Numerical calculations of the probabilities for quantum transitions in atoms and molecules by the path integral method

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Abstract. The probabilities of molecular quantum transitions induced by electromagnetic field are expressed as path integrals of a real alternating functional. We propose a new method for computing these integrals by means of recurrence relations. We apply this approach to description of the two-photon Rabi oscillations.

The nonlinear interactions of microsystems with the laser radiation of various configurations and levels of intensity (an excitation and dissociation of molecules, ionization of atoms etc.) are under active study now. We consider it reasonable to investigate the probabilities of transitions in multilevel quantum systems nonperturbatively and without any restrictions on the structure of laser radiation, as in the intensity and in the form of pulses. To this end a method of expressing probabilities of quantum transitions as path integrals of the real alternating functionals given in the energy representation is under investigation [1-3].

It was proved [2-3] that the probability of a quantum transition from the system quantum state $|n_{in}\rangle$ at time instant t_0 to the state $|n_f\rangle$ for the time interval $(t_f - t_0)$ under the interaction with the electromagnetic field of a frequency Ω can be represented as a path integral of a real functional in the energy representation:

$$P(n_f, t_f \mid n_{in}, t_0) = A\delta_{n_{K+1}, m_{K+1}}\delta_{n_{in}, m_{in}} \prod_{k=1}^{K} \mathbf{R}_k \cos\left(\sum_{k=1}^{K+1} \Delta S(n_k, m_k, n_{k-1}, m_{k-1}, \xi_{k-1}, \zeta_{k-1}) \right), \quad (1)$$

where

$$\mathbf{R}_k = \sum_{n_k=1}^{N} \sum_{m_k=1}^{N} \int_0^1 d\xi_k \int_0^1 d\zeta_k, \; k = 1, 2, ...$$

$$\Delta S(n_k, m_k, n_{k-1}, m_{k-1}, \xi_{k-1}, \zeta_{k-1}) = S(n_k, n_{k-1}, \xi_{k-1}) - S(m_k, m_{k-1}, \zeta_{k-1}),$$

$$S(n_k, n_{k-1}, \xi_{k-1}) = 2\pi(n_k - n_{k-1})\xi_{k-1} + \Omega^R_{n_k n_{k-1}} \cos(2\pi(n_k - n_{k-1})\xi_{k-1} - (\Omega - \omega_{n_k n_{k-1}})t_{k-1})\Delta t$$

is the action for the transition in the energy representation, n_i, m_i are the quantum numbers prescribing a state of the system, A is a constant that preserves the normalization condition

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$\sum_{n_f} P(n_f, t_f \mid n_{in}, 0) = 1$, $\Omega^R_{n_k n_{k-1}}$ is the Rabi frequency, $\omega_{n_k n_{k-1}}$ is a frequency of transitions between states $|n_k\rangle, |n_{k-1}\rangle$, $t_{k-1} = (k-1)\Delta t$.

The numerical calculation of probabilities according to (1) successfully describes the process for short intervals $(t_f - t_0)$, but leads to considerable errors for long time intervals $(t_f - t_0)$. In this work, we propose a method that makes it possible to calculate the probabilities of quantum transitions by formula (1) for any time interval using the recurrent representation method.

Let us consider functions

$$P_{\cos}(n_K, m_K, t; n_{in}, m_{in}, t_0) = \prod_{k=1}^{K-1} \mathbf{R}_k \cos\Big(\sum_{k=1}^{K} \Delta S(n_k, m_k, n_{k-1}, m_{k-1}, \xi_{k-1}, \zeta_{k-1})\Big),$$

$$P_{\sin}(n_K, m_K, t; n_{in}, m_{in}, t_0) = \prod_{k=1}^{K-1} \mathbf{R}_k \sin\Big(\sum_{k=1}^{K} \Delta S(n_k, m_k, n_{k-1}, m_{k-1}, \xi_{k-1}, \zeta_{k-1})\Big).$$

Then the quantum transition probability can be expressed as follows
$$P(n_f, t_f; n_{in}, t_0) = A\, \delta_{n_K = n_f, m_f} \delta_{n_{in} m_{in}} P_{\cos}(n_f, m_f, t; n_{in}, m_{in}, t_0).$$

The program for the numerical calculation of the integral (1) is based on the recurrence relations for the auxiliary probability functions $P_{\cos}$, $P_{\sin}$:

$$P_{\cos}(n_k, m_k, t_k; n_{in}, t_0) = \mathbf{R}_{k-1}\{\cos[\Delta S(n_k, m_k, n_{k-1}, m_{k-1}, \xi_{k-1}, \zeta_{k-1})]\times \\ \times P_{\cos}(n_{k-1}, m_{k-1}, t_{k-1}; n_{in}, t_0)\} - \\ -\mathbf{R}_{k-1}\{\sin[\Delta S(n_k, m_k, n_{k-1}, m_{k-1}, \xi_{k-1}, \zeta_{k-1})] P_{\sin}(n_{k-1}, m_{k-1}, t_{k-1}; n_{in}, t_0)\};$$

and

$$P_{\sin}(n_{k-1}, m_{k-1}, t_{k-1}; n_{in}, t_0) = \mathbf{R}_{k-2}\{\sin[\Delta S(n_{k-1}, m_{k-1}, n_{k-2}, m_{k-2}, \xi_{k-2}, \zeta_{k-2})]\times \\ \times P_{\cos}(n_{k-2}, m_{k-2}, t_{k-2}; n_{in}, t_0)\} + \\ +\mathbf{R}_{k-2}\{\cos[\Delta S(n_{k-1}, m_{k-1}, n_{k-2}, m_{k-2}, \xi_{k-2}, \zeta_{k-2})] P_{\sin}(n_{k-2}, m_{k-2}, t_{k-2}; n_{in}, t_0)\}.$$

Given the initial conditions we can determine functions $P_{\cos}(n_k, m_k, t_k; n_{in}, t_0)$ and $P_{\sin}(n_k, m_k, t_k; n_{in}, t_0)$, and then we calculate the probability of the quantum transition from the state $|n_{in}\rangle$ at the instant of time $t = t_0$ to the state $|n_k\rangle$ at the instant $t_k > t_0$.

The method successfully describes a behavior of a number of quantum systems, and in particular, the experimentally observed two-photon oscillations of Rabi [4].

References

[1] G.V. Ryazanov, Sov. J. Exp. Theor. Phys. **35**, 1 (1958)
[2] A.A. Biryukov, M.A. Shleenkov, Teoreticheskaia fizika **13** (2012) (in Russian)
[3] A.A. Biryukov, M.A. Shleenkov, Samara State Technical Univ. **19**, 2 (2015)
[4] T.R. Gentile, B.J. Hughey, D. Kleppner, et al., Phys.Rev. A **40**, 9 (1989)