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Analytic solution and pulse area theorem for three–level atoms

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We report a new analytic solution for three–level atom driven by arbitrary time-dependent electromagnetic pulses. In particular, we consider far–detuned driving pulses and show an excellent match between our analytic result and the numerical simulations. We use our solution to derive a pulse area theorem for three–level V and Λ–systems without making the rotating wave approximation. Formulated as an energy conservation law, this pulse area theorem can be used to understand pulse propagation though a three-level media.

The classic McCall-Hahn theorem [1] shows that a soliton with even-π pulse area propagates freely through a strongly interacting two–level atomic media. This fundamental result has generated much interest in studying pulse propagation in two-level systems [2]. In particular, in attempting to find accurate analytic expressions for a driven two–level system beyond the rotating wave approximation (RWA) [3–5], arbitrary pulse propagation through a two-level media [6–8], and control of a soliton propagation [9, 10].

The successes in two-level media have encouraged consideration of pulse propagation in three level media where rich phenomenon such as electromagnetically induced transparency in the Λ-scheme [11–13] and lasing without inversion in the V-scheme [14–16] represent modern quantum optics. Despite the advances in three level media, a theory of soliton propagation and in particular a pulse area theorem for three level systems is currently missing. So far soliton propagation in three level media has only been considered numerically [17] or analytically treated for limited special cases [18–21].

In this Letter we explore the dynamics of three-level systems driven by two arbitrary time-dependent electromagnetic fields, without using the RWA. First we introduce a new method based on the time evolution operator, that allows us to derive an analytic solution for three–level systems. Then we compare our analytic results with numerical simulations, to establish when our approach is useful. In particular, we show that the new solution works well for far-detuned pulses, when the ratio of the peak Rabi frequency to the detuning of central field frequency can be regarded as a small parameter. Based on this new solution we derive a pulse area theorem for three–level systems. In the special case of a single resonant field treated within the RWA, the new pulse area theorem reduces to the classic McCall-Hahn theorem.

The pulse area theorem was motivated by the recent development of the new type of laser, based on the quantum amplification by superradiant emission of radiation (QASER). [22, 23]. The two-level atomic media in the QASER description is driven by a strong far-off resonant field where the RWA breaks down. Therefore the classic McHall-Hahn theorem cannot be applied for this case. We have successfully applied the new pulse area theorem that is valid beyond the RWA and achieved the parametric excitation of a driven two–level atomic media [24].

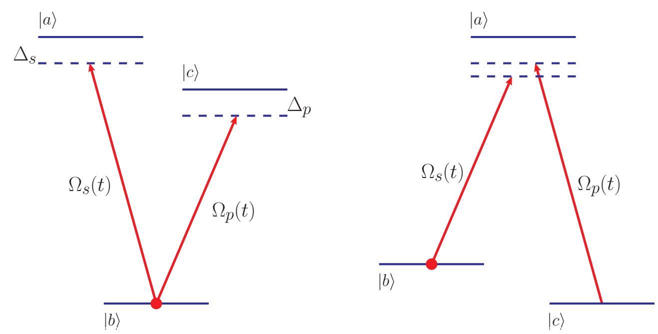

FIG. 1. The three level V and Λ–systems interacting with the electromagnetic pulses $\Omega_s(t)$ and $\Omega_p(t)$. The atomic transition frequencies are $\omega_{ab} = \omega_a - \omega_b$ and $\omega_{cb} = \omega_c - \omega_b$. The initial state is prepared in the ground state, $|\psi(t_0)\rangle = |b\rangle$. (Color online)

The exact Hamiltonian of a driven three-level V–system in the interaction picture is

$$H^V_{int}(t) = -\hbar \begin{pmatrix} 0 & \Omega_s(t)e^{i\omega_{ab}t} & 0 \\ \Omega^*_s(t)e^{-i\omega_{ab}t} & 0 & \Omega^*_p(t)e^{-i\omega_{cb}t} \\ 0 & \Omega_p(t)e^{i\omega_{cb}t} & 0 \end{pmatrix}. \tag{1}$$

Here the time-dependent Rabi amplitudes, $\Omega_s(t) = \Omega_s\Sigma_s(t)\cos(\nu_s t)$ and $\Omega_p(t) = \Omega_p\Sigma_p(t)\cos(\nu_p t)$ are defined for an arbitrary time–dependent electromagnetic pulse envelopes $\Sigma_s(t)$ and $\Sigma_p(t)$. The peak Rabi frequencies $\Omega_s = \wp_{ab}E_s/\hbar$ and $\Omega_p = \wp_{cb}E_p/\hbar$ are defined in terms of the maximal values of the electromagnetic fields E_s and E_p that drive the population between the ground state $|b\rangle$ and the excited states $|a\rangle$ and $|c\rangle$. The carrier driving frequencies are ν_s and ν_p. The matrix elements of the dipole moments are $\wp_{ab} = e\langle a|r|b\rangle$ and $\wp_{cb} = e\langle c|r|b\rangle$. The relative atomic transition frequencies $\omega_{ab} = \omega_a - \omega_b$ and $\omega_{cb} = \omega_c - \omega_b$ are defined in terms of the energies of the individual atomic levels ω_a, ω_b, and ω_c.

We start with the derivation of a new analytical solution for a three–level system interacting with an arbitrarily time-dependent electromagnetic field. Our approach is based on the time evolution operator $U(t)$ represented by the time-ordered T-exponent of the Hamiltonian for a driven three–level system. The time evolution operator $U(t)$ keeps both slowly and rapidly oscillating terms in the Hamiltonian and therefore leads to a solution valid for an arbitrary time-dependent electromagnetic field beyond the RWA. The time ordering procedure can be done by breaking the total time interval $(t_f - t_i)$ into N infinitesimal intervals $\Delta\tau = (t_f - t_i)/N$ and for each discrete time, $t_k = t_i + [(2k-1)/2]\,\Delta\tau$, we evaluate the original Hamiltonian $H_{int}(t_k)$. Then the time-evolution operator is represented by,

$$U(t) = \widehat{T}\exp\left[-\frac{i}{\hbar}\int_{t_i}^{t_f} dt' H_{int}(t')\right] = \exp\left[-\frac{i}{\hbar}\Delta\tau H_{int}(t_N)\right]\cdots\exp\left[-\frac{i}{\hbar}\Delta\tau H_{int}(t_1)\right]. \tag{2}$$

By collecting the infinitesimal contributions in the limit of an infinitely small time interval $\Delta\tau \longrightarrow 0$ and infinitely large number of steps $N \longrightarrow \infty$ we arrive at the Magnus expansion [25],

$$U(t) = \exp\left[\sum_{n=1}^{\infty} S^{(n)}\right]. \tag{3}$$

The first terms in the Magnus expansion can be obtained by means of the Baker–Campbell–Hausdorff formula,

$$S^{(1)}(t) = -\frac{i}{\hbar}\int_0^t dt_1 H_{int}(t_1), \tag{4}$$

and the second term is

$$S^{(2)}(t) = \left(-\frac{i}{\hbar}\right)^2 \frac{1}{2}\int_0^t dt_1 \int_0^{t_1} dt_2 [H_{int}(t_1), H_{int}(t_2)]. \tag{5}$$

This procedure can be further generalized to get an explicit expression for the n^{th} term in the Magnus expansion $S^{(n)}(t)$ in terms of integrals over sums of nested commutators, see for example [26].

Now we introduce our only approximation for a far-detuned field. In this case we can perform perturbation theory by truncating the infinite series in the exponential keeping solely the leading order term in the Magnus expansion, given by Eq.(4). Here we will first focus on the V-scheme, and later apply the same method to the Λ-scheme. By projecting the time evolution operator onto the ground state,

$$|\psi(t)\rangle = U(t)|b\rangle \tag{6}$$

we obtain the new solution of a three-level V-scheme beyond the rotating wave approximation for a general time-dependent pulse shape. In order to find the state given by Eq.(6) we need to diagonalize the matrix $S^{(1)}(t)$ which is a simple task after rewriting it in terms of the complex pulse areas

$$\theta_s(t) = \int_0^t dt\; \Omega_s(t)\exp\left[i\omega_{ab}t\right] \tag{7}$$

$$\theta_p(t) = \int_0^t dt\; \Omega_p(t)\exp\left[i\omega_{cb}t\right] \tag{8}$$

Here $\Omega_s(t)$ is the entire time dependence of the field including slow and fast oscillations. This matrix can be easily diagonalized using the standard method of eigenvalues and eigenvectors. Then the three eigenvalues of the matrix are zero, positive and negative effective pulse area,

$$\theta(t) = \sqrt{|\theta_s|^2 + |\theta_p|^2}. \tag{9}$$

With the initial condition for the population prepared in the ground state, the solution for a three-level V-scheme is,

$$|\psi^V(t)\rangle = i\theta_s(t)\frac{\sin\left[\theta(t)\right]}{\theta(t)}|a\rangle + \cos\left[\theta(t)\right]|b\rangle + i\theta_p(t)\frac{\sin\left[\theta(t)\right]}{\theta(t)}|c\rangle. \tag{10}$$

The same approach can be applied to the case of Λ-system, described by the exact Hamiltonian $H_{int}^{\Lambda}(t)$ in the interaction picture,

$$H_{int}^{\Lambda}(t) = -\hbar\begin{pmatrix} 0 & \Omega_s(t)e^{i\omega_{ab}t} & \Omega_p(t)e^{i\omega_{ac}t} \\ \Omega_s^*(t)e^{-i\omega_{ab}t} & 0 & 0 \\ \Omega_p^*(t)e^{-i\omega_{ac}t} & 0 & 0 \end{pmatrix}. \tag{11}$$

Following the same method as for the V-scheme, we obtain the solution for the Λ-scheme in terms of the complex field areas

$$|\psi^\Lambda(t)\rangle = \frac{i\theta_s(t)\sin\left[\theta(t)\right]}{\theta(t)}|a\rangle + \frac{\theta_s\theta_p^*}{|\theta(t)|^2}\left(\cos\left[\theta(t)\right] - 1\right)|c\rangle + \frac{\left(|\theta_p(t)|^2 + |\theta_s(t)|^2\cos\left[\theta(t)\right]\right)}{|\theta(t)|^2}|b\rangle, \tag{12}$$

where the pulse area θ_s is given by Eq.(7) while the pulse area θ_p is now modulated by the atomic transition frequency $\omega_{ac} = \omega_a - \omega_c$ for a Λ–scheme compared to ω_{cb} as in V–scheme,

$$\theta_p(t) = \int_0^t dt\; \Omega_p(t)\exp\left[i\omega_{ac}t\right] \tag{13}$$

In the special case of resonant CW driving field treated within RWA, i.e. $\Omega_s(t) \simeq \Omega_s e^{-i\nu_s t}$ and $\Omega_p(t) \simeq \Omega_p e^{-i\nu_p t}$, we can simplify the complex area variables into

$$\theta_s(t) = \frac{\Omega_s t}{2}, \tag{14}$$

$$\theta_p(t) = \frac{\Omega_p t}{2}. \tag{15}$$

and then from Eq.(10), we immediately obtain the conventional RWA solution for a three-level $V-$system,

$$|\psi(t)\rangle = \frac{i\Omega_s}{\Omega}\sin\left[\frac{\Omega t}{2}\right]|a\rangle + \cos\left[\frac{\Omega t}{2}\right]|b\rangle + \frac{i\Omega_p}{\Omega}\sin\left[\frac{\Omega t}{2}\right]|c\rangle \tag{16}$$

where the effective Rabi frequency is

$$\Omega = \sqrt{\Omega_s^2 + \Omega_p^2}. \tag{17}$$

Likewise for the Λ-scheme, the solution given by Eq.(12) simplifies under the resonant CW driving field to the conventional RWA solution,

$$\begin{aligned} |\psi(t)\rangle = \frac{i\Omega_s}{\Omega}\sin\left[\frac{\Omega t}{2}\right]|a\rangle + \frac{\Omega_s\Omega_p}{\Omega^2}\left(\cos\left[\frac{\Omega t}{2}\right] - 1\right)|c\rangle \\ + \frac{1}{\Omega^2}\left(\Omega_p^2 + \Omega_s^2\cos\left[\frac{\Omega t}{2}\right]\right)|b\rangle \end{aligned} \tag{18}$$

In order to compare our new solution with the exact numerics we will consider pulses of the form

$$\Omega(t) = \Omega_R\Sigma(t)\cos(\nu t), \tag{19}$$

where ν is the central frequency of the pulse, $\Omega_R = \wp_{ab}E_0/\hbar$ is the peak pulse Rabi frequency, and $\Sigma(t)$ is the pulse envelope. Here we will restrain our examples to pulses with the varying envelope function

$$\Sigma(t) = \tanh\left[10^q(t+\tau_p)/\tau_p\right] - \tanh\left[10^q(t-\tau_p)/\tau_p\right], \tag{20}$$

which gives pulse envelopes of duration $2\tau_p$ and with a shape controlled by the parameter q. For $q \geq 1$ we essentially have a square pulse as shown in Fig.(3). With $q < 1$ the square pulse smooths out until for $q = 0$ the envelope is nearly Gaussian as can be seen in Fig.(2).

Now we can examine the approximation of dropping all but the first term of the Magnus expansion. Considering the fields defined by Eq.(19) it can be seen that the n-th term in the Magnus expansion is proportional to the n-th power of the integral over the CW square pulse in the RWA

$$\int_0^t dt\;\Omega(t)\exp\left[i\omega t\right] \simeq \frac{\Omega_R}{\Delta}\sin\left[\frac{\Delta t}{2}\right]\exp[i\Delta t/2]. \tag{21}$$

Thus the approximation of dropping higher order terms in the Magnus expansion of the evolution operator is essentially a perturbation theory in the evolution operator with the small parameter, $\Omega_R/\Delta \ll 1$. Here $\Delta = \omega - \nu$ is the detuning between central field frequency and the transition frequency (either ω_{ab} or ω_{cb}). To get higher accuracy one should keep the higher order terms given by Eq.(5), but for the far-detuned case the leading term describes the dynamics of the light-matter interaction exceptionally well as one can see from the agreement between the analytic solution and numerical simulations. For both the case of adiabatic Gaussian-like pulse as shown in Fig.2 and non-adiabatic square pulse as shown in Fig.3 there is a strong match between our analytic expression and the exact numerical simulations.

The obtained solution allows us to formulate the pulse area theorem for a driven three-level $V-$scheme. The Maxwell-Schrödinger equations in the slowly varying amplitude approximation are given by

$$\left(\frac{\partial}{\partial t} + c\frac{\partial}{\partial z}\right)\frac{\partial}{\partial t}\theta_s(t) = -i\Omega_a^2\rho_{ab} \tag{22}$$

$$\left(\frac{\partial}{\partial t} + c\frac{\partial}{\partial z}\right)\frac{\partial}{\partial t}\theta_p(t) = -i\Omega_c^2\rho_{cb} \tag{23}$$

Here Ω_a, and Ω_c are the collective frequencies proportional to the dipole moments associated with the transitions between the levels a and b and the levels c and b correspondingly,

$$\Omega_a^2 = \frac{3}{8\pi}n\lambda_{ab}^2\gamma c, \tag{24}$$

$$\Omega_c^2 = \frac{3}{8\pi}n\lambda_{cb}^2\gamma c \tag{25}$$

Here n is the atomic density, λ_{ab} and λ_{cb} are the atomic wavelengths, and γ is the spontaneous decay rates.

If we disregard space propagation and combine the Maxwell–Schrödinger equations for $\theta_s(t)$ and $\theta_p(t)$ and their complex conjugates to form a full time derivative the left hand side of Eq.(22, 23) becomes,

$$\frac{\partial}{\partial t}\left(\frac{1}{\Omega_a^2}\left|\frac{\partial}{\partial t}\theta_s(t)\right|^2 + \frac{1}{\Omega_c^2}\left|\frac{\partial}{\partial t}\theta_p(t)\right|^2\right) \tag{26}$$

Due to the density matrix equations the right hand side of Eq.(22, 23) turns into a full–time derivative of the population in the ground state,

$$\begin{aligned} &-i\left[\rho_{ab}^*(t)\frac{\partial\theta_s}{\partial t} - \rho_{ab}(t)\frac{\partial\theta_s^*}{\partial t}\right] \\ &-i\left[\rho_{bc}(t)\frac{\partial\theta_p}{\partial t} - \rho_{bc}^*(t)\frac{\partial\theta_p^*}{\partial t}\right] = \frac{\partial\rho_{bb}}{\partial t} \end{aligned} \tag{27}$$

With the population in the ground state,

$$\rho_{bb}(t) = \cos^2\left(\sqrt{|\theta_s|^2 + |\theta_p|^2}\right) \tag{28}$$

we obtain the conservation law,

$$\begin{aligned} &\frac{\partial}{\partial t}\left(\frac{1}{\Omega_a^2}\left|\frac{\partial}{\partial t}\theta_s(t)\right|^2 + \frac{1}{\Omega_c^2}\left|\frac{\partial}{\partial t}\theta_p(t)\right|^2\right) = \\ &-\frac{\partial}{\partial t}\cos^2\left(\sqrt{|\theta_s|^2 + |\theta_p|^2}\right) \end{aligned} \tag{29}$$

Taking into account the initial conditions for both Ω_s and Ω_p pulses,

$$\left|\frac{\partial}{\partial t}\theta(t_0)\right|^2 = |\theta(t_0)|^2 = 0 \tag{30}$$

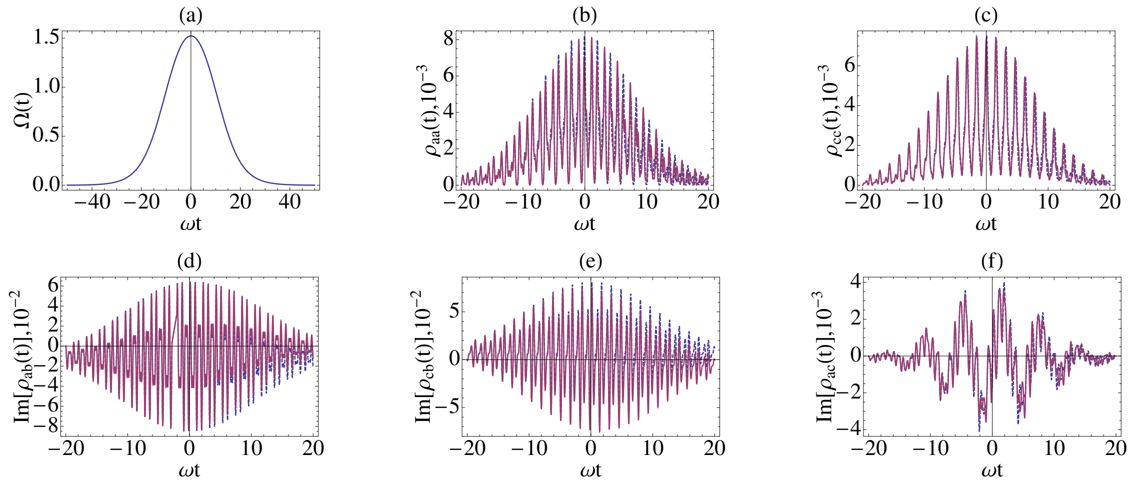

FIG. 2. Atomic three-level V-system interacting with a far detuned electromagnetic continuous wave pulses, $\Omega_s(t) = \Omega_s\Sigma(t)\cos(\nu_s t)$ and $\Omega_p(t) = \Omega_p\Sigma(t)\cos(\nu_p t)$ with $\nu_s = 3$, $\Omega_s = 3/5$ and $\nu_p = 2$, $\Omega_p = 1/2$. The pulse envelope is given by Eq.(20) with parameters $q = 0$ and $\tau_p = 10$. The driven atom has $\omega_{ab} = 12$ and $\omega_{cb} = 10$. Here the solid (red) line is the exact numerical solution and the dashed (blue) line is our approximate analytic solution from Eq.(10). For this case the two are almost perfectly matched. For this system we plot several key variables: (a) The pulse envelope. (b) The population in level a. (c) The population in level c. (d) The imaginary part of the coherence ρ_{ab}. (e) The imaginary part of the coherence ρ_{cb}. (f) The imaginary part of the coherence ρ_{ac}. (Color online)

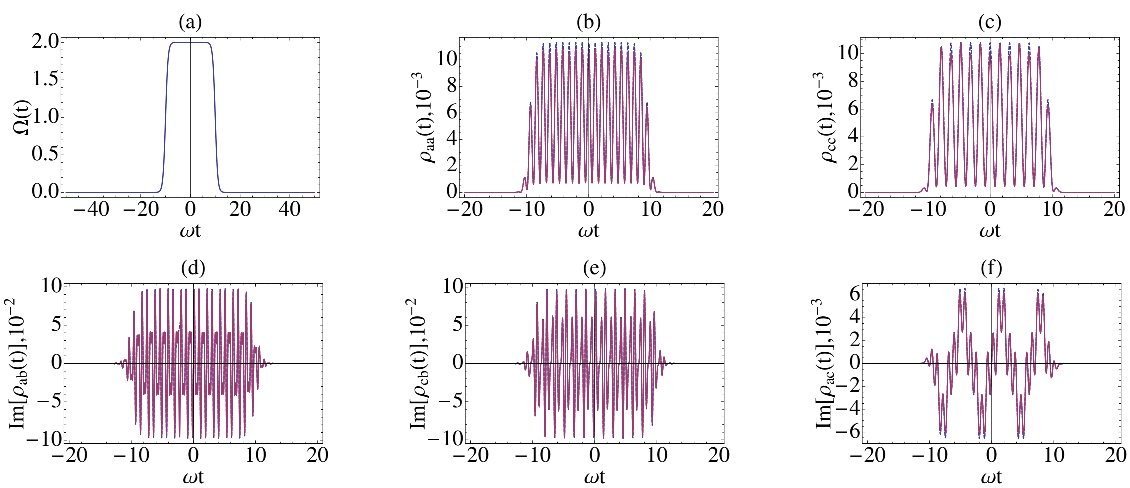

FIG. 3. Atomic three-level V-system interacting with a far detuned electromagnetic continuous wave pulses, $\Omega_s(t) = \Omega_s\Sigma(t)\cos(\nu_s t)$ and $\Omega_p(t) = \Omega_p\Sigma(t)\cos(\nu_p t)$ with $\nu_s = 3$, $\Omega_s = 3/5$ and $\nu_p = 2$, $\Omega_p = 1/2$. The pulse envelope is given by Eq.(20) with parameters $q = 1$ and $\tau_p = 10$. The driven atom has $\omega_{ab} = 12$ and $\omega_{cb} = 10$. Here the solid (red) line is the exact numerical solution and the dashed (blue) line is our approximate analytic solution from Eq.(10). For this case the two are almost perfectly matched. For this system we plot several key variables: (a) The pulse envelope. (b) The population in level a. (c) The population in level c. (d) The imaginary part of the coherence ρ_{ab}. (e) The imaginary part of the coherence ρ_{cb}. (f) The imaginary part of the coherence ρ_{ac}. (Color online)

we arrive at the Pulse Area Theorem for a three–level V–system in terms of the pulse areas (7, 8),

$$\frac{1}{\Omega_a^2}\left|\frac{\partial}{\partial t}\theta_s(t)\right|^2 + \frac{1}{\Omega_c^2}\left|\frac{\partial}{\partial t}\theta_p(t)\right|^2 = \sin^2\left(\sqrt{|\theta_s|^2 + |\theta_p|^2}\right) \tag{31}$$

In terms of the energy densities of the fields we obtain,

$$\left|\frac{\Omega_s(t)}{\Omega_a}\right|^2 + \left|\frac{\Omega_p(t)}{\Omega_c}\right|^2 = \sin^2\left(\sqrt{|\theta_s|^2 + |\theta_p|^2}\right) \tag{32}$$

The new pulse area theorem states that energy density of the fields $\Omega_s(t)$ and $\Omega_p(t)$ is transferred to the atomic population in the excited state of the system that in turn is expressed in terms of the pulse areas $\theta_s(t)$ and $\theta_p(t)$.

The preceding procedure applied for a Λ–scheme yields the new Pulse Area Theorem for a Λ–system, expressed in terms of the pulse areas (7, 13),

$$\frac{1}{\Omega_a^2}\left|\frac{\partial}{\partial t}\theta_s(t)\right|^2+\frac{1}{\Omega_c^2}\left|\frac{\partial}{\partial t}\theta_p(t)\right|^2=\frac{|\theta_s|^2\sin^2\left(\sqrt{|\theta_s|^2+|\theta_p|^2}\right)}{|\theta_s|^2+|\theta_p|^2}. \tag{33}$$

We see that the pulse evolution $\theta_s(t)$ affects the evolution of the $\theta_p(t)$ and vice versa, through the modulation of the population in the excited state. Thus the new pulse area theorem can be used to control and analyze pulse propagation though a three-level media.

In order to take into account the influence of decay mechanism and broadening phenomena one should solve the density matrix equations that includes the decay matrix Γ,

$$\frac{\partial}{\partial t}\rho(t)=-\frac{i}{\hbar}[H(t),\rho(t)]-\frac{1}{2}\{\Gamma,\rho\} \tag{34}$$

in terms of the commutator $[H(t),\rho(t)]$ and anticommutator $\{\Gamma,\rho\}$. Now we introduce effective non-Hermitian Hamiltonian

$$\mathscr{H}(t)=H(t)-\frac{i}{2}\hbar\Gamma \tag{35}$$

that is given in terms of the Hermitian Hamiltonian $H^\dagger=H$ and decay matrix $\Gamma^\dagger=\Gamma$. Using the standard definition of the density matrix, $\rho(t)=\sum P_\psi|\psi(t)\rangle\langle\psi(t)|$, one can show that the Schrödinger equation with the effective non-Hermitian Hamiltonian $\mathscr{H}(t)$,

$$\frac{\partial}{\partial t}|\psi(t)\rangle=-\frac{i}{\hbar}\mathscr{H}(t)|\psi(t)\rangle \tag{36}$$

is equivalent to the density matrix equation (34 that includes decay matrix Γ. Therefore in order to take into account the decay mechanism and broadening phenomena into the current approach based on the time evolution operator $U(t)$, one should consider the effective non-Hermitian Hamiltonian (35).

In conclusion we investigated the dynamics of three-level systems driven by two arbitrarily time-dependent electromagnetic fields, without referring to the rotating wave approximation (RWA). In particular, we considered far-detuned driving pulse envelopes where the rotating wave approximation breaks down and show excellent agreement between the new analytic solution and the numerical simulations. The new solution is based on the time evolution operator that operates with a time–dependent Hamiltonian and allows to keep both slowly and rapidly oscillating terms. With the new solution we formulate the new pulse area theorem for three–level V-system and Λ–system without making the rotating wave approximation. Formulated as the energy conservation law, the pulse area theorem for three–level systems provides a new tool for pulse manipulation and coherent control of three–level atoms.

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