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Allowed transitions between non-adjacent levels and analytical solutions for multilevel quantum systems

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Abstract

An analytical method is formulated to solve the Schrödinger equation for a number of multilevel systems with dipole transitions between adjacent and non-adjacent levels in multifrequency fields. It is shown that orthogonal polynomials can be used to construct exact analytical solutions. The excitation dynamics of the three non-equidistant levels system is analysed in detail.

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1. Introduction

The excitation of multilevel systems in multifrequency electromagnetic fields is one of the important problems of non-linear optics, spectroscopy and laser physics. Both numerical methods and analytical methods for fields with various *fixed* modulation are used mostly to solve multilevel systems excitation dynamics equations [1–8]. If the field is arbitrary or has *arbitrary* modulation, the indicated problem is solved analytically for the harmonic oscillator [4, 9] and special multilevel systems [10]. The use of various multifrequency exciting radiations is a consequence of the necessity of multilevel quantum system dynamics control. It is necessary to take into account the dipole transitions between non-adjacent energy levels in the general consideration of the excitation dynamics of multilevel molecular systems in multifrequency fields. Though such transitions have small probability, they play a significant role in strong multifrequency laser fields when the multiphoton resonances take place. The excitation of a molecule can require consideration of the resonant one-photon transitions between both adjacent and non-adjacent levels in two laser fields with essentially different frequencies.

In this paper an analytical method is defined to obtain the Schrödinger equation solutions for multilevel systems with adjacent and non-adjacent dipole transitions. The application of the orthogonal polynomials leads to multilevel models with different dependences of the spectroscopy characteristics (dipole moments and frequency detunings of the transitions) on

the energy. This analytical method is applied to a system of three non-equidistant levels. The obtained exact analytical solutions describe the excitation in the molecular model systems with both small and high probabilities of the non-adjacent transitions. Analysis of the analytical solutions will show that radiations with a modulation arbitrariness allow for improved quantum system excitation.

2. Analytical method of solving multilevel dynamics equations

Let us consider a molecular system with the Hamiltonian \hat{H}_0 possessing a discrete spectrum of eigenvalues E_n and eigenfunctions $\psi_n^0(t) = \varphi_n^0 \exp(-i(E_n/\hbar)t)$. Let the multifrequency fields be switched on at the moment $t = 0$

$$\begin{aligned} \mathcal{E}(t) &= \mathcal{E}_u u(t) \cos(\omega_u t) + \mathcal{E}_v v(t) \cos(\omega_v t) \\ \max(u(t)) &= \max(v(t)) \equiv 1. \end{aligned} \quad (1)$$

The laser fields are the superposition of the two components with any slow modulation functions $u(t)$ and $v(t)$. The spectrum of such radiation can have a band form. The arbitrarily modulated radiation is multifrequency and non-bichromatic. In the special case of non-modulated radiation the acting field is bichromatic [11–13].

The Schrödinger equation of the system has the following form:

$$i\hbar \frac{d\psi(t)}{dt} = (\hat{H}_0 - \hat{\mu} \mathcal{E}(t)) \psi(t) \quad (2)$$

where

$$\psi(t) = \sum_n a_n(t) \varphi_n^0 \exp(-i(E_n/\hbar)t) \quad (3)$$

$\hat{\mu}$ is the operator of the dipole interaction. Equation (2) can be written in the energy representation with the allowed transitions $n \rightarrow n \pm 1$, $n \pm 2$:

$$\begin{aligned} -i\hbar \frac{da_n}{dt} &= \frac{\mathcal{E}_u u(t)}{2} [\mu_{n-1,n} \exp(i(\omega_{n,n-1} - \omega_u)t) a_{n-1} \\ &\quad + \mu_{n,n+1} \exp(-i(\omega_{n+1,n} - \omega_u)t) a_{n+1}] \\ &\quad + \frac{\mathcal{E}_v v(t)}{2} [\mu_{n-2,n} \exp(i(\omega_{n,n-2} - \omega_v)t) a_{n-2} \\ &\quad + \mu_{n,n+2} \exp(-i(\omega_{n+2,n} - \omega_v)t) a_{n+2}] \end{aligned} \quad (4)$$

$$a_n(t=0) = \delta_{m,n}.$$

This equation was obtained in the rotating wave approximation with allowance made for the inequalities

$$|\omega_{n+1,n} - \omega_u| \ll \omega_u \quad |\omega_{n+2,n} - \omega_v| \ll \omega_v \quad (5)$$

superimposed on the laser fields (1). The non-equidistant character of the energy spectrum of a multilevel molecular system was taken in accordance with the conditions

$$|\omega_{n+2,n+1} - \omega_{n+1,n}| \ll \omega_{n+1,n} \quad \omega_{n+1,n} = (E_{n+1} - E_n)/\hbar. \quad (6)$$

Equations (4) can be solved analytically for a number of special cases. In the case of monochromatic excitation of multilevel systems, the application of orthogonal polynomials allows models with miscellaneous spectroscopic characteristics [14] to be investigated analytically. In this paper exact solutions for the multilevel systems excited by the multifrequency fields (1) with two imposed requirements are discussed. These requirements are:

(1) the multiplicity of the carrier frequencies

$$\omega_v = 2 \omega_u \tag{7}$$

(2) the absence of modulation of the field component with frequency ω_v

$$v(t) \equiv 1. \tag{8}$$

RWA approximation (5) leads to multilevel models with insignificant non-equidistance of the energy spectrum. Such models can be applied to describe the multiphoton excitation of molecular systems. The multiphoton molecular transitions between lower levels and in energy quasi-continuum are quasi-resonant. In accordance with the requirement (7), non-adjacent dipole transitions are successive adjacent dipole transitions. If the one-dimensional multiplication operator rx (r is a constant) corresponds to adjacent dipole transitions then the operator $(rx)^2$ as a result of the successive application rx leads to double successive adjacent dipole transitions. Further, orthogonal polynomials of one argument will be used as one-dimensional analogues of stationary Schrödinger equation wavefunctions. Operators and orthogonal polynomials, which will be used in this paper to solve the problem (4), do not allow the restriction (8) to be avoided. In the general case $v(t) \neq \text{const}$, a model Hamiltonian including the interaction operator $V(x, t) = u(t)rx + v(t)(rx)^2$ has diagonal matrix elements which depend on time. Dynamic equations with such a generalized Hamiltonian cannot be reduced to the problem (4), and are not considered here.

Let us construct an analytical method to solve equations (4) using the orthogonal polynomials with the continuous variable x . Polynomials $p_n(x)$ are orthogonal on a symmetric interval $(-A, A)$ with respect to an even weight function $w(x)$

$$\int_{-A}^A \frac{p_m(x)}{d_m} \frac{p_n(x)}{d_n} w(x) dx = \delta_{m,n} d_n^2 \tag{9}$$

where d_n is the norm. They obey the recurrence formula

$$rx \frac{p_n(x)}{d_n} = f_n \frac{p_{n-1}(x)}{d_{n-1}} + f_{n+1}^* \frac{p_{n+1}(x)}{d_{n+1}} \quad r = \frac{k_1}{k_0} \frac{d_0}{d_1} \quad f_n = r \frac{k_{n-1}}{k_n} \frac{d_n}{d_{n-1}} \tag{10}$$

in the general self-adjoint form (this form is known [14, 15] in the real symmetric case) as the Hamiltonian of equations (4). Here k_n is a leading coefficient of the polynomial $p_n(x)$. The five-term recurrence relation for the real polynomials

$$\begin{aligned} & \left(\mathcal{E}_v \mu_{0,2} \frac{(rx)^2}{f_2} + \mathcal{E}_u \mu_{0,1} u(t) rx + \mathcal{E}_v \mu_{0,2} s_n \right) \frac{p_n(x)}{d_n} \\ &= \mathcal{E}_u \mu_{0,1} u(t) \left(f_n \frac{p_{n-1}(x)}{d_{n-1}} + f_{n+1} \frac{p_{n+1}(x)}{d_{n+1}} \right) \\ &+ \mathcal{E}_v \mu_{0,2} \left(F_{n-1} \frac{p_{n-2}(x)}{d_{n-2}} + F_{n+1} \frac{p_{n+2}(x)}{d_{n+2}} \right) \end{aligned} \tag{11}$$

is obtained with the help of the trinomial equality (10). The coefficients F_n and s_n are defined by

$$F_n = f_n f_{n+1} / f_2 \quad s_n = -(f_n^2 + f_{n+1}^2) / f_2. \tag{12}$$

Comparing the equations (11) and (4), it is possible to obtain the solution of equations (4) for a particular case. Let the non-equidistant system of the energy levels be described as

$$E_n = E_0 + n\hbar\omega_u + (\mathcal{E}_v \mu_{0,2}) (s_n - s_0) \tag{13}$$

with matrix elements

$$\mu_{n-1,n} = \mu_{0,1} f_n \quad \mu_{n-2,n} = \mu_{0,2} F_{n-1}. \tag{14}$$

Then for the dipole transitions $n \rightarrow n \pm 1$, $n \pm 2$ with frequencies

$$\begin{aligned}\omega_{n,n-1} &= (\mathcal{E}_v \mu_{0,2} / \hbar) (s_n - s_{n-1}) + \omega_u \\ \omega_{n,n-2} &= (\mathcal{E}_v \mu_{0,2} / \hbar) (s_n - s_{n-2}) + 2\omega_u\end{aligned}\quad (15)$$

in laser fields (1), (7) and (8), one obtains the solution in the following form:

$$\begin{aligned}a_n(t) &= \int_{-A}^A \exp \left[i t \frac{\mathcal{E}_v \mu_{0,2}}{\hbar} \left(\frac{(r x)^2}{2 f_2} + s_n \right) \right. \\ &\quad \left. + i r x \frac{\mathcal{E}_u \mu_{0,1}}{2 \hbar} \int_0^t u(\tau) d\tau \right] \frac{p_m(x)}{d_m} \frac{p_n(x)}{d_n} w(x) dx.\end{aligned}\quad (16)$$

This solution has been found in [16, 17]¹ for the particular case $\mathcal{E}_u \equiv 0$.

Let us construct one more modification of the analytical method to solve equations (4) using the orthogonal polynomials $p_n(x_k)$ with the discrete variable x_k , $k = 0, 1, \dots, (N-1)$. It is proved [18] that those polynomials $p_n(x)$ with the continuous variable, which satisfy the orthogonality relation (9), have one more orthogonality relation

$$\sum_{k=0}^{N-1} \sigma(x_k) \frac{p_m(x_k)}{d_m} \frac{p_n(x_k)}{d_n} = \delta_{m,n} \quad m < N \quad n < N \quad (17)$$

$$\sigma(x) = A_{N-1} d_{N-1}^2 \left(p_{N-1}(x) \frac{d p_N(x)}{dx} \right)^{-1} \quad p_N(x_k) = 0. \quad (18)$$

Here $A_n = k_n / k_{n+1}$ is the ratio of the leading coefficients of the polynomials $p_n(x)$ and $p_{n+1}(x)$. The polynomials $p_n(x_k)$ with the discrete variable obey the recurrence relation (10) with the same coefficients r and f_n as those for the polynomials $p_n(x)$ with the continuous argument, except for the value

$$f_N = 0. \quad (19)$$

As the polynomials $p_n(x_k)$ obey the relation (11), it is possible to obtain the solution of equations (4)

$$\begin{aligned}a_n(t) &= \sum_{k=0}^{N-1} \exp \left[i t \frac{\mathcal{E}_v \mu_{0,2}}{\hbar} \left(\frac{(r x_k)^2}{2 f_2} + s_n \right) + i r x_k \frac{\mathcal{E}_u \mu_{0,1}}{2 \hbar} \int_0^t u(\tau) d\tau \right] \\ &\quad \times \frac{p_m(x_k)}{d_m} \frac{p_n(x_k)}{d_n} \sigma(x_k)\end{aligned}\quad (20)$$

for a particular case with the help of the orthogonality relation (17). This solution describes the excitation of the non-equidistant system of N levels (13). The transition characteristics $\mu_{n-1,n}$, $\mu_{n-2,n}$ and $\omega_{n,n-1}$ are defined by the formulae (12), (14) and (15) with the equality (19) taken into account. The fields acting on the system are given by expressions (1), (7) and (8). The solution (20) can also be obtained with the help of known polynomials with discrete variable which obey the recurrence formula (10) and the orthogonality relation in the form (17). The formula (18) for the weight function $\sigma(x_k)$ is not necessary if the polynomials $p_n(x_k)$ are known beforehand and polynomials $p_n(x)$ are not orthogonal on the known continuous interval. The zero matrix elements

$$\mu_{N-1,N} = 0 \quad \mu_{N-2,N} = 0 \quad \mu_{N-1,N+1} = 0 \quad (21)$$

occur in the recurrence equation (4) in accordance with equations (12), (14) and (19). Thus the analytical method, using the orthogonal polynomials with discrete variable, allows one to describe the dynamics of N -level systems.

¹ There is online access to the Latex source [16] in the Internet.

3. The analytical description for a three-level system

The constructed analytical method can be used to describe the excitation of the system which consists of three non-equidistant levels. The dynamics equations for the three-level quantum system in the *monochromatic* field have an analytical solution. In the *multifrequency* laser fields, the dynamics equations for the three-level model can be solved analytically only for a special spectrum of the excitation radiation. Further, exact analytical solutions are obtained for the three-level systems with adjacent and non-adjacent transitions in the multifrequency fields.

The dynamics of the quantum three-level model with any transitions is described by the system of three equations

$$\begin{aligned}
 -i\hbar \frac{da_0}{dt} &= \frac{\mathcal{E}_u u(t)}{2} \mu_{0,1} \exp(-i(\omega_{1,0} - \omega_u)t)a_1 + \frac{\mathcal{E}_v}{2} \mu_{0,2} \exp(-i(\omega_{2,0} - \omega_v)t)a_2 \\
 -i\hbar \frac{da_1}{dt} &= \frac{\mathcal{E}_u u(t)}{2} [\mu_{0,1} \exp(i(\omega_{1,0} - \omega_u)t)a_0 + \mu_{1,2} \exp(-i(\omega_{2,1} - \omega_u)t)a_2] \\
 -i\hbar \frac{da_2}{dt} &= \frac{\mathcal{E}_u u(t)}{2} \mu_{1,2} \exp(i(\omega_{2,1} - \omega_u)t)a_1 + \frac{\mathcal{E}_v}{2} \mu_{0,2} \exp(i(\omega_{2,0} - \omega_v)t)a_0
 \end{aligned} \tag{22}$$

where the initial condition is $a_n(t = 0) = \delta_{m,n}$. The system is characterized by the arbitrary dipole moments $\mu_{0,1}, \mu_{1,2}, \mu_{0,2}$ and the transition frequencies $\omega_{1,0}, \omega_{2,1}$.

Let us construct an analytical solution for the three-level model (22). The Gegenbauer polynomials

$$\begin{aligned}
 p_n(z) = C_n^\lambda(z) &\equiv \frac{(2\lambda)_n}{n!} {}_2F_1\left(-n, n + 2\lambda; \lambda + \frac{1}{2}; \frac{1-z}{2}\right) \\
 \lambda < -2 \quad \text{or} \quad \lambda > -\frac{1}{2}
 \end{aligned} \tag{23}$$

with the discrete variable x_k , the index $k = 0, 1, 2$, is used for this purpose. Application of the Luke theorem [18] allows one to establish the orthogonality of the polynomials (23) on the discrete range of the argument

$$x_k = -B, 0, B \quad k = 0, 1, 2 \quad B = (2(2 + \lambda)/3)^{-1/2} \tag{24}$$

with respect to the weight function

$$\sigma(x_k) = \frac{6 \sqrt{\pi} \Gamma(\lambda + \frac{3}{2})}{(\lambda + 2) \Gamma(\lambda + 3)} \{([\lambda + 1] x_k^2 - 2) ([\lambda + 2] x_k^2 - 2)\}^{-1} \tag{25}$$

and the norm

$$\begin{aligned}
 d_n &= 2^{-\lambda} \Gamma(1 - \lambda) \{|\lambda - n| n! \Gamma(1 - 2\lambda - n)\}^{-1/2} \quad \text{at} \quad \lambda < -2 \\
 d_n &= \left\{ \frac{\sqrt{\pi} (2\lambda)_n \Gamma(\lambda + \frac{1}{2})}{(\lambda + n) n! \Gamma(\lambda)} \right\}^{1/2} \quad \text{at} \quad \lambda > -\frac{1}{2}.
 \end{aligned} \tag{26}$$

The polynomials $C_n^\lambda(x_k)$ satisfy the recurrence formulae (10) and (11) with the coefficients

$$\begin{aligned}
 r &= \begin{cases} i\sqrt{2|\lambda + 1|} & \lambda < -2 \\ \sqrt{2|\lambda + 1|} & \lambda > -\frac{1}{2} \end{cases} \quad \lambda = \frac{2[f_2]^2 - 1}{2 - [f_2]^2} \\
 f_0 &= 0 \quad f_1 = 1 \quad f_2 = \frac{\mu_{1,2}}{\mu_{0,1}} \quad f_3 = 0
 \end{aligned} \tag{27}$$

and F_n, s_n are defined by (12). The explicit form of the solution (20) for equations (22) is found with the help of the substitutions of the formulae (12), (23)–(27). This solution is right

for the particular case of equations (22) describing the excitation of the three non-equidistant energy levels

$$\begin{aligned}
 E_0 \quad E_1 &= E_0 + \hbar\omega_u - \frac{\mathcal{E}_v\mu_{0,2}\mu_{1,2}}{\mu_{0,1}} \\
 E_2 &= E_0 + 2\hbar\omega_u + \mathcal{E}_v\mu_{0,2}\left(\frac{\mu_{0,1}^2 - \mu_{1,2}^2}{\mu_{0,1}\mu_{1,2}}\right)
 \end{aligned}
 \tag{28}$$

in the fields (1), (7) and (8). Dipole moments $\mu_{0,1}$, $\mu_{1,2}$ and $\mu_{0,2}$ are arbitrary. The transition frequencies are

$$\omega_{1,0} = \omega_u - \frac{\mathcal{E}_v\mu_{0,2}\mu_{1,2}}{\hbar\mu_{0,1}} \quad \omega_{2,1} = \omega_u + \frac{\mathcal{E}_v\mu_{0,2}\mu_{0,1}}{\hbar\mu_{1,2}}.
 \tag{29}$$

The formulae (28) and (29) are obtained from the expressions (12) and (27). If the field amplitude \mathcal{E}_v is not large for any values of the dipole moments $\mu_{0,1}$ and $\mu_{1,2}$, the RWA inequalities (5) and the conditions (6) are valid.

Let the zero level ($m = 0$) be only populated at initial time ($t = 0$). The problem of the maximal excitation of the top level ($n = 2$) will be investigated. The top-level population according to formulae (20) and (24) can be written as

$$\rho_2 = |\gamma_0 + \exp(i\alpha_B t + i\beta_B T)\gamma_B + \exp(i\alpha_{-B} t + i\beta_{-B} T)\gamma_{-B}|^2
 \tag{30}$$

where the constants are given as

$$\gamma_{x_k} = \frac{C_0^\lambda(x_k)}{d_0} \frac{C_2^\lambda(x_k)}{d_2} \sigma(x_k) \quad \alpha_{x_k} = \frac{\mathcal{E}_v\mu_{0,2}}{2\hbar} \frac{(r x_k)^2}{f_2} \quad \beta_{x_k} = r x_k \frac{\mathcal{E}_u\mu_{0,1}}{2\hbar}
 \tag{31}$$

and

$$T = \int_0^t u(\tau) d\tau.
 \tag{32}$$

Since the constants α_x and β_x are real numbers, and values t and T are arbitrary real numbers, the formula (30) determines a maximal possible population of the top level

$$\rho_2^{\max} = \left| \sum_{k=0}^2 |\gamma_{x_k}| \right|^2 \leq 1.
 \tag{33}$$

The dependence ρ_2^{\max} on the ratio $f_2 = \mu_{1,2}/\mu_{0,1}$ of the dipole moments is shown in table 1. The constants ρ_2^{\max} for inverse values $f_2 = \mu_{1,2}/\mu_{0,1}$, $\mu_{0,1}/\mu_{1,2}$ coincide.

Table 1. Maximal population of the top level ρ_2^{\max} at $\rho_0(t = 0) = 1$.

$\mu_{1,2}/\mu_{0,1}$	$\mu_{0,1}/\mu_{1,2}$	$\rho_2^{\max}(\mu_{1,2}/\mu_{0,1}) = \rho_2^{\max}(\mu_{0,1}/\mu_{1,2})$
1	1	1
0.9	1.111(1)	0.988 981
0.8	1.25	0.951 814
0.7	1.428 57	0.882 843
0.6	1.666 (6)	0.778 547
0.5	2	0.64
0.4	2.5	0.475 624
0.1	10	0.039 212
0.01	100	0.000 399 92

If the maximal population of the top level takes place, the function $T = T(t)$ (20) and a time value t should satisfy the following system of linear equations:

$$\begin{aligned}\alpha_{Bt} + \beta_B T &= -\arg(\gamma_B/\gamma_0) + 2\pi k \\ \alpha_{-Bt} + \beta_{-B} T &= -\arg(\gamma_{-B}/\gamma_0) + 2\pi l.\end{aligned}\quad (34)$$

The numbers k and l are integer and arbitrary. The infinite set of functions $u(\tau)$ in equation (32) and the miscellaneous values of the time t satisfy the system (34). It is the principal physical conclusion, defining further investigations, that the exact analytical selecting technique of any arbitrary form of the radiation modulation allows the best upper level excitation in accordance with the formulae (30), (32) and (34). However, table 1 is not the key result. It shows an extreme possible excitation and the restriction of the generally used dipole interaction approximation.

Thus, using a multifrequency excitation allows one to control the dynamics of the populations more effectively and also to reach the maximal population inversion in the system when both adjacent and non-adjacent transitions are taken into account.

4. Conclusions

The Schrödinger equation is solved analytically for a number of multilevel systems in the rotating wave approximation taking into account adjacent and non-adjacent transitions. The analytical method of obtaining the solutions for the investigation of the multilevel system dynamics in multifrequency fields is defined owing to the application of the orthogonal polynomials with continuous or discrete variables. The Schrödinger equation has been solved analytically for the three-level model within this approach. The energy spectrum of the system depends on the values of the dipole moments. The maximal inversion of the population at the transition $0 \leftrightarrow 2$ in the three-level system is reached when the ratio $\mu_{1,2}/\mu_{0,1}$ of the dipole moments of the adjacent transitions is equal to 1. The analytical expressions (1), (7), (8), (32) and (34) are obtained for the optimal forms of those multifrequency fields which effectively excite the system.

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