

Model of Interaction of Zero-Duration Laser Pulses With an Atom

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ABSTRACT

The soluble model of interaction of a finite series of zero - duration pulses with an atom is considered. The model is based on the nowadays laser techniques providing duration of pulses of a few femtoseconds and even less, and intensities higher than 10^{14} - 10^{20} Wt/cm².

Keywords: laser pulse interaction with atom

1. INTRODUCTION

Investigation of electromagnetic pulse interaction with quantum systems has led to discoveries of new phenomena and effects in the last two decades. One of them is the phenomenon of the above-threshold ionization of atoms¹ (a remarkable review of the theory of two-electron atom in the strong light field was given in the paper²). The other one is the adiabatic stabilization of atoms against photoionization in the intense laser pulse (cf. Ref.³ and references therein). Recently, the adiabatic stabilization of excitons in an intense THz laser pulse has been predicted in the case of GaAs system⁴. These particular findings illustrate just some of new interesting applications of modern laser technology⁵.

Nowadays laser facilities allow generation of pulses with durations of a few and even fractions of a femtosecond and intensities up to the order of 10^{14} - 10^{20} Wt/cm² (Ref.⁶). Recent proposals promise to achieve in the near future the durations of 10^{-17} s (Ref.⁷) and even 10^{-21} – 10^{-22} s (Ref.⁸). This makes possible to consider the models of zero-duration electromagnetic pulses interacting with a quantum system⁹⁻¹¹. Such a model belongs to so called soluble models what is of considerable importance for predictions of results which can be expected in the case of real short pulses. In particular, the effect of localization of the Rydberg atom in a train of ultrashort unidirectional weak electric-field pulses has been observed recently^{10,12}.

In the most of theoretical treatments the semiclassical and even classical methods are widely used, one - dimensional models are considered. However, the general problem of interaction of a many - electron quantum system with zero-duration electromagnetic pulses requires a proper quantum mechanical framework. In this paper we construct the model of interaction of a train of zero-duration pulses with an atom. The model allows, to some extent, the exact solution, what is important for future applications. Particular emphasis is put on the case of the single pulse and its effect on a hydrogen atom is investigated. Atomic units are used throughout the paper.

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2. THEORY

2.1. General formulas

The time - dependent Schrödinger equation including the interaction of electromagnetic radiation with the atom can be written, in the dipole approximation, in the form:

$$i\frac{\partial}{\partial t}|\Psi\rangle = H|\Psi\rangle + V(t)|\Psi\rangle. \quad (1)$$

In equation (1) H is the Hamiltonian of the atom. In the length gauge to be used further $V(t) = -\sum_i \vec{\mathcal{E}} \vec{r}_i$, where summation is performed over all electrons of the atom and $\vec{\mathcal{E}} = -\frac{1}{c} \frac{\partial \vec{A}(t)}{\partial t}$. With a definite choice of the short pulse shape of the vector potential $\vec{A}(t)$ we can write approximately

$$V(t) = V \sum_{n=0}^{N-1} A_n \delta(t - n\tau). \quad (2)$$

In (2) the potential spatial part $V = \vec{e} \sum_i \vec{r}_i$, \vec{e} is the polarization operator that is perpendicular to the wave vector and τ is the period between the pulses. The approximation (2) corresponds to the model of "kicked" excitations⁹ in the case of half-cycle pulses (HCPs). It is equivalent to that used in the theoretical treatment of 1D "kicked" Rydberg atoms^{10,12}. However in contrast to^{10,12}, we exploit below a fully quantum mechanical 3D approach.

Now we pass on to the interaction representation. For this purpose we transform

$$|\Psi\rangle = e^{-iHt} |\phi\rangle. \quad (3)$$

For $|\phi\rangle$ the following equation is obtained

$$i\frac{\partial}{\partial t}|\phi\rangle = e^{iHt} V(t) e^{-iHt} |\phi\rangle = \bar{V}(t) |\phi\rangle \quad (4)$$

with the initial and boundary conditions: $|\phi\rangle_{t \rightarrow -\infty} = |0\rangle$ is the ground state of the atom before the field begins its action, $|\phi\rangle_{t \rightarrow \infty} = |f\rangle$ is a final state when the action of the pulse series terminates. It is evident that

$$e^{-iHt} |s\rangle = e^{-iE_s t} |s\rangle,$$

where $|s\rangle$ is the eigenfunction of the operator H , describing a certain state of the atom.

The solution of (4) by means of its traditional transformation into the integral equation¹⁴ is not effective here, because we have to determine the values $|\phi(k\tau)\rangle$, which can not be calculated from the equation. In the points $t = k\tau$ the overlap between the step gap of the function $|\phi(t)\rangle$ and the strong singularity of the δ -function takes place*. To overcome this difficulty we introduce the evolution operator of the form

$$|\phi(t)\rangle = e^{S(t,t_0)} |\phi(t_0)\rangle \quad (5)$$

Inserting (5) into (4), we obtain

$$S' + \frac{1}{2!} [S, S'] + \frac{1}{3!} [S, [S, S']] + \dots = -i\bar{V}(t) \quad (6)$$

Let us consider first the interval $-\infty < t < \tau$. Inside this interval the obvious solution of eq. (6) looks like

$$S(t, -\infty) = -iA_0 V \theta(t) \quad (7)$$

*L.A. Melnikov paid our attention to this circumstance

and, consequently,

$$|\phi(t)\rangle = e^{-iA_0 V \theta(t)} |0\rangle \quad (8)$$

Consider now the interval $t_1 < t < 2\tau$ with $0 < t_1 < \tau$. In this interval we look for the solution in the form

$$S(t, t_1) = -iA_1 \exp(iH\tau)V \exp(-iH\tau)\theta(t - \tau) + P(t_1). \quad (9)$$

For the operator $P(t_1)$ we have the complex nonlinear differential equation from (6), but there is no sense to solve it if we remember well known properties of the evolution operator

$$e^{S(t, t')} = e^{S(t, t'')} e^{S(t'', t')}, \quad e^{S(t, t)} = 1.$$

It follows from the condition $S(t_1, t_1) = 0$ that $P(t_1) = 0$ and, besides that

$$\begin{aligned} e^{S(t, -\infty)} |0\rangle &= e^{S(t, t_1)} e^{S(t_1, -\infty)} |0\rangle = e^{-iA_1 \exp(iH\tau)V \exp(-iH\tau)\theta(t - \tau)} e^{-iA_0 V} |0\rangle \\ &\equiv e^{iH\tau} e^{-iA_1 V \theta(t - \tau)} e^{-iH\tau} e^{-iA_0 V} |0\rangle. \end{aligned} \quad (10)$$

Continuing this procedure of the solution of eq. (6) in different intervals we obtain at $t \rightarrow +\infty$

$$|\phi(\infty)\rangle = e^{S(+\infty, -\infty)} |0\rangle = \prod_{n=0}^{N-1} [e^{in\tau H} e^{-iA_n V} e^{-in\tau H}] |0\rangle \quad (11)$$

The solution (11) keeps the additivity property of the initial equation (4), i.e., if one puts there $\tau = 0$ (all the δ -functions combine in a single one), then the coefficient before the unified δ -function is $A = \sum A_i$. The same property we observe in (11). Note, that the solution (11) is exact and correct for all finite values A_i .

Here we have to say a few words about the result obtained in the paper of Klews and Schweizer¹³. That result coincides with ours if one uses the Pade approximation for the exponent

$$e^{-iAV} = \frac{1 - i\frac{A}{2}V}{1 + i\frac{A}{2}V},$$

and it, naturally, is valid only for small intensities A (see Fig. 2).

The amplitude t_{f0} of transition from the initial state of the atom into a certain final state assumes the form:

$$t_{f0} = \langle f | \prod_{n=0}^{N-1} [e^{in\tau H} e^{-iA_n V} e^{-in\tau H}] |0\rangle. \quad (12)$$

The probability density of such a transition is

$$w_{f0} = |t_{f0}|^2. \quad (13)$$

It is easy to verify that $\sum_f w_{f0} d\rho_f = 1$, with $d\rho_f$ being the density of final states. For convenience we shall further denote $\hat{F}_n = -1 + e^{-iA_n V}$ and use the formula

$$\prod_{s=0}^{N-1} (1 + a_s) = 1 + \sum_{s_1=0}^{N-1} a_{s_1} + \sum_{s_1 > s_2}^{N-1} a_{s_1} a_{s_2} + \dots$$

In this case the amplitude t_{f0} in (12) will be expressed by the sum

$$t_{f0} = \delta_{f0} + t_{f0}^{(1)} + t_{f0}^{(2)} + \dots + t_{f0}^{(N)} \quad (14)$$

where the term $t_{f0}^{(s)}$ describes the transition from the state $|0\rangle$ into the state $|f\rangle$ via $(s - 1)$ of intermediate states.

The characteristic period τ between the pulses determines the characteristic energy $\varepsilon = (\frac{2\pi}{\tau})$ of the photon being transferred by the field to the atom. If we measure this energy in electron-volts (eV) and the period in femtoseconds ($\text{fs} = 10^{-15}$ s), then $\varepsilon = 4.134/\tau$. Assume that very short pulses have a long period of τ (hundreds of femtoseconds and even picoseconds). In this case it is possible to consider the problem of influence of such a pulse train on transitions in the atom, taking all A_n to be equal to A . From equation (14) it follows that

$$t_{f0}^{(1)} = \frac{1 - e^{iN\tau(E_f - E_0)}}{1 - e^{i\tau(E_f - E_0)}} \langle f | \hat{F} | 0 \rangle \quad (15)$$

$$t_{f0}^{(2)} = \sum_{\nu} \frac{\langle f | \hat{F} | \nu \rangle \langle \nu | \hat{F} | 0 \rangle}{1 - e^{i\tau(E_{\nu} - E_0)}} \left[\frac{1 - e^{iN\tau(E_f - E_{\nu})}}{1 - e^{i\tau(E_f - E_{\nu})}} - \frac{1 - e^{iN\tau(E_f - E_0)}}{1 - e^{i\tau(E_f - E_0)}} \right] \quad (16)$$

and so on.

Let us first consider the direct transition term $t_{f0}^{(1)}$. The function

$$\chi = \frac{1 - e^{iN\tau(E_f - E_0)}}{1 - e^{i\tau(E_f - E_0)}} \quad (17)$$

has sharp peaks with sufficiently large N at $E_m = E_0 + m\varepsilon$, and at these finite energies $|\chi(\tau)| = N$ (Fig.1). At all other values of E_f the function $|\chi(\tau)|$ varies in the vicinity of unity. Thus, it is the states into which the atom has been transformed by absorbing an integer number of photons that are prominent in the whole spectrum of the atom.

If the state $|f\rangle$ lies within a bound part of spectrum, then in order to efficiently populate a particular bound state, it is necessary to exactly hit this state, i.e. to know an exactly determined energy of the photon.

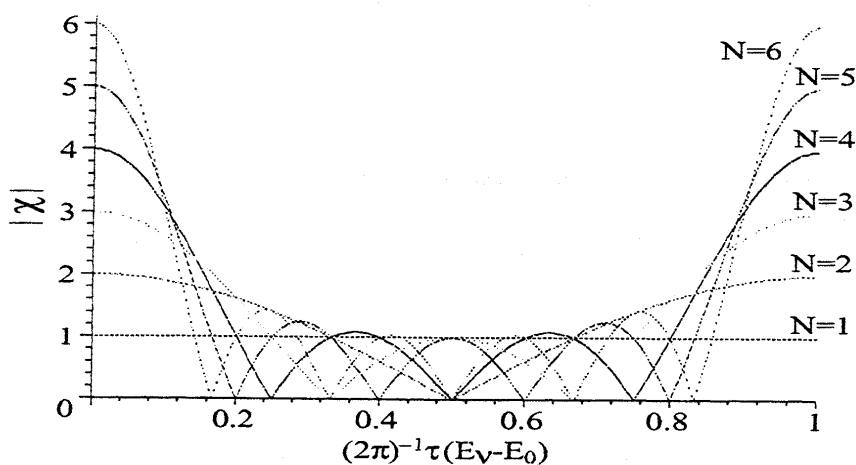


Fig 1. $|\chi(\tau)|$ as a function of the number of pulses $N = 1, 2, \dots, 6$.

It is easier to observe such "brightness peaks" in the ionization spectrum (in the literature the peaks in the atomic energy spectrum arising above the ionization threshold are referred to as ATI - above threshold ionization^{1,2}).

The structure of the τ -dependent function in the term $t_{f0}^{(2)}$ is rather complicated. However, it is also possible to show that if the final state satisfies again the condition $E_m = E_0 + m\varepsilon$, and the transition into the intermediate state $|\nu\rangle$ is onto the level $E_s = E_0 + s\varepsilon$, then at sufficiently high N we can write:

$$t_{m0}^{(2)} \simeq \frac{N(N-1)}{2!} \sum_s \langle m | \hat{F} | s \rangle \langle s | \hat{F} | 0 \rangle \quad (18)$$

Thus the amplitude in the "brightness peaks" assumes the form:

$$t_{m0} \simeq \delta_{f0} + N \langle m|\hat{F}|0 \rangle + \frac{N(N-1)}{2!} \sum_{s_1} \langle m|\hat{F}|s_1 \rangle \langle s_1|\hat{F}|0 \rangle + \frac{N(N-1)(N-2)}{3!} \sum_{s_1, s_2} \langle m|\hat{F}|s_1 \rangle \langle s_1|\hat{F}|s_2 \rangle \langle s_2|\hat{F}|0 \rangle + \dots \quad (19)$$

Since in the problem under consideration ε can amount to tenths and even hundredths of the electron-volt, the peaks are very frequent, beyond the measurement accuracy, and one should deal with certain averaged values. Thus, it is possible to write in an approximate way:

$$\sum_s |s\rangle \langle s| \approx \hat{I} \quad (20)$$

where \hat{I} is the unit operator. In this case, for large N equation (19) leads to the following approximate expression:

$$t_{m0} \simeq \langle m|(\hat{F} + 1)^N|0 \rangle = \langle m|e^{-iNAV}|0 \rangle. \quad (21)$$

Note, for a series of the pair pulses with equal intensity A but opposite sign, $A_{2s} = A$, $A_{2s+1} = -A$, $s = 0, \dots, [N/2]$, the transition amplitude (12) tends to zero, $t_{m0} \approx 0$, that assumes a stabilization effect at small τ .

2.2. The model of single and two pulses

This section describes the action of a zero-duration single pulse on the hydrogen atom. The transition amplitude is

$$t_{f0} = \langle f|e^{-iAV}|0 \rangle \quad (22)$$

To estimate the value of the coupling constant A it is useful to employ the ratio $A = 0.22 \times 10^{-6} \sqrt{IT}$, where I is the laser radiation intensity measured typically in Wt/cm^2 (radiation of such power is said to be intense), and T (in femtoseconds) stands for duration of the actual pulse that is approximated by the δ function. For example, if $I \approx 10^{14} \text{ Wt}/\text{cm}^2$ then $A \approx 2.2T$. If characteristic durations are a few femtoseconds, then A is measured within ten a.u.

It is seen from (22), that for hydrogen the transition operator is exactly equal to that used for the description of the first Born matrix elements for (e,2e) reactions if one puts the transferred momentum from the fast incoming electron to the atom being equal $\vec{Q} = A\vec{e}$ (see, for instance,¹⁵). Probability densities here are practically analytical functions¹⁶. The transition probability from the ground state to some excited state $|nl\rangle$ is described by the formula:

$$w_{nl} \equiv w_{nl,10} = 4(2l+1) \left| \int_0^\infty r^2 R_{nl}(r) j_l(Ar) e^{-r} dr \right|^2, \quad (23)$$

with

$$R_{nl} = 2 \left(\frac{1}{n} \right)^{3/2} \sqrt{\frac{(n-l-1)!}{n(n+l)!}} \left(\frac{2r}{n} \right)^l L_{n-l-1}^{2l+1} \left(\frac{2r}{n} \right) e^{-r/n}.$$

The following formula takes place¹⁷

$$w_n = \sum_{l=0}^{n-1} w_{nl} = \frac{256 n^7 (-1 + 3A^2 n^2 + n^2) A^2 (A^2 n^2 + n^2 - 2n + 1)^{(n-3)}}{3 (A^2 n^2 + n^2 + 2n + 1)^{(n+3)}}, \quad (24)$$

which for small $A \ll 1$ corresponds to that of Pade approximation

$$w_n \approx \frac{256 n^7 (n-1)^{(2n-5)}}{3 (n+1)^{(2n+5)}} A^2 \left[1 + A^2 \frac{n^2(n^2-9)}{(n+1)^2} \right] + O(A^6)$$

Fig. 2 represents the probabilities of several transitions into low - lying states of the hydrogen atom calculated by formula (23), depending on the coupling constant A . Two peculiarities are to be noted. It is known that in the case of transitions caused by ordinary dipole interaction it is the transitions with $\Delta l = \pm 1$ that "survive". In our case the transitions into any final state are represented. But the transitions that do not satisfy the rule of selection for the orbital quantum number are suppressed in comparison with the transitions that satisfy this rule. This can easily be seen when comparing w_{21} and w_{20} , w_{31} with w_{30} and w_{32} and so on.

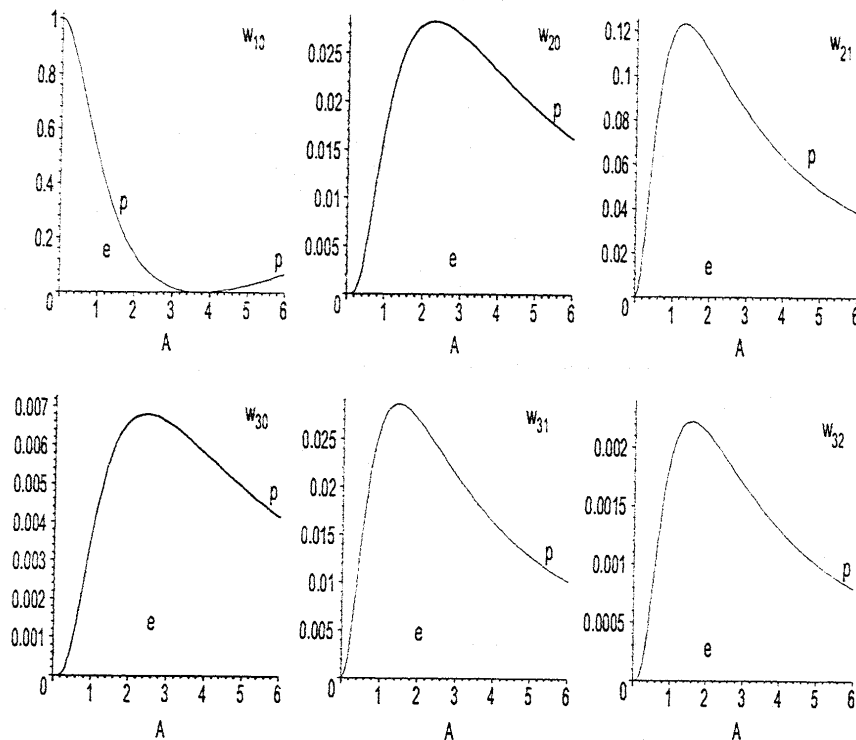


Fig.2. Dependence of the excitation probability w_{nl} on the laser pulse intensity A : (e) eq. (22); (p) Padé approximation of the evolution operator.

The differential transition probability to a continuum state with the momentum $|\vec{p}\rangle$ (photoionization) after integration over the azimuthal angle φ_p takes the form

$$dw_{\vec{p}} = \frac{128A^2p}{(1 - \exp(-\frac{2\pi}{p}))} \frac{[A^2 + 2Apx + (1 + p^2)x^2] \exp(-\frac{2}{p} \arctg \frac{2p}{A^2 - p^2 + 1})}{[A^2 + 2Apx + (1 + p^2)]^4 [(A^2 - p^2 + 1)^2 + 4p^2]} dp dx, \quad (25)$$

where $-1 < x = \cos \theta_{\vec{e}\vec{p}} < 1$ and the principal value of $\arctg(\dots)$ lies in $[0, \pi)$. Integrating (25) over x , we obtain¹⁷

$$dw_p = \frac{256A^2p}{3(1 - \exp(-\frac{2\pi}{p}))} \frac{(3A^2 + p^2 + 1) \exp(-\frac{2}{p} \arctg \frac{2p}{A^2 - p^2 + 1})}{(A^2 + 1 + 2Ap + p^2)^3 (A^2 + 1 - 2Ap + p^2)^3} dp. \quad (26)$$

Figs. 3 and 4 present the probability densities $w_{\vec{p}}$ and w_p calculated by eqs. (25) and (26), correspondingly. One can see significant differences from the case of ordinary dipole interaction: there is almost no scattering in the direction of the polarization vector, but only in the opposite direction, with the probability reaching the maximum at $A \approx 1.4$.

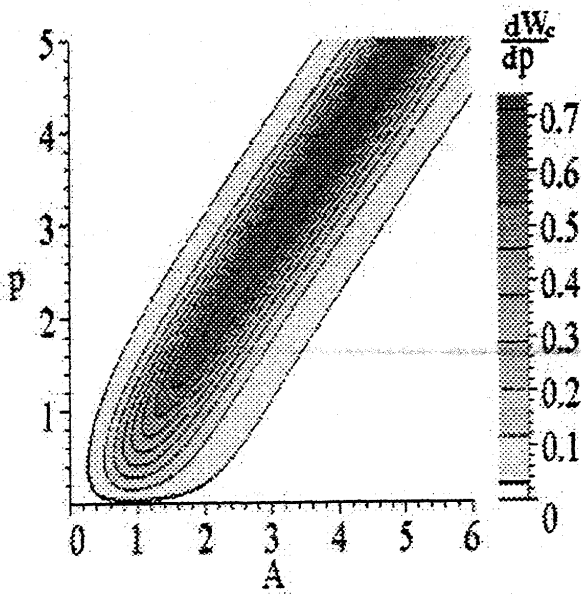


Fig 3. Plot of the ionization probability density w_p versus the pulse intensity A and the momentum p .

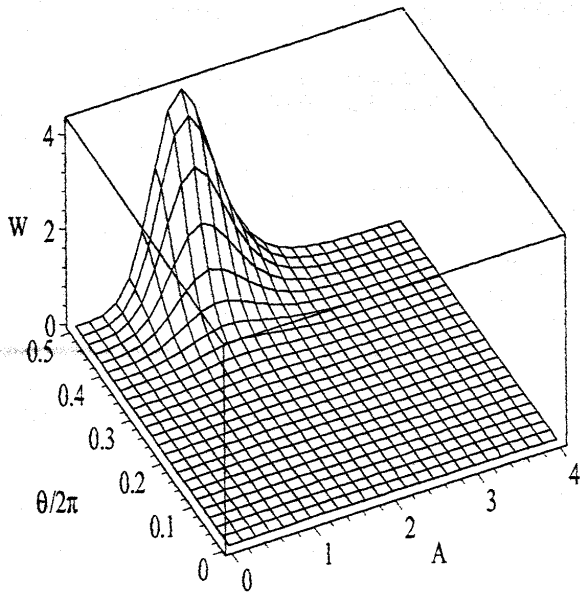


Fig 4. Ionization probability density $w_{\bar{p}}$ as a function of the pulse intensity A and the angle $\theta = \theta_{\bar{p}}$. $p = 1$ or $E_f = 13,6$ ev.

At last, integrating Eq. (26) over p , and summing up Eq. (24) by n , we obtain the full ionization probability w_c and the full excitation one $w_d = \sum_n w_n$ versus of the pulse intensity A , that sum equals unit, i.e. $w_c + w_d = 1$ (see, Fig. 3).

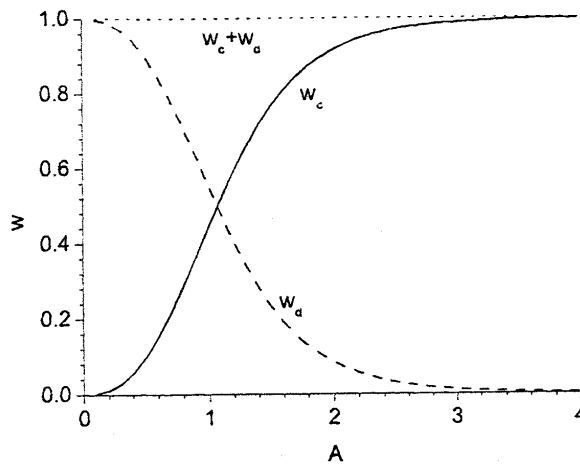


Fig. 5. Dependence of the full excitation w_d and ionization w_c probabilities on the pulse intensity A . Their sum $w_d + w_c = 1$.

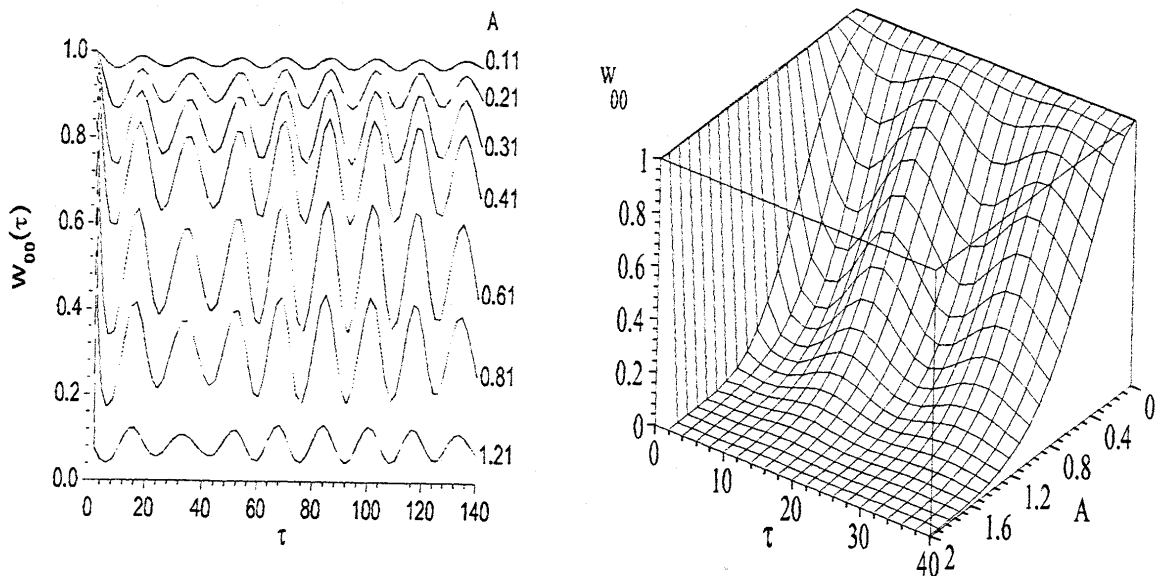
We can also ascribe to the subject of this subsection the interesting case $A_0 = -A_1 = A$ which models one

period of a real pulse. Eq. (12) gives for the transition $|0\rangle \rightarrow |0\rangle$

$$t_{00}(\tau) = e^{-i\frac{1}{2}\tau} \sum_{\nu} \langle 0 | e^{-iA_1 V} | \nu \rangle e^{-iE_{\nu}\tau} \langle \nu | e^{-iA_0 V} | 0 \rangle = e^{-i\frac{1}{2}\tau} \left[\int_0^{\infty} w_p e^{-i\frac{p^2}{2}\tau} dp + \sum_{n=1}^{\infty} w_n e^{i\frac{1}{2n^2}\tau} \right], \quad (27)$$

where w_p and w_n are defined in (24) and (26).

The probability $w_{00}(\tau) = |t_{00}(\tau)|^2$ for hydrogen are shown on Figs. 6-7. The ground state probability w_{00} decreases from 1 at $A = 0$ to values of the order of 10^{-3} at $A = 2$ for all chosen meanings of $\tau > 0$. Worthly of note that $w_{00}(\tau \rightarrow 0) \sim 1$ for any finite value of A . This means that we always can establish such a small value of τ that the probability is close to one. This fact justifies the above assumption on the stabilization effect for the series of pair pulses with equal intensity, but opposite sign, what follows from eqs. (12) and (21). The probability w_{00} like the function of τ oscillates and the oscillation magnitude increases from 0 at $A = 0$ to 0.3 at $A = 0.6$ and after that decreases up to 0 with increasing of A . On Figs. 6-7 the period τ is in atomic units. We remind that 1 a.u. of time is equal 0.024 fs.



Figs. 6-7. The ground state probability w_{00} for hydrogen versus the pulse intensity A and the time τ between two pulses.

2.3. Pulse series

In this section we shall outline the effect of pulse series with long period on the hydrogen atom. Formula (21) shows that the influence has a coherent character, and a pulse series enhance the effect of a single pulse. Even if the constant A is actually small the product NA can be rather big. Thus, due to a coherent character of the action the problem of interaction of series of relatively weak, short and sparse laser pulses with the hydrogen atom is reduced to a well known problem of interaction of the fast projectile electron with an atom in the first Born approximation. The constant A appears to become N times greater.

However, this condition is valid not for all the final states but only for those that satisfy the condition $E_m = E_0 + m\varepsilon$. It is evident that if the photon energy ε is fixed by the potentialities of the emitter then in the

general case it is rather difficult to satisfy the coherence conditions for transitions into the Rydberg states. It is easier to do so for transitions into the continuum states.

In Fig. 8 the full probability $w_c(N)$ for series of N identical pulses is presented. In calculations we used the approximate formulas (20)-(21). It is easy to see that there is no stabilization of Rydberg states within this model. This is not surprising, because our model of the pulse train assumes the large period between pulses compared with the relaxation time. In Fig. 9 the ground state probability w_{00} for hydrogen versus the pulse intensity A is displayed by the gray band for different values $2 < \tau < 140$ between two opposite pulses calculated by formula (27) and by two boundary lines $N = 1, 2$ calculated by approximated formula (21) at $N = 1$ and $N = 2$ for identical pulses.

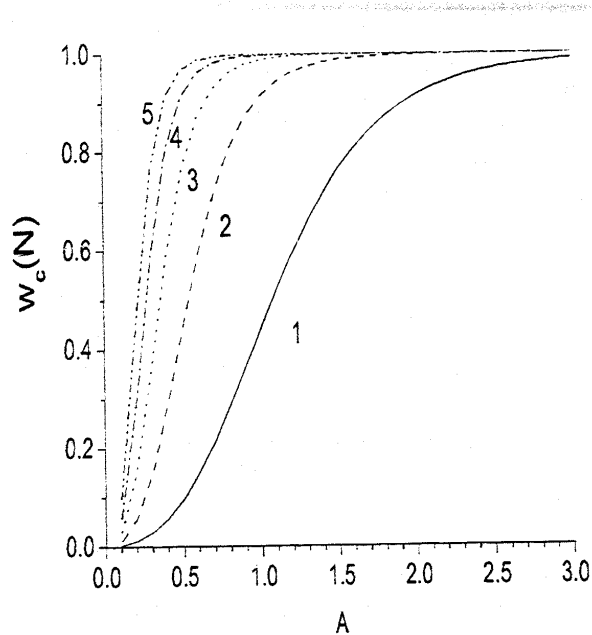


Fig. 8. The full ionization probability $w_c(N)$ for hydrogen from ground state versus the pulse intensity A at a different number of the identical pulses N .

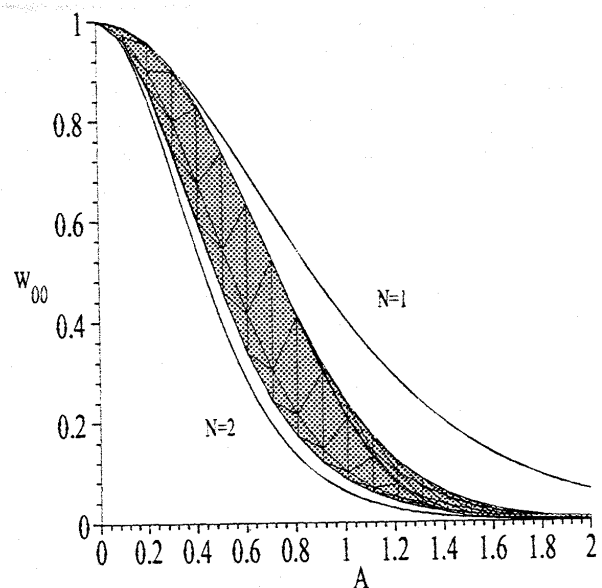


Fig. 9. The ground state probability w_{00} for hydrogen versus the pulse intensity A : the gray band calculated by formula (27) for different values $2 < \tau < 140$ between two opposite pulses ($A_0 = -A_1 = A$, $N = 2$), boundary lines $N = 1, 2$ calculated by approximated formula (21) with $A_0 = A_1 = A$.

3. BRIEF CONCLUSION

The presented exactly soluble model of the zero-duration pulses exhibits a number of peculiarities that distinguish the interaction of the ultrashort and powerful electromagnetic pulse with the atom from the interaction of, say, synchrotron radiation that is usually described in the framework of dipole approximation and one-photon exchange. It would be interesting to verify in experiment the results obtained for the case of a real short pulse.

The simple coincidence of the form of transition operator with that for $(e,2e)$ reactions is valid only for the hydrogen atom. Already for the helium one encounters the considerable difference. In this respect, it would

be interesting to investigate the multiple ionization reactions induced by laser pulse for the purposes of the spectroscopy of electron - electron correlations in quantum systems.

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