# **Two-color multiphoton resonant excitation of three-level atoms**

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The multiphoton resonant excitation of three-level atoms by the two laser fields of different frequencies is investigated. The time evolution of the system and analytical solutions expressing Rabi oscillations of the probability amplitudes at the two-color multiphoton resonant excitation are found using a nonperturbative resonant approach. The specific examples for experimental implementation of two-color multiphoton resonant excitation of hydrogen atoms are considered.

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# **I. INTRODUCTION**

The possibilities to control quantum systems, e.g., to obtain desired coherent superposition states using external electromagnetic fields have been studied intensively since the period of development of nonlinear optics  $[1,2]$  $[1,2]$  $[1,2]$  $[1,2]$  and is considered nowadays because of its significance in the contemporary problems of quantum informatics  $\lceil 3, 4 \rceil$  $\lceil 3, 4 \rceil$  $\lceil 3, 4 \rceil$ . Diverse schemes for the controlling of quantum systems with onephoton resonant excitation of atoms and associated cooperative processes have been described comprehensively in several review articles and books (see, e.g., Refs.  $[1,2]$  $[1,2]$  $[1,2]$  $[1,2]$ ). On the other hand, in the strong laser fields one can expect for multiphoton resonant excitation of atoms  $\lceil 5-7 \rceil$  $\lceil 5-7 \rceil$  $\lceil 5-7 \rceil$ . As was shown in Ref.  $[5]$  $[5]$  $[5]$  multiphoton resonant excitation of atoms is effective if the atomic system has a mean dipole moment in the excited states. Otherwise, the energies of the excited states of a three-level atomic system should be close enough to each other and the transition dipole moment between these states must not be zero.

The multiphoton resonant excitation of atoms will allow observing cooperative effects in high frequencies domain and it is also significant for high order harmonic generation [[8](#page-5-6)[,9](#page-5-7)]. Other interesting applications of multiphoton resonant excitation concern dipolar molecules with a permanent dipole moment  $\lceil 10 \rceil$  $\lceil 10 \rceil$  $\lceil 10 \rceil$ . Research on ultracold dipolar molecules is progressing very fast  $[11]$  $[11]$  $[11]$ . Especially interesting is the possibility to design a quantum information device  $\lceil 12 \rceil$  $\lceil 12 \rceil$  $\lceil 12 \rceil$ . In many cases for the implementation of quantum logic gates it is necessary to excite polar molecules and the transition frequency between the desired states lies in the uv domain and the problem reduces to resonant multiphoton excitation of such systems by actual laser fields.

In the present work we investigate the multiphoton resonant excitation of a three-level atom by the two laser fields of different frequencies towards the formation of coherent superposition states. The resonance condition for this scheme of the two laser fields of different frequencies:  $n_1\omega_1 + n_2\omega_2$  $=\omega_0$   $(n_{1,2}=0,\pm 1,\pm 2,...)$  expands the spectrum of possible combinations for the implementation of multiphoton resonance between the atomic  $(\omega_0)$  and waves  $(\omega_1$  and  $\omega_2)$  frequencies. First of all this is important from the point of view of experimental realization. As existing optical lasers operate on given frequencies associated with the difference of energetic levels of atoms, hence it will be difficult from the experimental point of view to realize multiphoton resonance with a given atomic system by a single laser radiation.

The set of equations for the probability amplitudes is solved using a nonperturbative resonant approach, which is a generalization of widely used rotating wave approximation  $(RWA)$   $[13-15]$  $[13-15]$  $[13-15]$  (for more recent discussions and related references on the subject see also Refs.  $[16-20]$  $[16-20]$  $[16-20]$ ). The considered method can be traced back to the averaging method for the Kapitza pendulum  $[21,22]$  $[21,22]$  $[21,22]$  $[21,22]$ , that is, a classical pendulum with a periodically moving point of suspension. In this method the motion is separated into a slow part and a fast part. The fast part results to effective potential. In our model fast parts are nonresonant terms (counter-rotating terms) and they account for dynamic Stark shifts. For a two-level system in the case of one photon resonance this is known as a Bloch-Siegert shift  $\lceil 14 \rceil$  $\lceil 14 \rceil$  $\lceil 14 \rceil$  which can be viewed as an extreme situation of the dynamic Stark splitting  $[16]$  $[16]$  $[16]$ .

The multiphoton resonant excitation problem is reduced to Rabi problem with a generalized "Rabi frequency" which has a nonlinear dependence on the amplitudes of electromagnetic waves. We have also performed numerical calculations considering the concrete examples of existing laser parameters.

The organization of the paper is as follows. In Sec. II we present analytical treatment of considered problem. In Sec. III we present numerical calculations. Finally, conclusions are given in Sec. IV.

### **II. BASIC MODEL AND RESONANT SOLUTION**

Let us consider a three-level atom interacting with the two radiation fields of frequencies  $\omega_1$  and  $\omega_2$  as it is shown in Fig.  $1(a)$  $1(a)$ . We assume an atom to be in a V configuration in which two upper levels  $|2\rangle$  and  $|3\rangle$  with mean dipole moments are coupled to a single lower level  $|1\rangle$ . We assume that only  $|1\rangle \rightarrow |2\rangle$  and  $|1\rangle \rightarrow |3\rangle$  transitions are dipole allowed. Usually the stationary states of atoms have definite parity and the diagonal matrix elements of dipole moment are zero for these states. For the hydrogen atom due to the random degeneration upon orbital moment there are stationary excited states which have not definite parity. Consequently, hy- \*Electronic address: avetissian@ysu.am drogen atom may have a mean dipole moment in the excited

<span id="page-0-0"></span>

<span id="page-1-0"></span>

FIG. 1. Three-level atomic structures for (a) V type with mean dipole moments in the excited states and (b)  $\Gamma$  configuration with the coupling transition between the excited states.

stationary states. The linear Stark shift for hydrogen atom is a well known example illustrating this fact  $\left[23\right]$  $\left[23\right]$  $\left[23\right]$  (for the other spherically symmetric atoms only quadratic Stark shift exists). For dipolar molecules the molecular states do not have definite parity as well, which results in a mean dipole moment in the stationary states.

Another possible three-level scheme is shown in Fig.  $1(b)$  $1(b)$ and one can refer it as a  $\Gamma$  configuration. In this case lower level  $|1\rangle$  is coupled to an upper level  $|2\rangle$  which in turn is coupled to an adjacent level  $|3\rangle$  ( $|1\rangle \rightarrow |3\rangle$  transition is dipole forbidden). If the energies of excited states in  $\Gamma$  configuration are close enough to each other (the frequency associated with the level's difference should be small compared with Rabi frequencies of the waves) then by the unitary transformation

$$
\hat{S} = \begin{pmatrix}\n1 & 0 & 0 \\
0 & \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} \\
0 & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}}\n\end{pmatrix}
$$
\n(1)

the problem can be reduced to the V configuration Fig.  $1(a)$  $1(a)$ ]. This is obvious for the hydrogen atom in parabolic [[23](#page-5-18)] and spheric coordinates. In the first case, the atom has a mean dipole moment in the excited states, while in the second case the mean dipole moment is zero for stationary states but due to the random degeneration upon orbital moment there is a transition dipole moment between degenerate states. Hence, we will consider the scheme of the V configuration.

The Hamiltonian for the system will be presented in the form

<span id="page-1-1"></span>
$$
\hat{H} = \varepsilon_1 |1\rangle\langle 1| + (\varepsilon_2 + V_{22})|2\rangle\langle 2| + (\varepsilon_3 + V_{33})|3\rangle\langle 3| + (V_{12}|1\rangle\langle 2| + V_{13}|1\rangle\langle 3| + \text{H.c.}),
$$
\n(2)

where

$$
V_{\eta\nu} = -d_{\eta\nu} [E_1 \cos(\omega_1 t) + E_2 \cos(\omega_2 t + \varphi)],\tag{3}
$$

 $d_{nv}$  is the matrix element of the electric dipole moment,  $E_{1,2}$ are slowly varying amplitudes of linearly polarized electromagnetic waves, and  $\varphi$ =const is the phase difference between the two waves. The terms  $V_{22}$  $V_{22}$  $V_{22}$  and  $V_{33}$  in Eq. (2) account for interaction due to the mean dipole moments and these terms are crucial for the multiphoton resonance.

<span id="page-1-3"></span>The wave function of the system can be written in the form

$$
|\Psi(t)\rangle = a_1(t)e^{-(i/\hbar)\varepsilon_1 t} |1\rangle + a_2(t)e^{-(i/\hbar)(\varepsilon_2 t + \int_0^t V_{22} dt)} |2\rangle + a_3(t)e^{-(i/\hbar)(\varepsilon_3 t + \int_0^t V_{33} dt)} |3\rangle.
$$
 (4)

From the Schrödinger equation

$$
i\hbar \frac{\partial |\Psi(t)\rangle}{\partial t} = \hat{H} |\Psi(t)\rangle
$$
 (5)

<span id="page-1-4"></span>one can obtain the equations for the probability amplitudes  $a_1(t)$ ,  $a_2(t)$ , and  $a_3(t)$ :

$$
i\frac{da_1}{dt} = F_{12}(t)a_2 + F_{13}(t)a_3,
$$
  

$$
i\frac{da_2}{dt} = F_{12}^{\dagger}(t)a_1,
$$
  

$$
i\frac{da_3}{dt} = F_{13}^{\dagger}(t)a_1,
$$
 (6)

where

$$
F_{12}(t) = \frac{V_{12}(t)}{\hbar} e^{(i/\hbar)[(\varepsilon_1 - \varepsilon_2)t - \int_0^t V_{22}(t)dt]},
$$
 (7)

$$
F_{13}(t) = \frac{V_{13}(t)}{\hbar} e^{(i/\hbar)[(\varepsilon_1 - \varepsilon_3)t - \int_0^t V_{33}(t)dt]},
$$
 (8)

and  $F^{\dagger}$  denotes the complex conjugation of *F*. With the help of expansion in Bessel function

$$
e^{ix\sin\alpha}\cos\alpha = \frac{1}{x}\sum_{s=-\infty}^{\infty} sJ_s(x)e^{is\alpha}
$$

<span id="page-1-2"></span>the functions  $F_{12}(t)$  and  $F_{13}(t)$  can be represented in the following form:

$$
F_{12}(t) = \sum_{s_1} \sum_{s_2} \eta_{12}(s_1, s_2) e^{(i/\hbar)(\varepsilon_1 - \varepsilon_2 + \hbar s_1 \omega_1 + \hbar s_2 \omega_2)t}, \qquad (9)
$$

$$
F_{13}(t) = \sum_{s_1} \sum_{s_2} \eta_{13}(s_1, s_2) e^{(i/\hbar)(\varepsilon_1 - \varepsilon_3 + \hbar s_1 \omega_1 + \hbar s_2 \omega_2)t}, \quad (10)
$$

<span id="page-1-5"></span>where

$$
\eta_{12}(s_1, s_2) = -\frac{d_{12}}{d_{22}}(\omega_1 s_1 + \omega_2 s_2)
$$

$$
\times J_{s_1} \left( \frac{d_{22} E_1}{\hbar \omega_1} \right) J_{s_2} \left( \frac{d_{22} E_2}{\hbar \omega_2} \right) e^{is_2 \varphi}, \qquad (11)
$$

<span id="page-1-6"></span>
$$
\eta_{13}(s_1, s_2) = -\frac{d_{13}}{d_{33}}(\omega_1 s_1 + \omega_2 s_2) J_{s_1} \left(\frac{d_{33} E_1}{\hbar \omega_1}\right) J_{s_2} \left(\frac{d_{33} E_2}{\hbar \omega_2}\right) e^{is_2 \varphi}
$$
\n(12)

represent the coupling of levels by  $s_1$  and  $s_2$  photons.

In this representation the quasienergy levels  $\varepsilon_{2,3}-s_1\omega_1$  $-s_2\omega_2$  ( $s_{1,2}=\pm 1, \pm 2,...$ ) close to the ground state arise. The <span id="page-2-2"></span>probabilities of multiphoton transitions between these levels will have maximal values for the resonant transitions

$$
\varepsilon_1 - \varepsilon_{2,3} + n_1 \omega_1 + n_2 \omega_2 \simeq 0
$$
,  $n_{1,2} = \pm 1, \pm 2,...$  (13)

<span id="page-2-0"></span>Hence, if resonant condition holds for any pair of photons numbers  $\{n_1 = n, n_2 = m\}$ , normal resonance, then the functions  $(9)$  $(9)$  $(9)$  and  $(10)$  $(10)$  $(10)$  can be represented in the following form:

$$
F_{12}(t) = [\Omega_{12} + f_{12}(t)]e^{i\delta_2 t},\tag{14}
$$

$$
F_{13}(t) = [\Omega_{13} + f_{13}(t)]e^{i\delta_3 t},\tag{15}
$$

<span id="page-2-6"></span><span id="page-2-1"></span>where

$$
\Omega_{12} = \eta_{12}(n,m), \quad \Omega_{13} = \eta_{13}(n,m) \tag{16}
$$

are resonant coupling constants and

$$
f_{12}(t) = \sum_{s_1 \neq n} \sum_{s_2 \neq m} \eta_{12}(s_1, s_2) e^{i[(s_1 - n)\omega_1 + (s_2 - m)\omega_2]t}, \qquad (17)
$$

$$
f_{13}(t) = \sum_{s_1 \neq n} \sum_{s_2 \neq m} \eta_{13}(s_1, s_2) e^{i[(s_1 - n)\omega_1 + (s_2 - m)\omega_2]t}, \qquad (18)
$$

are rapidly oscillating functions on the scale of waves' periods. In Eqs.  $(14)$  $(14)$  $(14)$  and  $(15)$  $(15)$  $(15)$  we have introduced resonance detunings

$$
\hbar \delta_{2,3} = \varepsilon_1 - \varepsilon_{2,3} + \hbar n \omega_1 + \hbar m \omega_2. \tag{19}
$$

The resonance condition  $(13)$  $(13)$  $(13)$  can also be satisfied by diverse pairs of photons numbers, degenerate resonance. Particularly if  $\omega_2 = k\omega_1$ , where *k* is an integer number, then there are many channels of resonance transitions and one should take into account all possible transitions. In this case resonant couplings are

$$
\Omega_{12} = \sum_{s_2} \eta_{12}(n - k s_2, s_2); \quad \Omega_{13} = \sum_{s_2} \eta_{13}(n - k s_2, s_2)
$$
\n(20)

and

$$
f_{12}(t) = \sum_{s_1 \neq n} \sum_{s_2} \eta_{12}(s_1 - k s_2, s_2) e^{i(s_1 - n)\omega_1 t}, \tag{21}
$$

$$
f_{13}(t) = \sum_{s_1 \neq n} \sum_{s_2} \eta_{12}(s_1 - k s_2, s_2) e^{i(s_1 - n)\omega_1 t}, \tag{22}
$$

$$
\hbar \delta_{2,3} = \varepsilon_1 - \varepsilon_{2,3} + \hbar n \omega_1. \tag{23}
$$

<span id="page-2-3"></span>Following the ansatz developed in Ref.  $[5]$  $[5]$  $[5]$  we will solve the system of equations in the resonant approximation. As consequence of separation of Eqs.  $(14)$  $(14)$  $(14)$  and  $(15)$  $(15)$  $(15)$  the probability amplitudes can be represented in the form

$$
a_1(t) = \overline{a}_1(t) + \alpha_1(t),
$$
  
\n
$$
a_2(t) = [\overline{a}_2(t) + \alpha_2(t)]e^{-i\delta_2 t},
$$
  
\n
$$
a_3(t) = [\overline{a}_3(t) + \alpha_3(t)]e^{-i\delta_3 t},
$$
\n(24)

where  $\bar{a}_j(t)$  are the time average of  $a_j(t)$  and  $\alpha_j(t)$  are rapidly oscillating functions. Substituting Eq.  $(24)$  $(24)$  $(24)$  into Eq.  $(6)$  $(6)$  $(6)$  and <span id="page-2-4"></span>separating slow and rapid oscillations, as well as taking into account Eqs.  $(14)$  $(14)$  $(14)$  and  $(15)$  $(15)$  $(15)$ , we obtain the following set of equations for the time average amplitudes  $\bar{a}_j(t)$ :

$$
i\frac{d\bar{a}_j}{dt} = \sum_{\nu=1}^{3} \bar{F}_{j\nu}\bar{a}_{\nu}; \quad j = 1, 2, 3,
$$
 (25)

<span id="page-2-7"></span>where

$$
\hat{F} = \begin{pmatrix}\n\Delta_2 + \Delta_3 & \Omega_{12} & \Omega_{13} \\
\Omega_{12}^* & -(\delta_2 + \Delta_2) & \Delta_{23} \\
\Omega_{13}^* & \Delta_{23}^* & -(\delta_3 + \Delta_3)\n\end{pmatrix}.
$$
 (26)

<span id="page-2-8"></span>Here terms  $\Delta_2$ ,  $\Delta_3$ , and  $\Delta_{23}$  describe dynamic Stark shifts. For the normal resonance we have

<span id="page-2-9"></span>
$$
\Delta_l = \sum_{s_1 \neq n} \sum_{s_2 \neq m} \frac{|\eta_{1l}(s_1, s_2)|^2}{(s_1 - n)\omega_1 + (s_2 - m)\omega_2}; \quad l = 2, 3, \quad (27)
$$

$$
\Delta_{23} = -\sum_{s_1 \neq n} \sum_{s_2 \neq m} \frac{\eta_{12}^*(s_1, s_2) \eta_{13}(s_1, s_2)}{(s_1 - n)\omega_1 + (s_2 - m)\omega_2},\tag{28}
$$

while for the degenerated resonance

$$
\Delta_{l} = \sum_{s_{1} \neq n} \frac{1}{(s_{1} - n)\omega_{1}} \left| \sum_{l_{2}} \eta_{1l}(s_{1} - kl_{2}, l_{2}) \right|^{2}; \quad l = 2, 3,
$$
\n(29)

$$
\Delta_{23} = -\sum_{s_1 \neq n} \sum_{l_2} \sum_{s_2} \frac{\eta_{12}^*(s_1 - k s_2, s_2) \eta_{13}(s_1 - k l_2, l_2)}{(s_1 - n)\omega_1}.
$$
\n(30)

Thus, we have a set of linear ordinary differential equations with fixed coefficients, the general solution of which is given by a superposition of three linearly independent solutions

$$
\bar{a}_j = \sum_{\nu=1}^3 C_{j\nu} \exp(i\lambda_i t), \qquad (31)
$$

<span id="page-2-5"></span>where  $C_{\mu\nu}$  are constants of integration determined by the initial conditions, and the factors  $\lambda_v$  are the solutions of the third-order characteristic equation

$$
\det(\hat{\vec{F}} - \lambda \hat{I}) = 0.
$$
 (32)

The set of equations  $(25)$  $(25)$  $(25)$  has been derived using the assumption that the amplitudes  $\bar{a}_{\eta}$  are slowly varying functions on the scale of the electromagnetic waves periods that puts the following restrictions:

$$
|\overline{F}_{j\nu}| \ll \min\{\omega_1, \omega_2\} \tag{33}
$$

on the characteristic parameters of the system considered.

The solution  $(31)$  $(31)$  $(31)$  is very complicated and in order to reveal the physics of multiphoton resonant excitation process let us consider the solution at the exact resonance when the dynamic Stark shift is small compared with the Rabi fre-quency. Then the solution ([31](#page-2-5)) for the system situated initially in the ground state is

<span id="page-3-0"></span>
$$
\overline{a}_1(t) = \cos \lambda t,
$$
  
\n
$$
\overline{a}_2(t) = \frac{\Omega_{12}^*}{i\lambda} \sin \lambda t,
$$
  
\n
$$
\overline{a}_3(t) = \frac{\Omega_{13}^*}{i\lambda} \sin \lambda t,
$$
\n(34)

where

$$
\lambda = \sqrt{|\Omega_{12}|^2 + |\Omega_{13}|^2}.
$$
 (35)

The solution ([34](#page-3-0)) expresses oscillations of the probability amplitudes at the multiphoton resonant excitation analogously to ordinary Rabi oscillations. However, in this case the generalized Rabi frequency has essentially nonlinear dependence on the amplitudes of the wave fields (through Bessel functions).

As is seen from Eqs.  $(11)$  $(11)$  $(11)$ ,  $(12)$  $(12)$  $(12)$ , and  $(16)$  $(16)$  $(16)$  the Rabi frequency is proportional to the ratio  $d_{1j}/d_{jj}$ , while dynamic Stark shift is proportional to  $|d_{1j}|^2/d_{jj}^2$ . Therefore, for the systems with  $|d_{ij}| \geq |d_{1j}|$  the dynamic Stark shift plays minor role. However, for the large photon numbers the dynamic Stark shift of atomic levels becomes comparable to Rabi frequency and takes the states off resonance. For compensation of dynamic Stark shift one should take an appropriate detuning. The solution can also be written in explicit form when by the appropriate detunings the Stark shifts are compensated. For simplicity we will present the solution for the normal resonance when  $\Omega_{12}$ ,  $\Omega_{13}$ , and  $\Delta_{23}$  are real functions and

$$
|\Omega_{12}| \simeq |\Omega_{13}| \equiv \Omega.
$$

<span id="page-3-1"></span>Then from Eqs.  $(25)$  $(25)$  $(25)$  and  $(26)$  $(26)$  $(26)$  it follows that the desired detunings are

$$
\delta_2 = \delta_{\text{S12}} = -2\Delta_2 - \Delta_3 + \frac{\Omega_{12}}{\Omega_{13}}\Delta_{23},\tag{36}
$$

$$
\delta_3 = \delta_{\text{S13}} = -\Delta_2 - 2\Delta_3 + \frac{\Omega_{12}}{\Omega_{13}}\Delta_{23}.
$$
 (37)

<span id="page-3-3"></span><span id="page-3-2"></span>In this case the solution  $(31)$  $(31)$  $(31)$  for the system situated initially in the ground state is

$$
\overline{a}_1 = e^{-i(\Delta_2 + \Delta_3)t} \cos \sqrt{2}\Omega t,
$$
  
\n
$$
\overline{a}_2 = \frac{1}{i\sqrt{2}} \frac{\Omega_{12}}{\Omega} e^{-i(\Delta_2 + \Delta_3)t} \sin \sqrt{2}\Omega t,
$$
  
\n
$$
\overline{a}_3 = \frac{1}{i\sqrt{2}} \frac{\Omega_{13}}{\Omega} e^{-i(\Delta_2 + \Delta_3)t} \sin \sqrt{2}\Omega t.
$$
\n(38)

For  $\{n,m\}$ -photon resonance the atomic inversion oscillates at a frequency

$$
\Omega_R = 2\sqrt{2}(\omega_1 n + \omega_2 m) \left| \frac{d_{12}}{d_{22}} J_n \left( \frac{d_{22} E_1}{\hbar \omega_1} \right) J_m \left( \frac{d_{22} E_2}{\hbar \omega_2} \right) \right|.
$$
\n(39)

## **III. NUMERICAL SOLUTION (HYDROGEN ATOM)**

In this section we will apply the obtained results for hydrogen atom and will present some numerical simulations with the specific parameters of available laboratory lasers. For the hydrogen atom 1*S*-2*P* transition falls in the vacuum ultraviolet range 3/8 a.u. (hereafter we use atomic units). This is a spectral domain where strong coherent radiation is difficult to generate and two or higher photon resonant excitation is of interest. Taking into account the problem symmetry it is more appropriate to consider hydrogen atom in parabolic coordinates  $\begin{bmatrix} 5 \end{bmatrix}$  $\begin{bmatrix} 5 \end{bmatrix}$  $\begin{bmatrix} 5 \end{bmatrix}$ . If the waves have parallel polarizations, then without loss of generality we can take the axis of polarizations as the *Z* axis of parabolic coordinates. Then within the two levels with the main quantum numbers [1](#page-1-0) and 2 we have V configuration illustrated in Fig.  $1(a)$ . The excited states have opposite mean dipole moments  $(d_{33})$  $=-d_{22}=3$  a.u.), and the transition matrix elements of the electric dipole moment are  $d_{12} = -d_{13} = 2^7 / 3^5$  a.u. Hence for the coupling parameters we have

$$
\eta_{13} = (-1)^{s_1 + s_2} \eta_{12} = \frac{2^7}{3^6} (\omega_1 s_1 + \omega_2 s_2)
$$
  
 
$$
\times J_{s_1} \left( \frac{3E_1}{\omega_1} \right) J_{s_2} \left( \frac{3E_2}{\omega_2} \right) e^{is_2 \varphi}.
$$
 (40)

<span id="page-3-6"></span>For the normal resonance the dynamic Stark shift is the same for the excited levels. The latter can be calculated from Eqs.  $(27), (28), (36),$  $(27), (28), (36),$  $(27), (28), (36),$  $(27), (28), (36),$  $(27), (28), (36),$  $(27), (28), (36),$  $(27), (28), (36),$  and  $(37)$  $(37)$  $(37)$ 

$$
\delta_{\rm St} = \frac{2^{14}}{3^{12}} \sum_{s_1 \neq n} \sum_{s_2 \neq m} \frac{[3 + (-1)^{n + m + s_1 + s_2}]}{(n - s_1)\omega_1 + (m - s_2)\omega_2} \times (\omega_1 s_1 + \omega_2 s_2)^2 J_{s_1}^2 \left(\frac{3E_1}{\omega_1}\right) J_{s_2}^2 \left(\frac{3E_2}{\omega_2}\right). \tag{41}
$$

<span id="page-3-5"></span>The solution ([38](#page-3-3)) is applicable for hydrogen atom (at  $\varphi = 0$ ) and the generalized Rabi frequency will be

$$
\Omega_R \simeq \sqrt{2} \frac{2^5}{3^5} \left| J_n \left( \frac{3E_1}{\omega_1} \right) J_m \left( \frac{3E_2}{\omega_2} \right) \right|.
$$
 (42)

<span id="page-3-4"></span>

FIG. 2. (Color online) Rabi oscillations at two-photon two-color resonance  $(n=1, m=1)$  for the hydrogen atom. Time in units of the low frequency wave period. The electric field strengths are *E*<sup>1</sup>  $=E_2=0.02$  a.u.; the dynamic Stark shift and the detuning are much smaller than the Rabi frequency.

<span id="page-4-0"></span>

FIG. 3. (Color online) As in Fig. [2,](#page-3-4) but for three-photon twocolor resonance  $(n=1, m=2)$  for hydrogen atom. The electric field strengths are  $E_1 = E_2 = 0.03$  a.u.

For the numerical solution of Eq.  $(6)$  $(6)$  $(6)$  with the goal of the two color resonant excitation of hydrogen atom we consider high power excimer lasers combined with optical and infrared lasers. Figure [2](#page-3-4) displays the temporal evolution of the state populations for two-photon two-color resonance with  $F_2$  excimer laser (157 nm,  $n=1$ ) and second harmonic of Nd:YAG laser (532 nm,  $m=1$ ) with  $E_1 = E_2 = 0.02$  a.u., which corresponds to laser intensities  $I \approx 1.4 \times 10^{13} \text{ W/cm}^2$ . For the chosen laser systems the detuning of resonance is  $\delta$ = 0.00084 a.u. The generalized Rabi frequency and the dynamic Stark shift calculated by Eqs.  $(42)$  $(42)$  $(42)$  and  $(41)$  $(41)$  $(41)$  are equal to  $\Omega_R$ = 0.0063 a.u. and  $\delta_{St}$ = 0.00039 a.u., respectively. As is seen, the Rabi frequency is much larger than the detuning and the dynamic Stark shift. Hence, we see Rabi oscillations with the frequency  $(42)$  $(42)$  $(42)$ . Figure [3](#page-4-0) displays three-photon resonance with ArF excimer laser (193 nm,  $n=1$ ) and Alexandrite (655 nm,  $m=2$ ) with  $E_1=E_2=0.03$  a.u., which corresponds to laser intensities  $I \approx 3.15 \times 10^{13} \text{ W/cm}^2$ . The detuning in this case is  $\delta = 0.00019$  a.u., the generalized Rabi frequency  $\Omega_R$ =0.00633 a.u., and the dynamic Stark shift  $\delta_{\text{St}}$ = 0.00084 a.u. The oscillation frequency coincides with high accuracy with the generalized Rabi frequency ([42](#page-3-5)).

Figure [4](#page-4-1) displays four-photon two-color resonant excitation of hydrogen atom with XeF excimer  $(351 \text{ nm}, n=2)$  and Ti:sapphire (780 nm,  $m=2$ ) laser systems with  $E_1=E_2$  $= 0.025$  a.u. In this case  $\delta = 0.0014$  a.u.,  $\Omega_R = 0.00135$  a.u., and  $\delta_{\rm St}$ = 0.00139 a.u. As is seen, for these parameters the dynamic Stark shift and detuning are comparable with the Rabi frequency. Nevertheless, for the chosen electric field amplitudes the detuning is compensated by the appropriate dynamic Stark shift. The calculations were also made for the finite wave pulses describing the envelope of low frequency laser field by Gaussian function  $\exp[-(t-\sqrt{2}\tau)^2/(2\tau^2)]$ . The state populations for finite wave pulses are shown in Fig.  $4(b)$  $4(b)$  for  $\omega_2 \tau / 2\pi = 12$ . For the chosen pulse length, in the final state we have  $\sim$ 100% overpopulation.

<span id="page-4-1"></span>

FIG. 4. (Color online) Two-color four-photon resonant excitation of hydrogen atom by XeF excimer  $(n=2)$  and Ti:sapphire  $(m)$  $= 2$ ) lasers. The electric field strengths are  $E_1 = E_2 = 0.025$  a.u.; the Rabi frequency is comparable with the dynamic Stark shift, but the latter is compensated by the appropriate detuning. Temporal evolution of the state populations for (a) continuous waves and (b) Ti:sapphire laser pulse of finite duration with  $\omega_2 \tau / 2\pi = 12$ .

### **IV. CONCLUSION**

We have presented a theoretical treatment of the multiphoton resonant excitation of three-level atoms by two laser fields of different frequencies. The solution of Schrödinger equation was found using nonperturbative resonant approach. The obtained solutions express oscillations of the probability amplitudes at the multiphoton two color resonant excitation analogously to Rabi oscillations. The generalized Rabi frequency has essentially nonlinear dependence on the amplitudes of the wave fields. We also made numerical calculations for hydrogen atom assuming specific parameters of available laboratory lasers. Our calculations for hydrogen atom suggest that by the appropriate uv and optical pulses with moderately strong intensities  $\sim 10^{13}$  W/cm<sup>2</sup> one can realize two-color multiphoton resonant excitation.

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