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Nonlinear responses of degenerate two-level systems to intense few-cycle pulses

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Population transfers in degenerate (or almost degenerate) two-level systems interacting with the few-cycle laser pulse are investigated. A simple and analytical formula of nonadiabatic transition probability is derived with completely degenerate condition, demonstrating the sensitive dependence of the transition probability on the phase of the few-cycle pulses. As one of the applications of this formula, a new way of controlling the nuclear wave packet dynamics at a potential curve crossing by 1 cycle laser pulse is proposed. © *2007 American Institute of Physics*.
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I. INTRODUCTION

Active control and manipulation of molecular processes and chemical dynamics by laser fields have been at the forefront of research for both chemistry and physics over the last couple of decades. The recent remarkable progress in laser technology has led to new possibilities to control molecule transitions. Control algorithms involve in manipulation of the phase and intensity of laser pulses including Rice-Tannor pump-dump scheme,[1] Brumer-Shapiro coherent control scheme,[2] optimal control scheme,[3,4] adiabatic rapid passage scheme,[5–7] and periodic sweeping of laser parameters.[8]

Recent advances of laser technology have made it possible to generate intense laser pulses shorter than 10 fs.[9] Unlike the conventional laser pulses, these pulses contain only few optical cycles. Therefore, it is expected that the phenomena induced by such the few-cycle laser pulses are sensitive to its phase. For examples, Chelkowski *et al.* have reported that angular distributions of the photoelectrons created by an intense few-cycle laser pulses clearly depend on the phase.[10] Recently, Kamta *et al.* have shown strong phase dependences of enhanced ionization in asymmetric diatomic molecules such as HeH^{2+} interacting with the few-cycle laser pulses.[11]

In this paper, we report the population dynamics in degenerate (or almost degenerate) two-level systems exposed to an intense few-cycle laser pulses. For a two-level system interacting with the continuous-wave (cw) laser field $F\cos(\omega t+\phi)$, the population dynamics is well known as Rabi oscillation.[12] Under the rotating wave approximation, the transition probability P_{12} is described by a simple formula

$$P_{12}=\frac{\Omega^2}{\Omega^2+\Delta^2}\sin^2\left(\frac{\sqrt{\Omega^2+\Delta^2}}{2}t\right), \tag{1}$$

where $\Omega=\mu_{12}F/\hbar$ denotes the Rabi frequency, μ_{12} is the transition dipole moment between the two levels, $\Delta=\omega-\omega_{12}$ is the detuning, and ω_{12} is the resonance frequency between the two levels. In this case, P_{12} does not depend on the phase ϕ at all. In the case of a conventional long pulse $F(t)\cos(\omega t+\phi)$ where $F(t)$ is the envelope function, the transition probability is derived with use of the semiclassical theory of the Rosen-Zener (RZ)-type nonadiabatic transition[13–15] and the Floquet (or dressed) state representation,[16,17] and it is given as[8]

$$P_{12}=4p(1-p)\sin^2\psi, \tag{2}$$

where p is the nonadiabatic transition probability at one complex crossing point, and ψ is the phase difference accumulated between the two adiabatic Floquet states. In this case, P_{12} does not depend on ϕ either. Since Eqs. (1) and (2) are valid for the cw laser fields and slowly varying long pulses, they cannot be applied to the few-cycle laser pulses. As there is no simple and analytical formula of transition probability for the few-cycle laser pulses, especially for 1 cycle laser pulse, we derive a simple formula of transition probability for two-level systems interacting with an intense 1 cycle laser pulse in this paper for the sake of the simplicity. Fortunately, when two-level system is completely degenerate we can derive an exactly analytical solution. As we know for nondegenerate two-level systems, we must utilize the semiclassical matrix propagation method[14,15] to obtain an analytical formula of nonadiabatic transition probabilities. These

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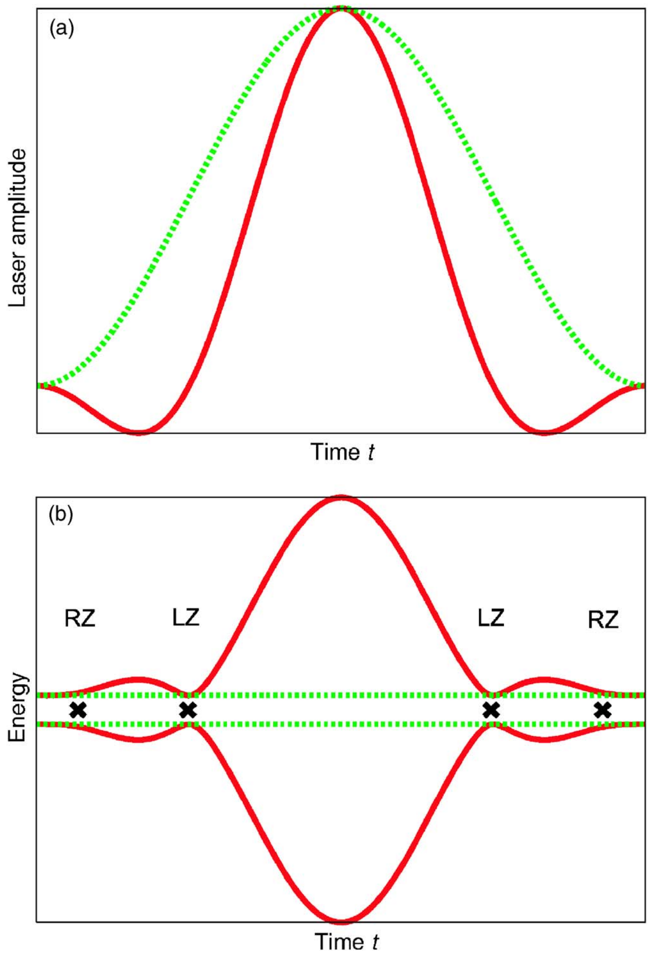

FIG. 1. (Color online) (a) Time variation of the laser amplitude for 1 cycle pulse. The phase ϕ is fixed at 0. (b) Time variation of the energy levels for 1 cycle pulse shown in (a). Solid line, adiabatic energy levels; dotted line, energy levels $\pm\hbar\omega_{12}/2$ of a field-free two-level system. $\times$ denotes the times when the LZ or RZ type nonadiabatic transitions are induced.

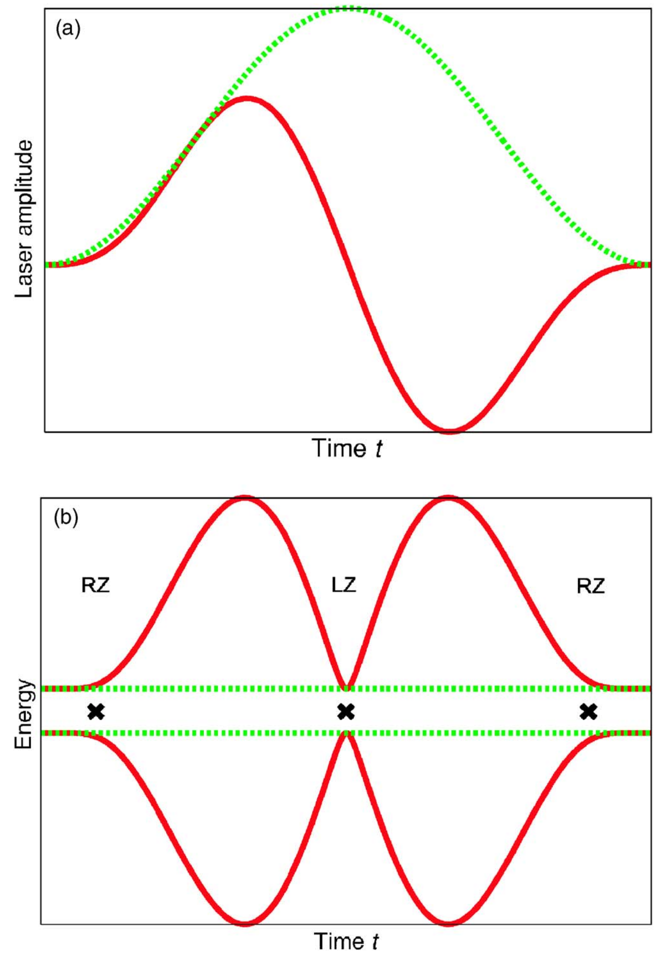

FIG. 2. (Color online) The same as Fig. 1 but the phase ϕ is fixed at $\pi/2$.

schemes have successfully been applied to the conventional long pulses along with the Floquet state representation.

This paper is organized as follows. In Sec. II, we derive a simple and analytical formula of nonadiabatic transition probability for degenerate (or almost degenerate) two-level systems interacting with 1 cycle laser pulse. This formula consists of $\sin^2$ function, including the field amplitude, frequency, and phase of the 1 cycle pulse as parameters. This analytical formula is compared with the numerical solution of the time-dependent Schrödinger equation in Sec. III. Furthermore, as one of the applications of this formula, we propose a new laser control of nonadiabatic dissociation dynamics of a diatomic molecule at a potential curve crossing, and agreement and limit of the formula are extensively analyzed as well. Concluding remarks are provided in Sec. IV.

II. THEORETICAL FRAMEWORK

In this section, we consider the interaction of the degenerate (or almost degenerate) two-level system with linearly polarized few-cycle laser pulses $\mathbf{E}(t)$. Throughout this paper, it is assumed that $\mathbf{E}(t)$ is defined as

$$\mathbf{E}(t) = \begin{cases} \mathbf{E}_0 E(t) & \text{for } 0 \leqslant t \leqslant 2t_c \\ 0 & \text{for } t < 0,\ t > 2t_c, \end{cases} \tag{3}$$

$$E(t) = \varepsilon(t)\cos[\omega(t - t_c) + \phi], \tag{4}$$

$$\varepsilon(t) = \varepsilon_0 \sin^2\left(\frac{\pi}{2t_c}t\right), \tag{5}$$

and

$$t_c = \frac{n\pi}{\omega}, \tag{6}$$

where $\mathbf{E}_0$ is the polarization vector (unit vector, $|\mathbf{E}_0| = 1$), $\varepsilon(t)$ is the pulse envelope, ω is the laser frequency, ϕ is the absolute phase of the laser pulse ($0 \leqslant \phi < 2\pi$), ε_0 is the peak amplitude, and n is the number of the optical cycle. Equation (3) represents the n-cycle laser pulses. It is also assumed that there is a dipole-allowed transition between the two levels $|1\rangle$ and $|2\rangle$. Therefore, the Hamiltonian $\hat{H}$ for this system under the dipole approximation is given by

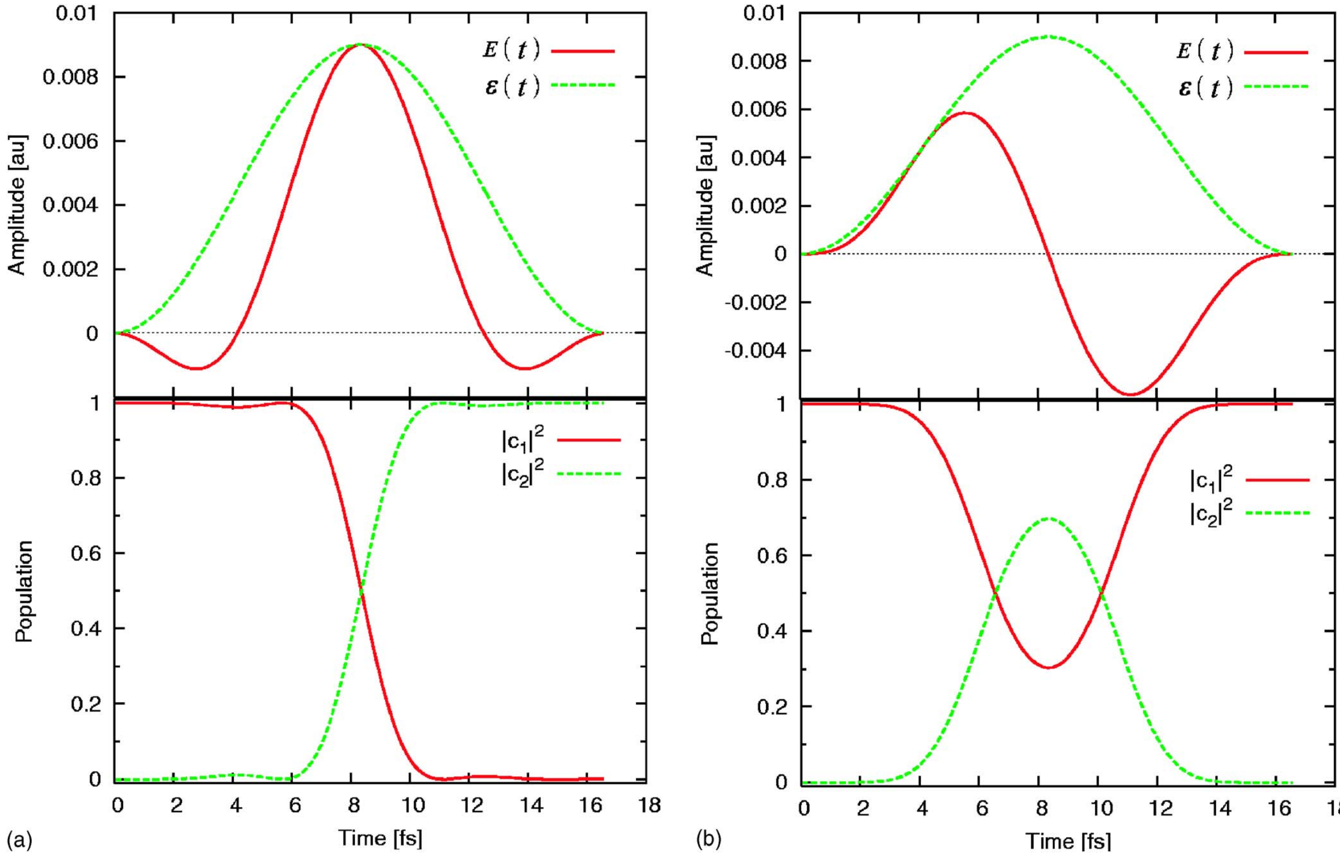

FIG. 3. (Color online) Calculated population dynamics (lower panel) of a degenerate two-level system ($\omega_{12}=0\ \mathrm{cm}^{-1}$, $\mu=1.0$ a.u.) interacting with 1 cycle pulse (upper panel: $\varepsilon_0=0.009$ a.u., $\omega=2000\ \mathrm{cm}^{-1}$) against time t. (a) $\phi=0$ and (b) $\phi=\pi/2$.

$$\hat{H}=\begin{bmatrix}-\hbar\omega_{12}/2 & -\mu E(t)\\ -\mu E(t) & \hbar\omega_{12}/2\end{bmatrix}, \tag{7}$$

where $\hbar\omega_{12}$ denotes the energy spacing between $|1\rangle$ and $|2\rangle$, and $\mu=\boldsymbol{\mu}_{12}\cdot\mathbf{E}_0$, where $\boldsymbol{\mu}_{12}$ is the transition dipole moment between $|1\rangle$ and $|2\rangle$. Time-dependent Schrödinger equation for the two-level system in Eq. (7) is given by[18]

$$i\hbar\frac{d}{dt}\begin{bmatrix}c_1(t)\\ c_2(t)\end{bmatrix}=\hat{H}\begin{bmatrix}c_1(t)\\ c_2(t)\end{bmatrix}. \tag{8}$$

The scattering matrix is defined by

$$\begin{bmatrix}c_1(+\infty)\\ c_2(+\infty)\end{bmatrix}=\begin{bmatrix}S_{11} & S_{12}\\ S_{21} & S_{22}\end{bmatrix}\begin{bmatrix}c_1(-\infty)\\ c_2(-\infty)\end{bmatrix}. \tag{9}$$

The off-diagonal matrix element represents the overall nonadiabatic transition probability $P_{12}=|S_{12}|^2$. In general, an exactly analytical solution cannot be obtained with Hamiltonian in Eq. (7). However, if we take completely degenerate case in which $\omega_{12}=0$, we can have

$$\begin{aligned} i\hbar\frac{d}{dt}[c_1(t)+c_2(t)]&=\Delta(t)[c_1(t)+c_2(t)],\\ i\hbar\frac{d}{dt}[c_1(t)-c_2(t)]&=-\Delta(t)[c_1(t)-c_2(t)], \end{aligned} \tag{10}$$

where $\Delta(t)=-\mu E(t)$. Equation (10) can be solved exactly as follows:

$$\begin{aligned} c_1(t)&=A_0\exp\left[-\frac{i}{\hbar}\int_0^t\Delta(t)dt\right]+B_0\exp\left[\frac{i}{\hbar}\int_0^t\Delta(t)dt\right],\\ c_2(t)&=A_0\exp\left[-\frac{i}{\hbar}\int_0^t\Delta(t)dt\right]-B_0\exp\left[\frac{i}{\hbar}\int_0^t\Delta(t)dt\right], \end{aligned} \tag{11}$$

from which the scattering matrix in Eq. (9) can be obtained as

$$\begin{aligned} S_{11}&=S_{22}=\cos\Psi,\\ S_{12}&=S_{21}=-i\sin\Psi, \end{aligned} \tag{12}$$

where

$$\Psi=\frac{1}{\hbar}\int_0^{2t_c}\Delta(t)dt. \tag{13}$$

The simple calculation leads to

$$\Psi=-\frac{\mu\varepsilon_0}{\hbar\omega}\left[1-\frac{1}{2((\pi/\omega t_c)+1)}+\frac{1}{2((\pi/\omega t_c)-1)}\right]\sin\omega t_c\cos\phi. \tag{14}$$

If cycles of laser pluses $n\geqslant 2$ in Eq. (6), $\Psi(n\geqslant 2)=0$. At 1 cycle laser pulse $n\rightarrow 1$, Eq. (14) turns to be

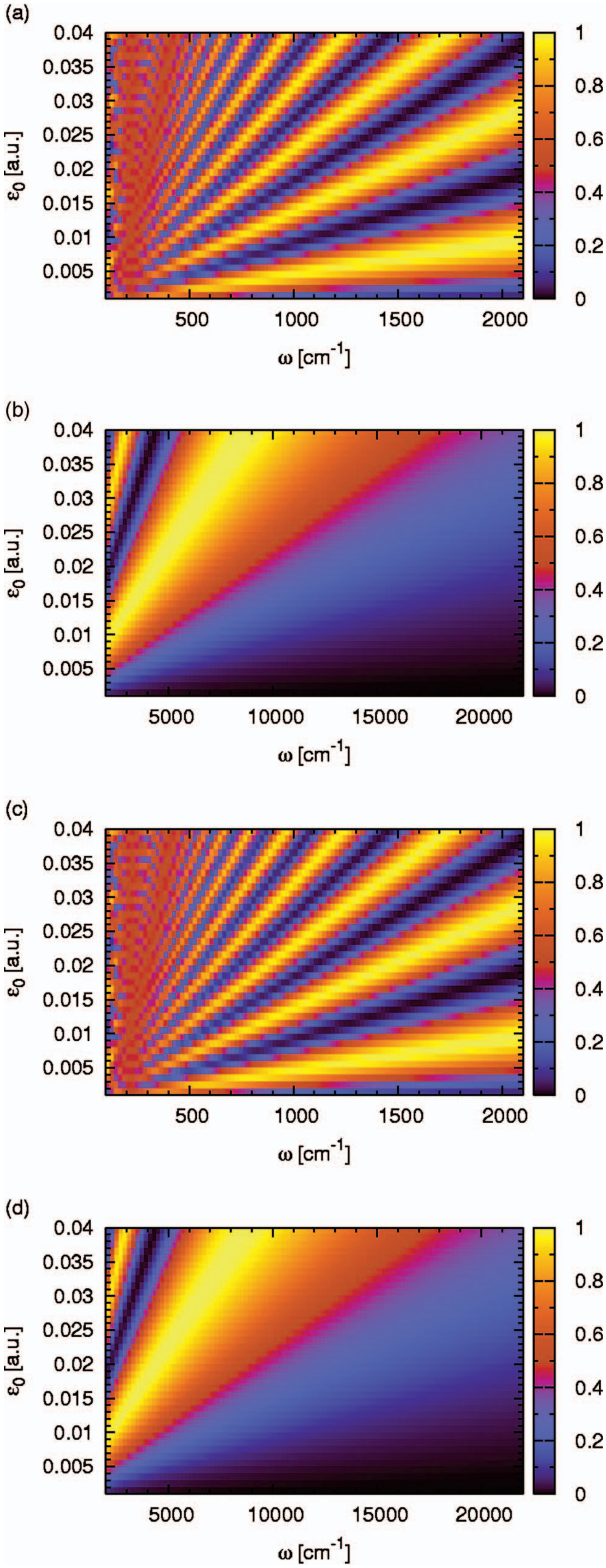

FIG. 4. (Color) Transition probabilities P_{12} for a degenerate two-level system ($\omega_{12}=0\ \mathrm{cm}^{-1}$, $\mu=1.0$ a.u.) interacting with 1 cycle pulse as a function of ω and ε_0. [(a) and (b)] Numerical solutions and [(c) and (d)] analytical formula, Eq. (16). ϕ is set to 0.

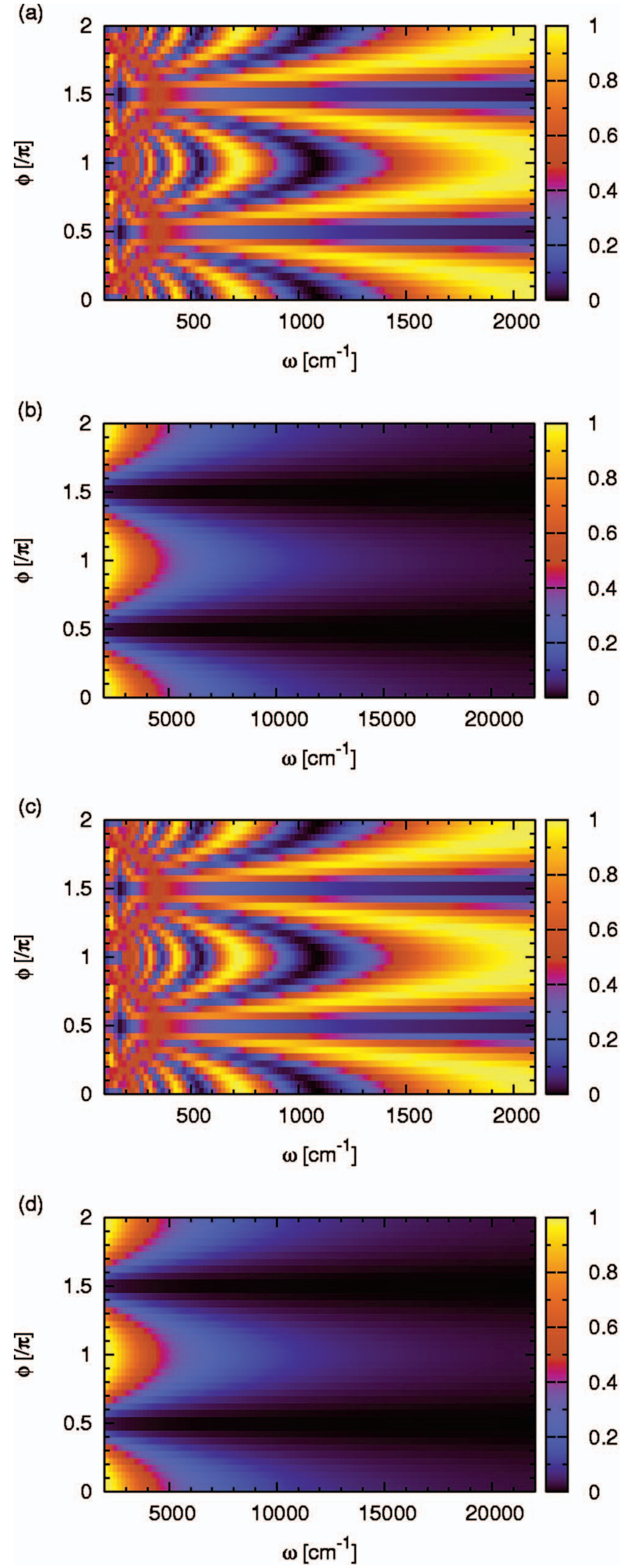

FIG. 5. (Color) Transition probabilities P_{12} for a degenerate two-level system ($\omega_{12}=0\ \mathrm{cm}^{-1}$, $\mu=1.0$ a.u.) interacting with 1 cycle pulse as a function of ω and ϕ. [(a) and (b)] Numerical calculations and [(c) and (d)] analytical formula, Eq. (16). ε_0 is fixed at 0.01 a.u.

$$\Psi = \frac{\pi|\mu|\varepsilon_0}{2\hbar\omega}\cos\phi. \tag{15}$$

The overall nonadiabatic transition probability is

$$P_{12} = \sin^2\left(\frac{\pi|\mu|\varepsilon_0}{2\hbar\omega}\cos\phi\right). \tag{16}$$

The condition for the population inversion reads

$$\frac{|\mu|\varepsilon_0}{\hbar\omega}\cos\phi = 1 + 2n(n=0,1,2,\ldots). \tag{17}$$

Let us define the pulse area of 1 cycle pulse in the same way as the conventional long pulses:

$$\text{pulse area} = \frac{|\mu|}{\hbar}\int_0^{2t_c} \varepsilon(t)dt = \frac{\pi|\mu|\varepsilon_0}{\hbar\omega}. \tag{18}$$

From Eqs. (17) and (18), the minimum pulse area for the population inversion is $\pi/\cos\phi$, which is equal to or larger than π. We note that it is impossible to make the pulse area smaller than π. This is the same as the conventional long pulse case.

It should be emphasized that for degenerate two-level system, only 1 cycle laser pulse gives nonzero transition for the two-level systems. For the nondegenerate system $\omega_{12} \neq 0$, Eq. (8) cannot be solved exactly in the analytical form, and the semiclassical propagation method has to be employed to derive analytical nonadiabatic transition probability. We briefly discuss semiclassical procedure for two representative cases, as shown in Figs. 1 and 2. We show the pulse envelope $\varepsilon(t)$ and the electric field $E(t)$ with $\phi=0$ in Eq. (4) as a function of time for the 1 cycle pulse in Fig. 1(a) and depict the adiabatic energy levels in Fig. 1(b) in which there are four complex crossing points [see $\times$ in Fig. 1(b)] where nonadiabatic transitions are taken place; the two outermost transitions are well described by RZ-type nonadiabatic transitions,[13–15] and the two inner transitions are Landau-Zener (LZ)-type nonadiabatic transitions.[14,15,19,20] Next we consider the case that electric field $E(t)$ in Eq. (4) with $\phi=\pi/2$ shown in Fig. 2(a) and depict the adiabatic energy levels in Fig. 2(b) in which there are three complex crossing points [see $\times$ in Fig. 2(b)] where nonadiabatic transitions are taken place; the outermost two are the RZ-type transitions and the central transition is the LZ-type transition. Based on those complex crossing points, we can derive overall nonadiabatic transition probability semiclassically, and its results are the same as those obtained in Eq. (16) for 1 cycle laser pulse with degenerate two-level system. We will discuss more details for nondegenerate two-level systems in the future publication. Furthermore, the above formulation can also be extended to the general m-level systems $(m>2)$.[21] In the next section, we demonstrate the validity of Eq. (16) by comparing it with the numerical solution of the time-dependent Schrödinger equation (8). Equation (16) is also applied to a new laser control of the nuclear wave packet dynamics at a potential curve crossing.

III. RESULTS AND DISCUSSIONS

A. Numerical demonstrations of Equation (16)

In this section, for the degenerate (or almost degenerate) two-level systems interacting with 1 cycle laser pulse, we compare the analytical formula in Eq. (16) with the numerical solutions of the time-dependent Schrödinger equation (8) in which $c_k(t)$ denotes the probability amplitude of the level $|k\rangle$, and Eq. (8) is solved in terms of the conventional fourth-order Runge-Kutta method.[22] Throughout this section, we assume $\mu=1ea_0$ with initial condition $c_1(0)=1$ and $c_2(0)=0$. In Fig. 3, the time evolution of the populations in a degenerate two-level system ($\omega_{12}=0$) under 1 cycle laser pulse is plotted for (a) $\phi=0$ and (b) $\phi=\pi/2$. The peak amplitude ε_0 and frequency ω of the 1 cycle pulse are fixed at 0.009 a.u. and 2000 cm^{-1}. Since $|\mu|\varepsilon_0/(\hbar\omega)$ is equal to 0.99, the condition of $P_{12}=1$, i.e., Eq. (17) is almost satisfied in the case of $\phi=0$. As the analytical solution predicts, the calculation result for $\phi=0$ clearly indicates the population inversion. On the other hand, in the case of $\phi=\pi/2$, although some population is transferred into $|2\rangle$ in the middle of the pulse, no population remains in $|2\rangle$ at the end of the pulse, which is also the same as the analytical prediction.

Two-dimensional plots of the transition probability are depicted against ω and ε_0 in Fig. 4. The phase ϕ is fixed at 0. As is easily found from Eq. (17), the population inversion takes place when the linear relation $\varepsilon_0=\hbar\omega(1+2n)/|\mu|$ is satisfied. This is clearly seen in Fig. 4. The numerical solutions completely agree with the analytical solution in a very wide range of laser parameters. As the laser frequency becomes larger, larger peak amplitudes are necessary for the population inversion. In Fig. 5, two-dimensional plots of the transition probability are shown as a function of ω and ϕ. The peak amplitude ε_0 is set to 0.01 a.u.. In this case, we also see that the numerical solutions are in perfect agreement with the analytical solution. It is noted that the transition probability is periodic as a function of ϕ at the intervals of $\pi/2$ and the complete inversion takes place unless $\phi=\pi/2$ $(3\pi/2)$. So far, we consider a degenerate two-level system. However, Eq. (16) is also valid for an almost degenerate two-level system. In Fig. 6, we show the population dynamics of a two-level system with $\omega_{12}=500$ cm^{-1} as a function of t for (a) $\phi=0$ and (b) $\phi=\pi/2$. The other laser parameters are the same as those used in Fig. 3. We note that the population in $|2\rangle$ at the end of the pulse slightly deviates from Eq. (16) because of the nondegeneracy of the system. In Fig. 7(a), two-dimensional plots of the calculated transition probability are depicted as a function of ω and ε_0 (ω and ϕ). The laser parameters are the same as those used in Fig. 4(a). The lower the laser frequency, the worse the agreement between the numerical and anlytical results becomes [compare Figs. 7(a), 7(b), 8(a), and 8(b) with Figs. 4(c), 4(d), 5(c), and 5(d), respectively]. At such low laser frequencies, we cannot regard the system as a degenerate two-level and thus it is necessary to derive semiclassical solution to model this situation. It can be seen, however, that as the laser frequency becomes higher, the numerical solutions almost follow the analytical one in Eq. (16). This is natural since the system can be regarded as degenerate two levels at the higher laser frequencies. In the next section, Eq. (16) is applied to the wave packet dynamics at a potential curve crossing under a 1 cycle pulse. It is demonstrated that controlling molecular nonadiabatic dissociation by a 1 cycle pulse can be achieved.

B. Application to the control of the wave packet dynamics at a potential curve crossing by 1 cycle laser pulse

Here, 1 cycle laser pulse is shined to the propagating wave packet to control its dynamics at a potential curve

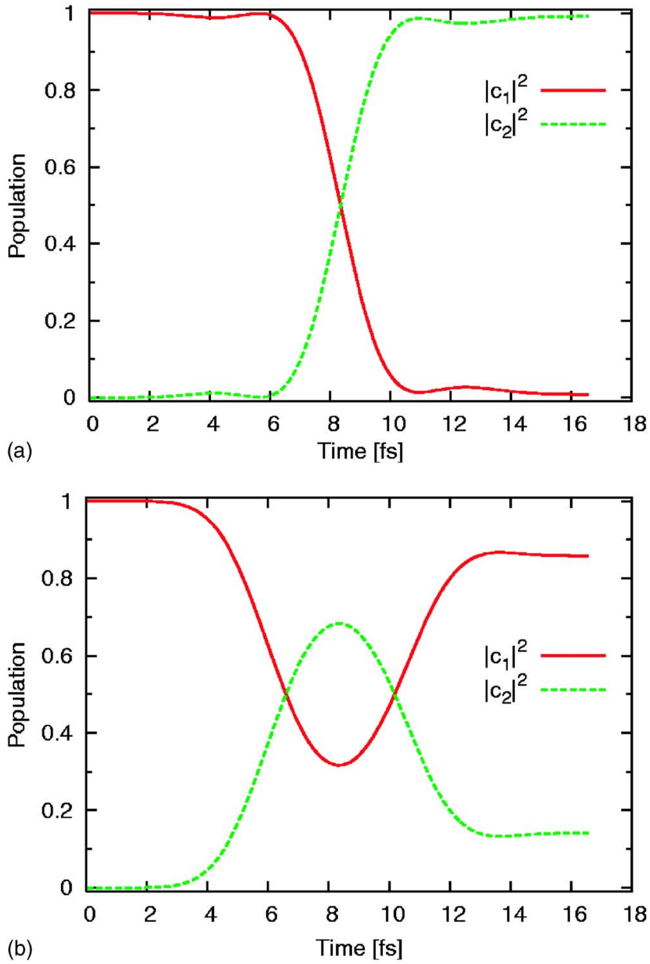

FIG. 6. (Color online) Calculated population dynamics of an almost degenerate two-level system (ω_{12}=500 cm^{-1}, μ=1.0 a.u.) interacting with 1 cycle pulse (ε_0=0.009 a.u., ω=2000 cm^{-1}) against time t. (a) ϕ=0 and (b) $\phi = \pi/2$.

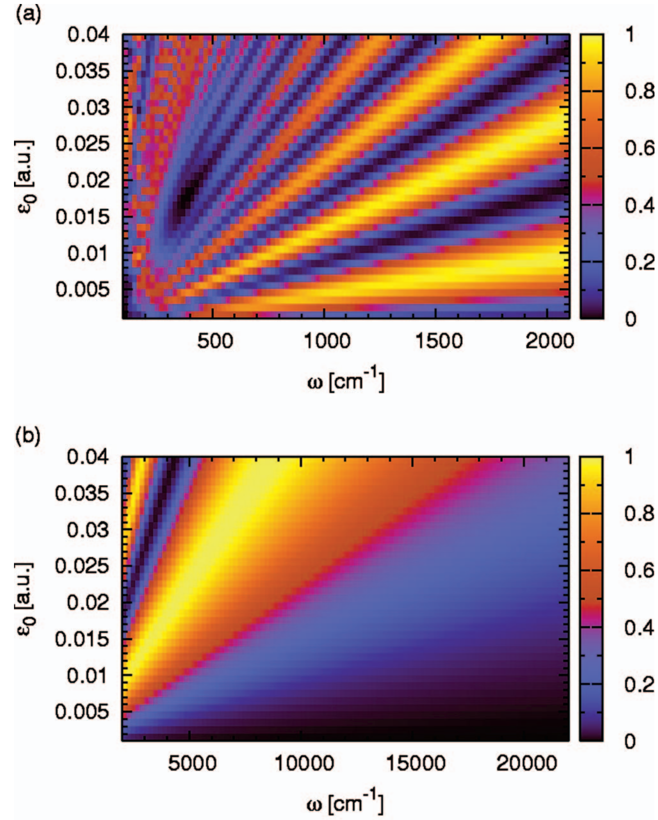

FIG. 7. (Color) Transition probabilities P_{12} obtained by the numerical calculations for an almost degenerate two-level system (ω_{12}=500 cm^{-1}, μ =1.0 a.u.) interacting with a 1 cycle pulse as a function of ω and ε_0. ϕ is set to 0.

crossing. We treat a simple model of diatomic molecules which consist of two dissociative linear potential energy curves (PECs), $V_1(R)$ and $V_2(R)$, where

$$V_i(R) = F_i(R - R_X) + E_X \quad (i = 1,2), \tag{19}$$

where R denotes the internuclear distance of diatomic molecules, and $F_2 < F_1 < 0$. At $R = R_X$, these PECs cross, and we have a LZ-type avoided crossing due to a diabatic potential coupling $V_{12}(R)$ (see Fig. 9). Let us consider a situation that a Gaussian wave packet is placed at $R = R_I$ on $V_2(R)$ with its initial momentum equal to zero. This wave packet propagates downward along $V_2(R)$ and bifurcates into two at R_X because of the diabatic potential coupling: one dissociates into channel 1 and the other into channel 2. The branching ratio depends on the coupling strength $V_X = V_{12}(R_X)$, $\Delta F = |F_1 - F_2|$, and the velocity υ_X of the wave packet at R_X because the dissociation probability into channel 2 is given by the LZ formula $\exp[-2\pi V_X^2/(\hbar \upsilon_X \Delta F)]$. We propose a control scheme to achieve selective dissociations into any one of the two channels with use of a 1 cycle laser pulse. The basic idea is as follows: when the wave packet reaches the crossing, the situation can be regarded as an almost degenerate two-level system. By applying 1 cycle laser pulse at the time when the wave packet reaches the crossing, it may be possible to control the wave packet transitions between the two states, leading to selective dissociations. Under the Born-Oppenheimer and the dipole approximation, the Hamiltonian $\hat{H}$ of the system is given as[18]

$$\hat{H} = \begin{bmatrix} \hat{T} + V_1(R) & V_{12}(R) - \boldsymbol{\mu}(R) \cdot \mathbf{E}(t) \\ V_{12}(R) - \boldsymbol{\mu}(R) \cdot \mathbf{E}(t) & \hat{T} + V_2(R) \end{bmatrix}, \tag{20}$$

where $\hat{T} = -(\hbar^2/2M)(d^2/dR^2)$ is the nuclear kinetic energy operator, M is the reduced mass of the system, $\boldsymbol{\mu}(R)$ is the transition dipole moment between $V_1(R)$ and $V_2(R)$, and $\mathbf{E}(t)$ is the electric field of 1 cycle laser pulse in Eq. (3). Throughout this paper, we assume the following: (i) molecular rotations can be neglected, (ii) molecules are aligned along the laser polarization $[\boldsymbol{\mu}(R)//\mathbf{E}_0]$, and (iii) $\boldsymbol{\mu}(R)$ is independent of R. For the Hamiltonian $\hat{H}$ in Eq. (3), using the fast Fourier transformation and split operator method,[18,23] we solve the time-dependent Schrödinger equation $i\hbar(\partial/\partial t)\Psi = \hat{H}\Psi$, where $\Psi = [\psi_1(R,t)\psi_2(R,t)]^t$, $\psi_i(R,t)$ is the nuclear wave function on $V_i(R)$, and the superscript t denotes the matrix transpose. The parameters of the system are given as follows: $F_1 = -0.005$ hartree$/a_0$, $F_2 = -0.01$ hartree$/a_0$, $R_X = 9.0a_0$, $E_X = 0.15$ hartree, $\mu = 1.0\ e\ a_0$, and $m = 20$ amu. We assume a Gaussian function for $V_{12}(R)$, i.e., $V_{12}(R) = V_X \exp[-(R - R_X)^2]$, where $V_X = 0.001$ hartree. The initial wave packet is

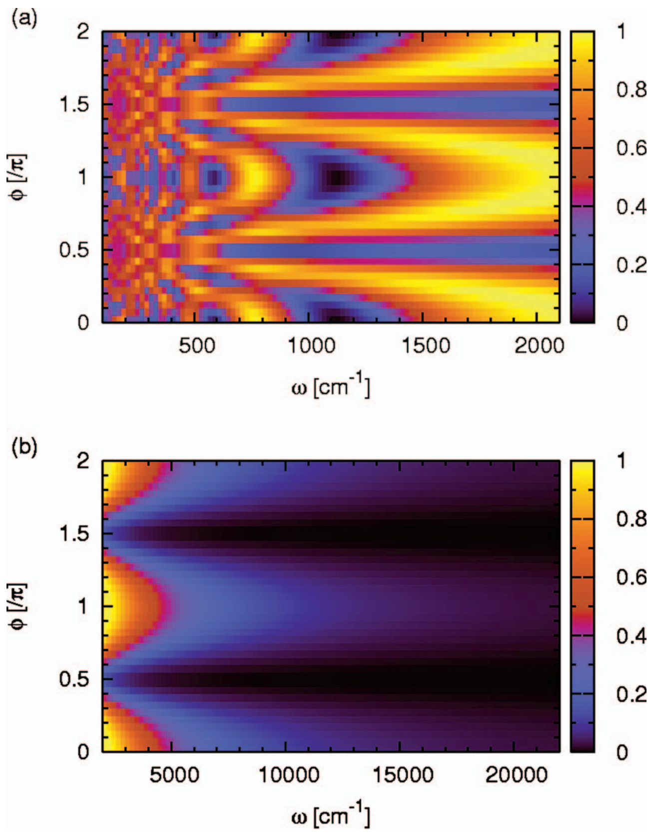

FIG. 8. (Color) Transition probabilities P_{12} obtained by the numerical calculations for an almost degenerate two-level system (ω_{12}=500 cm^{-1}, μ =1.0 a.u.) interacting with 1 cycle pulse as a function of ω and ϕ. ε_0 =0.01 a.u.

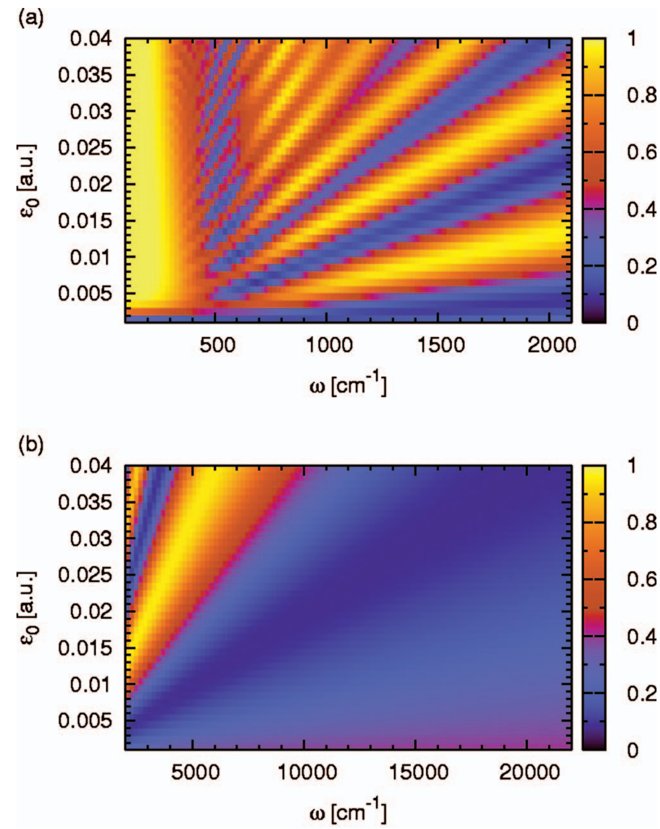

FIG. 10. (Color) For the two-state system shown in Fig. 9, the calculated dissociation probability into channel 1, $P_1^{(\mathrm{wp})}$, is plotted as a function of ω and ε_0 of 1 cycle pulse. ϕ is set to 0.

placed on $V_2(R)$, that is, $\psi_1(R,0)=0$ and $\psi_2(R,0)=(\pi\sigma_0^2)^{-1/4}\exp[-(R-R_I)^2/(2\sigma_0^2)]$. R_I is set to $2a_0$ and $\sigma_0 = 0.25a_0^{-1}$. Since this wave packet reaches the crossing around t=173.4 fs, the center of 1 cycle pulse is set to t_c =173.4 fs. The dissociation flux is integrated over time at $R=18a_0$ to obtain the corresponding dissociation probability. In order to prevent the unphysical reflection of the wave packet at the edge, the negative imaginary potential (absorption potential)[24] is set at $R=19a_0$. The grid sizes of t and R are δt=1.0 a.u. and $\delta R=0.0137a_0$, respectively. V_X

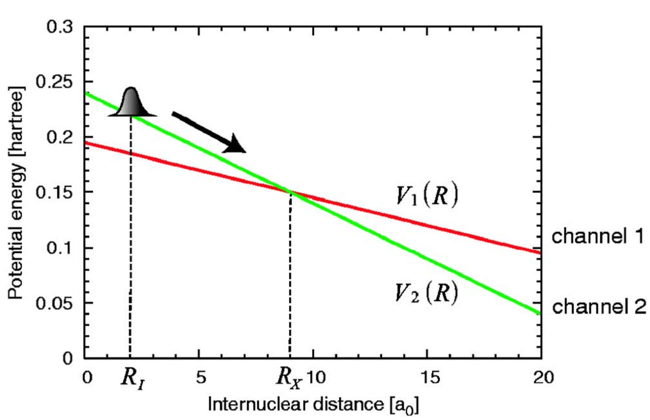

FIG. 9. (Color online) A two-state model of diatomic molecules is depicted. The initial wavepacket is placed on $V_2(R)$ at R_I. When the wave packet reaches the crossing of the potential energy curves at R_X, 1 cycle laser pulse is shined to control the branching ratio.

=0.001 hartree corresponds to an intermediate coupling strength, and thus the dissociation probability into the channel 1 is close to one-half (P_1=0.4206) in the field-free case. Figure 10 depicts $P_1^{(\mathrm{wp})}$ (wave packet calculation results of the dissociation probability into channel 1) as a function of ω and ε_0 of 1 cycle laser pulse. ϕ is fixed at 0. It is clearly seen that $P_1^{(\mathrm{wp})}$ drastically changes between 0 and 1. Thus, selective dissociations into any one of the channel can be achieved by properly choosing ω and ε_0. At the low frequencies of $\omega \leqslant 400$ cm^{-1}, we note that $P_1^{(\mathrm{wp})}$ is almost unity and does not depend on ω and ε_0. This is because 1 cycle pulse at such low frequencies does not change its amplitude during the passage of the wave packets around the crossing and the 1 cycle pulse acts as a strong static electric field, leading to the complete wave packet transfer from $V_2(R)$ to $V_1(R)$. On the other hand, at the higher frequencies of $\omega > 400$ cm^{-1}, it can be seen that the population inversion takes place when some linear relation between ε_0 and ω is satisfied. We note that the linear relation deviates a little from $\varepsilon_0 = \hbar\omega(1+2n)/|\mu|$, which is found from the analytical solution in Eq. (16). This seems to be due to the fact that, in the case of wave packet dynamics, the transition induced by the nuclear motion also takes place. Equation (16) does not take into account such transitions induced by the nuclear motion. Some scheme for treating both time-dependent and time-independent nonadiabatic transitions[25] is necessary to fully understand the transition mechanism. In Fig. 11, $P_1^{(\mathrm{wp})}$ is shown as a function of ω and ϕ of 1 cycle laser pulse. ε_0 is

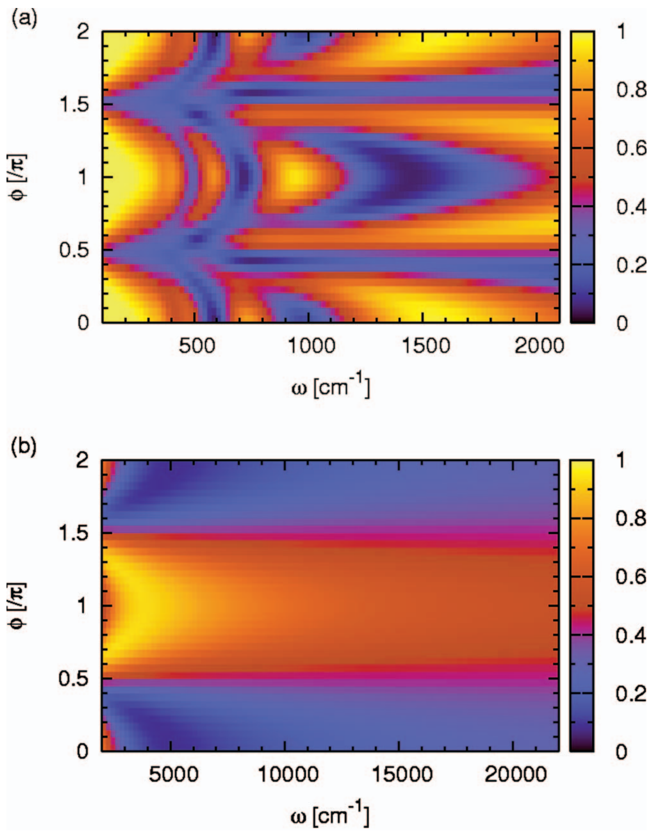

FIG. 11. (Color) For the two-state system shown in Fig. 9, the calculated dissociation probability into channel 1, $P_1^{(\mathrm{wp})}$, is plotted as a function of ω and ϕ of 1 cycle pulse. ε_0 is set to 0.01 a.u.

set to 0.01 a.u.. At the low frequencies of $\omega < 400\ \mathrm{cm}^{-1}$, as is mentioned above, $P_1^{(\mathrm{wp})}$ is almost unity except the case of $\phi=\pi/2$ or $3\pi/2$ because 1 cycle pulse is regarded as a strong static electric field. At the higher frequencies, the behavior of $P_1^{(\mathrm{wp})}$ is similar to but not the same as Eq. (16) [compare Figs. 11(a) and 11(b) with Figs. 5(c) and 5(d)]. The discrepancies arise from the fact that Eq. (16) does not include any transition induced by the nuclear motion. It is interesting to see that the behavior of $P_1^{(\mathrm{wp})}$ at $\phi=0$ is completely different from the one at $\phi=\pi$. If we use 1 cycle pulse of $\omega=950\ \mathrm{cm}^{-1}$ and $\varepsilon_0=0.01$ a.u., we can switch the dissociation channel by changing only the phase ϕ. The differences between $P_1^{(\mathrm{wp})}$ and Eq. (16) in Figs. 11 may contain some information on the potential coupling at the crossing. For example, the coefficient k for the linear relation $\varepsilon_0=k\omega$ where the population inversion takes place strongly depends on the coupling strength. This might be useful for extracting such information.

IV. CONCLUDING REMARKS

A simple and analytical solution of nonadiabatic transition probability for the degenerate (or almost degenerate) two-level systems interacting with 1 cycle laser pulse has been derived. The probability depends on the peak amplitude, frequency, and phase of 1 cycle pulse. The population inversion is possible unless the phase is $\pi/2$ nor $3\pi/2$ by properly choosing both the amplitude and frequency. It has also been found that the minimum pulse area for the population inversion never becomes smaller than π. As one of the applications of this formula, the new way of controlling the nuclear motions of diatomic molecules at a potential curve crossing by 1 cycle laser pulse has been proposed. We have treated a simple two-state model, in which two dissociative PECs cross at a certain internuclear distance. In the field-free case, since the potential coupling strength at the crossing is intermediate, the dissociation probability into one channel is almost one-half. It has been demonstrated that selective nonadiabatic dissociations into any one of the two channels can be achieved by properly choosing the laser frequency, amplitude, and phase of the 1 cycle laser pulse. The application to the real system such as Na I will be presented somewhere. Although we have concentrated on 1 cycle laser pulse in this paper, the extensions of the present semiclassical scheme to the general n-cycle pulses $(n>1)$ and/or complicated systems with more than two levels are straightforward. Since the amplitude is larger and the pulse duration is longer than the 1 cycle pulse used in Fig. 3, the population dynamics is much more complicated. However, the final transition probability is equal to unity. The general formula of the transition probability for n-cycle pulses with nondegenerate two-level system will be reported in the near future.

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