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Probability of absorption and emission by an atom interacting with ultrashort laser pulses

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We consider the process of transition of a two-level system to the excited state with subsequent photon emission in the presence of a laser pulse with a high degree of unipolarity. Within the framework of quantum electrodynamics, we obtain analytical expressions for the differential probability of the process depending on the temporal scales of the problem: laser pulse duration, excited-state lifetime, inverse transition frequency, and inverse frequency of the photon emitted. Besides, we calculate the total absorption probability by integrating over the three-dimensional photon momentum and summing over polarizations. We compare the results obtained for unipolar and bipolar (many-cycle) pulses.

Keywords: laser field, ultrashort pulses, two-level system, absorption, emission.

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1. Introduction

The theoretical description of the processes of absorption and emission of light by an atom was one of the most important tasks as part of quantum theory even at the stage of its formation at the beginning of the last century. First, a quantum mechanical approach to this problem [1,2] was proposed, and then a more rigorous formalism was developed within the framework of quantum electrodynamics (QED), which allowed, for example, to move on to the review of the spectral line profile [3]. As part of the modern theory, spontaneous emission of photons is accepted to occur when an atom transitions from an excited state due to interaction with a quantized electromagnetic field. The instability of the excited state is associated with an imaginary addition to the energy of the corresponding level, which arises due to the radiative QED corrections and determines the level width Γ. Thus, the process of spontaneous emission has been well studied as part of QED and described in many textbooks and monographs [4].

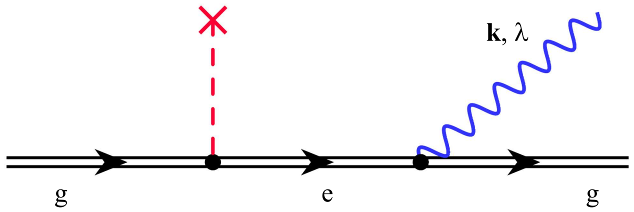
As is well known, for an atom to transition to an excited state, it must absorb energy through interaction with some other system. In many cases, such a system is a classical external field with a given amplitude and frequency, which simulates the laser radiation field. Since for non-stationary problems within QED energy states are introduced only asymptotically at large and small times, when there is no interaction, the absorption process must be considered together with radiation due to the presence of a finite lifetime $\sim 1/\Gamma$ of the excited state [5]. In this work, such a non-stationary formulation of the problem is reviewed and the dependence of the probability of the process on the parameters of the field and the atomic system in the regime of very short interaction times, i.e. short laser pulse durations, is studied. In this case, there is no concept of laser frequency and, as a consequence, there is no concept of resonance. In this mode, the probability depends in a nontrivial way on the time scales in the problem: the duration of the laser pulse τ, the lifetime of the excited state $1/\Gamma$, the inverse frequency of the transition $1/\omega_0$ and the inverse frequency of the emitted photon $1/\omega$. In this work, closed-form expressions for the probability will be obtained that describe this dependence.

Our research is motivated by ongoing developments in experimental capabilities in the generation of ultrashort laser pulses. In particular, obtaining pulses of attosecond duration is already possible [6]. These currently include an $\sim\tau$ width envelope, which contains several field oscillations. As the number of these oscillations decreases, it is possible to proceed to reviewing unipolar laser pulses of linear polarization, for which the corresponding projection of the electric field strength $E(t)$ predominantly has one sign [7–11]. In this case, the most important characteristic of the laser pulse is its electric-field area

$$S_E = \int\limits_{-\infty}^{\infty} E(t)dt. \tag{1}$$

It will be shown below that in the regime of short durations τ the probability of energy absorption followed by photon emission is largely determined by the value S_E.

In this work, the probability of interest to us will be calculated as the square of the modulus of the amplitude



Feynman diagram describing the process under study. An atom (two-level system) transitions from the ground state „g" to the excited state „e" due to the absorption of energy from the external laser field, and then emits a photon with momentum **k** and polarization λ, returning to the ground state. Photon $\omega = |\mathbf{k}|$ frequency. The double line denotes the wave function or Green's function, which accurately accounts for the Coulomb interaction in the atom.

of the process demonstrated in the figure. The interaction with the external classical field (vertex with a cross) is taken into account in the first order of perturbation theory (if the probability reaches values close to unity, then it is required to take into account higher orders, which imposes a limitation on the intensity of the laser field). The atom is considered as a two-level system, which is a reasonable approximation provided that the populations of the remaining levels in the process under study are small. The external field will mainly contain Fourier harmonics of relatively low frequencies, and we consider that these frequencies do not correspond to any resonances in the low-energy part of the system spectrum. Given the fact that the atom is initially in the ground state, we can take into account only one excited state „e". To calculate the required matrix elements, we will use the dipole approximation, implying that the size of the atom is significantly smaller than the characteristic spatial scale of the laser field (for a multi-cycle pulse this is the wavelength, and for a unipolar pulse — the parameter $c\tau$, where c is the speed of light).

The units used in this article are $\hbar = c = 1$ ($\hbar$ is the Planck constant). Charge units correspond to $\alpha = e^2/(4\pi)$ ($e < 0$ is electron charge, α is fine structure constant).

2. Calculating the probability of a process

Let us first obtain a general expression for the amplitude of the process presented in the figure, without specifying the explicit form of the 4-potential of the external field $\mathscr{A}(x)$. We will follow the conclusion from the work [5]. In accordance with Feynman rules, the amplitude is given by the following expression:

$$S_{\mathbf{k},\lambda} = e^2 \int d^4x_1 \int d^4x_2 \overline{\psi}_{\mathrm{g}}(x_2) \hat{\varepsilon}^*_{\mathbf{k},\lambda} \Phi^*_{\mathbf{k},\lambda}(x_2) \times S(x_2, x_1) \hat{\mathscr{A}}(x_1) \psi_{\mathrm{g}}(x_1), \tag{2}$$

where $\overline{\psi} \equiv \psi^\dagger \gamma^0$, $\hat{V} \equiv \gamma^\mu V_\mu$, γ^μ — Dirac matrices, $\varepsilon^\mu_{\mathbf{k},\lambda}$ — 4-vector of photon polarization (momentum and polarization of the photon are denoted by **k** and λ, respectively), $\Phi_{\mathbf{k},\lambda}$ — space-time part of the photon wave function and, finally, $S(x_2, x_1)$ — bound electron propagator. For 4-coordinate vectors the designation $x = \{t, \mathbf{r}\}$ is used. The wave function of the ground state of an atom can be written as

$$\psi_{\mathrm{g}}(x) = \psi_{\mathrm{g}}(\mathbf{r}) \mathrm{e}^{-iE_0 t}, \tag{3}$$

where E_0 — is the corresponding energy. The wave function of the excited state of a two-level system has the form

$$\psi_{\mathrm{e}}(x) = \psi_{\mathrm{e}}(\mathbf{r}) \mathrm{e}^{-iE_1 t}. \tag{4}$$

The transition energy is $\omega_0 = E_1 - E_0$. The wave function of a photon has the following form:

$$\Phi_{\mathbf{k},\lambda}(x) = \frac{1}{\sqrt{2\omega}} \mathrm{e}^{-i(\omega t - \mathbf{kr})}, \tag{5}$$

where $|\mathbf{k}| = \omega$. The bound electron propagator is given by

$$S(x_2, x_1) = \int \frac{d\tilde{\omega}}{2\pi} \mathrm{e}^{-i\tilde{\omega}(t_2 - t_1)} \sum_n \frac{\psi_n(\mathbf{r}_2)\overline{\psi}_n(\mathbf{r}_1)}{\tilde{\omega} - E_n(1 - i0)}, \tag{6}$$

the expression in which the summation is carried out over the entire spectrum of the atomic system, E_n — energies of states ψ_n. The integral over $\tilde{\omega}$ is calculated using the standard contour for the causal Green's function. The contribution from $n = g$ is significant only when reviewing soft photons, for which $\omega \to 0$. Since we will assume that the energy of the emitted photons lies in the region separated from zero, in the sum over n there remains only one term with $n = e$.

Integrating over time t_2 and variable $\widetilde{\omega}$ in expression (2), we obtain the following result ($t_1 = t$):

$$S_{\mathbf{k},\lambda} = \frac{4\pi\alpha}{\sqrt{2\omega}} \int_{-\infty}^{\infty} dt \int d\mathbf{r}\, \mathrm{e}^{i\omega t} \langle \mathrm{g}|\hat{\varepsilon}^*_{\mathbf{k},\lambda} \mathrm{e}^{-i\mathbf{kr}}|\mathrm{e}\rangle \times \overline{\psi}_{\mathrm{e}}(\mathbf{x}) \hat{\mathscr{A}}(x) \psi_{\mathrm{g}}(\mathbf{x}) \frac{1}{\omega - \omega_0 + i\Gamma/2}. \tag{7}$$

For the matrix element involving spatial coordinates, the designation is used

$$\langle n|f(\mathbf{r})|m\rangle \equiv \int d\mathbf{r}\, \overline{\psi}_n(\mathbf{r}) f(\mathbf{r}) \psi_m(\mathbf{r}). \tag{8}$$

Also in equation (7) the standard substitution $E_1 \to E_1 - i\Gamma/2$ was made in order to regularize the denominator. The natural width Γ arises when radiative corrections to the electron wave function are taken into account and determines the lifetime of the excited state [4].

Let us consider the classical field of a laser pulse having an electric component $\mathscr{E}(x) = \mathbf{e}_{\mathrm{L}} \mathscr{E}(x)$, where

$$\mathscr{E}(x) = \mathscr{E}_{\mathrm{L}} \mathrm{e}^{-t^2/\tau^2} \sin(\omega_{\mathrm{L}} t - \mathbf{k}_{\mathrm{L}}\mathbf{r} + \varphi_0). \tag{9}$$

Here $\mathbf{e}_{\mathrm{L}}$ is unit vector defining the polarization of the laser pulse, $\mathscr{E}_{\mathrm{L}}$ — amplitude of the laser pulse, τ — its duration,

ω_{L} — „frequency" of the laser field and φ_0 — relative phase between the Gaussian envelope profile and „carrier". In the case of pulses with a high degree of unipolarity, the value of $\omega_{\mathrm{L}}\tau$ is small and the frequency does not have a standard definition as in the case of monochromatic radiation. In the $\mathscr{A}_0 = 0$ gauge we have $\mathscr{E}(x) = -\partial_t \mathscr{A}(x)$, so the vector potential has the form $\mathscr{A}(x) = \mathbf{e}_{\mathrm{L}}\mathscr{A}(x)$, where

$$\mathscr{A}(x) = \frac{\sqrt{\pi}}{2}\,\mathscr{E}_{\mathrm{L}}\tau\,\mathrm{e}^{-\omega_{\mathrm{L}}^2\tau^2/4}\mathrm{Re}$$

$$\times\left\{\mathrm{e}^{-i\mathbf{k}_{\mathrm{L}}\mathbf{r}+i\varphi_0}\left[\mathrm{erfi}\left(\frac{it}{\tau}+\frac{\omega_{\mathrm{L}}\tau}{2}\right)-\mathrm{erfi}\left(\frac{\omega_{\mathrm{L}}\tau}{2}\right)\right]\right\}. \quad (10)$$

In order to calculate the integral over t in expression (7), we note that asymptotically $\lim\limits_{t\to\pm\infty}\mathscr{A}(x) = \mathscr{A}_{\pm}(\mathbf{r})$, where $\mathscr{A}_{\pm}(\mathbf{r})$ are other than zero. Thus, this integral, which is essentially a Fourier transform, will contain a contribution with generalized functions that have support at the point $\omega = 0$. Since we are reviewing a situation where ω is far from zero, these contributions can be neglected. Taking this into account, integrating by parts in expression (7), we obtain

$$\int\limits_{-\infty}^{\infty} dt\,\mathrm{e}^{i\omega t}\mathscr{A}(x) = -\frac{i}{\omega}\int\limits_{-\infty}^{\infty} dt\,\mathrm{e}^{i\omega t}\mathscr{E}(x). \quad (11)$$

As a result, for amplitude (7) we have

$$S_{\mathbf{k},\lambda} = \frac{\sqrt{2}\pi^{3/2}\alpha}{\omega^{3/2}}\,\mathscr{E}_{\mathrm{L}}\tau\,\langle \mathrm{g}|\hat{\varepsilon}^*_{\mathbf{k},\lambda}\mathrm{e}^{-i\mathbf{kr}}|\mathrm{e}\rangle\,\frac{1}{\omega-\omega_0+i\Gamma/2}$$

$$\times\Big[\langle \mathrm{e}|\hat{\mathbf{e}}_{\mathrm{L}}\mathrm{e}^{i\mathbf{k}_{\mathrm{L}}\mathbf{r}}|\mathrm{g}\rangle\mathrm{e}^{-i\varphi_0}\mathrm{e}^{-(\omega-\omega_{\mathrm{L}})^2\tau^2/4} -$$

$$-\langle \mathrm{e}|\hat{\mathbf{e}}_{\mathrm{L}}\mathrm{e}^{-i\mathbf{k}_{\mathrm{L}}\mathbf{r}}|\mathrm{g}\rangle\mathrm{e}^{i\varphi_0}\mathrm{e}^{-(\omega+\omega_{\mathrm{L}})^2\tau^2/4}\Big]. \quad (12)$$

Let us note that the multiplier containing Γ determines the Lorentz contour, which has a maximum at $\omega = \omega_0$. Strictly speaking, in the current order of perturbation theory it is also required to take into account a diagram in which the order of the vertices is changed — first a photon is emitted, then interaction with the laser field occurs. However, such a diagram will contain a denominator $(-\omega-\omega_0+i\Gamma/2)$, which is significantly larger in magnitude than the denominator in expression (12) due to the realistic condition $\Gamma \ll \omega_0$ and the fact that we are not interested in the number density of photons with $\omega \to 0$, and the radiation will mainly occur at frequencies close to the transition frequency ($\omega \approx \omega_0$). Amplitude (12) contains two terms, the origin of which is related to the expansion of the sine in formula (9). In the mode of multicycle pulses with a well-defined frequency ($\omega_{\mathrm{L}}\tau \gg 1$), the second term is strongly suppressed, since an atom in the ground state cannot emit a photon and at the same time transfer energy to the laser field. The exponential corresponding to the absorption of the laser radiation quantum, on the contrary, makes a large contribution at $\omega \approx \omega_{\mathrm{L}}$, which, under the condition of resonance $\omega_{\mathrm{L}} = \omega_0$, leads to a high probability of the process. Various effects associated with the finite interaction time in the case of $\omega_{\mathrm{L}}\tau \gg 1$ were recently discussed in work [5]. Since in this work we are interested in the mode in which the laser pulse is close to unipolar, the $\omega_{\mathrm{L}}\tau \gg 1$ condition is violated and the concepts of laser frequency and laser field quanta cannot be introduced. In this case, it is required to take into account both terms in expression (12).

In order to evaluate the matrix elements in square brackets in formula (12), we will work in the dipole (nonrelativistic) approximation. Let us replace $\exp(\pm i\mathbf{k}_{\mathrm{L}}\mathbf{r})$ by one and use the fact that the matrix element of the Dirac matrices γ corresponds to the matrix element of the velocity operator $\hat{\mathbf{p}}/m$ in the non-relativistic approximation [4]. It is not difficult to obtain

$$\langle \mathrm{e}|\hat{\mathbf{e}}_{\mathrm{L}}\mathrm{e}^{i\mathbf{k}_{\mathrm{L}}\mathbf{r})}|\mathrm{g}\rangle \approx \langle \mathrm{e}|\hat{\mathbf{e}}_{\mathrm{L}}\mathrm{e}^{-i\mathbf{k}_{\mathrm{L}}\mathbf{r})}|\mathrm{g}\rangle \approx -i\omega_0\mathbf{e}_{\mathrm{L}}\mathbf{r}_{\mathrm{eg}}, \quad (13)$$

where $\mathbf{r}_{\mathrm{eg}} = \langle \mathrm{e}|\mathbf{r}|\mathrm{g}\rangle$.

Now, squaring the amplitude (12) modulo, we obtain the following expression:

$$|S_{\mathbf{k},\lambda}|^2 = 4\pi^3\alpha^2\,\frac{\omega_0^2\mathscr{E}_{\mathrm{L}}^2\tau^2}{\omega^3}\left|\langle \mathrm{g}|\hat{\varepsilon}^*_{\mathbf{k},\lambda}\mathrm{e}^{-i\mathbf{kr}}|\mathrm{e}\rangle\right|^2\left|\mathbf{e}_{\mathrm{L}}\mathbf{r}_{\mathrm{eg}}\right|^2$$

$$\times\left(\cosh\omega\omega_{\mathrm{L}}\tau^2-\cos 2\varphi_0\right)\,\frac{\mathrm{e}^{-(\omega^2+\omega_{\mathrm{L}}^2)\tau^2/2}}{(\omega-\omega_0)^2+\Gamma^2/4}. \quad (14)$$

This value specifies the probability density of the process in accordance with the rule

$$\frac{dW_{\mathbf{k},\lambda}}{d^3\mathbf{k}} = \frac{|S_{\mathbf{k},\lambda}|^2}{(2\pi)^3}. \quad (15)$$

In expression (14), the trigonometric cosine containing the phase φ_0 arises due to the interference of the terms in square brackets in formula (12). First of all, let us note that in the limit of large $\omega_{\mathrm{L}}\tau$ this cosine is small compared to the hyperbolic cosine, which can be replaced by a growing exponential, which will result in the square of the modulus of the first term in (12), and we obtain the result that was discussed in detail in work [5]. In the same work, it was shown how, in the dipole approximation, sum over polarizations λ, integrate over photon emission angles and frequency ω in order to obtain the total probability of the process W. We will first focus on analyzing the differential probability (14), (15) in the short laser pulse mode, and then find the total probability.

3. Unipolar and multi-cycle pulses

Expression (14) specifies the probability density for the process of energy absorption by an atom followed by photon emission depending on a number of parameters. Let us rewrite the square of the amplitude modulus in the following form:

$$|S_{\mathbf{k},\lambda}|^2 = 8\pi^2\alpha^2 \frac{\omega_0^2}{\omega^3} \left|\langle g|\hat{\varepsilon}^*_{\mathbf{k},\lambda} e^{-i\mathbf{kr}}|e\rangle\right|^2 \times \left|\mathbf{e}_L \mathbf{r}_{eg}\right|^2 \frac{e^{-\omega^2\tau^2/2}}{(\omega-\omega_0)^2+\Gamma^2/4} \times \left[\frac{1}{2}\Theta^2\left(\cosh\omega\omega_L\tau^2 - 1\right)e^{-\omega_L^2\tau^2/2} + S_E^2\right], \quad (16)$$

where we introduced the electric area of the envelope

$$\Theta = \int_{-\infty}^{\infty} \mathscr{E}_L e^{-t^2/\tau^2} dt = \sqrt{\pi}\mathscr{E}_L\tau. \quad (17)$$

The electric area of the pulse is equal to

$$S_E = \int_{-\infty}^{\infty} \mathscr{E}_L e^{-t^2/\tau^2} \sin(\omega_L t + \varphi_0) dt = \sqrt{\pi}\mathscr{E}_L\tau\, e^{-\omega_L^2\tau^2/4}\sin\varphi_0. \quad (18)$$

Let us analyze the square of the amplitude module at $\omega = \omega_0$, when the Lorentz contour gives the maximum value:

$$\left|S^{(0)}_{\mathbf{k},\lambda}\right|^2 \equiv |S_{\mathbf{k},\lambda}|^2\Big|_{\omega=\omega_0} = \frac{32\pi^2\alpha^2\tau}{\Gamma^2} \times \left|\langle g|\hat{\varepsilon}^*_{\mathbf{k},\lambda} e^{-i\mathbf{kr}}|e\rangle\right|^2 \left|\mathbf{e}_L\mathbf{r}_{eg}\right|^2 \mathcal{S}_0, \quad (19)$$

where we have introduced

$$\mathcal{S}_0 = \frac{e^{-\omega_0^2\tau^2/2}}{\omega_0\tau}\left[\frac{1}{2}\Theta^2\left(\cosh\omega_0\omega_L\tau^2 - 1\right)e^{-\omega_L^2\tau^2/2} + S_E^2\right]. \quad (20)$$

Let us consider in more detail several limiting cases.

Multi-cycle pulse. Case $\omega_L\tau \gg \omega_0\tau \gg 1$.

We have

$$\mathcal{S}_0 \approx \frac{1}{4}\frac{1}{\omega_0\tau}\Theta^2 e^{-\omega_L^2\tau^2/2} = \frac{\pi}{4}\frac{\mathscr{E}_L^2\tau^2}{\omega_0\tau} e^{-\omega_L^2\tau^2/2}. \quad (21)$$

Multi-cycle pulse. Case $\omega_0\tau \gg \omega_L\tau \gg 1$.

In this case, the small exponential factor will contain the frequency ω_0:

$$\mathcal{S}_0 \approx \frac{1}{4}\frac{1}{\omega_0\tau}\Theta^2 e^{-\omega_0^2\tau^2/2} = \frac{\pi}{4}\frac{\mathscr{E}_L^2\tau^2}{\omega_0\tau} e^{-\omega_0^2\tau^2/2}. \quad (22)$$

Multi-cycle pulse. Case $\omega_L\tau = \omega_0\tau \gg 1$.

Since the laser field frequency is in resonance with the atomic transition energy, there will be no exponential factor:

$$\mathcal{S}_0 \approx \frac{1}{4}\frac{1}{\omega_0\tau}\Theta^2 = \frac{\pi}{4}\frac{\mathscr{E}_L^2\tau^2}{\omega_0\tau}. \quad (23)$$

Let us note that in all these cases the result does not depend on the phase of φ_0, as noted above.

Let us now turn to consideration of extremely short pulses.

Unipolar impulse. Case $\omega_L\tau \ll 1$, $\omega_0 \lesssim \omega_L$.

The hyperbolic cosine can be replaced by one, so that the answer is proportional to the square of the electrical area of the laser pulse:

$$\mathcal{S}_0 \approx \frac{1}{\omega_0\tau} S_E^2 = \pi\frac{\mathscr{E}_L^2\tau^2}{\omega_0\tau}\sin^2\varphi_0. \quad (24)$$

Depending on the phase of φ_0, this quantity takes values from zero to $\pi\mathscr{E}_L^2\tau^2/(\omega_0\tau)$, i.e. may be significantly less than probability (23), or may exceed it four times.

Finally, note that for $\omega_L\tau \ll 1$ and $\omega_0\tau \gg 1$, the argument of the hyperbolic cosine in formula (20) can be either small or large. In appropriate limiting cases, the result is given by one of the expressions presented above.

Function (20) has already been discussed in work [12], where the excited state population of a two-level system during interaction with the classical field of a laser pulse was reviewed (see also works [13,14]). Formula (16) contains this expression, since the excitation of an atom by a laser field is part of the diagram in the figure (taking into account the approximations made). On the other hand, differential probability (16) also describes the process of emission of a photon with arbitrary energy ω, given the finite lifetime of the excited state. Next, we will calculate the total probability by integrating expression (16) in the case of an extremely short laser pulse $\omega_L\tau \ll 1$, taking into account $\omega_0 \lesssim \omega_L$ and $\Gamma\tau \ll 1$.

This calculation can be performed by analogy with work [5]. Let us write down the final result:

$$W \approx \frac{8\pi\alpha^2}{3\Gamma}\omega_0^3\left|\mathbf{r}_{ge}\right|^2\left|\mathbf{e}_L\mathbf{r}_{eg}\right|^2 S_E^2. \quad (25)$$

Using the relation for the matrix element [4]

$$\left|\mathbf{r}_{ge}\right|^2 = \frac{3\Gamma}{4\alpha\omega_0^3}, \quad (26)$$

we obtain

$$W \approx 2\pi\alpha\left|\mathbf{e}_L\mathbf{r}_{eg}\right|^2 S_E^2 = \frac{1}{2}\left|\mathbf{e}_L\mathbf{d}_{eg}\right|^2 S_E^2. \quad (27)$$

Here $\mathbf{d}_{eg}$ is the matrix element of the electric dipole moment operator $\hat{\mathbf{d}} = e\mathbf{r}$. Thus, as in expression (24), we see that in the case of extremely short pulses, the total probability of radiation when excited by a laser field is determined by the electrical area of the pulse. Since here we have integrated over the photon momentum and summed over polarizations, the total probability W can be associated with the population of the excited level, so that formula (27) reproduces one of the expressions obtained in the work [12] (a similar result was also obtained in work [15], where the probability of excitation of a hydrogen atom from the ground state was considered within non-relativistic quantum mechanics).

4. Conclusion

In this work, the process of photon emission by a two-level system under excitation by a laser field of arbitrary duration was studied. The main attention was focused on the analysis of extremely short pulses that do not have a well-defined frequency and have a non-zero electric-field area. Within the framework of QED, a closed expression was obtained for the differential probability of the process in the first order of perturbation theory, which allows one to obtain the spectrum of emitted photons. The total probability was also calculated by summing over the parameters of the emitted photon. It was shown that for extremely short laser pulses the probability is, to first order, proportional to the square of the electric-field area of the pulse.

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Conflict of interest

The authors declare that they have no conflict of interest.

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