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PAPER

Few-cycle excitation of atomic coherence: an analytical solution beyond the rotating-wave approximation

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Abstract

Developing an analytical theory for atomic coherence driven by ultrashort laser pulses has proved to be challenging due to the breakdown of the rotating wave approximation (RWA). In this paper, we present an approximate analytical solution that describes a two-level atom under the excitation of a far-off-resonance, few-cycle pulse of arbitrary shape without invoking the RWA. As an example of its applicability, a closed-form solution for Gaussian pulses is explicitly given, and the result is used to analyse the impact of carrier envelope phase on atomic population ratios. Comparisons with numerical solutions validate the accuracy our solution within the scope of the approximation. Finally, we outline an alternative approach that can lead to a more accurate solution by capturing the nonlinear behaviors of the system. The work lays out feasible theoretical paths toward analytically describing two-level atoms driven by ultrashort pulses.

1. Introduction

Quantum coherent control (QCC) is of great importance in fundamental physics as well as a breadth of emerging applications [1]. With the emergence of femtosecond and attosecond light sources, control of atomic coherence using ultrafast laser pulses with very few optical cycles has attracted growing interest in recent years [2–15]. Apart from its significance in quantum theories, ultrafast QCC has profound implications in practical applications. For example, in certain QCC schemes, using ultrashort, broadband pulses allows the first electronic states of molecules to be accessible and, at the same time, enables fast population transfer that occurs well within the typical collision times [16, 17]. Few-cycle pulses can also excite coherence on high-frequency transitions that enables efficient generation of extreme ultraviolet (XUV) radiations [12, 18].

Studying ultrafast QCC in the few-cycle regime faces unique challenges. The ultrashort pulse duration invalidates the slowly-varying envelope approximation (SVEA) [19], while the high peak field causes breakdown of the rotation-wave approximation (RWA) [13]. As a result, the well-established theoretical framework based on the optical Bloch equations and the area theorem ceases to apply [20, 21]. Theoretical analysis has to rely on the Bloch equations or the Schrödinger equation in their original forms without simplifications, which are often highly nonlinear. This significantly increases the difficulty of developing analytical theories. In most cases, numerical simulations have to be used when dealing with few-cycle light–matter interactions [21–24].

Meanwhile, there has been a continued effort to develop analytical theories for atomic coherence driven by few-cycle pulses [2, 7–15]. Such an effort is motivated by the fact that analytical theories are able to offer general pictures of the atomic responses, which is often lacking in numerical solutions. For example, carrier envelope phase (CEP) is an important factor in coherent excitation by few-cycle pulses [7]. It has been shown that a closed-form relation between atomic inversion and pulse CEP can be obtained under certain conditions [15], which provides valuable insight and general guidance in the study of CEP-sensitive quantum coherence. A notably successful theory, proposed first by Rostovtsev *et al*, considers the coherence of a two-level atom under the excitation of a far-off-resonance strong ultrashort pulse [9]. Through a perturbative scheme, the model gives rise to a general solution of the Schrödinger equation without invoking the RWA. The original solution, however, is not in closed form, and the analysis of its features useful for practical applications still has to rely largely on numerical computations. Several attempts have been made to derive more explicit solutions under specific conditions [11, 15, 25]. In particular, it has recently been shown that a simple, closed-form analytical solution of the Schrödinger equation can be obtained for few-cycle *square* pulses [15].

The apparent limitation of this solution is that it only works for a highly idealized pulse shape, which restricts its applicability. In the current paper, an analytical theory encompassing arbitrary pulse shapes is presented. Atomic inversion driven by a few-cycle *Gaussian* pulse is analysed as an example of the general solution, and an explicit, closed-form solution is given. The solution is then used to examine the impact of CEP on the population ratio between the two states for Gaussian pulses. The accuracy of the solution is verified by comparing it to the exact numerical solution of the general equations of motion. Finally, an alternative approach to simplify the theory is suggested and is shown to produce a potentially more accurate solution with a closer representation of the nonlinear behaviors of the system and a broader scope of applicability.

2. An analytical general solution

2.1. General model

Our general model follows the theoretical framework described in [9]. A quick outline is given below. We consider a two-level system (TLS) under the influence of an electromagnetic field. The Hamiltonian of the system is

$$\hat{H} = \hbar\omega_c|c\rangle\langle c| - \mu\mathcal{E}(t)|c\rangle\langle d| - [\mu\mathcal{E}(t)]^*|d\rangle\langle c|, \tag{1}$$

where $|c\rangle, |d\rangle$ are upper and lower levels, respectively, ω_c is the transition frequency, $\mathcal{E}(t)$ is the electric field, and μ is the dipole moment of the system. We are interested in the electric field of the form $\mathcal{E}(t) = E(t)\cos(\omega t + \phi)$, where $E(t)$ is the pulse envelope function and ϕ is optical phase, also called carrier-envelope phase(CEP). Note that ϕ is included here as an extra degree of freedom for the consideration of CEP, an important concept in the context of few-cycle excitation [15, 24]. It also allows the theory to potentially analyse *chirped pulses*, which can be described via a time-dependent $\phi(t)$.

With this Hamiltonian, the equations of motion for the system are given by

$$\dot{C}(t) = -i\Omega(t)\cos(\omega t + \phi)e^{i\omega_c t}D(t), \tag{2a}$$

$$\dot{D}(t) = -i\Omega^*(t)\cos(\omega t + \phi)e^{-i\omega_c t}C(t), \tag{2b}$$

where $C(t)$ and $D(t)$ are the amplitudes of the two states $|c\rangle$ and $|d\rangle$, respectively, i.e., $|\Psi\rangle = C(t)e^{-i\omega_c t}|c\rangle + D(t)|d\rangle$, and $\Omega(t) = \mu E(t)/\hbar$ is the Rabi frequency.

It proves useful to introduce the following quantity to simplify our equations at this point

$$\theta(t) = \int_{-\infty}^{t} \Omega(t')\cos(\omega t' + \phi)e^{i\omega_c t'}dt'. \tag{3}$$

With this definition, the equations (2a), (2b) become

$$\dot{C}(t) = -i\dot{\theta}(t)D(t), \tag{4a}$$

$$\dot{D}(t) = -i\dot{\theta}^*(t)C(t). \tag{4b}$$

In this paper, we also assume the ultrashort pulse excitation to be non-zero only within a finite time interval $t \in [-\tau, \tau]$, and to have a sharp cut-off outside of this interval. We will imply this throughout the paper, even when we use the limits of integration that start at $-\infty$.

By introducing the quantity

$$f = \frac{C(t)}{D(t)} \tag{5}$$

the equations of motion (4) can be simplified to

$$\dot{f}(t) = i\dot{\theta}^*(t)f^2(t) - i\dot{\theta} \tag{6}$$

The main objective of this paper is to analyse the equation (6), and specifically to find approximate analytical solutions to this equation.

2.2. First step: a sequence of approximate solutions

To accomplish our goal, we consider a sequence of successive approximate solutions. The zeroth-order approximate solution $f_0(t)$ of (6) is obtained by neglecting the $f^2(t)$ term in (6). To be specific, we assume that $f_0(t)$ satisfies the equation

$$\dot{f}_0(t) = i\dot{\theta}^*(t)f_0^2(t) - i\dot{\theta}, \tag{7}$$

and also the condition

$$f_0^2(t) \ll 1, \tag{8}$$

which simplifies the equation to

$$f_0(t) = -i\theta(t). \tag{9}$$

Next, we want to find a first-order approximate solution $f_1(t)$. Intuitively, we want this first-order solution to be close to the zeroth order: $f_1 \approx f_0$. Specifically, the condition that is of importance is that

$$(f_1(t) - f_0(t))^2 \ll f_0^2(t). \tag{10}$$

The strategy that makes use of this condition to simplify equation (6) was introduced in [9], where the identity

$$f_1^2 = (f_1 - f_0)^2 + 2f_0 f_1 - f_0^2 \tag{11}$$

is used to obtain the relation

$$f_1^2 \approx 2f_0 f_1 - f_0^2. \tag{12}$$

Hence, the first-order approximation must satisfy (6),

$$\dot{f}_1(t) = i\dot{\theta}^*(t)f_1^2(t) - i\dot{\theta}, \tag{13}$$

which can be simplified using (12) to yield

$$\dot{f}_1(t) = 2\theta(t)\dot{\theta}^*(t)f_1(t) + i\theta^2(t)\dot{\theta}^*(t) - i\dot{\theta}(t). \tag{14}$$

Note that unlike (13), this equation is linear. The general solution to (14) is, as was already found in [15],

$$f_1(t) = -\frac{i}{2}[\theta(t) + \int_{-T}^{t} \dot{\theta}(t')e^{\alpha(t',t)}dt'], \tag{15}$$

where $\alpha(t', t) = 2\int_{t'}^{t} \theta(t'')\dot{\theta}^*(t'')dt''$.

The above strategy based on the condition (10) and the identity (11) can be generalized to obtain further approximate solutions to the main equation (6). In fact, let us suppose that the kth-order approximate solution $f_k(t)$ is already found, and then assume that the $(k+1)$ th-order approximate solution $f_{k+1}(t)$ satisfies the condition

$$(f_{k+1}(t) - f_k(t))^2 \ll f_k^2. \tag{16}$$

Next, apply the identity

$$f_{k+1}^2 = (f_{k+1} - f_k)^2 + 2f_k f_{k+1} - f_k^2 \tag{17}$$

to find that

$$f_{k+1}^2 \approx 2f_k f_{k+1} - f_k^2. \tag{18}$$

Assuming that f_{k+1} satisfies the main equation (6),

$$\dot{f}_{k+1}(t) = i\dot{\theta}^*(t)f_{k+1}^2(t) - i\dot{\theta}, \tag{19}$$

we can use (18) to get a simplified equation

$$\dot{f}_{k+1} = 2if_k(t)\dot{\theta}^*(t)f_{k+1}(t) - if_k^2(t)\dot{\theta}^*(t) - i\dot{\theta}(t). \tag{20}$$

This recursive equation is once again linear in $f_{k+1}(t)$, and hence can be solved explicitly, although the solution is rather cumbersome:

$$\begin{aligned} f_{k+1} = {}& \exp\left(2i\int_{-\infty}^{t} f_k(t')\dot{\theta}^*(t')dt'\right) \\ & \times \left(\int_{-\infty}^{t} e^{-2i\int_{-\infty}^{t'} f_k(t'')\dot{\theta}^*(t'')dt''}[-if_k^2(t')\dot{\theta}^*(t') - i\dot{\theta}(t')]dt'\right). \end{aligned} \tag{21}$$

To simplify it, we integrate the first integrand by parts and bring the outside exponential inside the integral to get:

$$f_{k+1}(t) = \frac{1}{2}\left(f_k(t) - \int_0^t e^{\beta_k(t',t)}(\dot{f}_k(t') + 2i\dot{\theta}(t'))dt'\right), \tag{22}$$

where

$$\beta_k(t', t) = e^{2i\int_{t'}^{t} f_k(t')\dot{\theta}^*(t')dt'}. \tag{23}$$

2.3. Second step: analytical solutions

Although the first-order approximate solution (15) and the arbitrary kth-order approximate solution (22) are generic, the integral on the right-hand side of either equation cannot be evaluated in closed-form for any realistic pulse shape. As our main goal in this paper is to find such a solution, we set out to achieve it by making a series of further approximations to the functions $\beta_k(t', t)$ defined above in (23). Let us start by considering the first function $\alpha(t', t)$.

First, we expand $\alpha(t', t)$ in powers of $\delta = t - t'$ and approximate

$$\alpha(t', t) = 2\int_{t'}^{\delta+t'} \theta\dot{\theta}^* dt'' = 2\theta(t)\dot{\theta}^*(t)\cdot[t - t'], \tag{24}$$

to the leading term in δ. While this approximation may seem rather crude for large values of $\delta = t - t'$, as demonstrated in [15], the quantity $\alpha(t, t')$ is linear to a surprisingly high extent, and the approximation (24) proves to be quite valuable.

Secondly, using the fact that all quantities considered here are assumed to be limited to a finite time interval $t \in [-\tau, \tau]$, we can further simplify (24) by approximating the product $\theta(t)\dot{\theta}^*(t)$ as its average over the interval $[-\tau, \tau]$:

$$\theta(t)\dot{\theta}^*(t) = \frac{1}{2\tau}\int_{-\tau}^{\tau} \theta(t'')\dot{\theta}^*(t'')dt''. \tag{25}$$

Substituting this back into (24) yields

$$\alpha(t', t) = 2(t - t')\frac{1}{2\tau}\int_{-\tau}^{\tau} \theta(t'')\dot{\theta}^*(t'')dt'' = -i\alpha_0(t - t'), \tag{26}$$

where

$$\alpha_0 = \frac{i}{\tau}\int_{-\tau}^{\tau} \theta(t'')\dot{\theta}^*(t'')dt'' \tag{27}$$

is a constant, and τ is once again such that the ultrashort excitation is only non-zero on $[-\tau, \tau]$.

While the seemingly crude zeroth-order averaging (25) may look unwarranted, it is central to being able to simplify our solutions to an analytical form. On that path, let us now use the final form (26) to simplify the first-order approximate solution (15).

First, we note

$$\begin{aligned}
&\int_{-\infty}^{t} \dot{\theta}(t')e^{\alpha(t',t)}dt' \\
&= \int_{-\infty}^{t} \Omega(t')\cos(\omega t' + \phi)e^{i\omega_c t'}e^{-i\alpha_0(t-t')}dt' \\
&= e^{-i\alpha_0 t}\int_{-\infty}^{t} \Omega(t')\cos(\omega t' + \phi)e^{i(\omega_c+\alpha_0)t'}dt'.
\end{aligned} \tag{28}$$

At this point, it is convenient to introduce ω_c as an explicit parameter on which $\theta(t)$ depends, so that

$$\theta(\omega_c, t) = \int_{-\infty}^{t} \Omega(t')\cos(\omega t' + \phi)e^{i\omega_c t'}dt'. \tag{29}$$

With this notation, we can write (28) simply as

$$\int_{-\infty}^{t} \dot{\theta}(t')e^{\alpha(t',t)}dt' = e^{-i\alpha_0 t}\theta(\omega_c + \alpha_0, t). \tag{30}$$

Substituting (30) back into (15), we have a closed-form solution for pulses of arbitrary shapes:

$$\tilde{f}_1(t) = -\frac{i}{2}[\theta(\omega_c, t) + e^{-i\alpha_0 t}\theta(\omega_c + \alpha_0, t)], \tag{31}$$

where α_0 is a constant given by (27). From now on, tilde will be used to denote the simplified analytical solutions, while a notation without tilde denotes an approximate solution without further simplification.

2.4. Sequence of analytical solutions and their limit

We can repeat the steps we just carried out for the first-order approximate solution for an approximate solution of arbitrary order, and obtain a sequence of analytical solutions in this way. We can further take the limit $k \to \infty$ which will give us an even better approximation.

Using the same idea as with (31), we approximate β_k defined in (23) as a linear function of $(t - t')$:

$$\beta_k(t', t) = -i\alpha_k(t - t'), \quad \alpha_k = -\frac{2}{\tau}\int_{\tau}^{\tau} \dot{\theta}^*(t')\tilde{f}_k(t')dt'. \tag{32}$$

Notice that we have used $\tilde{f}_k$, which is yet to be found, in the expression for α_k above. Substituting this back into (22), and using the same trick as in (30), we have

$$\tilde{f}_{k+1}(t) = \frac{1}{2}\{\tilde{f}_k(t) - 2ie^{-i\alpha_k t}\theta(\omega_c + \alpha_k, t) - \int_{-\infty}^{t} e^{-i\alpha_k(t-t')}\dot{\tilde{f}}_k(t')dt'\}, \tag{33}$$

where we again note that all f_k are tilded.

From (33), it is possible to derive an accurate solution for $\tilde{f}_{k+1}$ and then to take the limit $k \to \infty$. Although this would be the cleanest way to derive the limiting solution, it is very tedious. Here, we offer a more elegant approach. Assume that all functions in (33) are continuous and that taking the limit commutes with integration. Take the limit $k \to \infty$ on both sides and denote $\tilde{f}_\infty = \lim_{k\to\infty}\tilde{f}_k$ and $\alpha_\infty = \lim_{k\to\infty}\alpha_k$. We then have

$$\frac{1}{2}\tilde{f}_\infty(t) = -ie^{-i\alpha_\infty t}\theta(\omega_c + \alpha_\infty, t) - \frac{1}{2}e^{-i\alpha_\infty t}\int_{-\infty}^{t} e^{i\alpha_\infty t'}\dot{\tilde{f}}_\infty(t')dt'. \tag{34}$$

Introducing $g(t) = e^{i\alpha_\infty t}\tilde{f}_\infty(t) + i\theta(\omega_c + \alpha_\infty, t)$ and integrating the exponential by parts lead to

$$\frac{1}{2}g(t) = -\frac{1}{2}\int_{-\infty}^{t} (\dot{g}(t') - i\alpha_\infty(g(t') - i\theta(\omega_c + \alpha_\infty, t')))dt', \tag{35}$$

and, after differentiation,

$$\dot{g}(t) = \frac{i\alpha_\infty}{2}g(t) + \frac{\alpha_\infty}{2}\theta(\omega_c + \alpha_\infty, t), \tag{36}$$

which yields

$$g(t) = e^{\frac{i\alpha_\infty}{2}t}\left(\int_{-\infty}^{t} e^{\frac{-i\alpha_\infty}{2}t'}\frac{\alpha_\infty}{2}\theta(\omega_c + \alpha_\infty, t')dt' + C\right). \tag{37}$$

Applying the initial condition $g(0) = 0$ and integrating by parts, $g(t)$ is simplified to

$$g(t) = i\theta(\omega_c + \alpha_\infty, t) - ie^{\frac{i\alpha_\infty}{2}t}\theta(\omega_c + \frac{\alpha_\infty}{2}, t). \tag{38}$$

Substituting $g(t)$ back to $\tilde{f}_\infty$, we have

$$\tilde{f}_\infty(t) = -ie^{-i\Delta t}\theta(\omega_c + \Delta, t), \tag{39}$$

where $\Delta = \alpha_\infty/2$ is a constant frequency that we will find next. At this point, let us immediately recognize that the final solution (39) we just obtained is essentially the same as the zeroth-order solution (9), but with an introduced frequency shift Δ (note that the phase factor does not have any bearing on the physically observable quantities) to the system resonance frequency ω_c. The most valuable part of our finding is the realization that the most accurate approximate analytical solution is of this form. Next, we must discuss how to obtain the specific value of this frequency shift Δ.

The downfall of the above shortcut derivation is that we do not immediately have an expression for Δ (or α_∞). However, a closer inspection reveals that Δ can be found in the following way. Consider Δ to be an arbitrary frequency shift and try to identify the value of Δ that would make our final approximate solution (39) most accurate. Substituting (39) back into (6), we find

$$\Delta = i\dot{\theta}^*(\omega_c, t)e^{-i\Delta t}\theta(\omega_c + \Delta, t), \tag{40}$$

which provides an equation the optimal Δ should satisfy. Notice that the right hand side is time-dependent, so averaging it and using $\dot{\theta}^*(\omega_c, t)e^{-i\nu t} = \dot{\theta}^*(\omega_c + \nu, t)$ lead us to the equation for the optimal Δ,

$$\Delta = \frac{i}{\tau}\int_0^{\tau} \dot{\theta}^*(\omega_c + \Delta, t')\theta(\omega_c + \Delta, t')dt'. \tag{41}$$

Admittedly, (41) does not provide an explicit expression for Δ. However, the role of Δ is that of a particularly suitable frequency shift that makes the approximate solution of the form (39) most accurate. For any arbitrary pulse shape, once the envelope profile $\Omega(t)$ is given, in principle one can always numerically solve (41) to extract Δ.

Overall, our general solution for the equations of motion (4) is given by (39) and (41), where the parameter θ depends on frequency ν and time t through the general relation

$$\theta(\nu, t) = \int_{-\infty}^{t} \Omega(t')\cos(\omega t' + \phi)e^{i\nu t'}dt'. \tag{42}$$

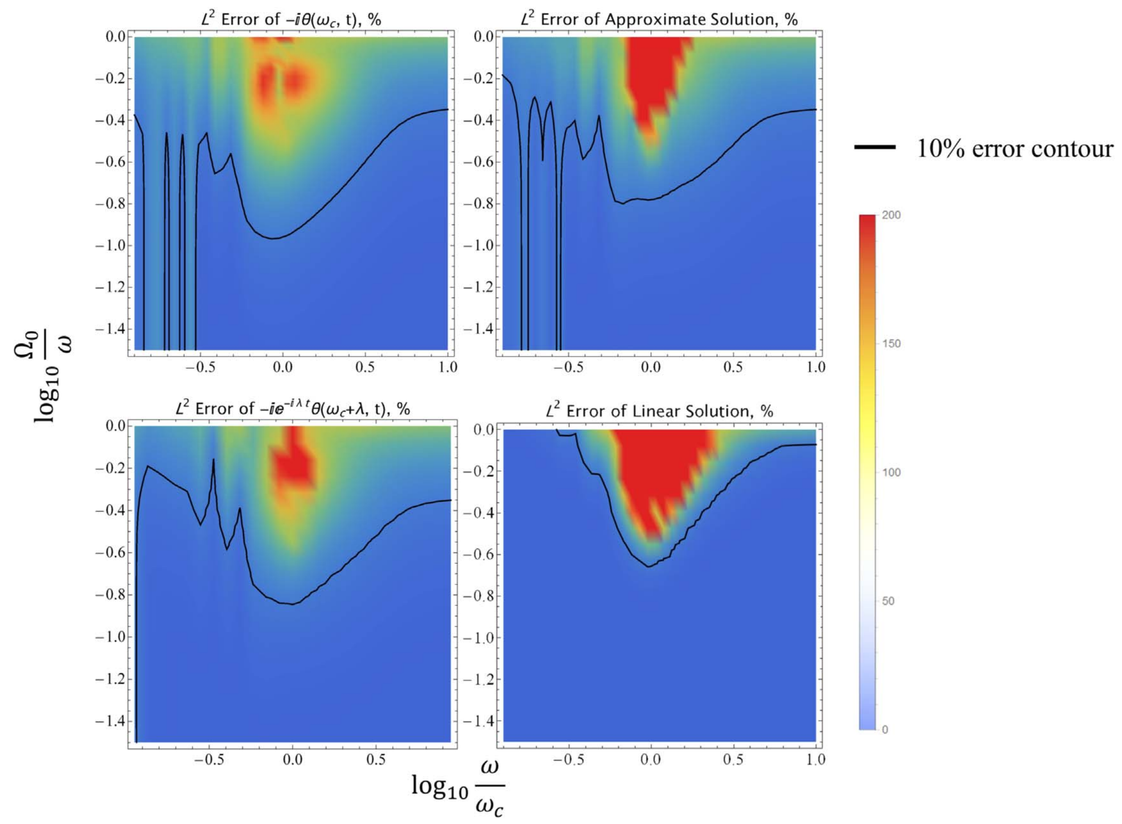

Figure 1. The percent L^2 error of the respective approximate solution as compared to the exact numerical solution of (6). Top left: the zero-order approximation (9). Top right: the first-order approximate solution (31). Bottom left: the infinite-limit approximate solution (39). Bottom right: the exact numerical solution of the linear equation (14). The solid black line is the contour on which the error is equal to 10%.

3. Gaussian pulse excitation

Let us study the conditions for which our solutions (31) and (39) are applicable, as well as their accuracy.

First of all, it should be pointed out that the main result of [15] can be obtained from our solution (31). For that, let us assume $\alpha_0 \ll \omega_c$ so that $\theta(\omega_c + \alpha_0, t) = \theta(\omega_c, t)$. In this case, denoting $\alpha_0 = \eta^2\omega_c$, equation (31) becomes

$$\tilde{f}_1(t) = -\frac{i}{2}(1 + e^{-i\eta^2\omega_c t})\theta(\omega_c, t), \tag{43}$$

which is exactly the equation (16) in [15]. The latter solution was shown to be accurate for a square pulse under the condition [15]

$$\left(\frac{\omega}{\omega_c}\right)^2 + \left(\frac{\Omega_0}{\omega}\right)^2 \ll 1. \tag{44}$$

Next, we examine how accurate the solution (31) and its generalization (39) are for more realistic pulses. As a quantity that expresses the accuracy of a solution, we choose the L^2-norm of the deviation of the solution from the exact solution. More specifically, the ratio of the latter quantity to the L^2-norm of the exact solution itself—we call this ratio the relative L^2 error. We numerically calculate this quantity for a Gaussian pulse with a Rabi frequency

$$\Omega(t) = \Omega_0 e^{-\frac{t^2}{2\sigma^2}}, \tag{45}$$

where we mainly explore the dependence of the relative error described above on the quantities Ω_0/ω and ω_c/ω, since they were shown in [15] to be the relevant parameters that determine applicability of a solution.

In figure 1, we plot the relative error described above for the approximate solutions (9), (31) and (39). As one can see, our most recent solution is the most accurate of the three, especially in the region where the previous solutions did not apply, namely, when $\omega_c \gg \omega$. At the same time, we can see that the current theoretical framework in general does not work well in the upper-middle regions of the plots, which exactly are the regions where the condition (44) is not satisfied. Therefore, these results also reconfirm the applicability condition of the current theory.

However, a slight modification of (44) is in order. Notice that our solution applies not only for the situations when $\omega/\omega_c \ll 1$, but also $\omega/\omega_c \gg 1$. As a result, the following condition is more appropriate

$$\min\left(\left(\frac{\omega_c}{\omega}\right)^2, \left(\frac{\omega}{\omega_c}\right)^2\right) + \min\left(\left(\frac{\Omega_0}{\omega}\right)^2, \left(\frac{\omega}{\Omega_0}\right)^2\right) \ll 1. \tag{46}$$

As demonstrated in figure 1, our solution accurately predicts the behavior of the TLS driven by a realistic Gaussian pulse. While the solution applies to arbitrary pulse shapes, a Gaussian pulse offers an exclusively convenient property under the current context, namely, the function $\theta(\omega_c, t)$ defined by (29) can be evaluated explicitly in closed-form for a Gaussian pulse,

$$\theta_{\text{Gaussian}}(\omega_c, t) = \frac{\sigma\Omega_0}{4}(\zeta(\omega_c - \omega, t)e^{-i\phi} + \zeta(\omega_c + \omega, t)e^{i\phi}), \tag{47}$$

$$\zeta(\nu, t) = e^{-\frac{1}{2}\sigma^2\nu^2}\left(\text{erf}\left(\frac{t}{\sqrt{2}\sigma} - \frac{i\sigma\nu}{\sqrt{2}}\right) + \text{erf}\left(\frac{t}{\sqrt{2}\sigma} + \frac{i\sigma\nu}{\sqrt{2}}\right)\right). \tag{48}$$

Such closed-form solutions have not been found for other common pulse shapes, such as the Lorentzian or the hyperbolic secant pulses. Using the closed form (47) and our main solution (39), we can also find the final population ratio between the upper and the lower state at the end of the pulse duration $t = \tau$, and we can study its dependence on the carrier-envelope phase ϕ.

$$\begin{aligned} I(\phi) = |f(\tau)|^2 = |\theta_{\text{Gaussian}}(\omega_c + \Delta, \tau)|^2 = \frac{\sigma^2\Omega_0^2}{16}\{|\zeta(\tilde{\omega}_c - \omega, \tau)|^2 + |\zeta(\tilde{\omega}_c + \omega, \tau)|^2 \\ + \zeta(\tilde{\omega}_c - \omega, \tau)\zeta(\tilde{\omega}_c + \omega, \tau)^* e^{-2i\phi} + \zeta(\tilde{\omega}_c - \omega, \tau)^*\zeta(\tilde{\omega}_c + \omega, \tau)e^{2i\phi}\}, \end{aligned} \tag{49}$$

where $\tilde{\omega}_c = \omega_c + \Delta$.

Note, however, that from (48), we have manifestly $\zeta(\nu, t)^* = \zeta(\nu, t)$. Therefore, (49) can be simplified to

$$I(\phi) = \frac{\sigma^2\Omega_0^2}{16}(\zeta(\tilde{\omega}_c - \omega, \tau)^2 + \zeta(\tilde{\omega}_c + \omega, \tau)^2 + 2\zeta(\tilde{\omega}_c - \omega, \tau)\zeta(\tilde{\omega}_c + \omega, \tau)\cos 2\phi). \tag{50}$$

Equation (50) provides a simple and explicit relation between the final population ratio and the CEP of the driving pulse, which is only possible with a closed-form solution such as (47). It offers some interesting insights into the CEP-dependence of the inversion. For example, the $\cos 2\phi$ dependence indicates that $I(\phi)$ has a period of π rather than 2π, and the maximum population ratio is achieved for the CEP value $\phi = 0$, regardless of the other parameters of the system. These results can be of important value in designing potential CEP-detection schemes based on atomic systems.

4. Discussion

Another insightful comparison of (39) is with the classical rotating-wave approximation(RWA) regime. Under the RWA, the equations of motion (2) become

$$\dot{C}(t) = -\frac{i}{2}\Omega(t)D(t), \tag{51}$$

$$\dot{D}(t) = -\frac{i}{2}\Omega(t)C(t). \tag{52}$$

Introducing the area of the pulse $A(t) = \int_0^t \Omega(t')dt'$, we can find a general solution of the TLS in the form of

$$C(t) = -i\sin\frac{A(t)}{2}, \tag{53}$$

$$D(t) = \cos\frac{A(t)}{2}. \tag{54}$$

Since $f(t) = C(t)/D(t)$, we have

$$f = -i\tan\frac{A}{2} \tag{55}$$

as the general solution under the RWA.

To appreciate the similarity of our solution, notice that we have the closed-form solution (39) and that for an arbitrary pulse, we can put $\theta(\omega_c, t)$ in the following form, just using the definition (3) and expanding $\cos(\omega t + \phi)$ in exponentials and simplifying:

$$\theta(\omega_c, t) = \frac{e^{i\phi}\tilde{A}(\omega_c + \omega, t) + e^{-i\phi}\tilde{A}(\omega_c - \omega, t)}{2}, \quad (56)$$

where

$$\tilde{A}(\nu, t) = \int_{-\infty}^{t} \Omega(t')e^{i\nu t'}dt', \quad (57)$$

can be considered as a generalization of the classical RWA pulse area $A(t)$.

Under RWA, $\tilde{A}(\omega_c + \omega, t) = 0$ and $\tilde{A}(\omega_c - \omega, t) = A(t)$, which means that up to a phase factor, our solution gives

$$\tilde{f}_\infty(t) = -i\frac{A}{2}. \quad (58)$$

This suggests that, near resonance (which is outside of its region of applicability), our solution corresponds to the linearization of the RWA solution. This apparently is the result of multiple linearizations we have taken in deriving $\tilde{f}_k$ to obtain the closed-form solution. With this observation in mind, we aim to overcome the oversimplification of our approximations and try to capture more of the nonlinear behaviors of the original equation (6). One approach is outlined as follows.

Consider again the general equation (6) and use $\dot{\theta}(t) = \Omega(t)\cos(\omega t + \phi)e^{i\omega_c t}$. Taking $g(t) = \Omega(t)\cos(\omega t + \phi)$ and multiplying through by $e^{-i\omega_c t}$, we arrive at

$$e^{-i\omega_c t}\dot{f}(t) = ig(t)e^{-2i\omega_c t}f^2(t) - ig(t). \quad (59)$$

Substituting $z(t) = e^{-i\omega_c t}f(t)$ into the above equation leads to

$$\dot{z}(t) = ig(t)z^2(t) - i\omega_c z(t) - ig(t). \quad (60)$$

Now expanding $z(t)$ as a perturbation series in ω_c, we obtain for the zeroth order

$$\dot{z}_0(t) = ig(t)(z_0^2(t) - 1), \quad (61)$$

which gives

$$z_0(t) = \cosh^{-1}\left(i\int_0^t g(t)dt\right). \quad (62)$$

Meanwhile, the nth-order can be written as

$$\dot{z}_n(t) = 2ig(t)z_n(t)z_0(t) + i\sum_{j=1}^{n-1} z_j(t)z_{n-j}(t) - iz_{n-1}(t). \quad (63)$$

The general solution to this linear equation, given the initial condition $z(0) = 0$, is:

$$z_n(t) = ie^{w(t)}\int_0^t e^{-w(t')}\left(\sum_{j=1}^{n-1} z_j(t')z_{n-j}(t') - z_{n-1}(t')\right)dt', \quad (64)$$

where $w(t) = 2i\int_0^t g(t')z_0(t')dt'$. Together with (62), (64) defines a sequence of functions that converge to the solution, similar to the sequence $f_k(t)$ discussed in section 2.

Note that, compared to the linearized sequence $f_k(t)$, the sequence of $z_n(t)$ is a much closer approximation to the accurate solution. Even the first-order approximation (62) captures the nonlinear behavior of the system. If a sequence of closed-form approximations to (64) could be found, similar to the sequence $\tilde{f}_k$ presented in section 2, the limit of such sequence z_∞ would be a promising approximation for capturing the nonlinear behaviors of the system. It seems plausible that a combination of such solution with the solution (39) would allow us to extend the conditions of applicability of (39) to the case when $\omega \sim \omega_c$, $\omega \sim \Omega_0$, and capture RWA better than a simple linear approximation. However, so far, we have been unable to find any approach to obtain a closed-form solution in this way. The difficulty is precisely the nonlinear features embedded into each (64).

5. Conclusion

In this work, we have developed an analytical general solution that describes a TLS driven by a far-off-resonance few-cycle pulse of *arbitrary* pulse shapes without using the RWA. We have identified the conditions under which our solution accurately predicts the behaviors of the system, and have demonstrated that the new solution offers improved accuracy compared to the previous solution under similar conditions. We have also applied the general solution to Gaussian pulses as a demonstration of its applicability, which results in an explicit closed-form solution. The solution is then used to examine the impact of CEP on the population ratio between the two states for Gaussian pulses. Finally, we suggest a possible alternative approach that can lead to a more accurate solution by capturing the nonlinear behaviors of the system and extending the applicability of the solution to the case of $\omega \sim \omega_c$, $\omega \sim \Omega_0$. It is our hope that this work can lay out a potential pathway toward an analytical theory for ultrafast QCC beyond the RWA.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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