Absorption of Ultrashort Electromagnetic Pulses on Broadened Dipole Transitions

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Abstract. The study is devoted to the theoretical analysis of ultrashort electromagnetic pulses (USP) absorption on broadened dipole transitions. Calculations are made in the frame of perturbation theory with the use of the basic formula for energy absorbed during all time of the action of USP on dipole transition. Dependences of absorbed energy upon pulse duration and carrier frequency are obtained and analyzed for different types of spectral line shape and USP parameters.

1. Introduction
The development of technology of generation of ultrashort electromagnetic pulses (USP) with controlled shape and duration down to several tens of attoseconds [1] make urgent the consideration of peculiarities of their interaction with a substance: atoms, clusters, nanoparticles, etc. The present study is devoted to the theoretical analysis of ultrashort electromagnetic pulses absorption on broadened dipole transitions.

2. General formulas for radiation absorption probability
Derivation of basic formula for total probability of photoabsorption for all time of pulse action is presented in paper [2]. Here we outline the main points of this derivation. Using standard perturbation approach, one can obtain the following expression for the photoexcitation probability by an electric field with amplitude $E(t)$ in the dipole approximation:

$$W_{tot} = \sum_{n} W_{n0} = \frac{1}{\hbar^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \langle 0 | \hat{d}_i(t') \hat{d}_k(t) | 0 \rangle E_i(t') E_k(t) dt dt'. \tag{1}$$

In terms of the the correlator of the dipole moments, $K_{ik}(\tau) = \langle 0 | \hat{d}_i(t') \hat{d}_k(t) | 0 \rangle$, Equation (1) can be rewritten as

$$W_{tot} = \frac{1}{\hbar^2} \int_{-\infty}^{\infty} \int K(\tau) E_i(t+\tau) E_k(t) dt d\tau. \tag{2}$$

After Fourier transformation of the functions under the integral in Eq. (2) we have
Here

\[ K(\omega) = \frac{1}{3} \int_{-\infty}^{\infty} \langle 0 | \hat{A}_i(t) \hat{A}_i(t+\tau) | 0 \rangle \exp(i \omega \tau) d\tau \]  \hspace{1cm} (4)

is the Fourier transform of the track of the dipole moment correlator.

It can be shown that function \( K(\omega) \) is connected with the photoionization cross-section \( \sigma(\omega) \) via the relation (see for details [2]):

\[ K(\omega) = \frac{\hbar c}{2\pi \omega} \sigma(\omega). \]  \hspace{1cm} (5)

Here \( c \) denotes the speed of light in free space. Substituting this expression for \( K(\omega) \) into Eq. (3) we obtain the basic formula

\[ W_{\text{tot}} = \frac{1}{2\pi \hbar^2} \int_{-\infty}^{\infty} K(\omega') |E(\omega')|^2 d\omega'. \]  \hspace{1cm} (3)

where \( E(\omega') \) is Fourier transform of electric field strength in USP. It is obvious that the expression (6) is meaningful only for \( W < 1 \), within the framework of applicability of the perturbation theory.

3. Numerical calculations

We consider three types of transition broadening: homogeneous (Lorentzian), Gaussian and Holtsmark. The spectral profiles of corresponding broadening are given by well-known expressions:

\[ G_{\text{hom}}(\omega, \Omega, \Delta) = \frac{\Delta/\pi}{(\omega - \Omega)^2 + \Delta^2}, \]  \hspace{1cm} (7)

\[ G_{\text{inh}}(\omega, \Omega, \Delta) = \frac{1}{\sqrt{2\pi}\cdot\Delta} \cdot \exp \left[ -\frac{(\omega - \Omega)^2}{2 \cdot \Delta^2} \right], \]  \hspace{1cm} (8)

\[ G_{\text{Hob}}(\omega, \Omega, \Delta) = \frac{1}{2\cdot\Delta} \cdot H_1 \left( \frac{\omega - \Omega}{\Delta} \right), \]  \hspace{1cm} (9)

\[ H_1(\beta) = 2 \cdot \frac{\beta}{\pi} \cdot \int_{0}^{\infty} x \cdot \sin(\beta \cdot x) \cdot \exp \left( -x^2 \right) dx, \]  \hspace{1cm} (10)

where \( \Omega \) is the central frequency, and \( \Delta \) is the spectral width of transition.

In this study we use a corrected Gaussian pulse [3], because it has a zero constant component (the Fourier transform equals to zero at zero frequency). The Fourier transform of the corrected Gaussian pulse equals

\[ E_{\text{cor}}(\omega', \omega, \tau, \varphi) = i E_0 \sqrt{\frac{\pi}{2}} \frac{\omega'^2}{1 + \omega'^2} \cdot \left[ e^{-i\varphi - (\omega - \omega')^2 \tau^2/2} - e^{i\varphi - (\omega + \omega')^2 \tau^2/2} \right]. \]  \hspace{1cm} (11)
where $\omega$ is the carrier frequency, $\omega'$ is the current frequency, $\tau$ is the pulse duration, and $E_0$ is the amplitude of electric field strength.

The absorption cross-section of the broadened transition $\sigma_{21}(\omega')$ is equal to

$$\sigma_{21}(\omega') = \frac{2e^2}{mc}f_{21}G_{21}(\omega'),$$  \hspace{1cm} (12)

where $f_{21}$ is the oscillator strength and $G_{21}(\omega')$ is the spectral profile of the transition.

Let us express the pulse duration in terms of a number of periods at the carrier frequency $\omega$ that fit into the effective pulse duration $\Delta t_p = \tau \cdot \Delta \tau$ to obtain

$$n = \frac{\Delta t_p}{T} = \frac{\omega \cdot \tau}{2\sqrt{\pi}}. \hspace{1cm} (13)$$

Thus the dimensionless parameter $n$ (the number of cycles in a pulse), together with the time $\tau$, can be used as measure of the pulse duration.

The expression for the absorbed energy during the USP action on broadening transitions follows from Equations (6) and (12), and one finds

$$\Delta E(n) = \frac{e^2f_{21}}{2m}\int_0^\infty G_{21}(\omega')\left|E_{cor}(\omega',\omega,n)\right|^2 d\omega'. \hspace{1cm} (14)$$

The spectral function $G_{21}(\omega')$ is given by Eqs. (7) to (10) for the different types of line broadening.

4. Results and conclusions

The results of the calculation of the absorbed energy dependence as a function of pulse duration for different line shapes (Lorentzian, Gaussian and Holtsmark) are shown in Figure 1 for nonresonant and Figure 2 for resonant carrier frequency of USP. One can see from these figures that the considered dependence is non-linear in contrast with the long pulse limit.

![Figure 1](image-url)  
**Figure 1.** Absorbed energy dependence upon USP duration for Gaussian (solid line), homogeneous (Lorentzian) (dotted line) and Holtsmark broadening (dashed line). Where $n$ is number of carrier frequency cycles, spectral width of the transition is 0.01 a.u., relative detuning is 0.15 and amplitude of USP electric field strength is assumed 0.1 a.u.
Figure 1 shows that for sufficiently large detuning of the carrier frequency from the central frequency of the transition the dependence is a response curve with a maximum. It is established that for the same parameters the maximum is reached when the number of cycles of carrier frequency $n$ is equal to 2...2.5, and for sufficiently large value of $n$ that are considered, the dependence reduces to a linear function. Figure 1 further illustrates that for the case of Gaussian broadening the considered dependence goes to zero in the limit of a long pulse.

![Figure 2](image)

**Figure 2.** Absorbed energy dependence upon USP duration for Gaussian (solid line), homogeneous (dotted line) and Holtsmark broadening (dashed line). Parameters are the same as in Fig.1, but the amplitude of incident pulse is 0.01 in atomic units, and the relative detuning equals zero.

In summary, we computed the absorption of USP energy by the dipole transition with different spectral shapes. Three types of broadening are considered: Lorentz type, Gaussian type and Holtsmark type. The dependence of the absorbed energy upon pulse duration is analyzed for nonresonant and resonant cases. We conclude that for sufficiently large detuning of the USP carrier frequency from resonance, this dependence is described by a response curve with a maximum, and the form of the response is determined by the type of broadening.

**References**

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[2] Astapenko V A 2010 *Phys. Lett. A* **374** 1585
[3] Lin Q, Zheng J, Becker W *Phys. Rev. Lett.* **97** 253902