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Excitation of a Quantum System by High-power Ultrashort Pulses of Various Shapes

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ABSTRACT: The paper is devoted to the theoretical investigation of special features of quantum system excitation by high-power ultrashort electromagnetic pulses (USP). The analysis is performed in frame of Bloch vector formalism for the excitation of two-level system by USP without constant component of electric filed strength (corrected Gaussian pulses, sine and cosine wavelet pulses). The main attention is given to the excitation dependences on pulse duration, carrier-envelope phase, value of coupling parameter and spectral detuning from the resonance. It is shown that in ultra-short regime above-mentioned dependences have peculiarities which are absent in the case of conventional long pulses.

Keywords: Ultra-short Laser Pulse; Two-Level System, Photo-excitation, Bloch vector formalism

The development of technology for generation of ultrashort pulses (USP) of a prescribed shape has opened up a new field of physics of interaction of radiation and a substance [1].

The peculiarities of interaction of USP of the traditional Gaussian shape, including chirped pulses, with elementary classical and quantum systems are considered in the book [2] (see also references herein). Using the traditional Gaussian shape to describe USP is a debatable question since such pulses contain a constant component of an electric field.

The present paper is dedicated to the theoretical study of excitation of an elementary quantum object (a two-level system (TS)) by high-power USP that do not contain the above constant component: by corrected Gaussian and wavelet pulses (see below).

In case of the action of ultrashort radiation pulses on a two-level system (with the relation $\Delta t \approx 1/\omega$ fulfilled, Δt is the pulse duration, ω is the pulse carrier frequency), the traditional rotating wave approximation [2] may be found to be inadequate. In this case the spectral width of radiation is of the order of the carrier frequency, so, strictly speaking, the following resonant condition is not satisfied:

$$\left|\omega - \omega_0\right| << \omega_0 , \tag{1}$$

where $\omega_0 \equiv \omega_{21} = \left(\mathrm{E}_2 - \mathrm{E}_1\right)/\hbar > 0$ is the eigenfrequency of a quantum transition in the two-level system, and neglecting the influence of a counter-propagating wave becomes incorrect. Then it is necessary to proceed from the exact vector equation for the Bloch vector [2]

$$\frac{d\mathbf{R}}{dt} = \mathbf{R} \times \mathbf{W} - \frac{\mathbf{R}_\perp}{T_2} + \frac{\mathbf{R}_3^e - \mathbf{R}_3}{T_1}, \tag{2}$$

where $\mathbf{W} = \left(2\Omega(t), 0, \omega_0\right)$ is the generalized angular velocity vector, $\mathbf{R}_3^e$ is the equilibrium value of the third component of the optical Bloch vector. In writing (2) it was taken into account that the thermodynamically equilibrium value $\mathbf{R}_{\perp}^{e}$ is equal to zero according to the basic principles of statistics, and $\mathbf{R}_{3}^{e} \neq 0$. The equation (2) is equivalent to the following system written in dimensional variables:

$$\begin{aligned} \frac{dR_1}{dt} &= \omega_0 R_2 - \frac{R_1}{T_2} \\ \frac{dR_2}{dt} &= -\omega_0 R_1 - \frac{R_2}{T_2} + 2\Omega(t) R_3 \\ \frac{dR_3}{dt} &= \frac{R_3^e - R_3}{T_1} - 2\Omega(t) R_2, \end{aligned} \tag{3}$$

where $\Omega(t) = \frac{d_0 E(t)}{\hbar}$ is the instantaneous Rabi frequency, ω_0, d_0 are the eigenfrequency and the matrix element of the electric dipole moment of the two-level system. Two relaxation times are introduced into the system (3): T_2 is the phase (transverse) relaxation time, T_1 is the longitudinal relaxation time (energy or population relaxation time for the two-level system), R_3^e is the equilibrium value of the third component of the Bloch vector in the absence of an electromagnetic field pulse.

In case of thermodynamic equilibrium: $R_3^e = th(\hbar \omega_0 / 2T)$, where T is the temperature in energy units. For room temperature and optical frequencies one has: $R_3^e \cong 1$. Let us introduce the dimensionless time-dependent electric field strength $\tilde{E}(t)$ by the formula:

$$E(t) = E_0 \tilde{E}(t), \tag{4}$$

where E_0 is an amplitude of electric field strength in a pulse. Then we have for the instantaneous Rabi frequency:

$$\Omega(t) = \frac{d_0 E_0}{\hbar} \tilde{E}(t), \tag{5}$$

and the system of the Bloch equations (3) in dimensionless variables will be rewritten as follows:

$$\begin{aligned} \frac{dR_1}{d\nu} &= R_2 - \frac{R_1}{\tilde{T}_2} \\ \frac{dR_2}{d\nu} &= -R_1 - \frac{R_2}{\tilde{T}_2} + 2\xi \tilde{E}(\nu) R_3 \\ \frac{dR_3}{d\nu} &= \frac{R_3^e - R_3}{\tilde{T}_1} - 2\xi \tilde{E}(\nu) R_3, \end{aligned} \tag{6}$$

where the dot means differentiation with respect to the dimensionless time $\nu = \omega_0 t$, $\xi = d_0 E_0 / \hbar \omega_0$ is the dimensionless amplitude of electric field strength (or coupling parameter), $\eta = \omega_0 \tau$ is the dimensionless pulse duration, $\tilde{T}_{1,2} = \omega_0 T_{1,2}$

In the general case (with the carrier frequency in a pulse) the electric field strength $\tilde{E}$ can also depend on φ (the carrier phase with respect to the pulse envelope (the CE (carrier-envelope) phase)) and $r = \omega / \omega_0$. The latter ratio can be expressed in terms of the relative carrier frequency detuning from the two-level system eigenfrequency:

$$\delta = \frac{\omega - \omega_0}{\omega_0} = r - 1. \tag{7}$$

If from first two equations of the system of equations (6) the second component of the Bloch vector is excluded, it is possible to obtain the equation for the first component of the Bloch vector R_1, the left side of which coincides with the equation for a harmonic oscillator with damping:

$$\ddot{R}_1 + \frac{2}{\tilde{T}_2}\dot{R}_1 + \left(1 + \frac{1}{\tilde{T}_2^{\,2}}\right)R_1 = \gamma\,\tilde{E}, \tag{8}$$

where $\gamma = 2\xi R_3$ is the constant of the force of binding of the two-level system with the electric field of a pulse that can be represented (in view of determination of the third component of the Bloch vector) as

$$\gamma = 2\xi\left(N_1 - N_2\right). \tag{9}$$

Thus coupling with the field is absent if populations of upper and lower levels are equal. Curiously, this is true for a coherent state, when the dipole moment of the two-level system has a maximum.

The oscillator (8) (at $R_3 = 1$), referred to as the transition oscillator, defines the evolution of the dipole moment of the system under the action of an external electric field.

The system of equations (6) is of a universal nature since it describes the evolution of an *arbitrary* two-level system that is connected by a dipole-allowed transition ($\xi \neq 0$).

In case of weak excitation $\xi << 1$ (the perturbation theory limit) of an originally unexcited two-level system, in the zero approximation it can be considered that $N_2 << 1$, $N_1 \approx 1$ and $R_3 \approx 1$, then $\gamma \approx 2\xi$, and the formula (8) is transformed to the equation for a harmonic oscillator.

It is possible to characterize excitation of a two-level system that was originally at a level with lower energy by the value of population of the upper energy level N_2 after termination of an electric field pulse. This population is related by a simple relation to the third component of the Bloch vector:

$$N_2 = \frac{1 - R_3}{2}. \tag{10}$$

In derivation of (10) the determination of the third component of the Bloch vector and the normalization condition $N_1 + N_2 = 1$ were used. Thus, determining numerically R_3 from the system (6), it is possible by the formula (10) to find the population of the upper level of the two-level system at an arbitrary instant of time.

The results of calculations of the upper energy level population after the termination of the action of an electromagnetic pulse (asymptotic population) presented in Figs. 1–8 are obtained for relaxation times $T_{1,2}$ = 480 fs.

The dependence of the population of the upper level of the two-level system excited by a half-cycle (n=0.5, where n is the number of periods (cycles) at the carrier frequency in a pulse) corrected Gaussian pulse [3] satisfying the equation

$$\int_{-\infty}^{\infty} E(t)\,dt = 0, \tag{11}$$

$$E_{cor}(t) = \mathrm{Re}\left[-i\,E_0\,\frac{\left(1 + i\,t/\omega\tau^2\right)^2 + 1/(\omega\tau)^2}{1 + 1/(\omega\tau)^2}\exp\left(-t^2/2\tau^2\right)\exp\left(i\,\omega t + i\varphi\right)\right] \tag{12}$$

on the value of the CE phase φ is shown in Fig. 1 in case of the zero detuning (δ = 0) for different values of the binding force constant ξ = 0.1, 1, 3.

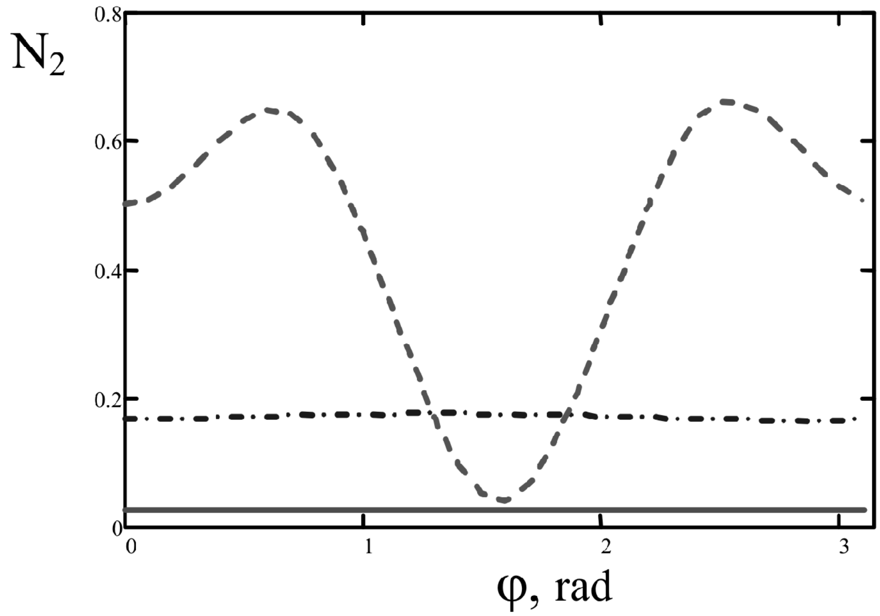

Figure 1: The dependence of the asymptotic population of the upper level on the CE phase in excitation by a half-cycle corrected Gaussian pulse for different values of the coupling parameter and the zero detuning δ = 0: solid curve – ξ = 0.1; dash-and-dot curve – ξ = 1, dashed curve – ξ = 3

Analogous dependences are given in Fig. 2 for a quarter-cycle corrected Gaussian pulse. It is seen that with growing parameter ξ and decreasing pulse duration the phase dependence of the population N_2 becomes stronger. In the perturbation theory limit (ξ = 0.1) the dependence of the upper energy level population on the CE phase is practically absent.

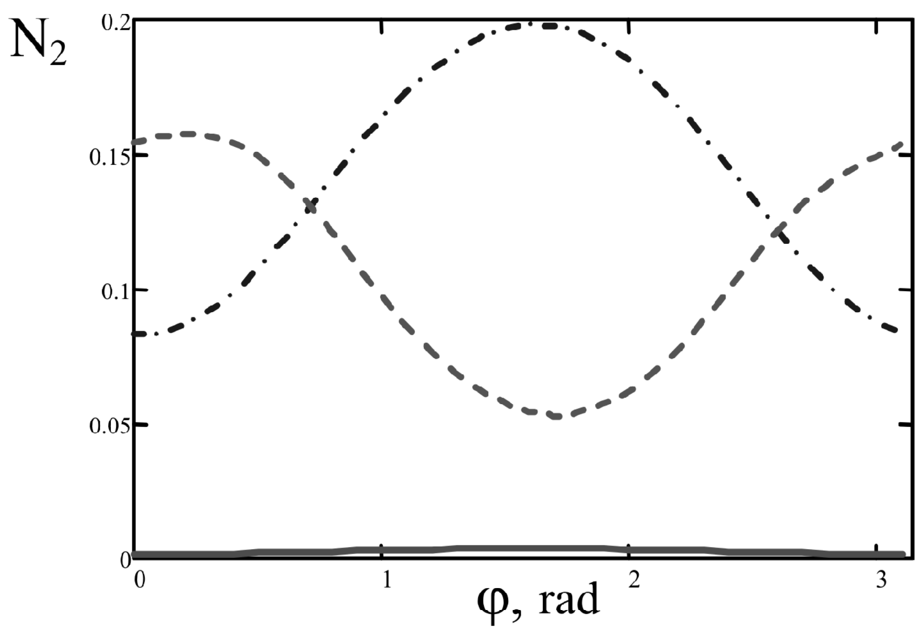

Figure 2: The dependence of the asymptotic population of the upper level on the CE phase in excitation by a quarter-cycle corrected Gaussian pulse for different values of the binding force parameter and the zero detuning δ = 0: solid curve – ξ = 0.1; dash-and-dot curve – ξ = 1, dashed curve – ξ = 3

In case of a half-cycle pulse the dependence on the CE phase is significant only for $\xi = 3$, while for a quarter-cycle pulse (Fig. 2) it is observed also for $\xi = 1$. And in the latter case with growing ξ the behavior of the phase dependence changes: the maximum at the point $\varphi = \pi/2$ (at $\xi = 1$) changes into the minimum for $\xi = 3$.

The spectra of excitation of the two-level system by single-cycle corrected Gaussian pulses calculated in the Bloch vector formalism are presented in Figs. 3 and 4.

In Fig. 3 calculation was carried out for sine and cosine pulses for the coupling constant $\xi = 1$ and $\delta > -0.4$. The asymmetry of the excitation spectrum in both cases is seen. The difference in the population value N_2 is relatively small and is observed for negative values of the parameter of the relative detuning δ of the pulse carrier frequency from the two-level system eigenfrequency.

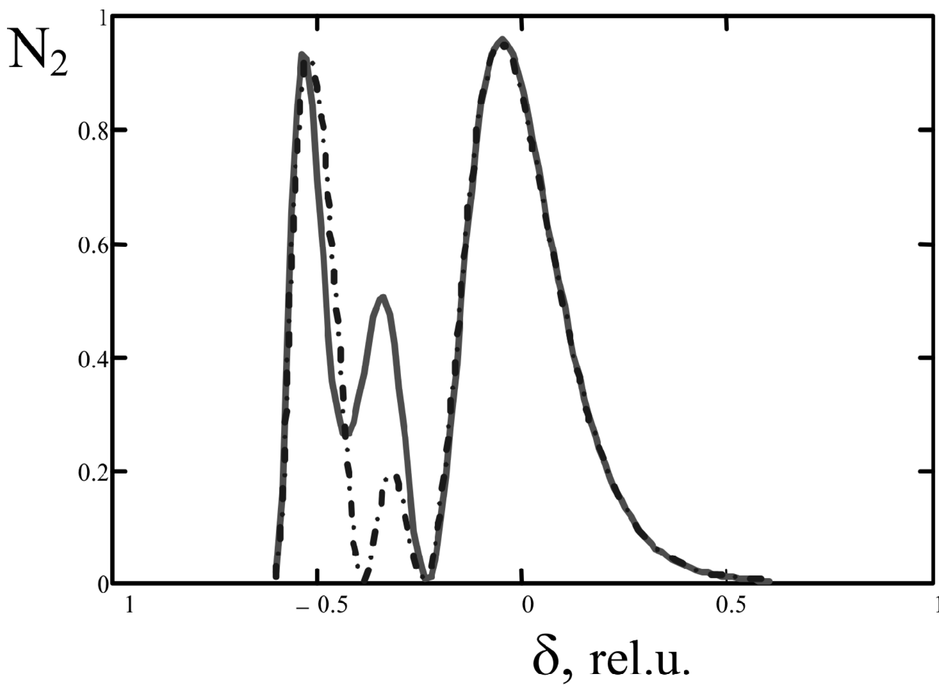

Figure 3: The dependence of the asymptotic population of the upper level excited by corrected Gaussian sine (solid curve) and cosine (dash-and-dot curve) single-cycle pulses on the relative detuning for $\xi = 1$

Presented in Fig. 4 is the population of the upper energy level N_2 excited by a single-cycle Gaussian sine pulse for different values of the coupling constant ξ and high negative detunings. It is seen that with growing parameter ξ the spectrum becomes more and more asymmetric: in its low-frequency region ($\omega < \omega_0$) additional maxima appear that were absent at the low value $\xi = 0.1$.

The appearance of additional maxima in the spectrum of excitation of the two-level system by intensive electromagnetic pulses can be explained by nonlinearity of electromagnetic interaction that results in occurrence of multiphoton processes of excitation. This circumstance is confirmed by the fact that the principal additional maximum at $\xi = 1$ in the low-frequency spectral region takes place for the detuning value $\delta = -0.5$, that is, excitation is of a two-photon nature, a maximum at $\delta = -0.3$ is also visible that corresponds to three-photon absorption. Analogous maxima take place for $\xi = 0.5$.

In case of strong coupling $\xi = 1$, besides principal maxima, in the excitation spectrum additional maxima of a lesser amplitude appear that are connected with the complex nature of interaction of an intensive ultrashort pulse and the two-level system, which cannot be described only in terms of multiphoton absorption.

Shown in Figs. 5 and 6 are the dependences of the population of the upper level of the two-level system excited by a corrected Gaussian pulse on the pulse duration for different values of the coupling constant. The curves of Fig. 5 are calculated for the relative frequency detuning equal to zero, and in the second case $\delta = 5\%$ is assumed.

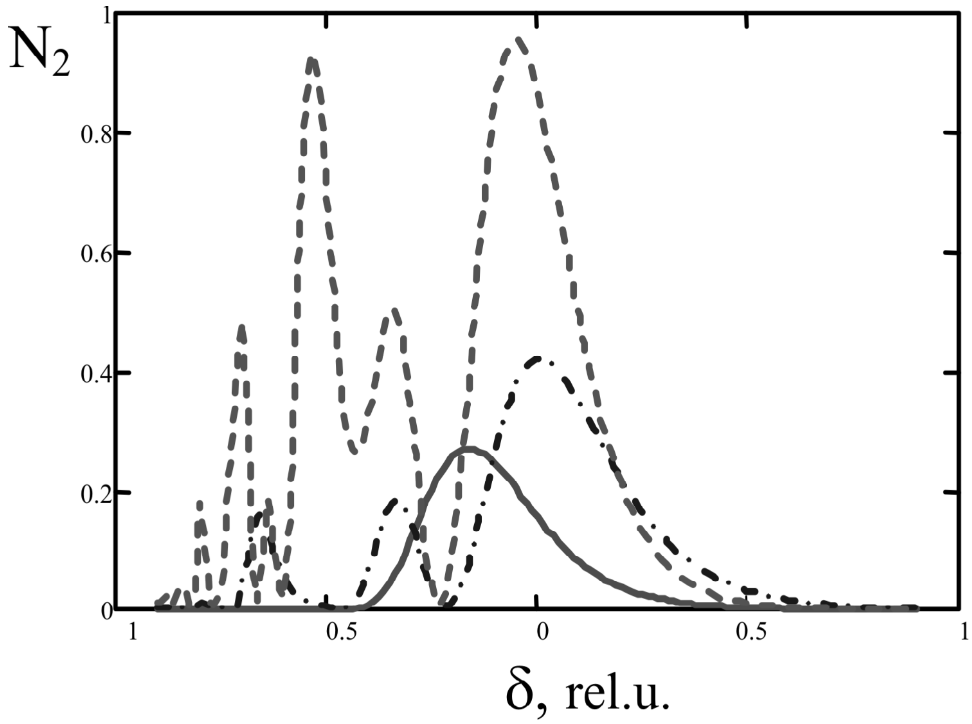

Figure 4: The dependence of the asymptotic population of the upper level excited by a corrected single-cycle Gaussian sine pulse on the relative detuning for different values of the binding parameter: solid curve – $\xi = 0.1$; dash-and-dot curve – $\xi = 0.5$, dashed curve – $\xi = 1$

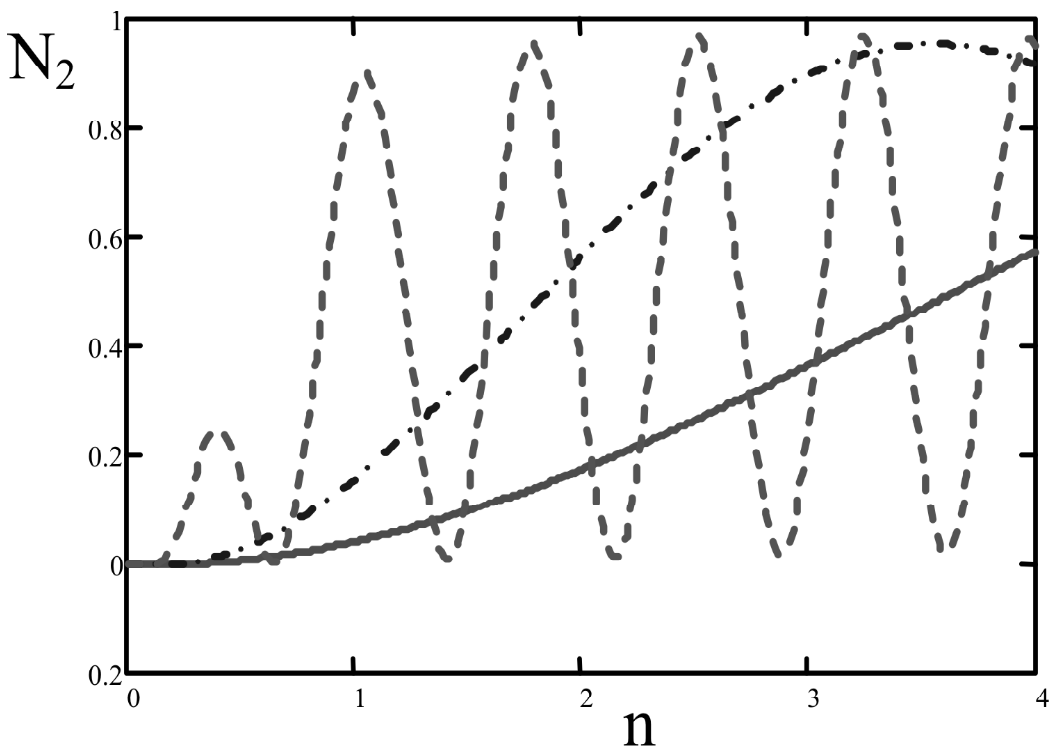

Figure 5: The asymptotic population of the upper level of the two-level system as a function of the number of cycles in a corrected Gaussian pulse for different values of the binding force for $\delta = 0$: solid curve – $\xi = 0.05$; dash-and-dot curve – $\xi = 0.1$, dashed curve – $\xi = 1$

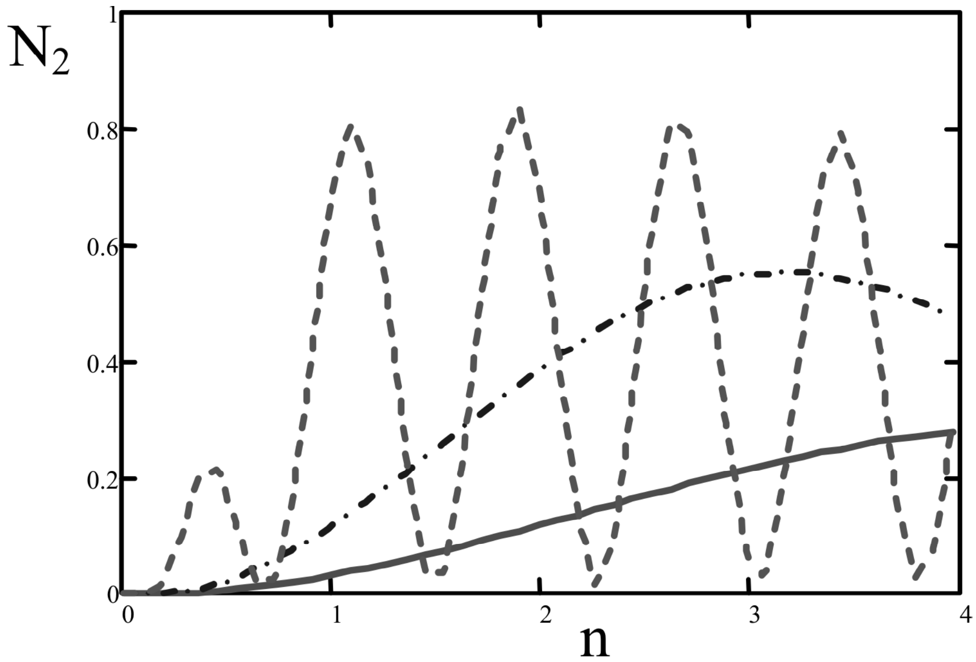

Figure 6: The asymptotic population of the upper level of the two-level system as a function of the number of cycles in a corrected Gaussian pulse for different values of the coupling parameter and the relative detuning $\delta = 5\%$: solid curve – $\xi = 0.05$; dash-and-dot curve – $\xi = 0.1$, dashed curve – $\xi = 1$

It is seen that in both cases at the high value of the coupling parameter ($\xi = 1$) oscillations of the upper level population with growing pulse duration are observed that represent the known Rabi oscillations. At low values of the parameter ξ these oscillations for ultrashort pulses do not show themselves.

With frequency detuning (Fig. 6) the qualitative picture remains the same as in resonance. The distinction is in some decrease of the amplitude and frequency of Rabi oscillations in the case $\xi = 1$.

Presented in Fig. 7 are the results of calculations of the asymptotic population of the upper level of the two-level system excited by a wavelet sine pulse

$$E_s(t) = \frac{\sqrt{2}}{\pi^{1/4}} E_0 \frac{t}{\tau} \exp\left(-\frac{t^2}{2\tau^2}\right) \tag{13}$$

and a wavelet cosine pulse [4]

$$E_c(t) = \frac{2}{\sqrt{3}\,\pi^{1/4}} E_0 \left(1 - \frac{t^2}{\tau^2}\right) \exp\left(-\frac{t^2}{2\tau^2}\right) \tag{14}$$

as functions of the pulse duration τ.

From Fig. 7 it is seen that there is a certain difference in the shape of curves corresponding to sine and cosine pulses that are on the whole of a qualitatively similar nature.

Presented in Fig. 8 are the dependences of the upper level population on the duration of a cosine wavelet pulse τ for different values of the coupling parameter ξ. From the figure it follows that with growing parameter ξ the dependence of N_2 on τ gets the behavior of Rabi oscillations, while in the perturbation theory limit ($\xi = 0.3$) the function $N_2(\tau)$ has one maximum.

The results of the numerical analysis of influence of relaxation times on the population of the upper level of the two-level system excited by a corrected Gaussian pulse as function of the pulse duration are presented in Fig. 9 for

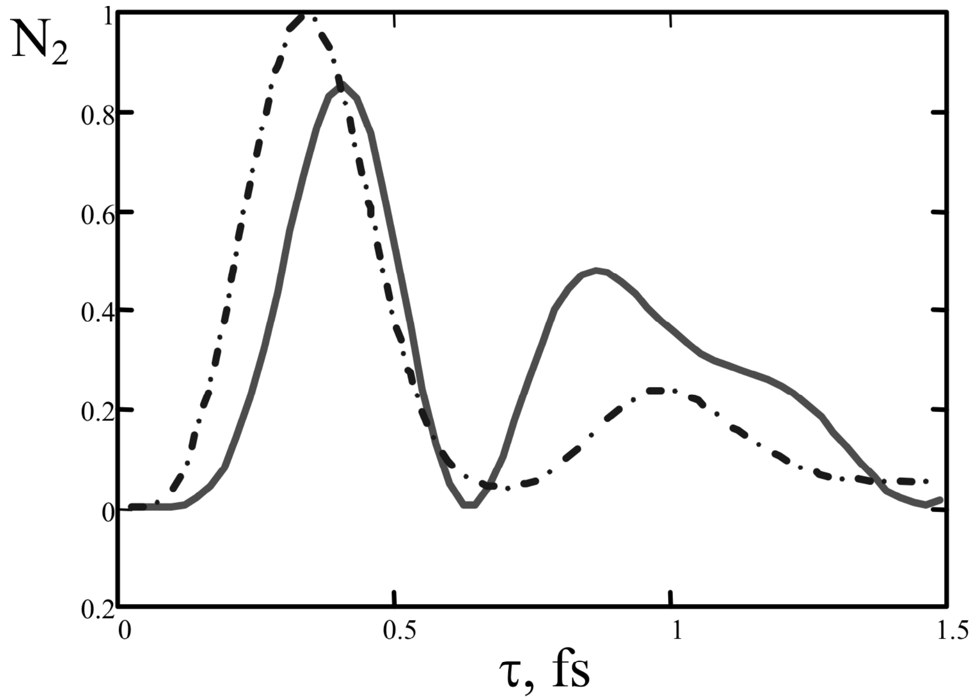

Figure 7: The dependence of the asymptotic population of the upper level on the pulse duration in excitation of the two-level system by cosine (solid curve) and sine (dash-and-dot curve) wavelet pulses for ξ =1

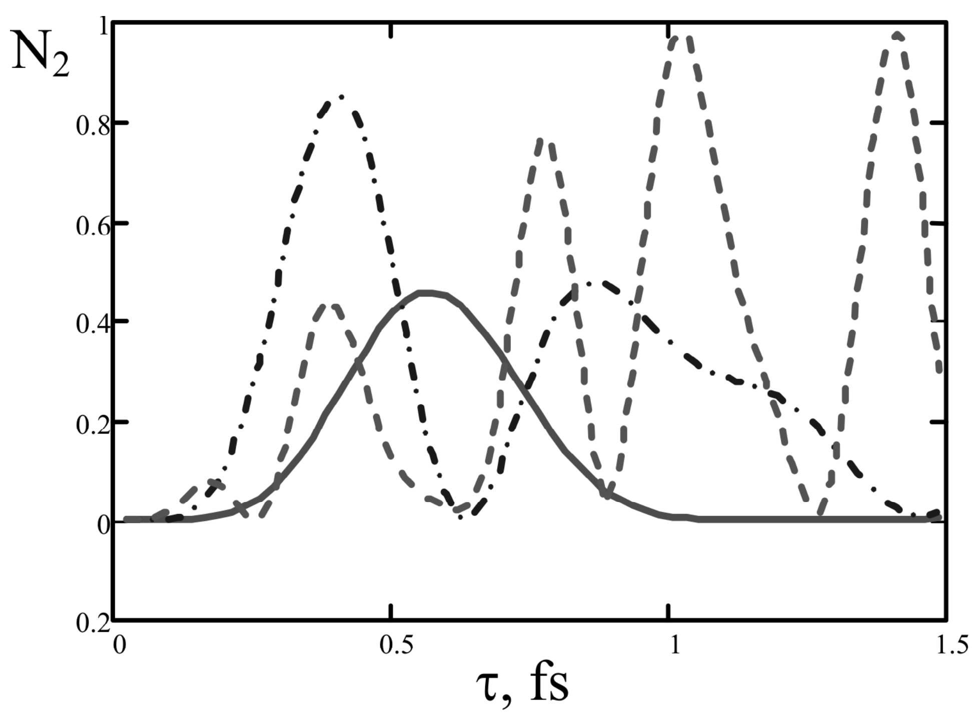

Figure 8: The dependence of the asymptotic population of the upper level on the duration of a cosine wavelet pulse for different values of the electron-phonon coupling parameter: solid curve – ξ =0.3, dash-and-dot curve – ξ =1, dotted curve – ξ =3

the relative frequency detuning $\delta = 5\%$. The calculation was carried out for equal times of longitudinal and transverse relaxation. In case of long times $T_{1,2}$ the function $N_2(n)$ after reaching the maximum value monotonically tends to zero. This can be explained by narrowing of the pulse spectrum, due to which all monochromatic components of an exciting pulse are beyond the width of an electron transition in the two-level system. For lesser relaxation times, when the spectral form of a TS line becomes wider, after reaching the maximum, the upper level population at first sharply decreases, and then at high values of the parameter *n* begins to grow linearly in conformity with the result of the perturbation theory.

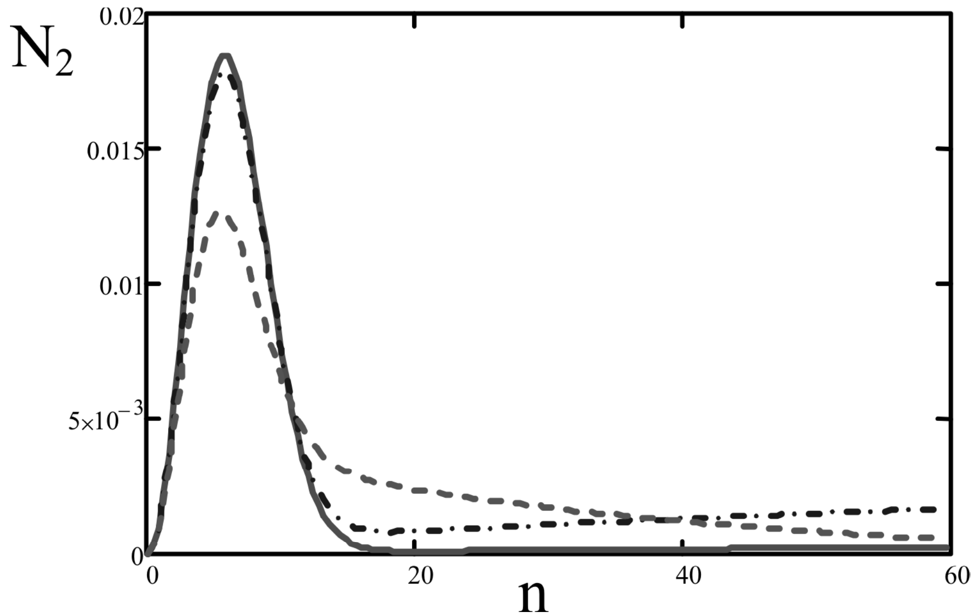

Figure 9: The dependence of the asymptotic population of the upper level of the two-level system on the duration of a corrected Gaussian cosine pulse for different relaxation times and ξ = 0.01: solid curve –$T_{1,2}$ = 4.8 ps, dash-and-dot curve – $T_{1,2}$ = 480 fs, dotted curve – $T_{1,2}$ = 48 fs

For lesser relaxation times ($T_{1,2}$ = 48 fs), however, the linear growth of the population N_2 at long pulse durations changes into decrease in contrast to the result of the perturbation theory. This fact is explained by decreasing population of the lower level during excitation of the two-level system with a wide spectrum that is not taken into account within the framework of the perturbation theory.

The analysis shows that for the zero frequency detuning (δ = 0) the maximum of the dependence $N_2(n)$ is shifted to the region of long pulse durations and widens considerably. The change of sign of the frequency detuning has little influence on the shape of the curves in Fig. 9, resulting in insignificant increase of the maximum value of the upper energy level population. In this case the behavior of the curves $N_2(n)$ at long pulse durations practically does not change.

In summary, by using the numerical solution of the system of equations for the components of the optical Bloch vector, the dependence of the population of the upper level of the two-level system after termination of USP was analyzed for various values of the problem parameters: the value of the CE phase, the pulse duration, the value of the electron-photon coupling parameter, and the relaxation times.

The action of two types of ultrashort pulses was considered: corrected Gaussian and wavelet pulses, which have no a constant component of electric field strength.

It was shown that (in case of a corrected Gaussian pulse) there is a noticeable dependence of the asymptotic population N_2 on the CE phase only for subcycle pulses at high enough value of the electron-photon coupling parameter.

The spectra of excitation of the TS by high-power (ξ = 1) cosine ($\varphi = \pi/2$) and sine (φ = 0) corrected Gaussian pulses are of an asymmetric nature with a small distinction in the region of negative detunings of the pulse carrier frequency from the TS eigenfrequency. With the parameter ξ growing from low values to one in the low-frequency spectral region additional maxima appear that reflect the complex dynamics of USP interaction with the TS in the nonlinear mode.

The analysis of the dependence of the TS upper level population on the duration of a corrected Gaussian pulse has shown that the increase of electron-photon coupling results in oscillating dependence of N_2 on the number of cycles in a pulse both in case of exact resonance and in a nonresonance case. Analogous results occur also in excitation of the TS by wavelet pulses with the only difference that for the latter there is no notion of resonance.

The calculation of the asymptotic population of the upper level of the TS excited by a corrected Gaussian pulse as a function of the pulse duration for different TS relaxation times at low values of the parameter ξ has shown that, in contrast to the result of the perturbation theory [2], the function $N_2(n)$ does not go to the linear dependence in the limit of short relaxation times, which can be explained by decreasing population of the TS lower level during excitation.

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