PHYSICAL REVIEW A **67**, 041404(R) (2003)

Validity of the two-level approximation in the interaction of few-cycle light pulses with atoms

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(Received 18 December 2002; published 28 April 2003)

The validity of the two-level approximation (TLA) in the interaction of atoms with few-cycle light pulses is studied by investigating a simple V-type three-level atom model. Even the transition frequency between the ground state and the third level is far away from the spectrum of the pulse; this additional transition can make the TLA inaccuracy. For a sufficiently large transition frequency or a weak coupling between the ground state and the third level, the TLA is a reasonable approximation and can be used safely. When decreasing the pulse width or increasing the pulse area, the TLA will give rise to non-negligible errors compared with the precise results.

DOI: 10.1103/PhysRevA.67.041404 PACS number(s): 42.50.Md, 42.65.Re

Recent advancements in ultrafast optical techniques have resulted in the generation of intense laser pulses as short as only a few optical cycles in the visible region of the spectrum [1]. At a peak power of 0.1 TW 5 fs laser pulses have been produced experimentally [2]. The interaction of such intense few-cycle laser pulses with matter have received a lot of interest [3–14].

Laser-atom interaction is one of the key problems in laser physics and quantum optics [15]. Generally, two-level approximation (TLA) model is used to describe the atom resonantly interacting with a single electromagnetic field. Under the rotating-wave approximation (RWA) and slowly varying-envelope approximation (SVEA), this simple model has led to a number of fascinating phenomena including the self-induced transparency [16], Rabi flopping [17], and photon echo [18] for pulses with duration of many cycles. When the pulse duration approached only a few optical cycles, the RWA and SVEA have been shown to be incorrect [6–14]. Based on the finite-difference time-domain (FDTD) simulation of the propagation of a few-cycle pulse in two-level atoms, Ziolkowski *et al.* have found the time-derivative effects in the evolution of the system [6], Hughes predicted the breakdown of the area theorem and the occurrence of carrier-wave Rabi flopping [7], and Tarasishin *et al.* have discussed how to amplify a few-cycle pulse in the inverted mediums [8]. If the pulse area is not very large, the theory of McCall and Hahn agrees reasonably well with the FDTD simulations [9,10]. Also some proposals of generating subfemtosecond pulse have been given in Refs. [11,12]. Some initially experimental results have been reported in Refs. [13,14].

All these mentioned works are studied in the framework of the TLA model. It is believed that the TLA is valid if the two atomic levels are resonant with the driving light pulse and all other levels are highly detuned [15]. However, there are no quantitative studies on the validity of the TLA in the interaction of few-cycle light pulses with atoms.

To discuss the validity of the TLA, we consider a simple V-type three-level atom model as illustrated in Fig. 1. The ground state $|g\rangle$ and the excited state $|e\rangle$ constitutes a two-level system (TLS) with transition frequency ω_e. This TLS resonantly interacts with a few-cycle light pulse $E(t)$. The ground state also couples to a higher level $|h\rangle$ with transition frequency ω_h. Inclusion of this additional $|g\rangle\rightarrow|h\rangle$ transition leads to non-TLA effects. We want to understand the influence of the non-TLA on the TLS-field interaction. Obviously, if ω_h is within the spectrum of the light pulse, the non-TLA effects cannot be neglected. However, if ω_h is detuned much far away from the pulse spectrum, can we still neglect the $|g\rangle\rightarrow|h\rangle$ transition? Using a numerical calculation, the conditions for the TLA are studied quantitatively.

Assume that the electric field of the few-cycle laser pulse takes the form

$$E(t)=AV(t)\cos(\omega_0 t), \tag{1}$$

where A is the peak amplitude, ω_0 is the carrier frequency, and $V(t)$ is the pulse envelope. An optical cycle is $T_0=2\pi/\omega_0$. The Hamiltonian of such an atom-field interacting system is

$$\hat{H}=\hbar\omega_e|e\rangle\langle e|+\hbar\omega_h|h\rangle\langle h|+\mu_e E(t)(|e\rangle\langle g|+|g\rangle\langle e|)$$
$$+\mu_h E(t)(|h\rangle\langle g|+|g\rangle\langle h|), \tag{2}$$

where μ_e and μ_h are electric dipole moments for $|g\rangle\rightarrow|e\rangle$ and $|g\rangle\rightarrow|h\rangle$ transitions. The dynamic evolution can be described with the Schrödinger equation for the state amplitudes $\{c_g,c_e,c_h\}$,

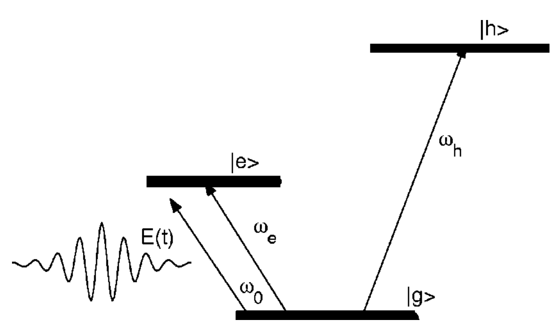

FIG. 1. The V-type three-level atom. The ground state $|g\rangle$ is coupled to the excited state $|e\rangle$ and a higher level $|h\rangle$ with transition frequencies ω_e and ω_h. A few-cycle pulse $E(t)$ with carrier frequency ω_0 near resonantly interacts with the $|g\rangle\rightarrow|e\rangle$ transition.

$$i\frac{d}{dt}\begin{pmatrix} c_g \\ c_e \\ c_h \end{pmatrix} = \begin{pmatrix} 0 & \Omega(t)\cos(\omega_0 t)\exp(-j\omega_e t) & \eta\Omega(t)\cos(\omega_0 t)\exp(-j\omega_h t) \\ \Omega(t)\cos(\omega_0 t)\exp(j\omega_e t) & 0 & 0 \\ \eta\Omega(t)\cos(\omega_0 t)\exp(j\omega_h t) & 0 & 0 \end{pmatrix}\begin{pmatrix} c_g \\ c_e \\ c_h \end{pmatrix}, \quad (3)$$

where $\Omega(t)=\mu_e V(t)/\hbar$ is the Rabi frequency and $\eta=\mu_h/\mu_e$ is the ratio of the transition dipole moments.

A two-level approximation is obtained by assuming $\eta=0$ in Eq. (3). Under the TLA, the evolution of $\{c_g, c_e\}$ obeys

$$i\frac{d}{dt}\begin{pmatrix} c_g \\ c_e \end{pmatrix} = \Omega(t)\cos(\omega_0 t)\begin{pmatrix} 0 & e^{-j\omega_e t} \\ e^{j\omega_e t} & 0 \end{pmatrix}\begin{pmatrix} c_g \\ c_e \end{pmatrix}. \quad (4)$$

Equation (4) can be divided into two parts: the RWA and the non-RWA. The RWA part has the form

$$i\frac{d}{dt}\begin{pmatrix} c_g \\ c_e \end{pmatrix} = \frac{1}{2}\Omega(t)\begin{pmatrix} 0 & e^{-j(\omega_e-\omega_0)t} \\ e^{j(\omega_e-\omega_0)t} & 0 \end{pmatrix}\begin{pmatrix} c_g \\ c_e \end{pmatrix}. \quad (5)$$

Previous works have shown that the RWA [Eq. (5)] cannot correctly describe the evolution of a two-level atom interacting with a few-cycle pulse [6–12]. However, the influence from other levels ($|h\rangle$ in our case) has not been examined. Equation (3) contains four oscillating frequencies. One is the RWA part ($\omega_e-\omega_0$), another is the non-RWA part ($\omega_e+\omega_0$), and the other two terms are from the $|g\rangle\to|h\rangle$ transition ($\omega_h\pm\omega_0$). If ω_h is not very large such that one of ($\omega_h\pm\omega_0$) is close to ($\omega_e+\omega_0$), there is no reason to only consider the contribution of the non-RWA part, but neglecting the influence from the $|g\rangle\to|h\rangle$ transition.

Assume that the envelope of the few-cycle laser pulse is hyperbolic secant, i.e.,

$$E(t)=A\,\mathrm{sech}(t/\tau)\cos(\omega_0 t), \quad (6)$$

where τ determines the pulse width and 1.76τ is the full width at half maximum duration of this kind of a few-cycle pulse. When $\tau\leqslant 2$ and $\omega_h>2\omega_0$, it is easy to show that no frequency content of the pulse will resonantly interact with the transition $|g\rangle\to|h\rangle$. But we will show that even in this case, the third level ($|h\rangle$) will have a significant influence on the pulse-atom interaction.

For this kind of driving pulse, we numerically solve Eqs. (3) and (4) to quantitatively study the validity of the TLA. For comparison, we also solve Eq. (5). The real part and the imaginary part of the polarization and the population inversion are obtained according to

$$u_k=2\,\mathrm{Re}[c_g c_e^*],$$

$$v_k=2\,\mathrm{Im}[c_g c_e^*],$$

$$w_k=|c_e|^2-|c_g|^2, \quad (7)$$

where $k=3,2,r$ corresponding to the results obtained from the three-level atom model [Eq. (3)], the TLA [Eq. (4)], and the RWA [Eq. (5)], respectively. Compared with the results obtained from the TLA model, the non-TLA corrections due to the $|g\rangle\to|h\rangle$ coupling are $\delta u_3=u_3-u_2$, $\delta v_3=v_3-v_2$, $\delta w_3=w_3-w_2$, and the non-RWA corrections are $\delta u_r=u_r-u_2$, $\delta v_r=v_r-v_2$, $\delta w_r=w_r-w_2$. In our calculations, the pulse takes the form of Eq. (6) with $\omega_0=\omega_e$.

A typical example is shown in Fig. 2. In this example, the driving field is a 4π hyperbolic secant pulse with $\tau=T_0$. The atom parameters are $\omega_h=2.2\omega_0$ and $\eta=1$. In this case, ω_h is much farther away from the spectrum of this few-cycle pulse. The upper plot gives the non-RWA correction δu_r (dotted line) and the non-TLA correction δu_3 (solid line). The oscillating amplitude of δu_3 is comparable to the amplitude of δu_r, and both are not negligible. The middle and bottom plots compare the effects of the non-TLA with the effects of the non-RWA on the imaginary part of the polarization and the population inversion. The non-TLA effects are more significant than the non-RWA effects. Under the TLA, a 4π pulse can lead to two Rabi flops, but the inclusion of the $|g\rangle\to|h\rangle$ transition makes the Rabi flops incomplete. In this example, the contributions of the non-TLA are larger than the contributions of the non-RWA, although ω_h is far away from the spectrum of $E(t)$. Thus, at least in this example, the TLA does not correctly describe the interaction of an intense few-cycle pulse with atoms.

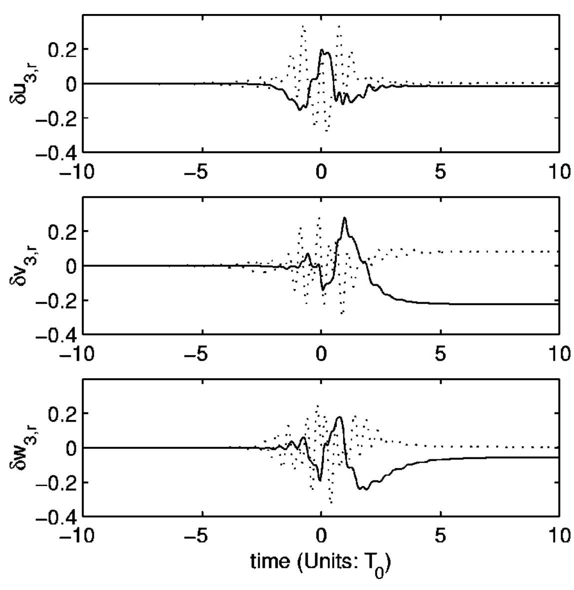

FIG. 2. Upper: evolution of $\delta u_3(t)$ (solid line) and $\delta u_r(t)$ (dotted line). Middle: evolution of $\delta v_3(t)$ (solid line) and $\delta v_r(t)$ (dotted line). Bottom: evolution of $\delta w_3(t)$ (solid line) and $\delta w_r(t)$ (dotted line). A 4π hyperbolic secant pulse with $\tau=T_0$ interacts with an atom with $\omega_h=2.2\omega_0$ and $\eta=1$.

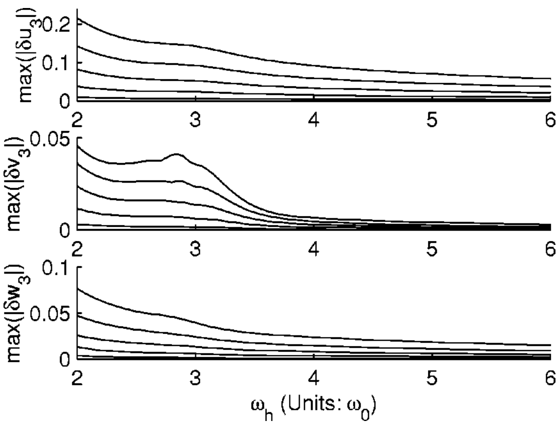

FIG. 3. Upper: max[$|\delta u_3(t)|$] as the function of ω_h. From bottom to top: $\eta=0.25,0.5,0.75,1,1.25$. Middle: max[$|\delta v_3(t)|$]. Bottom: max[$|\delta w_3(t)|$] as the function of ω_h. A 2π hyperbolic secant pulse with $\tau=T_0$ is used.

Different parameters of the atom and the pulse can modify the effects of the non-TLA. We use the maximum values of the non-TLA corrections, max[$|\delta u_3(t)|$], max[$|\delta v_3(t)|$], and max[$|\delta w_3(t)|$] as the measure to characterize the significance of the non-TLA. In Fig. 3, we plot max[$|\delta u_3(t)|$], max[$|\delta v_3(t)|$], and max[$|\delta w_3(t)|$] as functions of ω_h for five different ratios of the transition dipole moments, $\eta=0.25,0.5,0.75,1,1.25$. The driving field is a 2π hyperbolic secant pulse with $\tau=T_0$. We can see that increasing ω_h will decrease the non-TLA corrections. This means that the TLA will be a good approximation when level $|h\rangle$ is detuned, far enough, away from ω_0. Also, given ω_h, we find that a large η will lead to large non-TLA corrections. The non-TLA corrections are decreasing functions of η. The effects of the non-TLA are very important for large η and small ω_h, so the TLA is not correct. However, if $\eta<0.5$, one can use the TLA safely for $\omega_h>2\omega_0$.

The effects of the non-TLA will be significant for the increased few-cycle pulse intensity. To show this, the corresponding results with a 4π hyperbolic secant pulse and other parameters, the same as Fig. 3, are presented in Fig. 4. Here, max($|\delta u_3(t)|$) is about two times, max($|\delta v_3(t)|$) is about seven

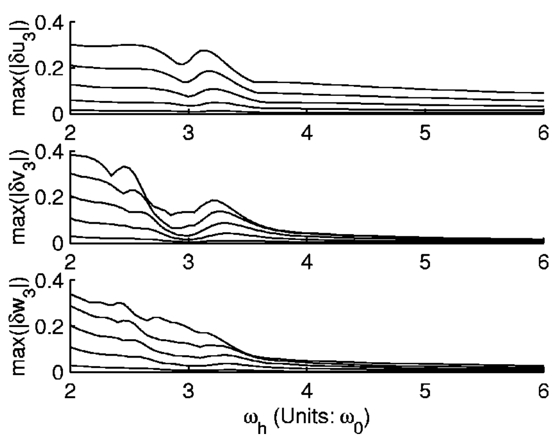

FIG. 4. The same as Fig. 3, but a 4π hyperbolic secant pulse with $\tau=T_0$ is used.

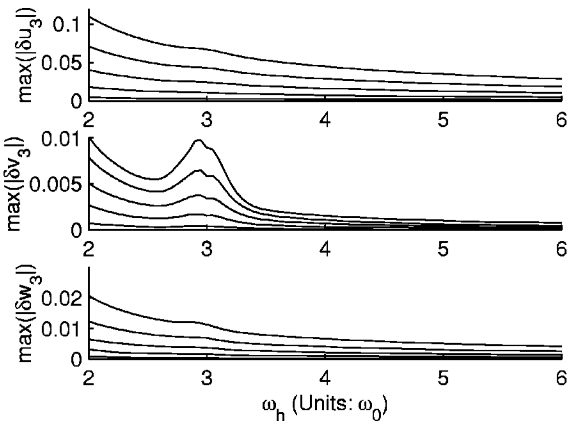

FIG. 5. The same as Fig. 3, but a 2π hyperbolic secant pulse with $\tau=2T_0$ is used.

times, and max($|\delta w_3(t)|$) is about four times large as compared with the corresponding curves in Fig. 3. Only when $\eta=0.25$, $\omega_h>2\omega_0$ can ensure the validity of the TLA. If $\eta=1$, the non-TLA correction max($|\delta u_3(t)|$) is not negligible even for $\omega_h=6\omega_0$.

The influence of the non-TLA will be small if the width of the pulse increases. As shown in Fig. 5, for a 2π hyperbolic secant pulse, the contributions of the non-TLA are much smaller than the results in Fig. 3. $|\delta v_3(t)|$ and $|\delta w_3(t)|$ are very small and close to 0. max($|\delta u_3(t)|$) is not negligible only for large η ($\eta\geqslant 0.75$). So the TLA correctly describes the resonant interaction of 3.5 cycles light pulse with the V-type three-level atoms, if the ratio of the transition dipole moments is not very large.

Generally, for a given level spacing between the ground state and third state ω_h, we have found that the small pulse width, or large pulse area, or the large ratio of the transition dipole moments will lead to more significant effects of the non-TLA. In fact, we have used the perturbation method to

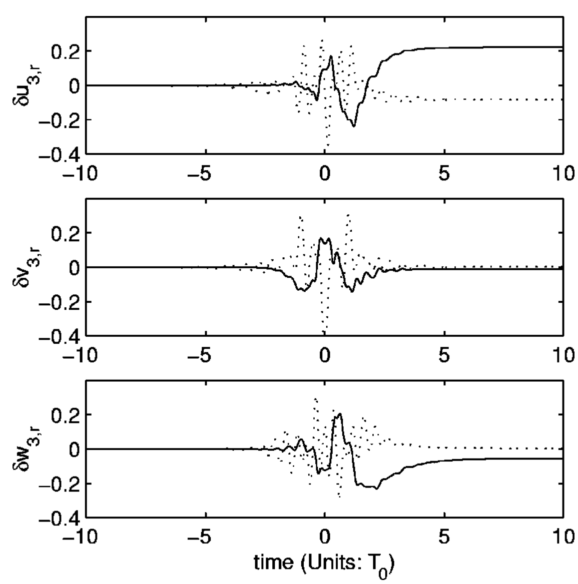

FIG. 6. The same as Fig. 2, but the absolute phase of the pulse is $\phi_0=\pi/2$.

study the effects of the non-TLA. Although the first-order corrections we obtained cannot give quantitatively consistent results with our numerical calculation, it really points out that the non-TLA corrections are proportional to $(\eta\Omega)/[\tau(\omega_h \pm \omega_0)]$, which agrees with our findings qualitatively. Some proposals of generating a subfemtosecond pulse [11,12] require propagation of the intense few-cycle light pulse in two-level atoms. The used pulse width is less than two cycles and the area is larger than 4π. If there are other levels coupling to these two levels, careful considerations are needed to prevent the influence from the other levels. We will study the propagation effects in future works.

In the end, we have performed some simulations to see the dependence of the results on the absolute phase ϕ_0. It is well known that the effects of the absolute phase may be very important for few-cycle pulses [1]. We can introduce the absolute phase by replacing $\omega_0 t$ in Eqs. (1)–(6) with $\omega_0 t + \phi_0$. An example is shown in Fig. 6. Compared with Fig. 2 in which $\phi_0 = 0$, it is clear that changing the absolute phase from 0 to $\pi/2$ really changes the results. Qualitatively, the main difference is that the shape of δu and δv are exchanged. More detailed studies will be done in the future.

In conclusion, the validity of the two-level approximation in the interaction of atoms with few-cycle light pulses is studied. Using a simple V-type three-level atom model, the influences of the transition between the ground state and the other highly excited state are investigated. Specifically, for driving pulses with width less than two cycles and area larger than 4π, the non-TLA corrections are significant and the TLA is not a good approximation. For strong coupling and large level spacing between the ground state and the third level, the TLA cannot describe the system evolution correctly. Increasing the pulse width or decreasing the pulse area can lower the influence of the non-TLA and make the TLA correct.

Support from the project by the National Key Basic Research Special Foundation (Grant No. G1999075200), the Chinese National Natural Science Foundation (Grant No. 90201027), and the Guangdong education office is acknowledged.

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