Strong-Coupling Theory of Periodically Driven Two-Level Systems

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We present a simple but highly efficient iterating approach for strong-coupling periodically driven twolevel systems. The obtained explicit approximating analytical solution reproduces accurately the exact numerical solution in the strong-coupling regime for a wide frequency range including resonance, far-off resonance, harmonic, and subharmonic cases. Our theory is suitable for singe- and multiperiod periodic driving and for the periodic driving with a few-cycle pulse as well, and it gives a general formula for calculating the strong-field ac Stark effect in such diverse situations.

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The periodically driven two-level model is an important prototype in diverse phenomena in nearly every subfield of optics and physics [1-6] such as the nuclear magnetic resonance technique and its quantum information applications [6-8], acoustically induced transparency [9], Josephson-junction devices [10], the Cooper-pair box coupled to a nanomechanical resonator [11], laser-nucleus interaction [12], various effects of a few-cycle pulses [5,13], high harmonic generation [14], and so on.

Though much studied over more than 60 years [15], analytically solving this model even approximately for a wide parameter range has proven to be an extremely hard and complex task [2,4,11,15-17]. However, there has been some promising progress recently in this regard [8,11,16,17]. It is well known [1,2,11] that the most successful and widely used rotating-wave approximation is applicable only to the cases of near resonance and very weak coupling. The former constraint is successfully removed to a certain extent [11] but the restriction of very weak coupling still persists. It is found that there may be some complexities at large detuning and strong coupling [11,17], while the strong-coupling regime is an indispensable regime in various applications of strong-field physics [5,12-14]. In this context, we note that significant progress has been achieved in a recent strong-coupling theory [17] by finding a way to eliminate secular terms plagued by the usual series. What is more, some of the system's features can be described accurately for both high-frequency and resonance situations, though in an extremely strong coupling regime. There are some shortcomings in this important theory in that it cannot accurately describe levels' probability amplitudes (particularly their phases) even in an extremely strong coupling regime, and its method to eliminate secular terms is still somewhat too complicated. On the other hand, the above-mentioned theories deal with single-period purely periodically driven cases and no one seems to know how to handle analytically the single-period pulse-type field, say a Gaussian pulse-type field, not to mention multiperiod purely or pulse-type periodically driven cases, which already have had a multitude of applications [5,13,14,18].

In this Letter, we develop a simple but highly efficient iterating approach for strong-coupling periodically driven two-level systems uniformly valid for a universal form of the periodic field such as the single-period and multiperiod pulse-type and purely periodic type for a very wide frequency range including resonance, near and far-off resonance cases, and the harmonic and subharmonic cases as well. Besides, we also derive a general formula for calculating strong-field modified transition frequency due to the ac Stark effect in such diverse situations. Our approach is innately free of the secular-term problem and hence naturally avoids the very complicated process of eliminating secular terms.

The explicit analytical solution obtained only after the second iteration is shown to reproduce accurately the exact numerical solution in every aspect for the case of purely single-period periodic driving in the strong-coupling regime for a wide frequency range including resonance, near and far-off resonance cases as well as harmonic and sub-harmonic cases. It is our conjecture that the accurate agreement between the second-iterating solution and the exact solution is also true for other situations such as singe-period pulse-type, multiperiod purely periodic, and pulse-type driving.

We begin with the periodically driven two-level systems of the following general form:

$$\dot{\Psi}(t) = i \left[\frac{\omega_a}{2} \sigma_z + \Omega f(t) \sigma_1 \right] \Psi(t), \tag{1}$$

where $\dot{\Psi} = d\Psi/dt$, $\Psi(t) = (a(t), b(t))^T$ with *T* denoting a transpose, ω_a the transition frequency of the two-level system in the absence of the driving field, *a* (*b*) the probability amplitude of the ground (excited) level, the real Rabi frequency $\Omega > 0$ describes the coupling between the driving field and the two-level system, and its sign can be absorbed into the field's real shape function f(t), and σ_z , $\sigma_1 \equiv \sigma_x$ and $\sigma_2 \equiv \sigma_y$ are the usual Pauli matrices.

We shall develop an efficient iterating approach to find approximately the solution of Eq. (1) under the strongcoupling condition $\varepsilon = \omega_a/\Omega \ll 1$ for a quite arbitrary form of the periodic field. For instance, the real shape function can be a single-period type of the form f(t) = $g(t)\cos(\omega t + \phi)$ with the frequency ω and the initial phase ϕ while g(t) = 1 describes a purely periodic driving and $g(t) = e^{-(t/T_w)^2}$ for a Gaussian pulse-type periodic driving of the finite width T_w . It can also be a multiperiodic (or quasiperiodic) type $f(t) = \sum_k g_k(t) \cos(\omega_k t + \phi_k)$. Besides, ω or ω_k 's are not necessarily equal to or near the transition frequency.

Our iterating approach is described by the formulas

$$\Psi_n(t) = U_n(t)\Psi_{n+1}(t), \qquad \Psi_0(t) \equiv \Psi(t),$$
 (2a)

$$\dot{\Psi}_n(t) = i \left[\frac{\omega_a^{(n)}(t)}{2} \sigma_z + s_n \Omega_n(t) \sigma_{n+1} \right] \Psi_n, \tag{2b}$$

$$\Psi(t) = U_0(t)U_1(t)\cdots U_{n-1}(t)U_n(t)\Psi_{n+1}(t),$$
 (2c)

$$U_n(t) = e^{is_n\tau_n(t)\sigma_{n+1}} = \cos\tau_n + is_n\sigma_{n+1}\sin\tau_n, \qquad (2d)$$

$$\omega_a^{(0)}(t) \equiv \omega_a, \qquad \omega_a^{(n+1)}(t) = \omega_a^{(n)}(t) \cos[2\tau_n(t)], \qquad (2e)$$

$$\Omega_0(t) = \Omega f(t), \qquad 2\Omega_{n+1}(t) = \omega_a^{(n)}(t) \sin[2\tau_n(t)], \quad (2f)$$

$$\tau_{0} = \tau = \Omega \int_{0}^{t} f(t')dt', \qquad \tau_{n+1} = \int_{0}^{t} \Omega_{n+1}(t')dt',$$
(2g)

where $n = 0, 1, 2, ..., \sigma_n = \sigma_1$ ($\sigma_n = \sigma_2$) for odd (even) integers *n* or $\sigma_{n+2} = \sigma_n$, s_n denotes a sign sequence of period 4, i.e., $s_{n+4} = s_n$ with $s_0 = s_3 = 1$ and $s_1 = s_2 =$ -1 or $s_n = (-1)^{n(n+1)/2}$ derived from $s_{n+1} = (-1)^{n+1} s_n$.

Except for Eqs. (2b) and (2c), all the others in Eq. (2) are nothing but definitions. Equation (2c) is the trivial consequence of Eq. (2a), while Eq. (2b) can readily be shown by induction as follows. The case of n = 0 is simply Eq. (1) in different notation and is hence true. The only thing left is to show that Eq. (2b) for n = k + 1 holds true if it is so as n = k. Substituting $\dot{\Psi}_k = \dot{U}_k \Psi_{k+1} + U_k \dot{\Psi}_{k+1}$ into Eq. (2b) for n = k, and noting $\dot{U}_k = is_k \sigma_{k+1} \dot{\tau}_k U_k$ and $\dot{\tau}_k = \Omega_k$, we obtain $\dot{\Psi}_{k+1} = i(\omega_a^{(k)}/2)U_k^{-1}\sigma_z U_k \Psi_{k+1}$. It is then straightforward by using $U_k^{-1}\sigma_z U_k =$ $\sigma_z \cos(2\tau_k) + (-1)^{k+1}s_k\sigma_{k+2}\sin(2\tau_k)$, $s_{k+1} =$ $(-1)^{k+1}s_k$, Eqs. (2e) and (2f) to show that Eq. (2b) indeed holds true for n = k + 1.

One essential ingredient of our approach (2) is that one more iteration decreases by an order of ε the coefficient Ω_n of the nondiagonal term in the differential equation sequence (2b), i.e., $\Omega_{n+1} = \mathcal{O}(\varepsilon \Omega_n)$ uniformly valid in the whole interval $t \in (-\infty, \infty)$. This conclusion is readily seen from Eqs. (2e) and (2f) by using repeatedly the following argument: $2\tau_1 = \omega_a \int_0^t \sin[2\tau(t')] dt' \equiv \varepsilon I(t)$ leads to $\tau_1 = \mathcal{O}(\varepsilon)$ and $\Omega_2/\omega_a = \mathcal{O}[\sin(2\tau_1)] = \mathcal{O}(\tau_1)$ valid for any time because $I(t) \equiv \int_0^{\Omega_t} \sin[2\tau(t_0/\Omega)] dt_0$ is always finite in the whole time interval.

The approximating solution is obtained by performing only m iterations and solving approximately Eq. (2b) for

Denoting $V(t) = e^{i\beta_m(t)\sigma_z}$ with $2\beta_m =$ n = m. $\int_{0}^{t} \omega_{a}^{(m)}(t') dt'$, and substituting $\Psi_{m} = V \Phi_{m}$ into Eq. (2b) for n = m, we obtain $\dot{\Phi}_m = iW(t)\Phi_m$ with W(t) = $s_m \Omega_m(t) V^{-1}(t) \sigma_{m+1} V(t)$. It is solved by the perturbation theory up to the order $\mathcal{O}(\Omega_m)$ or $\Psi_m(t) \approx$ $V(t)e^{i\int_{0}^{t}W(t')dt'}\Phi_{m}(0) = e^{i\int_{0}^{t}V(t)W(t')V^{-1}(t)dt'}V(t)\Psi(0)$ bv $\Phi_m(0) = \Psi(0).$ using formula Then the $e^{i\gamma\sigma_z}\sigma_{m+1}e^{-i\gamma\sigma_z} = \sigma_{m+1}\cos(2\gamma) + (-1)^{m+1}\sigma_m\sin(2\gamma)$ leads to $\Psi_m(t) \approx A(t)B(t)V(t)\Psi(0)$ with $A = e^{i\varphi_m(t)\sigma_{m+1}}$, $B = e^{i\psi_m(t)\sigma_m}$ and φ_m and ψ_m given by Eq. (4) below. Here we have used $\dot{\tau}_m \equiv \Omega_m$, $(-1)^{m+1}s_m = s_{m+1}$ and $e^{i\varphi_m\sigma_{m+1}+i\psi_m\sigma_m} \approx AB \approx BA$ by neglecting $\mathcal{O}(\Omega_m^2)$ terms; i.e., A and B can be considered as mutually commutative at the required precision $\mathcal{O}(\Omega_m)$. Besides, these two matrices can also be considered as commutative with U_k (k = $1, 2, \ldots$) because the noncommutativity introduces at most the $\mathcal{O}(\varepsilon \Omega_m)$ difference. The *m*th iteration approximating the solution of Eq. (1) is thus

$$\Psi(t) \approx U_0 \cdots U_{m-1} e^{i\varphi_m \sigma_{m+1}} e^{i\psi_m \sigma_m} e^{i\beta_m \sigma_z} \Psi(0), \qquad (3)$$

where $m = 1, 2, ..., U_k$'s and τ_k 's are given by Eqs. (2d) and (2g), respectively, $s_n = (-1)^{n(n+1)/2}$ and

$$\beta_m(t) = \frac{1}{2} \int_0^t \omega_a^{(m)}(t') dt', \qquad \frac{\omega_a^{(m)}(t)}{\omega_a} = \prod_{k=0}^{m-1} \cos[2\tau_k(t)],$$
(4a)

$$\varphi_m(t) = s_m \int_0^t \frac{d\tau_m(t')}{dt'} \cos[2\beta_m(t) - 2\beta_m(t')]dt', \quad (4b)$$

$$\psi_m(t) = s_{m+1} \int_0^t \frac{d\tau_m(t')}{dt'} \sin[2\beta_m(t) - 2\beta_m(t')]dt'.$$
 (4c)

The second-iteration approximating analytical solution is explicitly written as follows:

$$a(t) = a_0 c(t) + i b_0 s^*(t),$$
 $b(t) = i a_0 s(t) + b_0 c^*(t),$
(5a)

$$c(t) = [\cos\tilde{\tau}(t)\cos\eta(t) + i\sin\tilde{\tau}(t)\sin\eta(t)]e^{i\beta(t)},$$
 (5b)

$$s(t) = [\sin\tilde{\tau}(t)\cos\eta(t) - i\cos\tilde{\tau}(t)\sin\eta(t)]e^{i\beta(t)}, \qquad (5c)$$

where $a_0 = a(0)$, $b_0 = b(0)$, $\tilde{\tau}(t) = \tau(t) + \varphi_2(t)$, $\eta(t) = \tau_1(t) - \psi_2(t)$ and $\beta = \beta_2$ or

$$\tau(t) = \int_0^t \Omega f(t') dt', \qquad \tau_1(t) = \frac{\omega_a}{2} \int_0^t \sin[2\tau(t')] dt',$$
(6a)

$$\beta(t) = \frac{\omega_a}{2} \int_0^t \cos[2\tau(t')] \cos[2\tau_1(t')] dt',$$
 (6b)

$$\tilde{\tau}(t) = \tau(t) - \frac{\omega_a}{2} \int_0^t q(t') \cos[2\beta_m(t) - 2\beta_m(t')] dt', \quad (6c)$$

$$\eta(t) = \tau_1(t) - \frac{\omega_a}{2} \int_0^t q(t') \sin[2\beta_m(t) - 2\beta_m(t')] dt',$$
(6d)

with $q(t) = \sin[2\tau_1(t)]\cos[2\tau(t)]$.

Let us take the most important and fundamental case of a periodic driving $f(t) = \cos(\omega t)$ or $\tau(t) = (\omega_a/\Omega) \sin(\omega t)$ as an example to illustrate the second-iteration approximating solution in Eq. (5). It is found via extensive numerical computations of various parameters' range that the approximating solution is quite accurate for a very wide frequency range from $\omega_a/\omega \ll 1$ to $\gg 1$ including the resonance, near and far-off resonance cases as well as harmonic cases in the strong-coupling regime $\varepsilon = \omega_a / \Omega \ll 1$. Figure 1 clearly illustrates this point by demonstrating that the approximating solution (5) reproduces accurately the exact numerical solution of Eq. (1) for quite different frequencies. It is pointed out that all the quantities Re[v(t)], $\operatorname{Im}[v(t)]$ (v = a, b), $\operatorname{Re}[a^*(t)b(t)]$, and $\operatorname{Im}[a^*(t)b(t)]$ (some of them not shown here) demonstrate this accurate agreement between Eq. (5) and the exact numerical solution. A general trend is that the parameter $\varepsilon = \omega_a / \Omega$ needs not to be very small ($\varepsilon \sim 2-4$ is enough) for this accurate agreement if the field's frequency $\omega \ge 0.8\omega_a$ which includes the resonance, near resonance and the high-frequency driving, while the low frequency driving $(\omega < 0.8\omega_a)$ needs relatively smaller ε to reach such an



FIG. 1. $\operatorname{Re}[a(t)]$ (lower curves), $\operatorname{Im}[a(t)]$ (middle curves, offset a constant), and $2\operatorname{Re}[a^*(t)b(t)]$ (upper curves, offset a constant) versus dimensionless time $\overline{t} = \omega t/2\pi$ under the initial condition a(0) = 1 and b(0) = 0 for four different field frequencies and the corresponding Rabi frequency as designated in the plots. The thick and thin curves show little difference and they correspond to the exact numerical solution and the approximating solution in Eq. (5), respectively.

accurate agreement. The lower the frequency ω , the smaller the parameter ε .

Figure 2 further illustrates this kind of accurate agreement between Eq. (5) and the exact numerical solution for other four (Gaussian type pulses and bichromatic type) forms of the driving field, all of them are beyond the rotating-wave approximation.

The results in Eq. (6) for the case of $f(t) = \cos(\omega t)$ can be further simplified at the expense of narrowing the suitable parameter range. For instance, using the formulas for the Bessel functions of the first kind $J_n(x)$ [19], and making some further approximations and calculations to Eq. (6), we then obtain

$$\tilde{\tau}(t) \approx \tau(t) = \frac{\Omega}{\omega} \sin(\omega t),$$
(7a)

$$\beta(t) \approx \beta_1(t) = \frac{\bar{\omega}_a t}{2} + \frac{\omega_a}{\omega} \sum_{m=1}^{\infty} \frac{J_{2m}(\xi) \sin(2m\omega t)}{2m}, \quad (7b)$$

$$\eta(t) \approx \sum_{m=0}^{\infty} \frac{\omega_a J_{2m+1}(\xi) \{\cos(\bar{\omega}_a t) - \cos[(2m+1)\omega t]\}}{(2m+1)\omega},$$
(7c)

where $\xi = 2\Omega/\omega$ and $\bar{\omega}_a = \omega_a J_0(\xi)$. It can readily be checked by numerical computations that Eqs. (5) and (7) represent a fairly accurate approximating solution to Eq. (1) in the range of $\Omega \ge 8\omega_a$ and $\omega \ge 0.8\omega_a$. This range is of course smaller than the suitable range (ω from $\omega \ll \omega_a$ to $\omega \gg \omega_a$) of Eqs. (5) and (6).

If only the populations but not the coherence properties are concerned just as the previous literature [17], much simpler results can derived from our approach. For example, $|a(t)|^2 = 1 - |b(t)|^2 = \cos^2[\frac{\Omega}{\omega}\sin(\omega t)]$ represents a fairly accurate result for a(0) = 1 and b(0) = 0 within the above mentioned parameter regime.

Let us now mention an important byproduct of our theory. It is well known that the transition frequency in the presence of a strong periodic field is significantly modified due to the strong-field ac Stark effect. The



FIG. 2. Same as Fig. 1 except for $\omega = \omega_a$ and (a) $f(t) = (1 + e^{-t^2/4})\cos(\omega t)$ with $\overline{t} = \omega t/2\pi$, (b) $f(t) = \cos(\omega t) + \cos(0.95\omega t)$, (c) $f(t) = e^{-t^2/16}\cos(\omega t)$, and (d) $f(t) = \cos(\omega t) + \cos(1.5\omega t)$.

strong-field modified transition frequency $\bar{\omega}_a$ for a singleperiod field in the resonance case has been obtained before [2], but no one seems to know how to calculate it in much wider conditions. We have accomplished this difficult task by obtaining the following quite universal formula to calculate $\bar{\omega}_a^{(m)}$, i.e., $\bar{\omega}_a$ by using the result of the *m* iterations:

$$\bar{\omega}_{a}^{(m)} = \langle \omega_{a}^{(m)} \rangle = 2 \left\langle \frac{d\beta_{m}(t)}{dt} \right\rangle = \left\langle \prod_{k=0}^{m-1} \cos[2\tau_{k}(t)] \right\rangle, \quad (8)$$

where τ_k 's, related to the strong periodic field, are given by Eq. (2g), $\langle \cdots \rangle$ denotes the average over the period of the field, and m denotes the result after m iterations. This formula is the direct consequence of Eq. (2b) by comparing with Eq. (1). This is obvious by noting that $\omega_a^{(m)}(t)$ is the coefficient of the diagonal term in Eq. (2b) and represents the simultaneous (or time-dependent) strong-field modified transition frequency if the magnitude of Eq. (2b)'s nondiagonal term (Ω_m term) is negligibly small. In the case of a single-period field $f(t) = \cos(\omega t)$, Eqs. (7b) and (8) immediately lead to the result $\bar{\omega}_a^{(2)} \approx \bar{\omega}_a^{(1)} = \omega_a J_0(\xi)$ identical to the previous result [2,17]. However, it is emphasized that our result $\bar{\omega}_a = \omega_a J_0(\xi)$ has much wider suitable regimes for the frequency ω and the Rabi frequency Ω than the previous result [2,17]. What is more important, our ac Stark formula (8) is quite universal because it is suitable for single- and multiperiod driving as well as pulse-type periodically driving for a wide frequency range.

In summary, we have put forward a theory free of the annoying secular-term problem by developing a highly efficient iterating unitary transformation approach for strong-field periodically driven two-level systems. The explicit analytical approximating solution (5) in the case of single-period periodically driving shows an accurate agreement with the exact numerical solution for wide frequency range including the resonance, near resonance, far-off resonance, harmonic, and subharmonic cases in the strong-coupling regime. Consequently, our strongcoupling theory applies not only to the single-photon process but also to the multiphoton (or harmonic generation) and subharmonic processes as well, including those of the strong-field pulses with a few cycles, quite important and hot topics of wide applications. In addition we have obtained a quite universal formula to calculate the strongfield modified transition frequency or the strong-field ac Stark effect in such diverse situations.

The explicit analytical solution (5) forms a good starting point for further seeking simpler solutions such as Eq. (7) under various more restricted regimes so that more physical insight can be sought. For instance, Eq. (7) clearly reflects multiphoton processes and their relative contributions in the single-period periodic deriving of the two-level system, and how the strong-field ac Stark effect affects phase coherence. Note that the term involving the strongfield modified transition frequency $\bar{\omega}_a$ in Eq. (7c) is indispensable to describing accurately the phase coherence. It is emphasized that our strong-coupling theory differs from previous ones in that it naturally avoids the annoying and very complicated processes for eliminating secular terms in the previous literature.

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