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ON SOLVING COHERENT DYNAMICS EQUATIONS WITH DISCRETE MATHEMATICS METHOD FOR QUANTUM SYSTEMS UNDER LASER EXCITATION

Molecular coherent excitation calculations are performed using a simple model of quantum $N+1$-levels systems. An exact solution of the corresponding system of differential equations is obtained without their integration. For this, the discrete Fourier transform is applied: the sought-for functions – probability amplitudes $a_n(t)$ of a quantum system are represented with Fourier images $F_n(\omega)$, i.e. spectra that are described by some corresponding system of discrete orthogonal polynomials. Fourier spectra are calculated using the polynomials constructed. We find the required $a_n(t)$ by calculating the final sum from 0 to N. Based on a one-to-one correspondence: polynomial characteristics vs equations coefficients, we find all the characteristics of quantum systems, the dynamics of which are described by the obtained solution. The construction of various polynomial systems of a discrete variable makes it possible to obtain solutions for quantum systems with various characteristics, including systems with non-equidistant arrangement of energy levels, which are typical for real molecules.

Key words: coherent laser excitation of quantum systems, Fourier spectra, discrete orthogonal polynomials in Fourier space, exact solutions of differential equations systems.

Introduction. One of the unique properties of laser radiation is its ability, acting on molecules, atoms, crystals, to translate them into a special state of quantum coherence, which differs sharply from classical states. New states are obtained by exciting the medium with ultrashort pulses. Media in a quantum coherent state can be used for a number of new technologies: quantum computing, quantum communication lines, reliable encryption of information, laser control of chemical reactions, to create selectively excited molecules in order to study their spectral properties and rates of intramolecular redistribution energy in them etc. One of the problems in the implementation of these technologies is the preservation of quantum coherence of the medium, since coherence is rapidly destroyed. Decoherence occurs during the interaction of a quantum system with the environment, including measuring setup. A lot of literature is devoted to theoretical and experimental investigation of media in states of quantum coherence, methods for their production, conservation and control of their characteristics [1–4]. Nevertheless, many unsolved problems remain, in particular, with obtaining exact solutions of quantum equations and developing analytical methods for constructing solutions that describe the process of coherent excitation of quantum systems under various conditions.

This paper describes a new approach to the construction of exact solutions of equations of a certain type, describing the indicated process occurring under the action of a constant amplitude laser pulse. It allows one to obtain an analytical solution for the dynamics of various quantum systems, including those having a non-equidistant arrangement of energy levels and when excited by radiation with a carrier frequency that is not in exact resonance with the transition eigenfrequencies of the quantum system. In the work, the solution algorithm is illustrated with a simple example – a system with a few energy levels, but it allows a natural generalization. The method is "spectral". It is based on the transition from the sought-after functions $\{a_n(t)\}_0^N$ of time, the probability amplitudes of a $N+1$-level quantum system to their Fourier images $\{F_n(\omega)\}_0^N$, Fourier spectra. In this case, the discrete Fourier transform is used, that is, the spectral space is discrete. This is natural, since the amplitudes are periodic functions of time when $N < \infty$. The paper considers systems with a uniform space, i.e. spectra are function of a discrete argument given on a uniform grid. This restriction is not fundamental. It is also shown that the Fourier spectra are expressed in terms of orthogonal polynomials; they are used to construct a solution of differential equations of the type under consideration. Polynomials are given in the Fourier space of the probability amplitudes of a quantum system. The algorithm for constructing the solution is simple. Constructing a certain sequence of discrete orthogonal polynomials corresponding to a quantum system, we construct spectra, calculate probability amplitudes, and find the distribution of the quantum system by energy levels. Further, using the connections between the characteristics of the polynomials and the coefficients of the differential equations, we find these coefficients and determine the characteristics of the excited quantum systems, for which a solution is obtained, that describes their coherent dynamics.

The excited medium model and dynamic equations. The model is a quantum system containing $N+1$ energy levels $E_0, E_1, ..., E_N$ (N is a natural number). Radiation $\mathscr{E}_\ell \cos\omega_\ell\tau$ turned on at the moment $\tau = 0$ has constant amplitude $\mathscr{E}_\ell$ and frequency ω_ℓ and causes transitions between neighboring levels. Each transition $E_{n-1} \rightleftarrows E_n$ is characterized by its eigenfrequency $\omega_n = (E_n - E_{n-1})/\hbar$ and dipole moment $\mu_{n-1,n} \equiv \mu_n$ describing the intensity of the interaction of the transition with radiation. The radiation frequency may not coincide with the eigenfrequency of the transition, so the transition may also have a frequency detuning $\Delta\omega_n = \omega_n - \omega_\ell$. Thus, each n-th transition is described by quantities μ_n, $\omega_n, \Delta\omega_n$.

The Schrödinger equation, which describes the dynamics of coherent excitation, for such problems is written not for the wave function, but for complex-valued probability amplitudes $a_n(t)$ of detecting a system at a level n at a time τ during excitation [5]. In dimensionless variables, such a system of equations has the form

$$-\frac{da_n}{dt} = f_{n+1}e^{-i\varepsilon_{n+1}t}a_{n+1} + f_n e^{i\varepsilon_n t}a_{n-1}; \qquad (1)$$

$$a_n(t=0) = \delta_{0,n};\ n = 0, 1, ..., N.$$

This widely used method of the semiclassical description of the process by equations (1) is called the resonance approximation, or the rotating field method [4–6]. In this model, the terms containing the frequencies $\omega_n + \omega_\ell$ are omitted, which practically does not affect the dynamics. Here, the dimensionless function of dipole moments f_n; $f_1 = 1$ with respect to the moment of the first transition $0 \leftrightarrow 1$ is introduced, $\mu_n = \mu_1 f_n$. It characterizes all the transitions of a quantum system interacting with radiation. The usage of the Rabi frequency $\Omega_R = \mu_1\mathscr{E}_\ell / 2\hbar$ of the first transition allows the introduction of dimensionless time $t = \Omega_R\tau$ and frequency detunings $\varepsilon_n = (\omega_n - \omega_\ell)/\Omega_R$ at the transitions. As a result, a quantum system is characterized by a parameter N – the number of transitions interacting with radiation and two sets of quantities f_n, ε_n; they are the coefficients of equations (1); the number of equations (1) is $N+1$. The experimentally measured quantities are the populations of energy levels $\rho_n(t) = a_n^*(t)a(t)_n$ forming a discrete statistical distribution. Building a discrete distribution is the ultimate goal of calculations. The study of its dependence on the characteristics of the process, the properties of the quantum systems and radiation opens the way to controlling the process of coherent excitation of molecules and atoms and to its technological use.

Differential equation system solving. The algorithm for solving the problem is simple and physically clear. We apply the discrete Fourier transform to equations (1), passing from the functions $a_n(t)$ with the continuous argument to the discrete Fourier space of these functions, that is to their spectra $F_n(\omega)$ – the functions of the discrete argument. Indeed, $a_n(t)$ are time-periodic functions of time if $N < \infty$. The solution to the system of differential equations (1) is sought in the form

$$a_n(t) = e^{is_n t}\sum_{\omega=\omega_0}^{\omega_N}\tilde{F}_n(\omega)e^{i\omega t} =$$

$$= e^{is_n t}\sum_{x=0}^{N}F_n(x)e^{irxt}. \qquad (2)$$

Further simplest case – a uniform Fourier space is considered, i.e. Fourier frequencies are equidistant: $\omega = rx$; $x = 0, 1, ..., N$. Constants s_n and r in (2) will be defined below. We also assume that discrete Fourier spectra are as follows

$$F_n(x) = \sigma(x)\hat{p}_0\hat{p}_n(x);\quad n, x = 0, 1, ..., N, \qquad (3)$$

i.e. they are expressed in terms of a certain sequence of discrete polynomials $\{p_n(x)\}_{n=0}^N$ in this space, and they correspond, are adequate to the quantum systems under consideration as well as to the equations (1). The presence $\hat{p}_0$ in (3) is due to the initial conditions for equations (1) – at the moment $t = 0$ the systems are in the ground energy state. The polynomials are normalized and orthogonal

$$\sum_{x=0}^{N}\sigma(x)\hat{p}_m(x)\hat{p}_n(x) = \delta_{m,n} \qquad (4)$$

with discrete weight function $\sigma(x)$.

The most important property of orthogonal polynomials is the three-term recurrence relation

$$\overline{f}_{n+1}\hat{p}_{n+1}(x) + \overline{f}_n\hat{p}_{n-1}(x) = (rx + s_n)\hat{p}_n(x) \qquad (5)$$

which is satisfied by the polynomials of both continuous and discrete arguments [7–9]. Here it is written in an unconventional, but equivalent form, with normalization $\overline{f}_1 = 1$. To study or construct a sequence of orthogonal polynomials means to know not only their explicit form, but also the weight function and the recurrence relation. This information is available in the corresponding sources for polynomials. To date, a huge "zoo" of various orthogonal polynomials, classical and new ones, has been created [9]. Continuous polynomials are widely used in various fields of science and technologies, discrete polynomials are used much less often.

Earlier, we used some classical polynomials of continuous and discrete variables to construct analytic solutions of the equations describing coherent excitation of multilevel quantum systems [10–12]. A number of results were obtained, including interesting ones for physical applications. The classical discrete polynomials (Charlier, Kravchuk) turned out to be especially meaningful in comparison with the polynomials of a continuous argument. So, the solutions describing the excitation of both basic models of quantum physics – a harmonic oscillator and a two-level system, were constructed using Kravchuk polynomials. The dynamics of these systems is described by the same statistical (binomial) energy distribution (populations of energy levels) at any time while radiation is in effect, and with any detuning of the radiation frequency from the eigenfrequencies of the quantum system [12].

A "drawback" of using classical polynomials for the problem under discussion was that they led to solutions describing the excitation of quantum systems with equidistant energy levels. However, it is important to try to build exact solutions for the dynamics of more real models, since molecules and atoms, as a rule, have levels that are non-equidistant. In addition, radiation interacts with a finite number of transitions between levels. This, in particular, explains the motivation for the work presented here.

If well-known polynomials are used, then their weight function and the coefficients of the recurrence relation (5) are known. But it is possible to construct new discrete polynomials by defining, setting in advance, for example, a weight function, and, using the well-known procedure, and determine the coefficients of the recurrence relation [7–9]. Then the probability amplitudes are found by calculating the final sum (2). The desired solution is obtained without integration, using finite discrete mathematics. Thus, by setting a discrete set of Fourier frequencies and a weight function, we construct a sequence of polynomials, calculate the Fourier spectra according to formula (3) and find a solution of equations (1) according to formula (2). It can be seen that the probability amplitudes are spectrally bounded functions if $N < \infty$. The constructed solution describes the coherent dynamics of some $N + 1$-level quantum systems.

Dynamical equations coefficients and quantum systems characteristics. Now it is easy to determine the coefficients of equations (1), the solution of which is constructed, and to determine the characteristics of systems whose dynamics are described by this solution (see Example below). That is the assumption (3) can be confirmed. Substituting (2), (3) into the equations (1) gives:

$$\sum_{x=0}^{N}\sigma(x)\hat{p}_0 e^{irxt}\left\{\begin{array}{c} e^{-i(\varepsilon_{n+1}-(s_{n+1}-s_n))t} f_{n+1}\hat{p}_{n+1}(x) \\ +\, e^{i(\varepsilon_n-(s_n-s_{n-1}))t} f_n\hat{p}_{n-1}(x) \\ -(rx+s_n)\hat{p}_n(x) \end{array}\right\} = 0. \quad (6)$$

Expression (6) is satisfied if

$$\begin{gathered} \varepsilon_n = s_n - s_{n-1}; \quad \varepsilon_{n+1} = s_{n+1} - s_n; \\ f_{n+1}\hat{p}_{n+1}(x) + f_n\hat{p}_{n-1}(x) = (rx+s_n)\hat{p}_n(x). \end{gathered} \quad (7)$$

A comparison of recurrence relations (5) and (7) proves that (2), (3) is a solution of equations (1) if the polynomials in (3) that determine the Fourier spectra are related to the coefficients of equations (1) as follows

$$f_n = \overline{f}_n; \quad \varepsilon_n = s_n - s_{n-1}, \quad (8)$$

where $\overline{f}_n$, ε_n and r in (2) are the coefficients of the recurrence relation for polynomials. The one-to-one correspondence between the coefficients of the dynamical equations and the characteristics of the polynomials shows that the system of equations corresponds to a certain sequence of polynomials with its weight function and recurrence relation. This allows one to solve the inverse problem – based on polynomials one can construct a solution that describes the coherent excitation of the corresponding quantum systems, the dynamics of which are described by equations (1). Next example shows how this method works, using discrete orthogonal polynomials and without using integration of quantum dynamical equations (1).

Example. Here we use the well-known method for constructing a sequence of orthogonal polynomials based on a preset weight function [7–9]. Consider the simplest case – three-level quantum systems ($N = 2$). Let the weight function of polynomials have the form

$$\sigma(x) = \{0.2;\ 0.3;\ 0.5\};\ \sum_{x=0}^{N=2}\sigma(x) = 1. \quad (9)$$

The weight function moments $c_k = \sum_{x=0}^{N+1=3} x^k\sigma(x) = \{1;\ 1.3;\ 2.3;\ 4.3\}$ are special determinants elements that give rise to the sequence of three non-normalized orthogonal polynomials

$$\begin{gathered} p_0 \equiv 1; \\ p_1(x) = x - 1.3; \\ p_2(x) = 0.61x^2 - 1.31x + 0.3. \end{gathered}$$

The squares of their norms are as follows:

$$d_0^2 = 1;\ d_1^2 = 0.61;\ d_2^2 = 0.0732;$$

these are exact results. The normalized polynomials have the following form $\hat{p}_n(x) = p_n(x) / d_n$. Next,

the coefficients of the recurrence relation (5) can be obtained

$$r = \frac{1}{d_1}; \quad \bar{f}_2 = \frac{d_2}{d_1^4}; \\ s_0 = -\frac{1.3}{d_1}; \; s_1 = -\frac{0.517}{d_1^3}; \; s_2 = -\frac{0.52}{d_1^3}. \tag{10}$$

These exact data determine the parameters of quantum systems:

$$f_1 = 1; \; f_2 = \frac{\sqrt{0.0732}}{0.3721} \approx 0.727,$$

i.e. the second transition interacts with radiation slightly. Frequency detunings at transitions are non-equal

$$\varepsilon_1 = s_1 - s_0 = +\frac{0.276}{d_1^3}; \; \varepsilon_2 = s_2 - s_1 = -\frac{0.003}{d_1^3}. \tag{11}$$

The radiation frequency almost coincides with the eigenfrequency of the second transition and differs noticeably from the frequency of the first transition. The energy levels are located non-equidistantly, and the detunings have different signs.

Equations solution and a discrete level distribution function of particles. Using formulas (3) and (2), we calculate the Fourier spectra and probability amplitudes, i.e. obtain the solution of equations (1)

$$a_0 = e^{is_0 t} d_0^{-1}(0.20 + 0.30e^{irt} + 0.50e^{i\cdot 2rt}); \\ a_1 = e^{is_1 t} d_1^{-1}(-0.26 - 0.09e^{irt} + 0.35e^{i\cdot 2rt}); \\ a_2 = e^{is_2 t} d_2^{-1}(0.06 - 0.12e^{irt} + 0.06e^{i\cdot 2rt}). \tag{12}$$

Energy level populations

$$\rho_0 = 0.38 + 0.42\cos rt + 0.2\cos 2rt; \\ \rho_1 = \frac{1}{0.61}(0.1982 - 0.0162\cos rt - \\ - 0.1820\cos 2rt); \\ \rho_2 = \frac{1}{0.0732}(0.0216 - 0.0288\cos rt + \\ + 0.0072\cos 2rt), \tag{13}$$

i.e. energy distribution discrete function can be presented in an equivalent, more convenient form

$$\rho_0 = 0.2r^2 \cdot 0.61(0.9 + 2.1\varphi + 2\varphi^2); \\ \rho_1 = 0.2r^2(1 - \varphi)(1.901 + 1.82\varphi); \\ \rho_2 = 0.2r^2 \cdot 0.6(1 - \varphi)^2; \\ \varphi = \cos rt. \tag{14}$$

Figure shows time dependence of populations of energy levels.

Three-level quantum systems, the dynamics of which are described by the solution (12) and distribution function (14). Let us consider in more detail what the proposed and used algorithm gives, and the dynamics of which three-level systems it describes for a given weight function $\sigma(x)$ (9) that does not contain parameters.

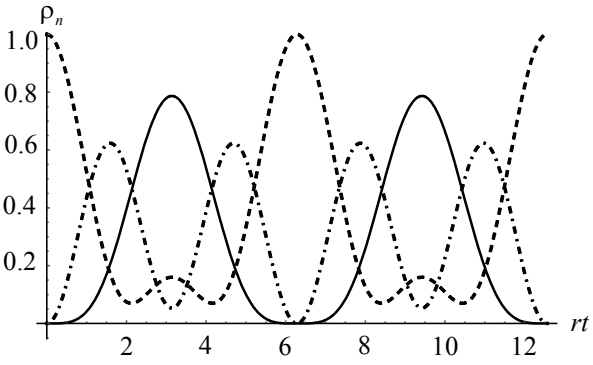

Figure. The dependence of the populations of a three-level quantum system on time: ρ_0, ρ_1, ρ_2 – dashed, dot-dashed, solid lines

Let some system (#1) with dipole moments $\mu_1^{(\#1)}$ and $\mu_2^{(\#1)} = f_2\mu_1^{(\#1)}$ and frequencies $\omega_1^{(\#1)}$ and $\omega_2^{(\#1)}$ of transitions be excited by radiation with amplitude $\mathscr{E}_\ell^{(\#1)}$ and carrier frequency $\omega_\ell^{(\#1)}$. And let the dynamics of this system be described by the distribution function (14). We take radiation of a different amplitude $\mathscr{E}_\ell^{(\#2)} = k\mathscr{E}_\ell^{(\#1)}$, changed at k times. What should be the characteristics of a system (#2) so that its dynamics is identical to the dynamics of the system (#1)? The answer is obvious: the system (#2) must have $\mu_1^{(\#2)} = \mu_1^{(\#1)} / k$, $\mu_2^{(\#2)} = \mu_2^{(\#1)} / k$. Indeed, then the Rabi frequency $\Omega_R = \frac{\mu_1 \mathscr{E}_\ell}{2\pi\hbar}$ of the systems (#1) and (#2) will be the same, as well as the "proper" time $rt = r\Omega_R\tau$ (where τ is the time in seconds), and the process speeds are the same. The population ρ_2 peaks occur at the same moment $\tau_{\max}$ in both processes, when $r\,\Omega_R\tau_{\max} = \pi$. And the value

$$\rho_{2\,\max} = 0.48r^2 = \frac{0.48}{0.61} \simeq 0.7869$$

will also be the same. There are infinitely many such systems. They have different μ_1, each is excited by radiation of the corresponding amplitude, have the same Rabi frequencies, the same (identical) dynamics. An algorithm implemented with a given function $\sigma(x)$ that does not contain free parameters "selects" many systems with identical dynamics.

In addition to dipole, power characteristics all systems and radiation have frequency characteristics. They form two dimensionless quantities

$$\frac{\omega_1 - \omega_\ell}{\Omega_R} = \varepsilon_1; \quad \frac{\omega_2 - \omega_\ell}{\Omega_R} = \varepsilon_2, \qquad (15)$$

where $\varepsilon_1, \varepsilon_2$ are constants. We will show now that the algorithm and solution (14) contain many systems whose dynamics are similar with the coefficient of similarity k to the dynamics of the system (#1). Systems (#2) have Rabi frequency $\Omega_R^{(\#2)} = k\Omega_R^{(\#1)}$ changed several times due to changes in the field amplitude, but the dipole moments coincide with the moments of the system (#1). There are also infinitely many such systems, expression (14) is still their distribution function, and its plot in coordinates population – time is compressed $(k > 1)$ or stretched $(k < 1)$ compared to the plot in Figure. What are the characteristics of these systems with *k*-like dynamics? These are systems with modified natural frequencies ω_1 and ω_2. Since $\varepsilon_1, \varepsilon_2$ are constants, when the Rabi frequency changes, condition (15) can be fulfilled in this way. For example, for ε_1 we obtain

$$\varepsilon_1^{(\#1)} = \frac{\omega_1^{(\#1)} - \omega_\ell}{\Omega_R^{(\#1)}} =$$

$$= \varepsilon_1^{(\#2)} = \frac{\omega_1^{(\#2)} - \omega_\ell}{k\Omega_R^{(\#1)}}.$$

This implies

$$\begin{aligned} \omega_1^{(\#2)} &= \omega_1^{(\#1)} + (k-1)(\omega_1^{(\#1)} - \omega_\ell); \\ \omega_2^{(\#2)} &= \omega_2^{(\#1)} + (k-1)(\omega_2^{(\#1)} - \omega_\ell). \end{aligned} \qquad (16)$$

The second expression in (16) is obtained similarly. Systems (#2) with such a change in the transition frequencies have a *k*-like dynamics, regardless of which of the two methods the Rabi frequency has changed. The similarity coefficient can be any positive number $0 < k < \infty$ if conditions (15) and (16) are satisfied. More complicated cases are possible when the systems have a *k*-like dynamics, and condition (15) is fulfilled while changing both the frequencies of the transitions and the frequency of the radiation.

Thus, the proposed and used algorithm leads to the consideration of quantum systems "dressed" by the radiation field, since the coefficients of equations (1) include physical parameters that characterize both the quantum system and electromagnetic radiation. The algorithm allows us to construct a solution, determine systems with similar dynamics, and shows that there are infinitely many such systems, their dynamics is described by solution (12) and distribution function (14). There is an extensive family containing quantum systems having different dipole moments, level arrangements, excited by different frequencies and amplitudes of radiation, possessing similar (universal) coherent dynamics. The algorithm made it possible to discover the property of similarity of dynamics, to distinguish such families, to build a single analytical solution for their dynamics.

The algorithm can be generalized with using other discrete functions as a weight function or introducing additional parameters for $\sigma(x)$. In [13] for the first time an additional parameter a was introduced into the weight function. A corresponding analytical solution is constructed that describes the dynamics of a one-parameter family of quantum three-level systems with non-equidistant energy levels. In addition the family has three systems with equidistant energy levels when $a = 0$ and $a = \pm 1 / (2\sqrt{3})$ as well.

Obviously, it is possible to include quantum systems whose probability amplitudes have a non-uniform Fourier space, which corresponds to orthogonal polynomials with a non-uniform grid.

Conclusion. For the first time it has been shown that to solve the system of equations under consideration it is possible not to be limited to known polynomials, but to build "any" discrete orthogonal polynomials, which significantly expands the capabilities of the proposed algorithm that does not require integration, but use discrete mathematics.

An example of constructing a sequence of discrete polynomials is given, a solution is obtained for a number of three-level quantum systems and their characteristics are determined. This solution describes the dynamics of quantum systems with a non-equidistant arrangement of energy levels. Such a model is closer to real molecules.

For quantum systems with a large number of transitions interacting with radiation a computer algebra system, for example, "Mathematica" can be used.

It is shown that there are many quantum systems with *k*-like dynamics among all systems described by equations (1).

The solution of the equations of coherent dynamics of quantum systems by the method described above has required knowledge in several areas of mathematics: not only the theory of differential equations, but also the Fourier transform and Fourier spectra, orthogonal polynomials of a discrete variable defined in Fourier space, and their properties, statistical distribution functions, i.e. probability theory. This applies equally to scientific work and to education, university and even school.

An educated person is a carrier of fundamental deep and versatile knowledge about nature and culture.

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