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Dynamics of Two-Level System Excitation by Ultra-Short Laser Pulses

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ABSTRACT: The paper is devoted to theoretical study of two-level system (TLS) excitation dynamics in the field of ultra-short laser pulse. Simple formula for dependence on time of excitation probability is derived in the framework of perturbation theory. This formula expresses excitation probability in terms of excitation cross section and incomplete Fourier transform of laser pulse. It generalizes previously obtained by author formula for probability of photo-process for all time of pulse action [1]. The calculation is made for Gaussian and Lorentz line shape of dipole-allowed transition in TLS.

The development of technology for generating ultra-short laser pulses (USP) [2] makes it relevant to study theoretically the photo-processes occurring under the action of such pulses. As was shown in a number of previous works (see e.g. [3]), an adequate description in this case requires a new approach based on using the probability of a photo-process for the entire duration of the pulse instead of the traditional approach based on probability per unit time.

The use of this method allowed us to identify an important distinctive feature of photo-processes in the field of an ultra-short pulse: namely, the nonlinear dependence of probability on the pulse duration even for weak laser field [4].

Our approach also makes it possible to describe the time dependence of the photo-process, the experimental observation of which is one of the most important areas of modern attosecond physics [5-6].

The purpose of the present paper is theoretical investigation of the dynamics of TLS excitation within framework of the perturbation theory with account for various spectral line shape of dipole-allowed transition in TLS.

Let us consider the excitation of TLS with dipole-allowed transition by USP in the framework of the first order of the perturbation theory.

For the probability of photo-excitation at a given time under the action of the field $E(t)$ (we assume that $E(t \to \pm\infty) = 0$) in the dipole approximation, one has

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

here angle brackets mean averaging over the initial state of the target.

There is a correlator of dipole moments in the formula (1), which for a stationary system can be represented as:

$$\left\langle \hat{d}(t') \hat{d}(t'') \right\rangle = K(t', t'') = K(t'' - t'). \tag{2}$$

After substitution of formula (2) into expression (1) one has

$$W(t) = \frac{1}{\hbar^2} \int_{-\infty}^{t} dt' \int_{-\infty}^{t} dt'' \, K(t'' - t') E(t') E(t'') . \tag{3}$$

Using the Fourier transform of dipole moment correlator

$$K(t'' - t') = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \exp(-i\omega(t'' - t')) K(\omega) \tag{4}$$

and it connection with excitation cross section [1]

$$K(\omega) = \frac{\hbar c}{2\pi\omega} \sigma(\omega) \tag{5}$$

we obtain

$$W(t) = \frac{c}{4\pi^2 \hbar} \int_{-\infty}^{t} dt' \int_{-\infty}^{t} dt'' \int d\omega \exp(-i\omega(t'' - t')) \frac{\sigma(\omega)}{\omega} E(t') E(t'') \tag{6}$$

and

$$W(t) = \frac{c}{4\pi^2} \int_{0}^{\infty} d\omega \frac{\sigma(\omega)}{\hbar\omega} \int_{-\infty}^{t} dt' \int_{-\infty}^{t} dt'' \exp(-i\omega(t'' - t')) \, E(t') E(t'') . \tag{7}$$

Using this formula we finally arrive at general expression for time dependence of excitation probability in the framework of validity of perturbation theory

$$W(t) = \frac{c}{4\pi^2} \int_{0}^{\infty} d\omega \frac{\sigma(\omega)}{\hbar\omega} \left| \int_{-\infty}^{t} dt' \exp(i\omega t') \, E(t') \right|^2 . \tag{8}$$

Note that for large time $t >> \tau$ (t is pulse duration) expression (8) transforms into formula for probability of photo-process for all time of pulse action [1].

For calculation of excitation probability of TLS we use the following formula for excitation cross section [7]

$$\sigma_{21} = \frac{2\pi^2 e^2 f_{21}}{m c} G_{21}(\omega) \tag{9}$$

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

Substituting (9) in the right-side of equality (8) we find

$$W_{21}(t)=\frac{e^2 f_{21}}{2m\hbar}\int_0^\infty d\omega\frac{G_{21}(\omega)}{\omega}D(t,\omega)\,. \tag{10}$$

Here

$$D(t,\omega)=\left|\int_{-\infty}^{t}dt'\exp(i\omega t')\,E(t')\right|^2 \tag{11}$$

is squared modulus of incomplete Fourier transform of electric field strength in laser pulse.

When the following inequality is valid ($\Delta\omega$ is spectral width of TLS line shape)

$$\tau << 1/\Delta\omega \tag{12}$$

one has $G_{21}(\omega)\approx\delta(\omega-\omega_{21})$ and expression (10) is simplified to the form:

$$W_{21}(t)=\frac{e^2 f_{21}}{2m\hbar\,\omega_{21}}D(\omega_{21},t) \tag{13}$$

here ω_{21} is own frequency of TLS.

Thus excitation probability does not depend on TLS spectral line shape in the short pulse limit (12).

For Gaussian pulse with carrier frequency ω_c and amplitude E_0 it is possible to obtain the following expression for envelope dependence of $D(t,\omega)$ function of time (11):

$$D_{env}(t,\omega)=\frac{\pi}{2}E_0^2\,\tau^2\,\widetilde{D}(t/\tau,|\omega-\omega_c|\tau), \tag{14}$$

$$\widetilde{D}(\widetilde{t},\Delta)=\exp(-\Delta^2)\left|erfc\left(-\frac{\widetilde{t}+i\Delta}{\sqrt{2}}\right)\right|^2, \tag{15}$$

here $erfc(z)$ is supplementary error function, $\widetilde{t}=t/\tau$ is normalized time, $\Delta=|\omega-\omega_c|\tau$ is non-adiabatic parameter.

Calculation results for the probability of TLS excitation dynamics which we obtained using formulas (10), (14), (15) are presented in figures 1-3 for Lorentz and Gaussian line shapes and various line width of TLS.

In our calculations we used the following parameters: ω_{21}=0.375 a.u., ω_c=0.33 a.u., E_0=0.01 a.u., t=60 a.u.

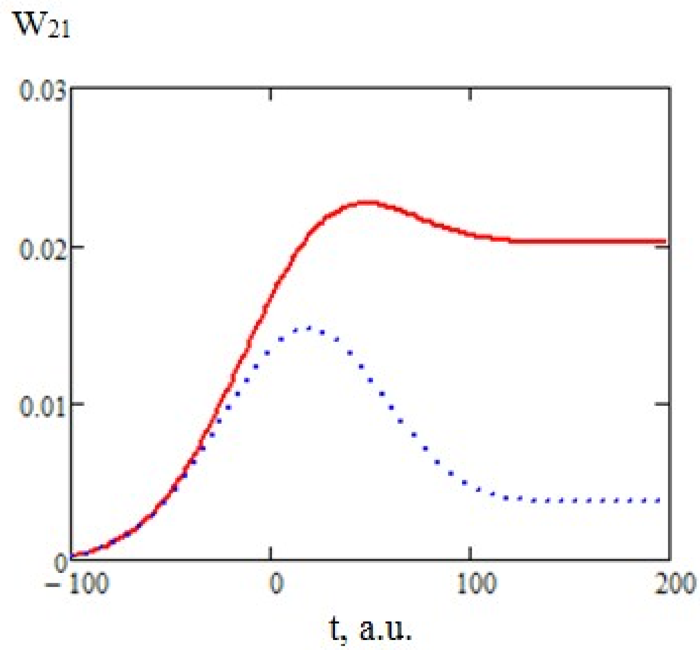

Fig.1. Envelope of TLS excitation probability as a function of time for TLS line width $\Delta\omega$=0.01 a.u. Solid line – Lorentz line shape, dotted line – Gaussian line shape.

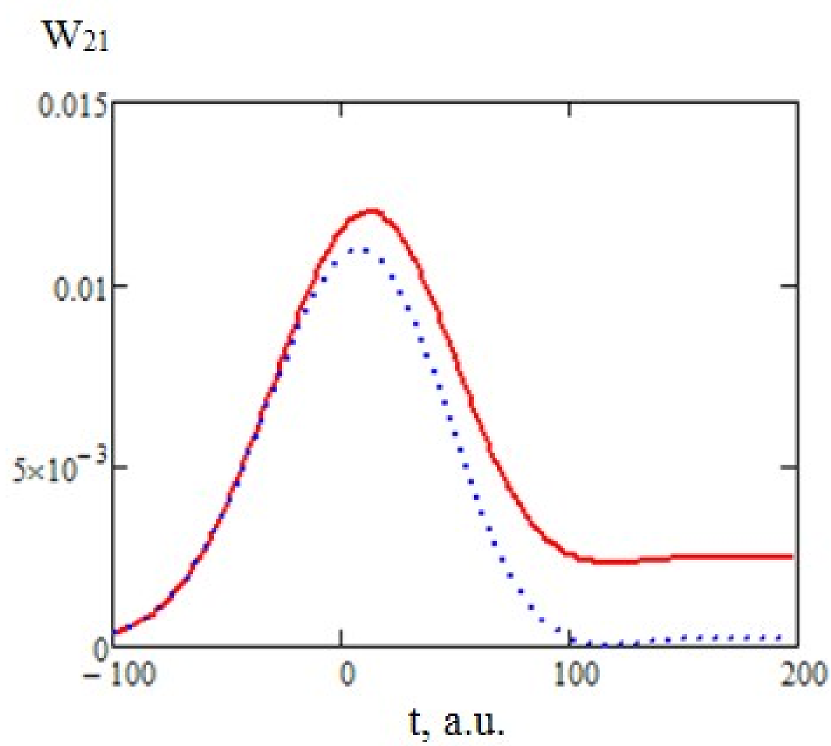

Fig. 2. The same as in Figure 1 for $\Delta\omega$=0.001 a.u.

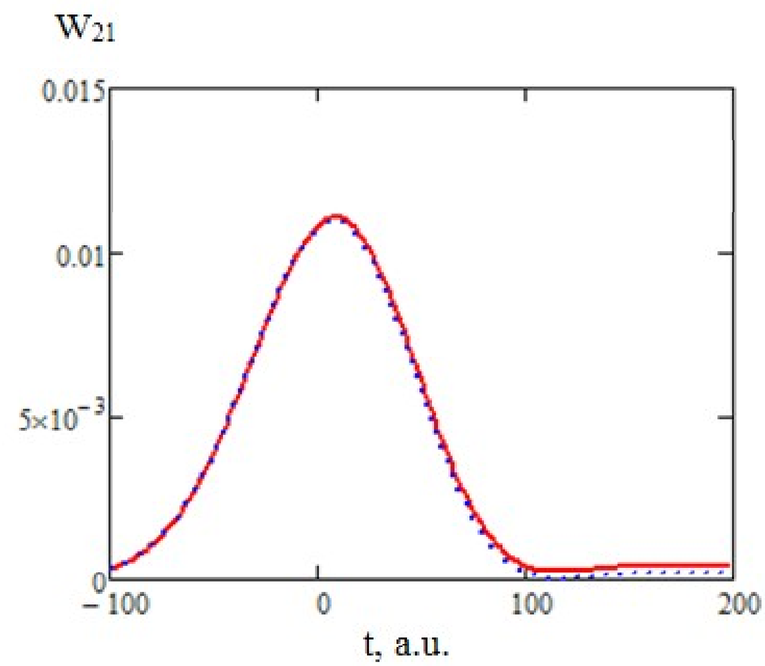

Fig. 3. The same as in Figure 1 for $\Delta\omega$=0.0001 a.u.

From these figures one can see that for given parameters function $W_{21}(t)$ has maximum which become more pronounced for smaller values of spectral line width $\Delta\omega$.

It should be noted that in considered case non-adiabatic parameter Δ=2.7>1. That means that the excitation probability for zero spectral line width turns to zero with increase of time. This is clearly shown by the figures 1-3.

Another important consequence of obtained results is the sensitivity of TLS excitation dynamics on spectral line shape in the case of relatively large value of spectral line width. Namely, in the case of Gaussian line shape the excitation probability is smaller than the same quantity for Lorentz line shape. This can be explained by the fact that Gaussian spectral line shape has small "wings" in comparison with Lorentz one.

Acknowledgments

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