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The effect of the shape of short electromagnetic pulses on the probability of quantum transitions

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Within the first order of perturbation theory without using the electric dipole approximation, the probability of quantum transitions of micro-objects under the action of extremely short electromagnetic pulses is analyzed. The selection rules are discussed and the dependence of the transition probability on the pulse parameters is determined.

Keywords: extremely short electromagnetic pulses, electric pulse area, probability of quantum transitions.

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1. Introduction

The progress in getting increasingly shorter radiation pulses up to the attosecond level has made it possible to observe the signs of electron motion in atoms, molecules and condensed media [1]. This also necessitates the solution of new theoretical problems concerning such duration of pulse impact on objects and corresponding relations between the object size and radiation packet. Besides direct numerical calculations, from which it is difficult to derive common patterns, two approaches are used in the literature. The first approach is based on the sudden perturbation approximation [2–4]; general expressions for the probability of transitions induced by extremely short pulses can be successfully derived on this way [5]. A disadvantage of this approach is in ignoring the signs of pulse edges for which field strength is insufficiently high to make this approach applicable. Second approach is the involvement of perturbation theory with respect to the field, as a matter of fact, first-order theory. The literature uses electric dipole approximation that is valid when the object dimensions are much smaller than the central radiation wavelength or radiation packet dimensions. Electric dipole approximation applicability conditions are violated for certain important cases. Rydberg atoms and molecules [6,7] whose sizes may achieve 1 mm, and quantum dots [8] can be mentioned here. In [9,10], it is noted that this approximation does not provide correct expressions for cutoff frequency during generation of higher harmonics of intense laser radiation when it travels through dilute gases.

The purpose of this report is to analyze the probability of quantum transitions of microobjects induced by extremely short electromagnetic pulses without involvement of the electric dipole approximation. The next section contains the statement of the problem and general relations for the probability of transitions. Given that short pulse duration is defined as the smallness of field packet dimensions that are longitudinal with respect to the radiation propagation direction, the discussion is limited to the plane-wave approximation for the field. This is justified if the lateral dimensions of the packet are much larger than those of the object. These relations are examined hereafter with the main focus made on detecting the dependence of transition probability on pulse characteristics.

2. Statement of the problem and general relations

Non-stationary non-relativistic Schrödinger equation is taken as the basis [11,12]

$$i\hbar \frac{\partial \Psi}{\partial t} = (\hat{H}_0 + \hat{V})\Psi. \tag{1}$$

Here, $\hbar$ is the reduced Planck constant. The unperturbed Hamiltonian $\hat{H}_0$ is stationary (independent of the time t). Wave functions of stationary states with unperturbed system (particle or quantum microobject) energy $\hbar\omega_n$ are written as $\Psi_n^{(0)}(\mathbf{r}, t) = \psi_n(\mathbf{r}) \exp(-i\omega_n t)$. The perturbation $\hat{V}$ describes interaction between a particle with the charge e and mass m and an electromagnetic pulse when spin effects are not available or neglected [12]

$$\hat{V} = -\frac{e}{mc} \mathbf{A}\hat{\mathbf{p}} + \frac{e^2}{2mc^2} \mathbf{A}^2. \tag{2}$$

In (2), c is the speed of light in vacuum, $\hat{\mathbf{p}} = -i\hbar\nabla$ is the particle pulse and $\mathbf{A}$ is the field vector potential during calibration with the zero scalar potential.

Perturbation theory is used on the assumption that the perturbation is small. The wave function is expanded in the basis of the stationary states of the unperturbed system $\Psi_n^{(0)}(\mathbf{r}, t)$ in a standard way. The probability of transition induced by the finite duration pulse from the unperturbed

system state i to the state f at first order in perturbation theory is generally given by [11]

$$w_{fi} = \frac{1}{\hbar^2\omega_{fi}^2}\left|\int_{-\infty}^{+\infty}\frac{\partial V_{fi}}{\partial t}\,\exp(i\omega_{fi}t)dt\right|^2. \qquad (3)$$

Here, $\omega_{fi} = \omega_{gf} - \omega_i$ and the perturbation operator matrix elements

$$V_{fi} = \int \psi_f^*\hat{V}\psi_i d\mathbf{r}.$$

In first-order perturbation theory, the term in (2) that is quadratic in the vector potential may be neglected. The probability of transition (3) is written as [12]

$$w_{fi} = \frac{e^2}{m^2\omega_{fi}^2}\left|\int_{-\infty}^{+\infty}\psi_f^*\mathbf{E}\nabla\exp(i\omega_{fi}t)\psi_i d\mathbf{r}dt\right|^2, \qquad (4)$$

where

$$\mathbf{E} = -\frac{1}{c}\frac{\partial\mathbf{A}}{\partial t}$$

is the electric field intensity during calibration with the zero scalar potential. Note that expression (4) doesn't use the electric dipole (long-wavelength) approximation.

Let's now consider that radiation is a linearly polarized plane wave that travels along the x axis:

$$\mathbf{E} = E\left(t - \frac{x}{c}\right)\mathbf{e}_z \qquad (5)$$

($\mathbf{e}_z$ is the unit vector along the z axis). The plane-wave approximation is justified if the lateral dimensions of the radiation structure exceed those of the object. Then

$$w_{fi} = \frac{e^2}{m^2\omega_{fi}^2}\left|\int_{-\infty}^{+\infty}\psi_f^*\frac{\partial}{\partial z}\,\psi_i E\left(t - \frac{x}{c}\right)\right.$$

$$\left.\times\exp(i\omega_{fi}t)d\mathbf{r}dt\right|^2 = \frac{e^2}{m^2\omega_{fi}^2}I_{fi}I_E, \qquad (6)$$

where

$$I_{fi} = \left|\int\psi_f^*\frac{\partial}{\partial z}\,\psi_i\exp\left(i\,\frac{\omega_{fi}}{c}\,x\right)d\mathbf{r}\right|^2,$$

$$I_E = \left|\int_{-\infty}^{+\infty}E(t)\exp(i\omega_{fi}t)dt\right|^2. \qquad (7)$$

Thus, the transition probability is factorized: besides the transition frequency ω_{fi} and radiation polarization direction, the dependence on the radiation pulse characteristics is completely concentrated in I_E, while the dependence on the microobject states between which the transition takes place is concentrated in I_{fi}.

3. Transition probability analysis

Let's look first at the factor I_{fi}. By introducing the wavenumber $k_{fi} = \omega_{fi}/c$, it can be seen that $k_{fi}|x \ll 1$ is satisfied for transitions between the „ordinary" (not too highly excited) states, so $\exp(ik_{fi}x) \approx 1$ may be assumed. Then we arrive at a conventional dipole approximation [12]. In this case, the conventional selection rules are certainly kept.

For highly excited states (Rydberg atoms) and nanoparticles, the value of $k_{fi}|x|$ may be much higher than 1. Then, the following expansion may be involved

$$\exp(ik_{fi}x) = J_0(k_{fi}r\sin\theta) + 2\sum_{j=1}^{\infty}J_j(k_{fi}r\sin\theta)\cos(j\theta). \qquad (8)$$

This expansion is suitable for an important centrosymmetric (not necessarily Coulomb) potential case where the unperturbed Hamiltonian wave functions are written as [11]

$$\psi_{nlm} = R_{nl}(r)Y_{lm}(\theta,\varphi). \qquad (9)$$

Spherical (r, θ, φ), n, l coordinates and m — integer principal orbital and magnetic quantum numbers, $n = 1, 2, 3, \ldots$, $l = 0, 1, \ldots, n-1$ and $m = l, l-1, \ldots, -l$ are used here. The spherical functions are additionally factorized: $Y_{lm}(\theta,\varphi) = Q(\theta)\exp(im\varphi)$.

The quantum numbers are denoted as n, l, m for the initial state and as n', l', m' for the excited state. By substituting expansion (8) into the first equation (7) and integrating with respect to φ, it can be seen that a non-zero value of I_{fi} is possible in case of any variation of the magnetic number (in the range of values limited by the orbital quantum number):

$$m' - m = \pm j,\ \ j = 0, 1, 2, \ldots,\ \ |m'| \le l',\ \ |m| \le l. \qquad (10)$$

As indicated by two signs in the first equation (10), when $j > 0$, two states with different magnetic numbers are excited simultaneously.

Another selection rule states that the initial and excited states shall have different parity with respect to z (or to $\cos\theta$). Hence it follows that the orbital quantum number variation l shall be an odd number:

$$l' - l = \pm1, \pm3, \pm5, \ldots,\ \ 0 \le l' \le n' - 1,\ \ 0 \le l \le n - 1. \qquad (11)$$

For transitions between non-highly excited states of the „ordinary" atoms, the probabilities of „additional" transitions are low. In view of high accuracy of spectroscopic measurements, additional measurement data analysis is reasonable, however, this is not in the scope of this paper.

The main focus is made herein on the analysis of the pulse waveform factor I_E. For extremely short pulses having the duration τ_P much less than the optical transition period, $\omega_{fi}\tau_P \ll 1$, I_E coincides with the squared electric area of the pulse $\mathbf{S}_E = \int_{-\infty}^{+\infty}\mathbf{E}dt$: $I_E = S_E^2$. Wherein the transition fi may have an arbitrary multipolarity.

Generally, I_E is the squared absolute value of the spectral component of the electric intensity at ω_{fi}. Dependence of this factor on the pulse duration can be conveniently illustrated for a pulse with the waveform

$$E(t) = E_0 \exp(-t^2/\tau_p^2)\cos(\omega_p t + \varphi_p). \qquad (12)$$

For multicycle pulses, $\omega_p\tau_P \gg 1$, τ_p and ω_p have the meaning of a pulse duration and carrier frequency, for low-cycle pulses - such terminology is tentative. The pulse is purely unidirectional when $\omega_p = 0$. For the pulse with waveform (12), the spectrum is

$$E_\omega = \int_{-\infty}^{\infty} E(t)\exp(-i\omega t)dt$$

$$= \frac{\sqrt{\pi}}{2} E_0\tau_p \left\{ \exp\left[-\frac{1}{4}(\omega_p - \omega)^2\tau_p^2\right] e^{i\varphi_p} \right.$$

$$\left. + \exp\left[-\frac{1}{4}(\omega_p + \omega)^2\tau_p^2\right] e^{-i\varphi_p} \right\}, \qquad (13)$$

$$I_\omega = |E_\omega|^2 = \frac{\pi}{4}(E_0\tau_p)^2 \left\{ \left[\exp\left(-\frac{1}{4}(\omega_p - \omega)^2\tau_p^2\right) \right.\right.$$

$$\left. + \exp\left(-\frac{1}{4}(\omega_p + \omega)^2\tau_p^2\right)\right]^2 - \left[\exp\left(-\frac{1}{4}(\omega_p - \omega)^2\tau_p^2\right)\right.$$

$$\left.\left. + \exp\left(-\frac{1}{4}(\omega_p + \omega)^2\tau_p^2\right)\right]^2 \sin^2\varphi_p \right\}, \qquad (14)$$

the electrical area of the pulse is

$$S_E = E_{\omega=0} = \sqrt{\pi}E_0\tau_p \exp(-\omega_p^2\tau_p^2/4)\cos\varphi_p, \qquad (15)$$

the pulse energy (more precisely, a quantity proportional to it) is

$$W = \int_{-\infty}^{+\infty} E^2(t)dt$$

$$= \frac{\sqrt{\pi}}{2\sqrt{2}} E_0^2\tau_p \left[1 + \exp(-\omega_p^2\tau_p^2/2)\cos(2\varphi_p)\right]. \qquad (16)$$

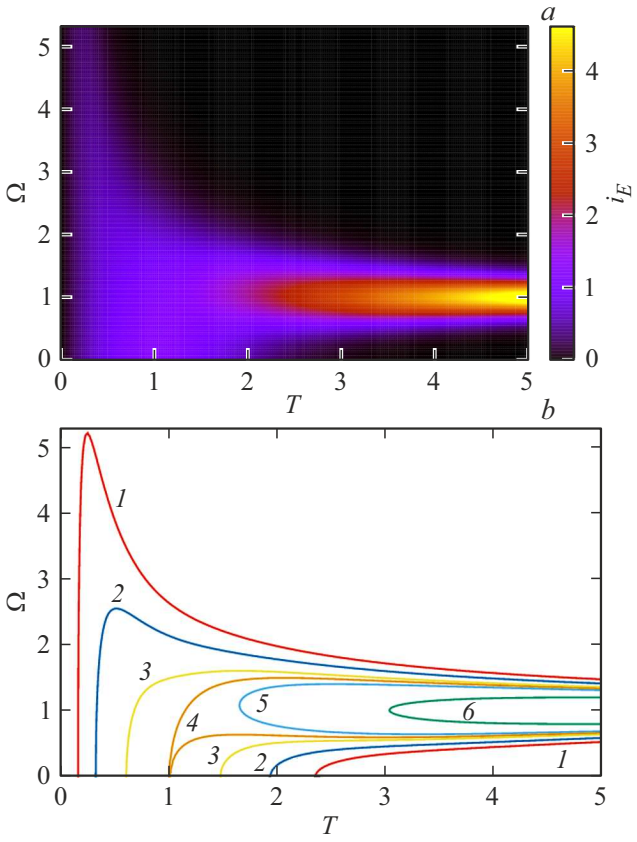
Finally,

$$I_E = I_{\omega_{fi}} = \frac{\pi}{4}(E_0\tau_p)^2 \left\{ \left[\exp\left(-\frac{1}{4}(\omega_p - \omega_{fi})^2\tau_p^2\right) \right.\right.$$

$$\left. + \exp\left(-\frac{1}{4}(\omega_p + \omega_{fi})^2\tau_p^2\right)\right]^2 - \left[\exp\left(-\frac{1}{4}(\omega_p - \omega_{fi})^2\tau_p^2\right)\right.$$

$$\left.\left. + \exp\left(-\frac{1}{4}(\omega_p + \omega_{fi})^2\tau_p^2\right)\right]^2 \sin^2\varphi_p \right\}. \qquad (17)$$

I_ω, S_E^2 and I_E are maximum when $\sin\varphi_p = 0$, therefore this condition is adopted hereinafter. Then, the spectral density I_ω and factor I_E have their maximum when $\omega = 0$, if $\omega_p\tau_p < 1/\sqrt{2}$, and minimum in the opposite case. It is more correct to compare the efficiency of microobject excitation by pulses with the coinciding energy W. For this, dimensionless „pulse duration“ $T = \omega_{fi}\tau_p$ and „carrier frequency“ $\Omega = \omega_p/\omega_{fi}$ are introduced. Thus, $I_E = \sqrt{\pi/2}(W/\omega_{fi})i_E$. W/ω_{fi} may be associated with the number of photons with ω_{fi} whose total energy is equal to W. The dimensionless pulse waveform factor is



(a) Dependence of the field factor i_E on the dimensionless pulse duration T and pulse frequency Ω. (b) The same shown in the form of lines of the level $i_E(T, \Omega) = 0.3$ (curve *1*), 0.6 (*2*), 1 (*3*), 1.21 (*4*), 1.5 (*5*) and 3 (*6*).

$$i_E = \frac{T}{1 + \exp(-\Omega^2T^2/2)}$$

$$\times \left\{ \exp\left[-(\Omega - 1)^2T^2/4\right] + \exp\left[-(\Omega + 1)^2T^2/4\right] \right\}^2. \qquad (18)$$

For multicycle resonance pulses $\Omega = 1$, $\Omega T \gg 1$, hence $i_E = T$. This corresponds to the asymptotically constant probability of object excitation in a unit of time. In other cases, as the pulse duration increases, i_E first grows reaching its maximum and then decreases as shown in the figure. Resonance excitation appears to be more effective than in the purely unidirectional pulse case ($\Omega = 0$), when $T > 1.1$, while for shorter pulses the situation is reversed.

Discussion

Under the assumptions made in the paper, the standard dipole transition approximation is justified if the microobject size is much greater than the wavelength associated with this transition ($\lambda_{fi} = 2\pi c/\omega_{fi}$) and for pulse durations that are less than the transition period ($2\pi/\omega_{fi}$). It is reasonable that for perturbation theory to be applicable, the transition probability shall remain small, $w_{fi} \ll 1$. Moreover, the pulse duration shall be lower than the relaxation times.

The described factorization of the role of quantum microobject and exciting radiation packet characteristics appears to be possible in the plane-wave pulse waveform case (5). Deviations from this waveform are possible for field packets focused into a region smaller than the object dimensions, for example, of the Rydberg atom. This would allow microobject tomography to be made. However, then separation of the microobject and radiation factors is no longer possible, which leads to removal of the remaining prohibitions on quantum transitions. In this case, only numerical calculations of these processes are probably available. This entanglement is a sign of a pronounced non-point-like nature of the microobject leading to the spatiotemporal dispersion effects that are important at the macrolevel for crystals [13,14].

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Conflict of interest

The author declares that he has no conflict of interest.

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