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Temporal dependence of quantum system excitation by exponential pulse

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ABSTRACT: We investigate analytically the excitation probability of quantum system by exponential pulse as function of time and pulse duration in the frame of the first order of perturbation theory and rotating wave approximation. Long and short time limits are considered for monochromatic (MP) and ultra-short pulses (USP). For long time limit (after the termination of pulse) analytical expressions are derived for excitation probability for Lorentz and Gaussian profiles of excitation cross section. Particularly it is shown that in short time limit for USP the excitation probability is oscillating function of time with period inversely proportional to carrier frequency detuning from eigenfrequency of the quantum system while for MP the probability is linear function of time. Results of numeric calculations of excitation time dependence are also presented.

Development of laser pulse generation technology in broad range of durations and carrier frequencies as well as new methods of photo-processes registration in the real time in femtosecond and attosecond scale [1-2] require the adequate theoretical description of the behavior of various target under the action of laser pulses with given parameters.

The purpose of the present paper is the investigation of the excitation of quantum system by laser pulse with exponential envelope (exponential pulse – EP) as function of time and pulse duration. EP is widely used model for laser radiation generated in Q-switching regime [3]. Important advantage of EP from the point of view of theoretical treatment is the possibility to derive analytical formulas which make transparent main dependences of photo-process which are hidden under numerical calculations for another pulse shapes.

Let us consider the excitation of a quantum system under the action of laser pulse. The probability of this process in the first order of perturbation theory and dipole approximation is given by well-known expression:

$$W(t)=\frac{1}{\hbar^2}\int_{-\infty}^{t}dt'\int_{-\infty}^{t}dt''\left\langle \hat{d}(t')\hat{d}(t'')\right\rangle E(t')E(t''). \tag{1}$$

here $\hat{d}(t)$ is time dependent operator of dipole moment, angle brackets mean averaging over the initial state of the target and $E(t)$ is electric field strength in the pulse. We suppose that $E(t\to\pm\infty)\to 0$.

It was shown in papers [4-5] that formula (1) can be transformed to the following one

$$W(t,\tau)=\frac{c}{4\pi^2}\int_{0}^{\infty}d\omega\frac{\sigma(\omega)}{\hbar\omega}D(t,\tau,\omega). \tag{2}$$

Here $\sigma(\omega)$ is total photo-excitation cross section of the target and

$$D(t,\tau,\omega)=\left|\int_{-\infty}^{t}dt'\exp(i\omega t')\,E(t',\tau)\right|^2 \tag{3}$$

is squared modulus of incomplete Fourier transform of electric field strength in laser pulse. In this paper we use the designation “D-function” for (3).

Dynamics of two-level system excitation by ultra-short pulses with Gaussian envelope was investigated in the paper [6] for Lorentz and Gaussian spectral profiles.

In this paper we consider excitation of quantum system by exponential pulse (EP) with arbitrary duration. Exponential pulse (EP) has the form:

$$E_{EP}(t,\tau)=\theta(t)E_0\exp(-t/\tau)\cos(\omega_c t) \tag{4}$$

here E_0 is amplitude of electric field strength, τ, ω_c are pulse duration and carrier frequency, $\theta(t)$ is Heaviside step-function.

For EP it is possible to obtain simple analytical representation of D-function (3). In what follows we use rotating wave approximation for D-function of EP (4):

$$D_{EP}(t,\tau,\omega)\cong\frac{1}{4}\theta(t)E_0^2\,\tau^2\,\frac{1+\exp(-2t/\tau)-2\exp(-t/\tau)\cos[(\omega-\omega_c)t]}{1+\tau^2(\omega-\omega_c)^2} \tag{5}$$

It should be noted that formula (5) is valid for quasi-resonance $|\omega-\omega_c|<<\omega_c$ and multi-cycle pulses $\omega_c\,\tau>>1$.

Further we consider the resonance excitation when the cross section has the form:

$$\sigma_{21}=\frac{2\pi^2e^2\,f_{21}}{m\,c}G_{21}(\omega) \tag{6}$$

here e, m are charge and mass of electron, f_{21}, $G_{21}(\omega)$ are oscillator strength and spectral profile of resonance transition, *c* is light velocity.

After pulse termination (long time limit) $t>>\tau$ we have in view of (5):

$$D(\tau,\omega)\cong\frac{1}{4}E_0^2\,\tau^2\,\frac{1}{1+\tau^2(\omega-\omega_c)^2}. \tag{7}$$

So D-function is constant in time and only the dependence on pulse duration τ remains. Substituting (6) and (7) into (2) we obtain:

$$W(\tau)=\frac{\pi e^2 f_{21}}{8m\hbar}E_0^2\,\tau\int_0^\infty d\omega\frac{G_{21}(\omega)}{\omega}\frac{1/(\pi\tau)}{\tau^{-2}+(\omega-\omega_c)^2}. \tag{8}$$

Let us first consider Lorentz spectral profile for excitation cross section (6):

$$G_L(\omega)=\frac{\gamma/\pi}{(\omega-\omega_0)^2+\gamma^2} \tag{9}$$

here ω_0 and γ is eigenfrequency and spectral width of resonant transition. Then integral in right-side of equation (8) can be evaluated analytically and we obtain

$$W_L(\tau)=\frac{e^2\,f_{21}}{8\,m\hbar\omega_0}\frac{E_0^2}{\gamma^2}F_L(y,\rho). \tag{10}$$

Here we introduce “F-function” for Lorentz profile:

$$F_L(y,\rho)=\frac{y^2(1+y)}{(1+y)^2+\rho^2 y^2}. \tag{11}$$

This function defines the dependence of the excitation probability on pulse duration and frequency detuning via following dimensionless variables:

$$y=\gamma\tau;\qquad \rho=(\omega_0-\omega_c)/\gamma. \tag{12}$$

Graphs of function (11) are presented in Figure 1 for various values of dimensionless detuning ρ.

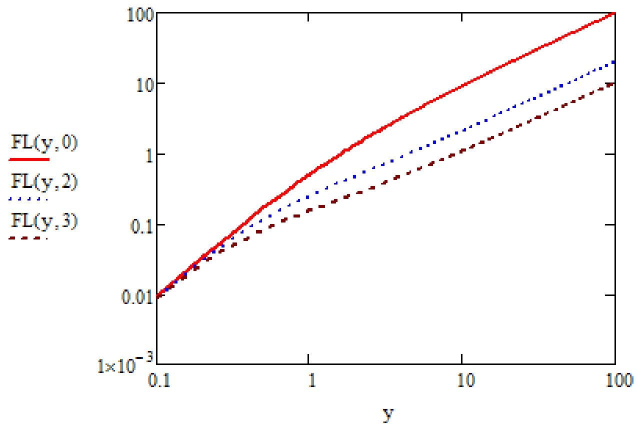

Fig.1. Lorentz F-function (11) for different ρ values: solid line - ρ=0, dotted line - ρ=2, dashed line - ρ=3.

For Gaussian profile of excited transition we have

$$G_G(\omega)=\frac{\exp\left(-(\omega-\omega_0)^2/2\sigma^2\right)}{\sqrt{2\pi}\,\sigma} \tag{13}$$

After integration in right side of equation (8) with function (13) we obtain the following equality

$$W_G(y,\rho)=\frac{\sqrt{\pi}\,e^2 f_{21}}{16 m\hbar\omega_0}\frac{E_0^2}{\sigma^2}F_G(y,\rho) \tag{14}$$

$$y=\sqrt{2}\,\sigma\tau;\ \rho=(\omega_0-\omega_c)/\sqrt{2}\,\sigma. \tag{15}$$

Here we introduce F-function for Gaussian profile:

$$F_G(y,\rho)=y\,\mathrm{Re}\left[w\left(\rho+\frac{i}{y}\right)\right] \tag{16}$$

$$w(z)=\exp(-z^2)\,erfc(-i z) \tag{17}$$

is complex error function, $erfc(z)$ is complimentary error function.

Gaussian F-function is presented in Figure 2 for different values of dimensionless frequency detuning.

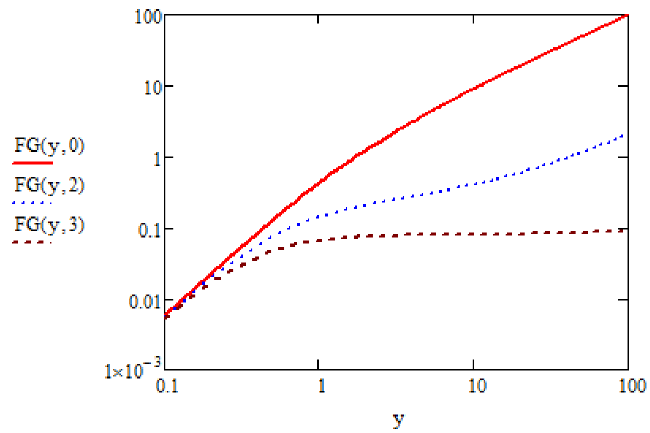

Fig.2. Gaussian F-function (16)-(17) for different ρ values: solid line - ρ=0, dotted line - ρ=2, dashed line - ρ=3.

It is important that Lorentz and Gaussian F-functions for EP monotonically increase with pulse duration for all values of frequency detuning. It is different from the case of Gaussian pulse and Gaussian profile when for sufficiently large detunings the excitation probability as function of pulses duration has maximum and minimum [7].

Note that in the limit $y \to \infty$ ($\tau >> 1/\gamma$ - monochromatic pulse) we have linear dependence on pulse duration as in the frame of traditional approach

$$F_L(y,\rho) \to \frac{y}{1+\rho^2} \text{ and } F_G(y,\rho) \to \exp(-\rho^2)\, y. \tag{18}$$

Formulas (18) also give the dependence of the excitation probability on dimensionless frequency detuning ρ.

In the opposite limit $y \to 0$ (USP case) we have quadratic dependence upon pulse duration:

$$F_L(y,\rho) \to y^2 \text{ and } F_G(y,\rho) \to y^2/\sqrt{\pi}. \tag{19}$$

Figures 1-2 demonstrate transition from linear to quadratic dependence of excitation probability as a function of dimensionless pulse duration.

In short time limit $t < \tau$ we have for D-function

$$D(t,\tau,\omega) \cong \theta(t) E_0^2 \frac{\tau^2}{1+\tau^2(\omega-\omega_c)^2} \sin^2[(\omega-\omega_c)t/2] \quad \omega \neq \omega_c \tag{20}$$

and

$$D(t) = (1/4)\theta(t) E_0^2 t^2 \quad \omega = \omega_c. \tag{21}$$

For ultra-short pulses $\tau << 1/\gamma$ when $\sigma(\omega) \propto \delta(\omega-\omega_0)$ in integral (2) the following expression for the excitation probability as function on time and pulse duration is hold

$$W_{USP}(t,\tau) = \frac{c}{4\pi^2} \frac{\sigma_{tot}}{\hbar\omega_0} D(t,\tau,\omega_0) \tag{22}$$

here

$$\sigma_{tot} = \int \sigma(\omega) d\omega = \frac{2\pi^2 e^2 f_{21}}{m\, c}.$$

Substituting (20) and (21) into (22) we obtain the excitation probability dependence on time for ultra-short pulses ($\tau << 1/\gamma$) in short time limit ($t < \tau$):

$$W_{USP}(t,\tau) \cong \frac{c}{4\pi^2}\frac{\sigma_{tot}}{\hbar\omega_0}\frac{\theta(t)E_0^2\tau^2}{1+\tau^2(\omega_0-\omega_c)^2}\sin^2[(\omega_0-\omega_c)t/2] \quad \omega_0 \neq \omega_c \tag{23}$$

Expression (23) demonstrates oscillation in time of the excitation probability. Period of these oscillations is inversely proportional to the modulus of the carrier frequency detuning from eigenfrequency of quantum system.

It is interesting to note that analogous oscillation was predicted for spectral probability of resonant scattering of USP by atom [8].

For $\omega_c = \omega_0$ we have instead of (23):

$$W_{USP}(t,\omega_0=\omega_c) = \frac{c}{16\pi^2}\frac{\sigma_{tot}}{\hbar\omega_0}\theta(t)E_0^2 t^2 \cdot \tag{24}$$

Thus in the resonance case the probability W increases quadratically with time.

In short-time limit ($t < \tau$) for monochromatic pulse $\tau >> 1/\gamma$ after substitution of formula (20) into (2) and performing integration with respect to frequency we obtain

$$W_{MP}(t) \approx \frac{c}{16\pi^2}\frac{\sigma_{tot}}{\hbar\omega_0}\theta(t)E_0^2\frac{\tau^2}{1+\gamma\tau}\left[1+\frac{\gamma\tau e^{-t/\tau}-e^{-\gamma t}}{1-\gamma\tau}\right] \approx \frac{c}{16\pi^2}\frac{\sigma_{tot}}{\hbar\omega_0}\theta(t)E_0^2 t^2 \cdot \tag{25}$$

Thus the excitation probability increases quadratically with time.

Figures below show the peculiarity of time dependence of the excitation probability for Lorentz profile and different values of parameters involved. Values of parameters are chosen in the intermediate range to demonstrate different behavior of $W(t)$. All parameters are in atomic units.

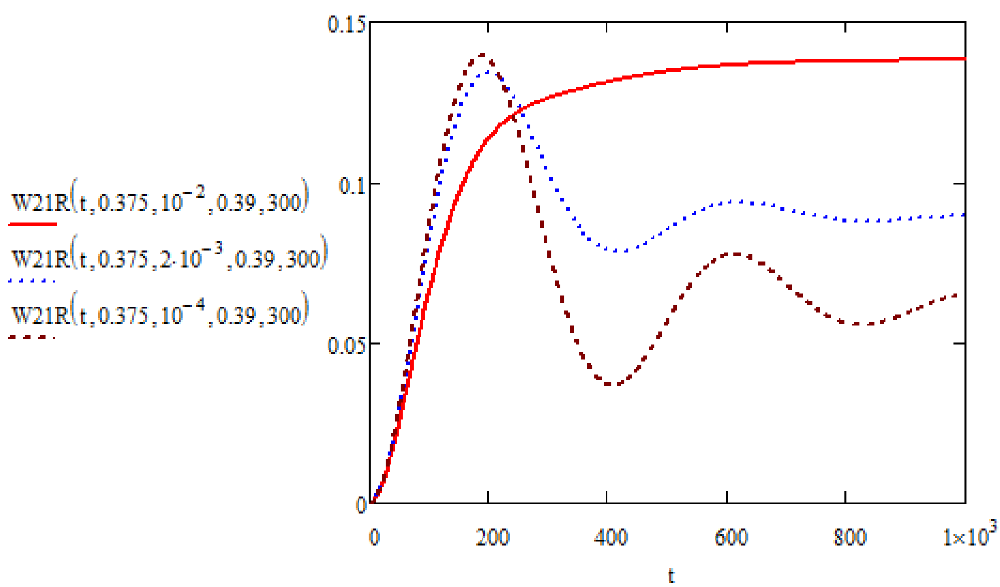

Fig.3. Time dependence of the excitation probability for Lorentz profile and various values of spectral width γ: solid curve – $\gamma=10^{-2}$, dotted curve – $\gamma=2\cdot10^{-3}$, dashed curve – $\gamma=10^{-3}$ and $\omega_0=0.375$, $\omega_c=0.39$, $\tau=300$, $E_0=10^{-2}$

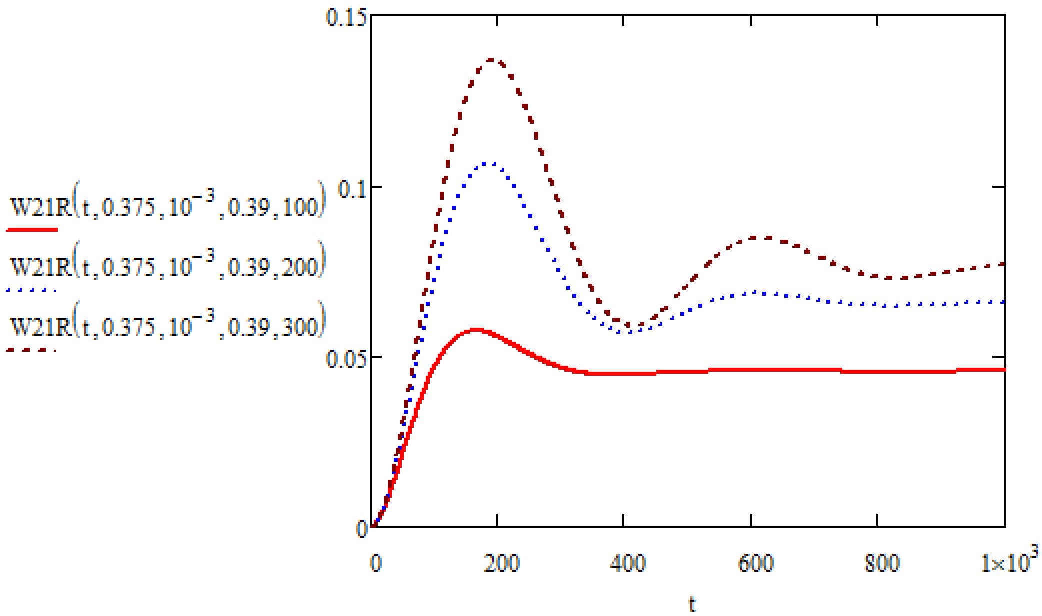

Fig.4. Time dependence of the excitation probability for Lorentz profile and various values of pulse duration: solid curve – τ=100, dotted curve – τ=200, dashed curve – τ=300 and ω_0=0.375, ω_c=0.39, $\gamma=10^{-3}$, $E_0=10^{-2}$

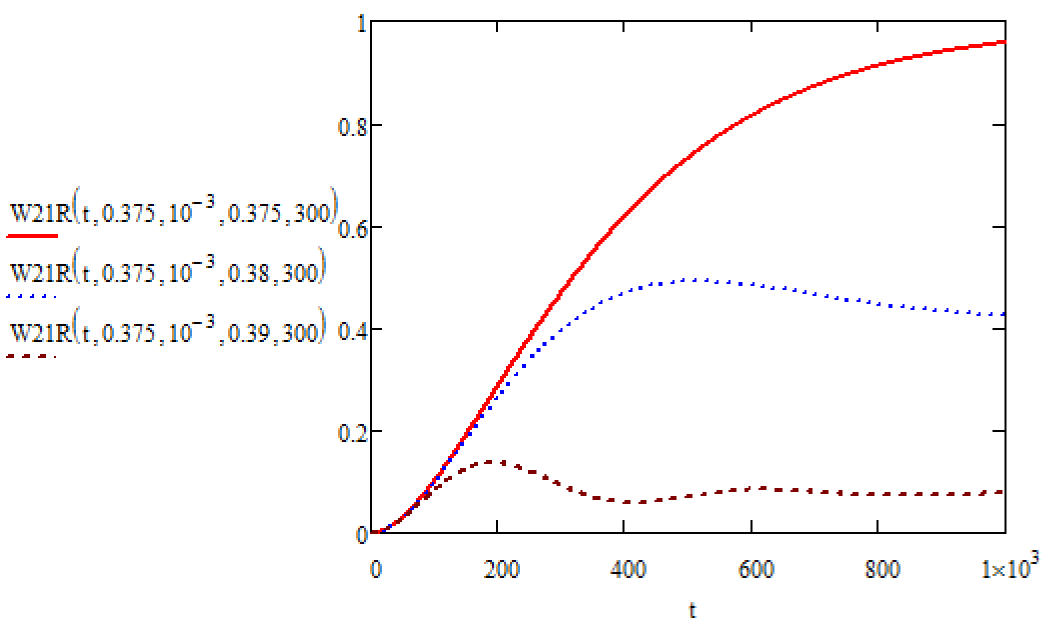

Fig.5. Time dependence of the excitation probability for Lorentz profile and various values of carrier frequency: solid curve – ω_c=0.375, dotted curve –ω_c=0.38, dashed curve – ω_c=0.39 and ω_0=0.375, τ=300, $\gamma=10^{-3}$, $E_0=10^{-2}$

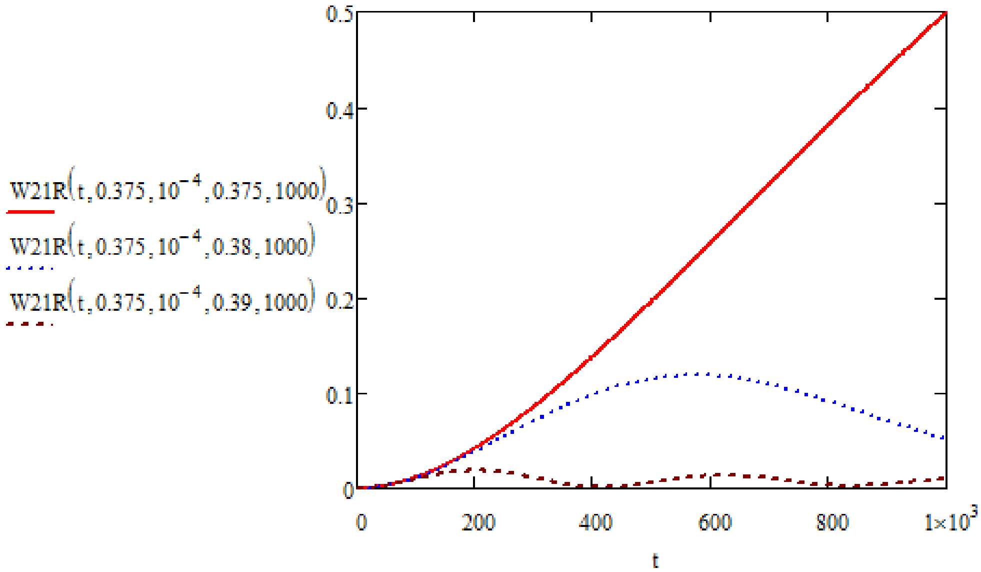

Fig.6. Time dependence of the excitation probability for Lorentz profile and various values of carrier frequency for longer pulse: solid curve –ω_c=0.375, dotted curve –ω_c=0.38, dashed curve – ω_c=0.39 and ω_0=0.375, τ=1000, $\gamma=10^{-4}$, $E_0=3\cdot10^{-3}$

It can be seen from above figures that character of excitation probability as a function of time changes from quadratic to oscillating and linear as the values of parameters change. These numerical results are in agreement with prediction of analytical formulas derived in this paper.

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