Numerical simulations of the quantum systems dynamics in the path integral approach

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Abstract

We study the dynamics of quantum system interacting with electromagnetic field. We present density matrix and transition probability as a path integrals in energy state space without resonance and rotating wave approxinations. By the use of obrained equations we develop an algorithm for numerical simulations of the dynamics of quantum system interacting with electromagnetic field. Using this approach we consider rotational dynamics of nitrogen molecules $^{14}N_2$ and $^{15}N_2$ which interact with a sequence of ultrashort laser pulses. Our computer simulations indicate the complex dependency of the high rotation states excitation probability upon ultrashort laser pulses sequence periods. We observe pronounced resonances, which correspond to the results of some experiments.

Keywords: Path integral formalism; Numerical simulation; Quantum optics; Non-resonance processes

1. Introduction

The modern development of laser radiation technologies induces theoretical and experimental investigations of the dynamics of quantum objects (such as atoms or molecules) under the action of intense electromagnetic field of different forms.

This dynamics is principally non-linear, because the probability is high of multiphoton processes (absorption and emission more the one photon) and nonresonant processes (electromagnetic field frequency is far from quantum transitions frequency). We note the recent studies of different rare gases multiphoton ionization [1, 2, 3], of multiphoton photoemission of the Au(111) surface state with 800-nm laser pulses [4], of multiphoton transitions in GaSb/GaAs quantum-dot intermediate-band solar cells [5], of three-photon electromagnetically induced absorption in a ladder-type atomic system [6].

There are certain difficulties for theoretical studies of these processes and for simulations of quantum objects dynamics that interact with laser field. Thus, different approximations are used. For example, there are two- or three-level quantum system models [7] and rotating wave approximation [8]. For high-intensity laser field the perturbation theory runs into problems. It is necessary to calculate the large number of terms. High-order perturbation theory for miltilevel quantum system dynamics was considered in [9]. For theoretical researches of this processes the numerical solution of time-dependent Schrödinger equation is used [10]. For this reason different schemes of space-time discretization is realized. The discretization parameter should be small enough for simulations of a minute error.

We present original non perturbative method of transition probability calculation in path integral approach [11]. In this paper we present original approach for numerical simulations of the dynamics of a quantum system, interacting with laser radiation by path integration in energy states space.

Recent experimental [12] and theoretical [13, 14] investigations point at possibilities of selective excitations of nitrogen isotopes by a sequence of ultrashort laser pulses (a pulse train). We have developed and are applying numerical algorithm to quantum resonances problem in molecule rotational excitation by ultrashort laser pulses.

2. Mathematical model of quantum system interacting with electromagnetic field

We consider interaction of multilevel quantum system (such as an atom or a molecule) with electromagnetic field. The Hamiltonian $\hat{H}_{full}$ describing our model is given as

$$\hat{H}_{full} = \hat{H}_{syst} + \hat{V}, \tag{1}$$

where $\hat{H}_{syst}$ is Hamiltonian of the investigated quantum system. We define stationary eigenstates $|l\rangle$ with energies E_l having the following properties:

$$\hat{H}_{syst} = \sum_{l=0}^{N-1} E_l |l\rangle\langle l|, \tag{2}$$

$$\sum_{l=0}^{N-1} |l\rangle\langle l| = 1, \qquad \langle l'|l\rangle = \delta_{l'l}; \tag{3}$$

$\hat{V}$ – the interaction operator.

Our main goal is to define the probability $P(l_f, t|l_{in}, 0)$ of investigated quantum system transition from eigenstate $|l_{in}\rangle$ at the moment $t = 0$ to the one $|l_f\rangle$ at the moment $t > 0$.

We describe the investigated system by statistical operator $\hat{\rho}(t)$. The evolution equation of $\hat{\rho}(t)$ in Dirac (interaction) picture [15] is as follows:

$$\hat{\rho}(t) = \hat{U}_D(t)\hat{\rho}(0)\hat{U}_D^+(t), \tag{4}$$

where $\hat{\rho}(0)$ — statistical operator at initial time moment $t = 0$,

$$\hat{U}_D(t) = T\exp[-\frac{\imath}{\hbar}\int\limits_0^t \hat{V}_D(\tau)d\tau] \tag{5}$$

– the evolution operator in Dirac picture,

$$\hat{V}_D(\tau) = \exp[\frac{\imath}{\hbar}\hat{H}_{syst}\tau]\hat{V}(\tau)\exp[-\frac{\imath}{\hbar}\hat{H}_{syst}\tau] \tag{6}$$

– the operator of quantum system and electromagnetic field interaction in Dirac picture.

Eq. (4) in energy representation on the base of eigenvectors Eq. (3) is

$$\rho_{l_f m_f}(t) = \sum_{l_{in},m_{in}} \langle l_f|\hat{U}_D(t)|l_{in}\rangle\rho_{l_{in}m_{in}}\langle m_{in}|\hat{U}_D^+(t)|m_f\rangle, \tag{7}$$

where

$$\rho_{l_f m_f}(t) = \langle l_f|\hat{\rho}(t)|m_f\rangle, \quad \rho_{l_{in},m_{in}} = \langle l_{in}|\hat{\rho}(0)|m'_{in}\rangle \tag{8}$$

are density matrix elements in energy representation at time moment $t = 0$.

The probability of a quantum state observation is to define as diagonal matrix element. At initial time moment $t = 0$ it is equal to $\rho_{l_{in}l_{in}}(t = 0) = \rho_{l_{in}}$. At final time moment t it is equal to $\rho_{l_f l_f}(t) = \rho_{l_f}(t)$.

Eq. (7) describes evolution of the probability of a quantum state observation:

$$\rho_{l_f}(t) = \sum_{l_{in}} \langle l_f|\hat{U}(t)|l_{in}\rangle\langle l_{in}|\hat{U}^\dagger|l_f\rangle\rho_{l_{in}}, \tag{9}$$

where $l_{in},\ l_f = 1, 2, \ldots$.

The quantum transition probability from state $|l_{in}\rangle$ or $\rho_{l_{in}}(0) = \delta_{l_{in}n_{in}}$) at time moment $t = 0$ to state $|l_f\rangle$ or $\rho_{l_f}(t) = \rho_{l_f m_f}(t)\delta_{l_f m_f}$ at time moment $t > 0$ is to describe as follows

$$P(l_f, t|l_{in}, t) = \langle l_f|\hat{U}_D(t)|l_{in}\rangle\langle l_f|\hat{U}_D(t)|l_{in}\rangle \tag{10}$$

By the use of eq. (10) we present eq. (9) in the following

$$\rho_{l_f}(t) = \sum_{l_{in}} P\left(l_f, t|l_{in}, 0\right)\rho_{l_{in}}(0) \tag{11}$$

For numerical calculation $\rho_{l_f m_f}(t)$, $\rho_{l_f}(t)$, $P(l_f, t|l_{in}, 0)$ by the use of eq. (7), eq. (9) and eq. (10) we need to know matrix elements $\langle l_f|\hat{U}(t)|l_{in}\rangle$ of evolution operator $\hat{U}(t)$.

For that reason we use evolution operator $\hat{U}$ group properties and express the evolution operator $\langle l_f|\hat{U}_D(t)|l_{in}\rangle$ as

$$\hat{U}_D(t) = \prod_{k=1}^{K+1} \hat{U}_D(t_k, t_{k-1}), \tag{12}$$

as long as $t_k > t_{k-1}$ and where

$$\hat{U}_D(t_k, t_{k-1}) = \exp[-\frac{\imath}{\hbar}\int\limits_{t_{k-1}}^{t_k} \hat{V}_D(\tau)d\tau], \tag{13}$$

here we introduce the notations $t_{K+1} = t$, $l_{K+1} = l_f$, $t_0 = 0$, $l_0 = l_{in}$, $\sum\limits_{k=1}^{K+1}(t_k - t_{k-1}) = t$.

By the use of eq. (12) and completeness condition Eq. (3) of eigenvectors $|l_k\rangle$ basis the kernel $\langle l_f|\hat{U}_D|l_{in}\rangle$ can be expessed as

$$\langle l_f|\hat{U}_D(t)|l_{in}\rangle = \sum_{l_1,\ldots l_K=0}^{N-1} \prod_{k=1}^{K+1}\langle l_k|\hat{U}_D(t_k, t_{k-1})|l_{k-1}\rangle, \tag{14}$$

where

$$\langle l_k|\hat{U}_D\left(t_k,t_{k-1}\right)|l_{k-1}\rangle = \langle l_k|\exp\left[-\frac{\imath}{\hbar}\int\limits_{t_{k-1}}^{t_k}\hat{V}_D(\tau)d\tau\right]|l_{k-1}\rangle \tag{15}$$

We consider the evolution operator kernel Eq. (15) as a series and for the time interval $(t_k - t_{k-1}) \to 0$ it is

$$\langle l_k|\hat{U}_D(t_k,t_{k-1})|l_{k-1}\rangle = \langle l_k|l_{k-1}\rangle - \frac{\imath}{\hbar}\int\limits_{t_{k-1}}^{t_k}\langle l_k|\hat{V}_D(\tau)|l_{k-1}\rangle d\tau. \tag{16}$$

Using eq. (6), interaction operator matrix element in Dirac picture $\langle l_k|\hat{V}_D(\tau)|l_{k-1}\rangle$ is expressed

$$\langle l_k|\hat{V}_D(\tau)|l_{k-1}\rangle = V_{l_k l_{k-1}}(\tau)\exp[\imath\omega_{l_k l_{k-1}}\tau]. \tag{17}$$

where $V_{l_k l_{k-1}}(\tau) = \langle l_k|\hat{V}(\tau)|l_{k-1}\rangle$ – interaction operator matrix element, $\omega_{l'l} = (E_{l'} - E_l)/\hbar$ – frequency of quantum transition between eigenstates with eigenvalues (energies) $E_{l'}$ and E_l.

It is possible to prove that for small time interval $(t_k - t_{k-1}) \to 0$ the evolution operator kernel $\langle l_k|\hat{U}_D(t_k,t_{k-1})|l_{k-1}\rangle$ eq. (16) can be expressed as

$$\langle l_k|\hat{U}_D(t_k,t_{k-1})|l_{k-1}\rangle = \int\limits_0^1 \exp[\imath S\left[l_k,l_{k-1};\xi_{k-1}\right]]d\xi_{k-1}, \tag{18}$$

where $S\left[l_k,l_{k-1}\xi_{k-1}\right]$ – dimensionless (in $\hbar$ units) action in energy representation during time interval $(t_k - t_{k-1})$

$$S\left[l_k,l_{k-1};\xi_{k-1}\right] = 2\pi(l_k - l_{k-1})\xi_{k-1} - \int\limits_{t_{k-1}}^{t_k}\frac{V_{l_k l_{k-1}}(\tau)}{\hbar}2\cos[2\pi(l_k - l_{k-1})\xi_{k-1} - \omega_{l_k l_{k-1}}\tau]d\tau, \tag{19}$$

where $V_{l_k l_{k-1}}(\tau) = \langle l_k|\hat{V}(\tau)|l_{k-1}\rangle$ – interaction operator matrix element.

For this proof, by using eq. (15) we transform eq. (18) into eq. (16).

By the use of eq. (15) we present eq. (14) in the following

$$\langle l_k|\hat{U}_D(t_k,t_{k-1})|l_{k-1}\rangle =$$

$$= \int\limits_0^1 \exp[2\pi\imath(l_k - l_{k-1})\xi_{k-1}]\exp[-\frac{\imath}{\hbar}\int\limits_{t_{k-1}}^{t_k}V_{l_k l_{k-1}}(\tau)2\cos[2\pi(l_k - l_{k-1})\xi_{k-1} - \omega_{l_k l_{k-1}}\tau]d\tau]d\xi_{k-1} \tag{20}$$

If $(t_k - t_{k-1}) \to 0$ then we write

$$\exp[-\frac{\imath}{\hbar}\int\limits_{t_{k-1}}^{t_k}V_{l_k l_{k-1}}(\tau)2\cos[2\pi(l_k - l_{k-1})\xi_{k-1} - \omega_{l_k l_{k-1}}\tau]d\tau] \simeq$$

$$\simeq 1 - \frac{\imath}{\hbar}\int\limits_{t_{k-1}}^{t_k}V_{l_k l_{k-1}}(\tau)2\cos[2\pi(l_k - l_{k-1})\xi_{k-1} - \omega_{l_k l_{k-1}}\tau]d\tau \tag{21}$$

where

$$2\cos[2\pi(l_k - l_{k-1})\xi_{k-1} - \omega_{l_k l_{k-1}}\tau] = e^{-\imath[2\pi(l_k - l_{k-1})\xi_{k-1} - \omega_{l_k l_{k-1}}\tau} + e^{+\imath[2\pi(l_k - l_{k-1})\xi_{k-1} - \omega_{l_k l_{k-1}}\tau}. \tag{22}$$

Using eq. (21) and eq. (22) we present eq. (20) in the following

$$\langle l_k|\hat{U}_D(t_k,t_{k-1})|l_{k-1}\rangle =$$

$$= \int\limits_0^1 \exp[2\pi\imath(l_k - l_{k-1})\xi_{k-1}]d\xi_{k-1} -$$

$$-\frac{\imath}{\hbar}\int\limits_{t_{k-1}}^{t_k}V_{l_k l_{k-1}}(\tau)\int\limits_0^1(\exp[4\pi\imath(l_k - l_{k-1})\xi_{k-1} - \imath\omega_{l_k l_{k-1}}\tau] + \exp[\imath\omega_{l_k l_{k-1}}\tau])d\xi_{k-1}d\tau =$$

$$= \int\limits_0^1 \exp[2\pi\imath(l_k - l_{k-1})\xi_{k-1}]d\xi_{k-1} -$$

$$-\frac{\imath}{\hbar}\int\limits_{t_{k-1}}^{t_k}V_{l_k l_{k-1}}(\tau)\int\limits_0^1\{\exp[\imath\omega_{l_k l_{k-1}}\tau] + \exp[4\pi\imath(l_k - l_{k-1})\xi_{k-1}]\cdot\exp[-\imath\omega_{l_k l_{k-1}}\tau]\}\,d\tau d\xi_{k-1} \tag{23}$$

We note that

$$\int_0^1 \exp[4\pi\imath(l_k - l_{k-1})\xi_{k-1}]d\xi_{k-1} = \delta_{l_k l_{k-1}}, \tag{24}$$

if $n = 1, 2, \ldots$ is integer.

Using eq. (24) we transform eq. (23) to the following

$$\langle l_k|\hat{U}_D(t_k, t_{k-1})|l_{k-1}\rangle = \delta_{l_k l_{k-1}} - \frac{\imath}{\hbar}\int_{t_{k-1}}^{t_k} V_{l_k l_{k-1}}(\tau)\exp[\imath\omega_{l_k l_{k-1}}\tau]d\tau - \frac{\imath}{\hbar}\int_{t_{k-1}}^{t_k} V_{l_k l_{k-1}}(\tau)\exp[-\imath\omega_{l_k l_{k-1}}\tau]d\tau \tag{25}$$

By the use of eq. (3) and $V_{l_k l_{k-1}} = 0$ for $l_k = l_{k-1}$ we prove that eq. (25) is the same as eq. (16)

We note that using Eq. (14), Eq. (18), Eq. (12) quantum transition amplitude $U_D(l_f, t|l_{in}, 0)$ for any t can be expressed as path integral in energy eigenstates space

$$\langle l_f|\hat{U}_D(t)|l_{in}\rangle = U_D(l_f, t|l_{in}, 0) = \lim_{K\to\infty}\sum_{l_1,\ldots,l_K=0}^{N-1}\int_0^1 .. \int_0^1 \exp[\imath S[l_f, l_K, \xi_K; ..; l_k, l_{k-1}, \xi_{k-1}; ..; l_1, l_{in}, \xi_0]]d\xi_0..d\xi_K, \tag{26}$$

where

$$S[l_f, l_K, \xi_K; ..; l_k, l_{k-1}, \xi_{k-1}; ..; l_1, l_{in}, \xi_0] = \sum_{k=1}^{K+1} S[l_k, l_{k-1}, \xi_{k-1}] \tag{27}$$

– dimensionless action. It is a functional, which is defined on a path set in discrete variables l_k space of size N (quantum system levels number) and continuous c-number variables ξ_k space $[0, 1]$.

The quantum transition amplitude eq. (26) with eq. (27) and eq. (19) describes transition of quantum system under electromagnetic field influence. It is possible to use for high-iintensity and an arbitrary structure of field in space and time. Parameters $\omega_{l_k l_{k-1}}$ and $V_{l_k l_{k-1}}$ must be defined for investigated model.

However analytical calculation eq. (26) can not be realized on practice. Then we develop numerical approach to amplitude eq. (26) calculation as well as for eq. (11), (10), (7).

3. Algorithm of numerical simulation of quantum system dynamics

We consider algorithm for numerical calculation of quantum transition amplitude $U(l_f, t|l_{in}, 0)$ and probability $P(l_f, t|l_{in}, 0)$.

Using eq. (14) the quantum transition amplitude calculation was made by recurrence relation

$$U(l_K, t_K|l_{in}, 0) = \sum_{l_{k-1}} U(l_K, t_K|l_{k-1}, t_{k-1})U(l_{k-1}, t_{k-1}|l_{in}, 0), \tag{28}$$

where we introduce

$$U(l_K, t_K|l_{in}, 0) = \langle l_K|\hat{U}_D(t_K)|l_{in}\rangle,$$
$$U(l_K, t_K|l_{k-1}, t_{k-1}) = \langle l_K|\hat{U}_D(t_K, t_{k-1})|l_{k-1}\rangle,$$
$$U(l_{k-1}, t_{k-1}|l_{in}, 0) = \langle l_{k-1}|\hat{U}_D(t_{k-1})|l_{in}\rangle.$$

We define transition amplitude $U(l_0, t_0|l_{in}, 0)$ for first iteration with $t_0 = \Delta\tau_0$. By the use of eq. (28) and eq. (18) we obtain transition amplitude for an arbitrary time moment $t = \sum_{k=0}^{K}\Delta\tau_k$.

For any t_k, t_{k-1} the transition amplitude $U(l_k, t_k|l_{k-1}, t_{k-1})$ is a complex number. Then for numerical calculation we need to express real and imaginary part of amplitude:

$$U(l_k, t_k|l_{k-1}, t_{k-1}) = \Re[U(l_k, t_k|l_{k-1}, t_{k-1})] + \imath\Im[U(l_k, t_k|l_{k-1}, t_{k-1})] \tag{29}$$

For these parts eq. (28) transform into two equations:

$$\Re[U(l_k, t_k|l_{in}, 0)] = \sum_{l_{k-1}=0}^{N-1}(\Re[U(l_k, t_k|l_{k-1}, t_{k-1})]\Re[U(l_{k-1}, t_{k-1}|l_{in}, 0)] - \Im[U(l_k, t_k|l_{k-1}, t_{k-1})]\Im[U(l_{k-1}, t_{k-1}|l_{in}, 0)]) \tag{30}$$

$$\Im[U(l_k, t_k|l_{in}, 0)] = \sum_{l_{k-1}=0}^{N-1}(\Im[U(l_k, t_k|l_{k-1}, t_{k-1})]\Re[U(l_{k-1}, t_{k-1}|l_{in}, 0)] + \Re[U(l_k, t_k|l_{k-1}, t_{k-1})]\Im[U(l_{k-1}, t_{k-1}|l_{in}, 0)]) \tag{31}$$

We present eq. (30) and eq. (31) in matrix form

$$\begin{pmatrix} \Re[\tilde{U}(l_k,t_k|l_{in},0)] \\ \Im[\tilde{U}(l_k,t_k|l_{in},0)] \end{pmatrix} =$$

$$= \sum_{l_{k-1}=0}^{N-1} \int_0^1 \begin{pmatrix} \Re[U(l_k,t_k|l_{k-1},t_{k-1})] & -\Im[U(l_k,t_k|l_{k-1},t_{k-1})] \\ \Im[U(l_k,t_k|l_{k-1},t_{k-1})] & \Re[U(l_k,t_k|l_{k-1},t_{k-1})] \end{pmatrix} \begin{pmatrix} \Re[U(l_{k-1},t_{k-1}|l_{in},0)] \\ \Im[U(l_{k-1},t_{k-1}|l_{in},0)] \end{pmatrix}, \tag{32}$$

The initial condition for pure quantum state $|l_{in}\rangle$ is as follows

$$\begin{pmatrix} \Re[U(l_0,0|l_{in},0)] \\ \Im[U(l_0,0|l_{in},0)] \end{pmatrix} = \begin{pmatrix} \delta_{l_0 l_{in}} \\ 0 \end{pmatrix}. \tag{33}$$

Quantum transition probability $P(l_k,t_k|l_{in},0)$ of investigated system from the state $|l_{in}\rangle$ at moment $t = 0$ to the state $|l_k\rangle$ at moment t_k can be expressed as

$$P(l_k,t_k|l_{in},0) = (\Re[U(l_k,t_k|l_{in},0)])^2 + (\Im[U(l_k,t_k|l_{in},0)])^2, \tag{34}$$

The transition probability $P(l_k,t_k|l_{in},0)$ must be normalized for each time moments t_k

$$\sum_{l_k=0}^{N-1} P(l_k,t_k|l_{in},0) = 1. \tag{35}$$

For this we calculate $\Re[U(l_k,t_k|l_{in},0)]$ and $\Im[U(l_k,t_k|l_{in},0)]$ using eq. (34) and product them on normalizing factor A:

$$\begin{pmatrix} \Re[U(l_k,t_k|l_{in},0)] \\ \Im[U(l_k,t_k|l_{in},0)] \end{pmatrix} = A \begin{pmatrix} \Re[U(l_k,t_k|l_{in},0)] \\ \Im[U(l_k,t_k|l_{in},0)] \end{pmatrix}. \tag{36}$$

The normalizing factor A is calculated by the following formula:

$$A = \left(\sum_{l_k=0}^{N-1} (\Re[\tilde{U}(l_k,t_k|l_{in},0)]^2 + \Im[\tilde{U}(l_k,t_k|l_{in},0)]^2) \right)^{-1/2}. \tag{37}$$

Using Eq. (30)–(37) we calculate the amplitude $U(l_f,t|l_{in},0)$, the transition probability $P(l_f,t|l_{in},0)$ and the probability of quantum state observation for any t.

4. Rotational dynamics of $^{14}N_2$ and $^{15}N_2$ interacting with laser pulses sequences

Recent results of experimental observation of $^{14}N_2$ and $^{15}N_2$ high rotational states excitation were published in [12]. Detailed discussions of the results were in [14, 13].

In the experiments the groups of $^{14}N_2$ and $^{15}N_2$ molecules were investigated. At the initial moment the distribution of rotational population is thermal and corresponds to $T = 6.3$ K. Molecules interact with a sequence of ultrashort laser pulses with period from 6.5 ps to 9.5 ps. Each laser pulse has duration equal 500 fs. Laser radiation intensity reaches the value $I = 5 * 10^{12}$ W/cm^2. The relative populations were measured of the rotational levels of $^{14}N_2$ and $^{15}N_2$ and the functional dependence of the populations on the pulse train period was obtained.

The results of these experiments show that there are quantum nonlinear resonances i.e. the nonlinear increase of rotational excitation efficiency under specific values of the pulse train period. The most efficient population transfer up the rotational ladder occurs around 8.4 ps for $^{14}N_2$ and 9 ps for $^{15}N_2$.

We analyse these experiments using the method developed by us which is based on path integral formulation in energy states space.

We calculate the energy E_l of investigated molecules rotational levels for quantum rigid rotor model [16]

$$-\frac{\hbar^2}{2I}\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}(\sin\theta\frac{\partial}{\partial\theta})Y_l(\theta) = E_l Y_l(\theta), \tag{38}$$

where $I = \mu R^2$ – moment of inertia,
μ – molecule reduced mass,
R – atom distances,
$Y_l(\theta) = Y_l^0(\theta,\phi)$, where $Y_l^m(\theta,\phi)$ – spherical harmonics.

Eq. (38) defines the rotational energy spectrum of a diatomic molecule

$$E_l = \frac{\hbar^2}{2I} l(l+1), \tag{39}$$

where l – azimuthal quantum number.

It is known, that nonpolar molecule dipole moment is equal to zero. However, strong laser fields induce the molecular dipole by exerting an angle-dependent torque.

The interaction is described by the potential [17, 18]

$$V(\tau) = -\frac{1}{4}\Delta\alpha E^2(\tau)\cos^2\theta, \tag{40}$$

where $\Delta\alpha$ describes the molecular polarizability, θ is the angle between the molecular axis and the field polarization.

Matrix elements of interaction operator are

$$V_{l'l}(\tau) = -\frac{1}{4}\Delta\alpha E^2(\tau)\langle l'|\cos^2\theta|l\rangle, \tag{41}$$

where

$$\langle l'|\cos^2\theta|l\rangle = 2\pi\int_0^\pi Y^*_{l'}(\theta)\cos^2\theta Y_l(\theta)\sin\theta d\theta. \tag{42}$$

Matrix elements $\langle l'|\cos^2\theta|l\rangle$ were numerically calculated by Eq. (41) and Eq. (42).

The investigated molecules parameters are [19]: $\Delta\alpha = 1.97 * 10^{-40}$ C$*$m^2/V, $I = 1.4 * 10^{-46}$ kg$*$m^2 for $^{14}N_2$, $I = 1.5 * 10^{-46}$ kg$*$m^2 for $^{15}N_2$.

We consider a sequence of ultrashort laser pulses which was used in [12]. The electric field value is as follows

$$E(\tau) = \sum_{n=-3}^{3} J_n(A)E_0\exp[-\frac{(\tau - n\tau_{per})^2}{\tau^2_{pul}}], \tag{43}$$

were $J_n(A)$ is Bessel function of the first kind,
$A = 2.5$ is the spectral phase modulation amplitude,
$E_0 \approx 6 \times 10^9$ V/m is electric field value,
$\tau_{pul} \approx 500$ fs is each laser pulse duration,
7.98 ps $\leq \tau_{per} \leq$ 9.38 ps is pulse train period.

We are considering the model of N_2 with $N = 8$ rotational levels ($l = 0, 1, \ldots, 7$). This model is a good approximation, because in experiments [12] higher rotational states are practically not excited.

The initial distribution of rotational population is thermal and corresponds to $T = 6.3$ K:

$$P_{l_{in}} = \frac{1}{Z}\exp[-\frac{E_{l_{in}}}{k_BT}], \tag{44}$$

where

$$Z = \sum_{l_{in}=0}^{N-1}\exp[-\frac{E_{l_{in}}}{k_BT}] \tag{45}$$

— particle function,
k — Boltzmann factor,
T — absolute temperature,
N — rotational states number in the theoretical model.

By the use of Eq. (44)-(45) and numerical simulation algorithm we calculate the probability of excitation from the initial state (Boltzmann distribution) to different rotational states having interacted with a sequence of ultrashort laser pulses as a function of pulse train period. The absolute error of our probability calculation was not more than 10^{-3}. The results of our numerical simulations are given in fig. 1, fig. 2 and agree well with experimental data as for $^{14}N_2$ both for $^{15}N_2$ molecules.

In fig. 1 we present the population of $^{14}N_2$ molucels on diffent rotational quantum level l after interaction with pulse train. For the pulse train period equal to 2.79 ps, 5.58 ps and 8.38 ps for $^{14}N_2$ the population is efficiently transferred from the initial (thermal distribution) states $l = 0, 1, 2$ to the higher states $l = 3, 4, 5, 6, 7$. The resonanse train period value $\tau = 8.38$ ps was observed in experiment [12].

In fig. 2 we present normalized probability of $^{14}N_2$ molecules rotational state observation after they have interacted with 7 laser pulses with period $\tau = 8.38$ ps and different values of laser pulses maximum intensity $0.5I_0$, I_0 and $2I_0$, where $I_0 = 5 * 10^{12}$ W/cm^2. We note the population of high rotational state is depend on intensity of laser pulses non-linearly.

5. Conclusion

In this paper we present new method of calculating the transition probability of a quantum system interacting with electromagnetic field by the path integral formalism. We construct the amplitude and probability of quantum transition as path integrals

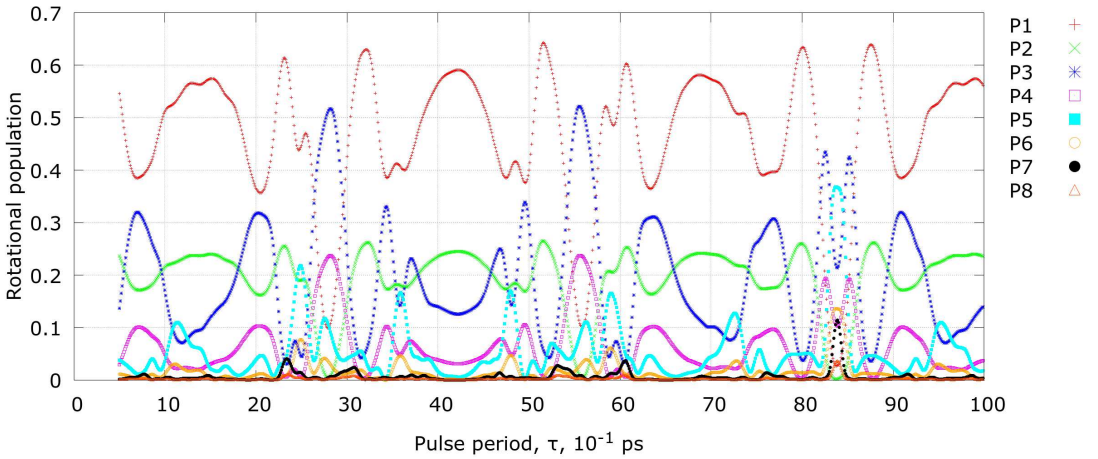

Figure 1: Rotational population of $^{14}N_2$ molucels on diffent rotational quantum level l after interaction with pulse train with period τ

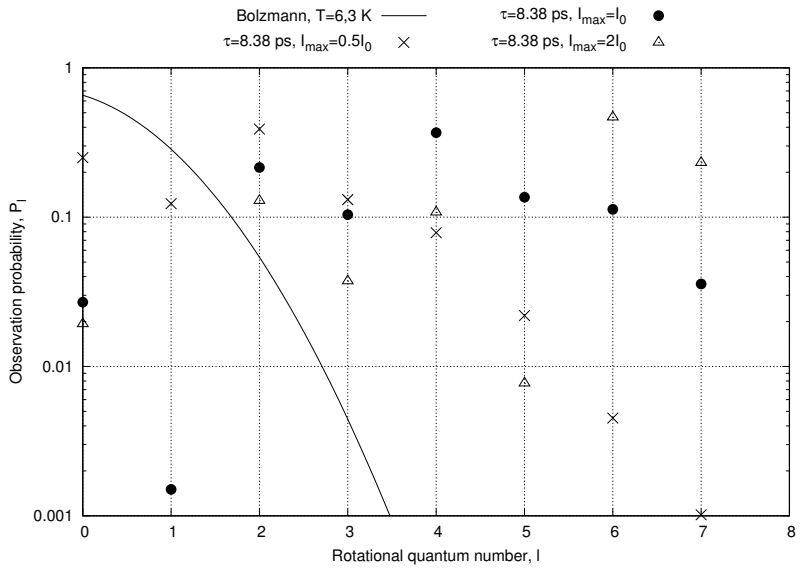

Figure 2: The distribution of observation probabilities of $^{14}N_2$ molucels on diffent rotational quantum level l after interaction with pulse train (τ = 8.38 ps)

in energy states space. The algorithm of path integral calculation was developed. This approach enables us to perform computer simulations of molecule dynamics induced by a laser field.

By the deduced formulas we describe quantum resonances in dynamics of nitrogen molecules, that interact with a sequence of ultrashort laser pulses. The obtained results are in good agreement with the experimental data [12] and the theoretical investigations [14, 13] by Schrödinger equation numerical solution.

The approach developed is appliable to nonperturbative studies of different multiphoton and nonresonant processes.

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