison of approaches

The homogeneous broadening of the spectral line is consistent with the Bloch Eqs. (17). We must, thus, employ the Lorentzian for the spectral profile of cross-section to compare the results of our model with those of traditional methodologies.

$$G(\omega) = \frac{1}{\pi T_2} \frac{1}{(\omega - \omega_0)^2 + T_2^{-2}}. \tag{20}$$

For calculation simplicity, consider excitation by an exponential pulse of the form

$$E_{EP}(t, \tau) = \theta(t) E_0 \exp(-t/\tau) \cos(\omega_c t). \tag{21}$$

Then for multi-cycle pulse ($\omega_c \tau \gg 1$), we have from definition (9), the following expression for D-function

$$\tilde{D}_{EP}(t, \tau, \omega) \cong \frac{1}{4} \theta(t) \tau^2 \frac{1 + \exp(-2t/\tau) - 2 \exp(-t/\tau) \cos[(\omega - \omega_c) t]}{1 + \tau^2(\omega - \omega_c)^2}. \tag{22}$$

In the context of the suggested model of generalized kinetics, we derive the excitation probability by substituting this function into formula (8). This excitation probability is then used to determine the population of the upper level (12).

Within framework of standard approaches (3) and (6), we have the following formulas for excitation probability

$$W_{st1}(t, \tau) = \int_{-\infty}^t w_{st}(t') dt' = \pi G(\omega_c) \Omega_0^2 \int_{-\infty}^t \tilde{E}^2(t', \tau) dt' \tag{23}$$

$$W_{st2}(t, \tau) = \int_{-\infty}^t w_E(t') dt' = \tau \Omega_0^2 \int_{-\infty}^t \tilde{E}^2(t', \tau) dt' \tag{24}$$

These probabilities should be substituted in the expression (12) to obtain population of an upper energy state of quantum system in the framework of standard approaches.

For exponential pulse (21), we have

$$\int_{-\infty}^t \tilde{E}^2(t', \tau) dt' = \theta(t) \frac{\tau}{4} \left\{ 1 - \exp\left(-\frac{2t}{\tau}\right) + \frac{1 + \exp(-2t/\tau) [\omega_c \tau \sin(2\omega_c t) - \cos(2\omega_c t)]}{1 + \omega_c^2 \tau^2} \right\} \cong (\omega_c \tau \gg 1) \cong \theta(t) \frac{\tau}{4} \left\{ 1 - \exp\left(-\frac{2t}{\tau}\right) \right\}. \tag{25a}$$

It is crucial to note that, in contrast to probability (23) which comprises the spectrum profile, probability (24) only faintly depends on the carrier frequency of the multi-cycle laser pulse.

Note that at resonance $\omega_c = \omega_0$ for the Lorentzian spectral profile (20), equality (23) gives

$$W_{st1}(t, \tau) = T_2 \Omega_0^2 \int_{-\infty}^t \tilde{E}^2(t', \tau) dt'. \tag{25b}$$

Thus, in this case, the probabilities (23) and (24) differ by the factor T_2/τ .

Comparing the photoprocess probabilities determined using different techniques in two limiting circumstances, namely the monochromatic and ultrashort limitations, is interesting. Analytically, such a comparison may be created for an exponential laser pulse (21).

In the monochromatic limit $\tau \gg T_2$ and for times $t \geq \tau$, using formulas (8), (9), and (22), we can obtain the following expression for the probability in the framework of our approach [6]:

$$W_{EP}(t \geq \tau, \tau \gg T_2) \approx \frac{\pi}{4} \Omega_0^2 G(\omega_c) \theta(t) \tau (1 - e^{-2t/\tau}). \tag{26}$$

The same expression is obtained in the framework of the first standard approach after substituting the integral (25) (for multi-cycle pulse $\omega_c \tau \gg 1$) into formula (23).

The second standard approach (24) overestimates the probability in this limit by τ/T_2 times.

The opposite situation occurs in the ultrashort limit, when $\tau \ll T_2$. Our approach in this case gives [6]

$$W_{EP}(t, \tau \ll T_2) \cong \frac{1}{4} \frac{\theta(t) \Omega_0^2 \tau^2}{1 + \tau^2(\omega_0 - \omega_c)^2} \times \left\{ (1 - e^{-t/\tau})^2 + 4 e^{-t/\tau} \sin^2\left(\frac{(\omega_0 - \omega_c) t}{2}\right) \right\}. \tag{27}$$

The second standard approach (24), taking into account (25), gives

$$W_{st2}(t) = \frac{1}{4} \theta(t) \Omega_0^2 \tau^2 \left(1 - \exp\left(-\frac{2t}{\tau}\right) \right). \tag{28a}$$

Hence, in the limit of long times, we obtain

$$W_{st2}(t \gg \tau) = \frac{1}{4} \Omega_0^2 \tau^2. \tag{28b}$$

The same asymptotic value for probability is obtained from (27) in the actual frequency range $|\omega_c - \omega_0| \leq T_2^{-1}$. The first standard approach in this case overestimates the probability of a photo process by T_2/τ times.

As a result, the first standard technique produces good results for long laser pulses, but the second standard approach is asymptotically correct for short laser pulses in the long time limit.

6 Numerical results and discussion

Let us compare the generalized kinetics, standard approaches, and the exact solution of the Bloch equations for the following values of parameters $T_2 = 0.1\text{--}0.2$ ps, $T_1 = 1$ ps, $\hbar\omega_0 = 0.155$ eV ($\lambda_0 = 8$ μm). These values are typical for transitions between energy states in quantum cascade lasers [17].

To compare the results of approximative computations with the precise solution of the Bloch equations, we assume that the spectral profile is Lorentzian (20).

The graphs shown in Figs. 1, 2, 3 illustrate how the population of the upper level of a two-level system changes over time in response to exponential pulses of different lengths, depending on the resonant carrier frequency and the above-mentioned relaxation times. The precise solution of the Bloch equations and the computation in the generalized kinetics model are extremely well in agreement for the values of the parameters employed, and this is true for all pulse lengths.

Figure 1 shows that for short pulses ($\tau < T_2$), the second standard approach gives a better result than the first one. In this case, the coincidence with the exact solution occurs for times greater than population relaxation time.

For the case $\tau = T_2$ (Fig. 2), the results of using the first and second standard approaches coincide (for $\omega_c = \omega_0$) and considerably exceed the exact result. For long times, all four approaches in this case give the same result.

Finally, as it can be seen from Fig. 3, in the case of long pulses ($\tau \gg T_2$), the best agreement with the exact solution and generalized kinetics model is given by the first standard approach, which is especially pronounced at

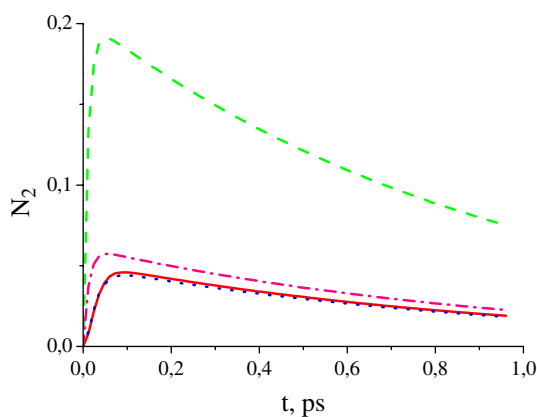


Fig. 1 Population of upper level of two-level system excited by exponential laser pulse as function of time: solid line—solution of Bloch equations, dotted line—generalized kinetics model, dashed line—first standard approach, dash-dotted line—second standard approach. Ultra-short pulse: $\tau = 0.024$ ps, $\omega_c = \omega_0 = 155$ meV, $T_2 = 0.1$ ps, $T_1 = 1$ ps, $\Omega_0 = 14$ meV

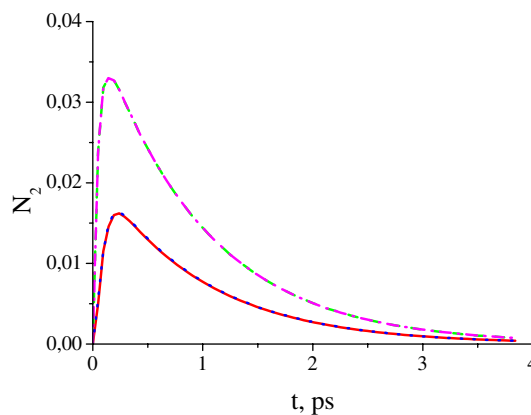


Fig. 2 The same as in Fig. 1 for $\tau = T_2 = 0.1$ ps and $\Omega_0 = 2.72$ meV

times of $t > \tau$. The second standard approach significantly overestimates the exact result, N_2 .

It should be noted that in all three cases considered, the standard approaches significantly overestimate the exact dependences $N_2(t)$ for times shorter than the time at which the maximum population of the upper level is reached. It follows from formulas (23)–(25) that, at short times $t \ll \tau$, the standard approaches give a linear dependence of the population of the upper level on time, while the exact solution is a quadratic function of time both for short pulses and long pulses.

The analysis of the performed calculations, in particular, shows that in the case of short pulses ($\tau < T_2$), the results of exact calculation and of the standard approach coincide at times several times longer than the pulse duration (see Figs. 1, 2). In the case of long pulses ($\tau > T_2$), this coincidence takes place already for times longer than the duration of the exciting pulse itself as one can see from Fig. 3.

Figure 4 displays the temporal dependencies $N_2(t)$ for a large detuning of the laser pulse's carrier frequency from

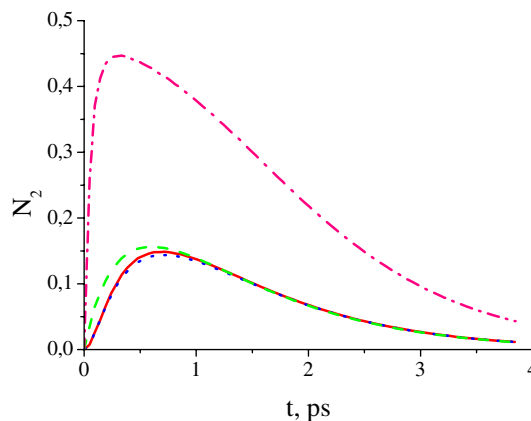


Fig. 3 The same as in Fig. 1 for long pulse case ($\tau \gg T_2$): $\tau = 1$ ps, $\omega_c = \omega_0 = 155$ meV, $\Omega_0 = 2.72$ meV

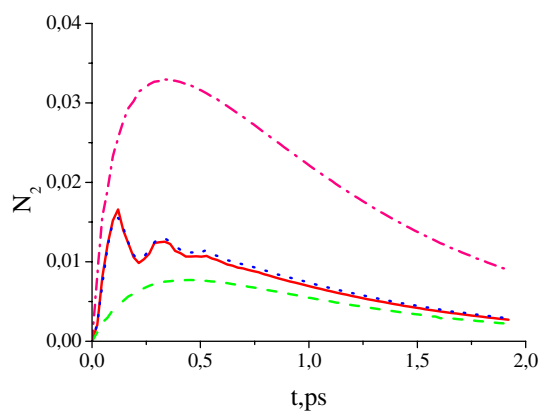


Fig. 4 The same as in Fig. 1 for off-resonance carrier frequency $\omega_c = 136$ meV ($\omega_c \neq \omega_0$), $\tau = 0.5$ ps, $T_2 = 0.2$ ps, the ordinate of the dash-dotted line is reduced by ten times

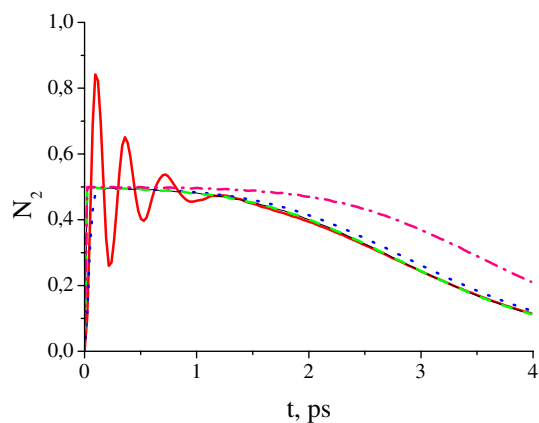


Fig. 5 Saturation effect for long pulse case: $\tau = 1$ ps, $T_2 = 0.2$ ps, $T_1 = 1$ ps, $\Omega_0 = 20.4$ meV

eigenfrequency of quantum system. The exact solution of the Bloch equations and the result of the generalized kinetics model also demonstrate excellent agreement, and in this case, population oscillations in time have been identified, whereas they are not in the data from conventional techniques. These oscillations, although they are not Rabi oscillations, are connected to the contribution to the quantum system's excitation at the different frequency $|\omega_c - \omega_0|$. Additionally, the Bloch equations' solutions include high-frequency oscillations at the sum frequency $|\omega_c + \omega_0|$, which are harder to identify in Fig. 4.

The case of strong saturation is shown in Fig. 5. At the value of large times, the three dependencies practically coincide, except for the one obtained in the framework of the second standard approach, which gives a slightly overestimated result. One can see the Rabi oscillations in the dependence obtained with the exact solution of the

Bloch equations for $t < \tau$. These oscillations decay at times determined by the phase relaxation time T_2 .

7 Conclusion

We developed and tested a generalized kinetics model that describes the dynamics of the population in a quantum system when a laser pulse acts on a dipole-allowed transition. It is demonstrated that this model is correct within the framework of perturbation theory's application to laser pulses of any duration and carrier frequency, including monochromatic and ultrashort pulses. In particular, it correctly reproduces the population oscillations at off-resonance carrier frequency of the laser pulse, which are caused by the perturbation of the quantum system at the difference frequency $\omega_{osc} = |\omega_c - \omega_0|$.

Our model is compared with the results of standard approaches for calculation of time dependence of upper level population of two-level system using two expressions for the rate of photo-induced processes. It is shown, that the first standard approach, which takes into account the spectral profile of the photoexcitation cross-section, corresponds to the exact solution in the monochromatic limit, when the pulse duration is much longer than the phase relaxation time of the excited transition. The second standard approach, based on using the Einstein's formula for the photoprocess rate, is adequate for the opposite limit of ultrashort pulses. Outside the ranges of their applicability, the standard approaches significantly overestimate the population of the upper energy level for $\omega_c = \omega_0$.

It is also demonstrated that, in the case of strong saturation, all procedures taken into consideration produce results that are about the same for periods of time that are considerably longer than the timeframes required for phase relaxation and population relaxation.

When using the Bloch equations is difficult or impossible, the proposed model can be used to calculate the time dependence of the populations of quantum systems in the field of short laser pulses for various types of spectral broadening of the excited transition and in the presence of other types of excitation/ionization of the quantum system.

Acknowledgements This work was performed with the financial support of the Russian Science Foundation (Agreement №22-22-00537).

Author contributions VAA wrote the main manuscript text and ESK prepared Figures 1–5. All authors reviewed the manuscript

Funding Russian Science Foundation, 22-22-00537.

Declarations

Conflict of interest The authors declare no competing interests.

References

1. V. Astapenko, *Interaction of Ultrafast Electromagnetic Pulses with Matter* (Springer, Heidelberg, New York, Dordrecht, London, 2013)
2. A. Pakhomov, M. Arkhipov, N. Rosanov, R. Arkhipov, *Phys. Rev. A* **105**, 043103 (2022)
3. R.M. Arkhipov, A.V. Pakhomov, M.R. Arkhipov et al., *Opt. Lett.* **44**(5), 1202 (2019)
4. V.A. Astapenko, *Phys. Lett. A* **436**, 128075 (2022)
5. R. Arkhipov, A. Pakhomov, M. Arkhipov et al., *Opt. Express* **28**, 17020 (2020)
6. V.A. Astapenko, *J. Exp. Theor. Phys.* **135**, 1–8 (2022)
7. M. Hassan, A. Wirth, I. Grguras and etc., *Rev. Sci. Instrum.* **83**, 111301 (2012)
8. F. Krausz, M. Ivanov, *Rev. Mod. Phys.* **81**, 163 (2009)
9. J. Xu, B. Shen, X. Zhang et al., *Scientific Rep.* **8**, 2669 (2018)
10. V. Prasad, B. Dahiya, K. Yamashita, *Phys. Scr.* **82**, 055302 (2010)
11. A.C. Brown, G. Armstrong, J. Benda et al., *Comp. Phys. Comm.* **250**, 107062 (2020)
12. V.A. Astapenko, *Phys. Lett. A* **374**, 1585 (2010)
13. V.A. Astapenko, *J. Exp. Theor. Phys.* **130**, 56 (2020)
14. O. Svelto, *Principles of Lasers* (Springer, NY, Dordrecht, Heidelberg, London, 2010)
15. A.V. Gorbunov, D.A. Shuvaev, I.V. Moskalenko, *Plasma Phys. Rep.* **38**, 574 (2012)
16. L. Allen, J.H. Eberly, *Optical Resonance and Two-level Atoms* (Wiley, NY, London, Sydney, Toronto, 1975)
17. C.R. Menyuk, M.A. Talukder, *Phys. Rev. Lett.* **102**, 023903 (2009)

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