A stationary treatment of time-dependent Hamiltonian by the many-mode Floquet formalism and its application to the study of effects of laser pulses in multiphoton processes

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Abstract

A general stationary nonperturbative treatment of time-dependent Hamiltonian system is presented by extending the many-mode Floquet formalism and a feature of initial value problem of the Schrödinger equation. The method is applied to the first analytical treatment of the effects of pulsed laser fields in multiphoton processes. Explicit analytical formulas for multiphoton transition probabilities and a generalized (multiphoton) pulse area concept for two-level quantum systems driven by pulse laser fields are obtained for the first time.

1. Introduction

Recently there has been an increased interest in nonlinear multiphoton processes in the presence of pulsed radiation fields. Almost all recent experiments on multiphoton and above threshold ionization of atoms and multiphoton and above threshold dissociation of molecules were performed by the use of short-pulse intense laser fields [1]. In general, compared to a field with constant amplitude, a pulse field has more degrees of freedom. Such a pulse property has been utilized in the solvent suppression [2] in nuclear magnetic resonance (NMR) so that the resolution can be greatly enhanced, and in selective excitation [3,4] in laser spectroscopy for controlling molecular motion. Since arbitrarily shaped laser pulses can now be generated in the 100 fs (or less) time scales in laboratories [5], it is important and necessary that the relation between the shape of pulses and excitation dynamics be understood both numerically and analytically. In this Letter, we present a new method, based on the many-mode Floquet formalism [6–8], for a nonperturbative stationary treatment (both analytically and numerically) of the pulse excitation of atoms and molecules.

The effect of a pulse has been extensively studied, limited, however, mainly to one-photon processes [9–11]. Among the theoretical studies, coherent averaging theory [12] and Magnus approximation [13] are often used. The pulse area [14], \( \int E(t) \, dt \), is an extremely useful concept in the understanding of one-photon processes. No such pulse-area concept exists for multiphoton processes. In this Letter, we advance a generalized pulse area expression for multiphoton processes for two-level systems. The two-level system is a prototype quantum system in nonlinear optics. It serves as a tool for exploring various nonlinear optical phenomena in the last several
decades, such as the population inversion by a $\pi/2$ pulse and the ‘soliton’ propagation of a pulse in the optical fiber associated with advanced optical devices.

In this Letter, a general stationary treatment of time-dependent Hamiltonian systems is presented based on an extension of the many-mode Floquet formalism [6–8] and a feature of the initial value problem of the Schrödinger equation. The generalized Floquet formalism [15–17], including the many-mode Floquet theory, have been developed and applied to non-perturbative treatments of a number of atomic and molecular multiphoton and nonlinear optical processes in intense laser fields in the past decade. Some recent applications of the methods include the prediction and explanation of some novel ‘high-intensity’ phenomena such as laser-induced stabilization of negative ions [18], chemical bond hardening [19,20] and multiple high-order harmonic generation [21] in intense laser fields. In this Letter, we derive a stationary form for the time-dependent Hamiltonian systems based on the many-mode Floquet formalism. We then use this stationary treatment to study the pulse effects on multiphoton excitation.

This Letter is organized as follows. In Section 2, we present the general stationary treatment of the time-dependent Hamiltonian in terms of the many-mode Floquet theory [6–8,15–17]. In Section 3, the stationary treatment is explicitly formulated in terms of the two-mode Floquet theory for pulsed laser fields. The fast oscillation with laser frequency $\omega$ is separated and the resulting equation contains the slowly varying laser pulse envelope. An application of this multiple time scale equation to an analytical study of the effects of laser pulses in multiphoton processes in two-level model is presented. A generalized pulse area expression is derived for the multiphoton processes for the first time. Such an analytical study is of considerable interest in the area of nonlinear optics and laser excitation of atoms and molecules. Results are given in Section 4. We conclude our study with a discussion in Section 5.

2. A stationary treatment of time-dependent Hamiltonian

The Schrödinger equation

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

is an initial value problem because of the first derivative of time $t$, involved in the equation. The wavefunction, $|\Psi(t)\rangle$, at time $t_1$, is uniquely determined by

$$|\Psi(t_1)\rangle = U(t_1, t_0) |\Psi(t_0)\rangle.$$  \hspace{1cm} (2)

Here $|\Psi(t_0)\rangle$ is the wavefunction at initial time, $t_0$, and $U(t_1, t_0)$ is the time-evolution operator and can be written formally in the time-ordering form.

$$U(t_1, t_0) = \mathcal{S} \exp \left( -\frac{i}{\hbar} \int_{t_0}^{t_1} H(t) \, dt \right),$$

where $\mathcal{S}$ is the time-ordering operator. The time-independent Hamiltonian $H(t)$ can be written as a sum of a time-independent part and time-dependent potential, i.e.

$$H(t) = H_0 + V(t).$$

From Eq. (3), it is obvious that $U(t_1, t_0)$ is uniquely determined by $H_0$, the time-independent part of the Hamiltonian, and $V(t)$, the time-dependent potential during the time $t_0$ and $t_1$ ($t_1 > t_0$). Consequently, the time-dependent potential, $V(t)$, before time $t_0$ and after time $t_1$ has no effect on the solution of the wavefunction $|\Psi(t_1)\rangle$ if the initial condition $|\Psi(t_0)\rangle$ is fixed. This physical significance of the initial value problem leads us to the following stationary approach of solving the time-dependent Schrödinger equation, Eq. (1).
The time-dependent potential, \( V(t) \), is assumed to start at time \( t_0 \) and end at time \( t_0 + T \). The wavefunction \( | \Psi(t_0) \rangle \) at time \( t_0 \) is assumed to be given either by preparation or by means of time-dependent methods. Justified by the feature of the initial value problem of the time-dependent Schrödinger equation, we impose a quasi-periodic condition to the time-dependent potential \( V(t) \). By the quasi-periodicity, we mean that the envelope of the time-dependent potential, \( V_0(t) \), is reproduced with the period \( T \) in the time domain while the other time variations of \( V(t) \) continue. In the case of pulsed laser fields, the envelope of \( V(t) \) is related to the pulse shape and the other time variations correspond to the oscillations with laser frequencies. The wavefunction, \( | \Psi(t) \rangle \), during \( t_0 \) and \( t_0 + T \) is independent to the artificial periodicity of \( V(t) \) provided that \( | \Psi(t_0) \rangle \) is fixed. Most importantly, this introduction of the quasi-periodicity of \( V(t) \) and \( H(t) \) suffices for one to cast the time-dependent Schrödinger equation in a stationary form using the many-mode Floquet formalism [6-8]. The generalized Floquet theorem [6-8,15-17] states that there are wavefunctions, \( | \Psi_A(t) \rangle \), for a quasi-periodic Hamiltonian system with incommensurate frequencies, which are the solutions of

\[
\mathcal{H} | \Psi_A(t) \rangle = \left( H_0 + V(t) - i\hbar \frac{\partial}{\partial t} \right) | \Psi_A(t) \rangle = 0 ,
\]

and \( | \Psi_A(t) \rangle \) can be written as

\[
| \Psi_A(t) \rangle = \exp(-i q_A(t)/\hbar) | q_A(t) \rangle ,
\]

where \( | q_A(t) \rangle \) is quasi-periodic in time \( t \). Substituting Eq. (6) to Eq. (5), one obtains the eigenvalue equation

\[
\mathcal{H} | q_A(t) \rangle = \left( H_0 + V(t) - i\hbar \frac{\partial}{\partial t} \right) | q_A(t) \rangle = \omega | q_A(t) \rangle ,
\]

where \( q_A \) and \( | q_A(t) \rangle \) are termed quasienergy and quasieigenstate respectively. Correspondingly, the solution, \( | \Psi_A(t) \rangle \), is called quasi-energy-state. The quasieigenstates \( | q_A(t) \rangle \) are defined in a space \( \otimes \) time extended space and form a complete subset, namely \( \sum_A | q_A \rangle \langle q_A | = I \), with the quasi-periodic condition. \( | q_A \rangle \) is defined as a vector in the extended space and \( | q_A(t) \rangle \) is defined as the projection of \( | q_A \rangle \) in the extended space into the normal space at time \( t \): \( | q_A(t) \rangle = \langle t | q_A(t) \rangle \). The time-evolution operator, \( U(t, t_0) \), can be represented, instead of the time-ordering form, by the quasi-energy-states, \( | \Psi_A \rangle \),

\[
U(t, t_0) = \sum_A | \Psi_A(t) \rangle \langle \Psi_A(t_0) | .
\]

Using Eqs. (6) and the completeness \( \sum_A | q_A \rangle \langle q_A | = I \), \( U(t, t_0) \) can be cast into a stationary form,

\[
U(t, t_0) = \sum_A | q_A(t) \rangle \exp[-i \mathcal{H} (t-t_0)/\hbar] \langle q_A(t_0) | = \langle t | \sum_A (| q_A \rangle \langle q_A | ) \exp[-i \mathcal{H} (t-t_0)/\hbar] | t_0 \rangle = \langle t | \exp[-i \mathcal{H} (t-t_0)/\hbar] | t_0 \rangle .
\]

We should point out that the term \( t-t_0 \) in Eq. (9) commutes with the extended Hamiltonian \( \mathcal{H} (=H(t)-i\hbar \partial/\partial t) \), and, to avoid misunderstanding, we write the Eq. (9) as

\[
U(t, t_0) = \langle t | \exp[-i \mathcal{H} (\tau-t_0)/\hbar] | t_0 \rangle |_{\tau=t} ,
\]

which is the projection of the super operator \( \exp[-i \mathcal{H} (\tau-t_0)/\hbar] \) in the extended space into the normal physical space at time \( t_0 \) and \( t \). Physically, the time evolution operator between time \( t_0 \) and \( t \) in the physical space is reflected by the propagation \( \exp[-i \mathcal{H} (\tau-t_0)/\hbar] \) from the supersurface at \( t_0 \) to the supersurface at \( t \) in the extended space, which bears resemblance to a physical propagation in the physical space for time-independent Hamiltonian. The very same expression for the \( U(t, t_0) \) has been obtained in a different context [22-24] using Howland's stationary scattering theory [25]. As a matter of fact, the derivation of the expression of the time-evolution operator is justified for any time \( t \) since \( t_0 \) and \( t_0 + T \) can be arbitrary. In a practical calculation, how-
ever, an extended dimension means more work. To be efficient and optimal, it is ideal to do the calculation with
the extra dimension during the application of the time-dependent potential only. In addition, it should be pointed
out that the extended Hamiltonian, \( \mathcal{H} (t) = H(t) - i \partial H / \partial t \), is always an unbound Hamiltonian in the space time
extended space, and a quasi-periodic boundary condition therefore confine the calculation performed in the
feature of \( \mathcal{L}^2 \) wavefunctions in a finite time domain. Eq. (10) can be solved expeditiously by the Faber polynomial
expansion [26], a general polynomial expansion for Hermitian and non-Hermitian Hamiltonians.

In the following, we shall focus on an analytical study of effect of the pulsed laser fields using the stationary
approach.

3. Effects of laser pulses in multiphoton processes: two-mode Floquet treatment

Atomic units, \( e = 1 \) and \( \hbar = 1 \), will be used in the rest of the Letter. In the presence of a pulsed laser field, \( E(t) \),
which can be expressed as the form
\[
E(t) = E_0(t) \cos \omega t, \quad \text{if } t_0 \leq t \leq t_0 + T,
\]
\[
E(t) = 0, \quad \text{otherwise},
\]
where \( E_0(t) \) is the envelope of the pulse field and \( \omega \) is the frequency of the laser field, the Hamiltonian of a
quantum system is
\[
H(t) = H_0 + \mu \cdot E(t) = H_0 + V(t),
\]
where \( \mu \) is the dipole operator, \( V(t) = V_0(t) \cos \omega t \) and \( V_0(t) = \mu \cdot E_0(t) \).

To solve the Schrödinger equation with the time-dependent Hamiltonian in Eq. (12) for a bound quantum
system, we begin with the stationary expression of time evolution operator, Eq. (10). In particular, the two-
mode Floquet formalism [15-17] is employed to facilitate the analytical study. The use of the two-mode Flo-
quet formalism serves the purpose of the separation of the fast oscillation, \( \cos \omega t \), from the slowly varying
envelope, \( V_0(t) \). The frequency, \( \omega \), is one of the modes. We further set the other mode, \( \Omega \), to be \( 2\pi / T \), where \( T \)
is the time when the pulse shape, \( E_0(t) \), is not zero. The time evolution operator, \( U(t, t_0) \), given in Eq. (10) yields
correct results for the wavefunction in the time interval \([t_0, t_0 + T]\), and can be written in a matrix form [6-8,15-17]
\[
U_{\alpha\beta}(t, t_0) = \sum_n \sum_{n_1 n_2} \langle \beta n_1 n_2 | q_{n_{12}}(t) \rangle \langle q_{n_{12}}(t_0) | \alpha 00 \rangle \exp \left[ i (n_1 \omega + n_2 \Omega) t \right] \exp \left[ i (n_1 \omega + n_2 \Omega) t_0 \right],
\]
where \( |\alpha\rangle \) and \( |\beta\rangle \) are two of the states of the unperturbed system \( H_0 \) \( (E_{\alpha} > E_{\beta}) \). \( \langle \beta n_1 n_2 | q_\lambda \rangle \) is a Fourier
component of the projection \( \langle \beta | q_\lambda(t) \rangle \) \( = \sum_n \langle \beta n_1 n_2 | q_\lambda \rangle \exp \left[ i (n_1 \omega + n_2 \Omega) t \right] \) because of the quasi-peri-
dericity of the quasieigenstate \( |q_\lambda(t)\rangle \), and satisfies the following equation [6-8,15-17],
\[
\sum_{\beta} \sum_{n_1 n_2} \langle H_F \rangle_{\alpha \beta}^{n_1 n_2} \langle \beta | q_{\lambda} \rangle = \sum_{\beta} \sum_{n_1 n_2} \left( \langle E_{\alpha} + n_1 \omega + n_2 \Omega \rangle \delta_{n_1 h \lambda} \delta_{n_2 h \lambda} \right.
\]
\[+ \left. \sum_n \langle V_n \rangle_{\alpha \beta} \left( \delta_{n_1 h \lambda + 1} + \delta_{n_1 h \lambda - 1} \right) \delta_{n_2 h \lambda + n} \right) \langle \beta | q_{\lambda} \rangle = q_\lambda \langle \alpha n_1 n_2 | q_\lambda \rangle,
\]
where \( H_F \) is the extended Hamiltonian \( \mathcal{H} \) and is called the (two-mode) Floquet matrix in the rest of the Letter,
\( V_n \) is the Fourier component of the time-dependent potential \( V(t) \). \( V(t) = \sum_n V_n (e^{(\omega + n \Omega) t} + e^{-i(\omega - n \Omega) t}) \),
in Eq. (12) with an imposed quasi-periodic condition.

The quasieigenstate \( |q_\lambda\rangle \) can then be solved by diagonalizing the Floquet matrix (14) and the time-evolution
operator can be evaluated via Eq. (13) in terms of quasienergy eigenvalues and eigenvectors. Alternatively, we
can use the two-mode Floquet formalism as a method to separate the fast oscillation, \( \cos \omega t \), from the slowly
time-varying envelope and hence transform the Schrödinger equation, Eq. (1), to a working equation. The resulting effective Schrödinger equation has the following form:

$$i \frac{d}{dt} \Psi(t) = \tilde{H}_F(t) \Psi(t),$$  

(15)

where $\tilde{H}_F(t)$ is given in a matrix form, i.e.

$$(\tilde{H}_F(t))_{\alpha\beta}^{m_0} = (E_{\alpha} + n_1 \omega)\delta_{\alpha\beta}\delta_{n_1,1} + \frac{1}{2} (V_0(t))_{\alpha\beta} (\delta_{n_1+1} + \delta_{n_1-1}) .$$  

(16)

The equivalence of Eqs. (14) and (15) can be easily proved by using one-mode ($\Omega$) Floquet theoretical approach to Eq. (15). As a result, the matrix element of time evolution operator, $U_{\alpha\alpha}(t, t_0)$ in Eq. (13), is

$$U_{\alpha\alpha}(t, t_0) = \sum_{n_1} \langle \beta n_1 | \tilde{O}(t, t_0) | \alpha 0 \rangle \exp(in_1 \omega t),$$  

(17)

where $\tilde{O}(t, t_0)$ is the time evolution operator of Eq. (15), which again can be represented in the extended space as

$$\tilde{O}(t, t_0) = <t| \exp[-i \tilde{H}(\tau - t_0)|t_0] >_{\tau \rightarrow \infty} ,$$  

(18)

where $\tilde{H} = \tilde{H}_F(t) - i \omega \partial / \partial t$. Eqs. (15), (17) and (18) are the working equations in which the fast oscillation with frequency $\omega$ is separated. This multiple time scale formulation of $U(t, t_0)$ in Eq. (17) provides a way to solve a quantum system in the presence of pulsed laser fields efficiently.

In this study, we apply the general formula, (15), (17) and (18) to a two-level model system. The Floquet system is nearly degenerate due to the resonance, i.e. $E_{\alpha} \approx E_{\beta} - (2n_1 + 1) \omega$, in a multiphoton transition regime. The infinite dimensional matrix in Eq. (16) can be blockly diagonalized by the generalized Van Vleck (GVV) degenerate perturbation method [27-29], and $\tilde{O}(t, t_0)$ can be obtained analytically in a reduced dimensional space. In terms of the GVV method, the two unperturbed nearly degenerate Floquet states, $|\alpha 0\rangle$ and $|\beta, N+1\rangle$, are systematically perturbed (modified) by the other states such that the coupling between the two perturbed states, the model space, and the other states, the external space, is systematically weakened until it is negligible. Finally, the GVV method transforms Eq. (15) unitarily into an effective equation

$$i \frac{d}{dt} \Psi_{\text{eff}}(t) = (\tilde{H}_F(t))_{\text{eff}} \Psi_{\text{eff}}(t)$$  

(19)

in the model space spanned by the two perturbed states, $|M_{\alpha}\rangle$ and $|M_{\beta}\rangle$, where the transformed Hamiltonian $(\tilde{H}_F(t))_{\text{eff}}$ is

$$(\tilde{H}_F(t))_{\text{eff}} = \begin{pmatrix} E_{\alpha} + \delta(t) & u(t) \\ u(t) & E_{\beta} - (2n_1 + 1) \omega - \delta(t) \end{pmatrix} ,$$  

(20)

where the terms $\delta(t)$ and $u(t)$ are the time dependent level shift and coupling respectively and are given as

$$\delta(t) = - \frac{2n_1 + 1}{2n_1 (n_1 + 1)} \frac{v^2(t)}{\omega} + \mathcal{O}(v^4), \quad u(t) = \frac{(-1)^{m_1} v^2(t) (2n_1 + 1)}{2^{2n_1} (n_1 + 1)^2 \omega^{2n_1}} + \mathcal{O}(v^{2n_1 + 3}) ,$$  

(21)

where $v(t) = \frac{1}{2} (V_0(t))_{\alpha\beta}$. To solve Eq. (19), another transformation for the wavefunction $|\Psi_{\text{eff}}\rangle = (\exp[-i \tilde{H}(t) \sigma_z] |\Psi_{\text{eff}}\rangle)$ and another GVV method are applied, where $\lambda(t) = \int_0^t [\delta(t') - \delta_0] dt'$, $\sigma_0 = \int_0^t \delta(t) dt$ and $\sigma_z$ is the Pauli matrix. The resulting $2\times2$ effective Hamiltonian is,

$$(h_F)_{\text{eff}} = \begin{pmatrix} h_{11} + \delta & f_{m_1} + \tilde{f} \\ f_{m_1} + \tilde{f} & -h_{11} - \delta + n_p \omega \end{pmatrix} ,$$  

(22)

where $h_{11} = \frac{1}{2} \delta + \delta_0$, $\delta = (2n_1 + 1) \omega - \omega_0$, $\omega_0 = E_{\beta} - E_{\alpha}$, and $f_{m_1}$ is the Fourier component defined in.
where the corresponding Fourier index \( n_p \) results from the resonance condition, i.e. \( h_1 + \delta = -h_1 - \delta + n_p \Omega \) in Eq. (22). The energy level shift \( \delta \) and coupling \( f \) are

\[
\delta = \sum_{n_p=0} \frac{f_{n_p}^2}{2h_1 - n_2 \Omega} \quad f = \sum_{n_p=0} \sum_{n_0} \frac{f_{m_2-n_p-n_2}}{(2h_1 - n_2 \Omega)(2h_1 + m_2 \Omega)}
\]

The time evolution matrix element in Eq. (17) now can be written explicitly as

\[
U_{\text{eff}}(t_0) = \left( \frac{\nu(t)}{\omega_0 - \omega} \exp[-i(2n_1 + 1)\omega t] - \frac{\nu(t)}{\omega + \omega_0} \exp(i2n_1 \omega t) \right) (\bar{U}_{\text{eff}})_{M,M'N} + (\bar{U}_{\text{eff}})_{M'MN} \exp[-i(2n_1 + 1)\omega t - i\lambda(t)]
\]

for multiphoton processes, where the time evolution matrix elements in Eq. (18) in the model space are

\[
(\bar{U}_{\text{eff}})_{M,M'N} = \sum_{j=1,2} \left( c_j - \sum_{n_p=0} \frac{f_{n_p}^2}{2h_1 + m_2 \Omega} \exp[i(m_2 + n_p)\Omega t] \right) d_j
\]

\[
(\bar{U}_{\text{eff}})_{M'MN} = \sum_{j=1,2} \left( \sum_{n_p=0} \frac{f_{n_p}^2}{2h_1 + m_2 \Omega} \exp[i(m_2 + n_p)\Omega t] c_j + d_j \right)
\]

and \( c, c_j \), and \( d_j \) are the eigenvalues and the components of the eigenvectors of the Hamiltonian (22) respectively.

Taking the lowest-order perturbation in the GVV approximation, one can solve Eq. (22) for the time evolution operator \( \bar{U}_{\text{eff}}(t, t_0) \) in the resonance,

\[
\Delta + 2\delta_0 N = (2n_1 + 1)\omega - \omega_0 + 2\delta_0 - n_p \Omega = 0,
\]

and the time evolution matrix element \( U_{\text{eff}}(t, t_0) \) in the lowest order becomes

\[
| U_{\text{eff}}(t, t_0) | = | (\bar{U}_{\text{eff}}(t, t_0))_{M,M'N} | = \left| \sin \left( \int_{t_0}^t \right) dt' u(t') \exp[i\lambda(t') \exp(-in_p \Omega t') \right] .
\]

Eq. (27) generalizes the pulse area concept [14] used in one-photon processes to multiphoton processes.

In Section 4, we shall present the results of the effective equation, Eq. (19), the time evolution matrix element, Eq. (25), and the generalized pulse area concept, Eq. (27), for three-photon processes.

4. Numerical study and analytical result

In this section we study the effects of laser pulses on the multiphoton dynamics of general two-level systems. We shall focus on the three-photon dominant regime. A Gaussian pulse starting at \( t_0 = 0 \) is used. The pulse ends at \( t = T \). At both ends, the value of the pulse decreases to essentially zero \( (10^{-10} \text{ of the maximum pulse amplitude}) \). The transition probability \( P_{\alpha \rightarrow \beta} P_{\alpha \rightarrow \beta}(t, 0) = |U_{\text{eff}}(t, 0)|^2 \), versus time \( t \) are plotted in the figures pre-
presented in this section. The energy level spacing of the two-level system under study is set to be one, i.e. $\omega_0 = 1$.

The figure in the bottom frame in Fig. 1 shows the result of the effective equation (19) for a pulse of duration of 70 optical cycles. The detuning $\delta$ is $2 \times 10^{-3}$ and the laser peak strength is 0.15. The result agrees very well with the exact result obtained by direct numerical solution of the Schrödinger Eq. (1) shown in the top frame.

The analytical result of the time evolution matrix element in Eq. (25) is shown in the bottom frame in Fig. 2 for a short pulse, 35 optical cycles. The laser peak strength is 0.1 and the detuning $\delta$ is $4 \times 10^{-2}$. The analytical result follows the exact solution in the top frame.

The analytical result, Eq. (25), in Fig. 3 shows an almost population inversion. The parameters used in Fig. 3 are the same as those in Fig. 2 except this pulse contains 350 optical cycles. To explain this phenomenon using the generalized pulse area expression given in Eq. (27), first, we notice that the approximated $|U_{\mu_0}(T, 0)|$ in Eq. (27) is $\sin(\lambda T)$ after the pulse is over. $f_{\mu_0}$ is a Fourier component defined in Eq. (23). The average level shift $\delta_0 = \int T \delta(t) \, dt$ is $5.26 \times 10^{-3}$. The three-photon resonance, Eq. (26), requires the index $n_p$ be $-29$. The Fourier component $f_{-29}$ is $2.5556 \times 10^{-1} \Omega$ and $f_{-29} \Omega = 1.02 \pi / 2$. The pulse is an almost $\pi / 2$ pulse and, therefore, a population inversion occurs after the pulse is over. Again, the analytical result in Fig. 3 follows the exact one, which is omitted.

The analytical results given here show that, in the multiphoton processes, the effects of a pulsed laser field on the excitation of a two-level system are the energy level shift through the average energy level shift $\delta_0$ and the Fourier component $f_{\mu_0}$ through the phase $\lambda(t)$.

5. Conclusion

We have presented a stationary treatment of time-dependent Hamiltonian by the many-mode Floquet formalism and an imposed quasi-periodic condition to $H(t)$. In particular, we have used the generalized two-mode Floquet theoretical approach to treat time-dependent pulsed laser fields. In this approach, the fast oscillation with frequency $\omega$ of the laser field is separated, and the resulting equations, Eqs. (15), (19) and (18), contain the slowly varying pulse envelope only. These effective formula are tested for three-photon processes in a two-level system. The analytical study presented in the work is the first attempt to study the effects of the pulses in multiphoton processes. We have applied the nearly degenerate GVV perturbation method to the explicit analytic

![Fig. 1.](image-url)
Fig. 2. (Top) a numerical exact direct solution of the Schrödinger equation (1). $\omega_0 = 1.0$, $\omega = 0.3466$ and $\delta = 4 \times 10^{-2}$. The laser peak strength is 0.1 and the pulse length $T$ is 35 o.c. (Bottom) an analytical solution of the GW method, Eq. (25). The parameters are the same as those in the top figure.

Fig. 3. An analytical solution of the GW method, Eq. (25). The parameters are the same as those in Fig. 2 except for $T = 350$ o.c.

treatment of the two-level system in multiphoton processes. We have found that the effects of the pulse result in a time-dependent energy level shift $\delta(t)$ and coupling $u(t)$. In the multiphoton processes, the energy level shift is the second-order perturbation and stronger than the coupling which is the $(2n + 1)$-order one. The analytical results of the GVV approximation have been shown to follow very closely the exact solutions in three-photon processes in the presence of pulses, which in turn show the power of the stationary treatment and the importance of the separation of fast oscillations from slow variations. Furthermore, we have derived the generalized pulse area expression and have used it to predict and explain the phenomenon of two-level population inversion in three-photon processes using a pulsed laser field. Our analytical results have shown that the main effect of a pulse envelope in multiphoton processes is the time-dependent energy level shift $\delta(t)$, which not only shifts the multiphoton resonant frequency by the average energy level shift $\delta_0$, but also controls the effective coupling $f_{\text{eff}}$. 

through an indefinite integration of the time-dependent energy level shift $\delta(t)$. The generalized pulse area expression provides a new insight in understanding the effects of the pulsed laser in multiphoton processes.

In summary, we have successfully developed and applied the new stationary formulation of time-dependent Hamiltonian system to study the effects of the pulsed laser fields in multiphoton processes analytically. We shall be applying the stationary approach to study some realistic time-dependent Hamiltonian systems numerically in the future.

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