Detuning-induced stimulated Raman adiabatic passage in two-level systems with permanent dipole moments

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The detuning-induced stimulated Raman adiabatic passage (D-STIRAP) in two-level systems with permanent dipole moments (PDMs) is investigated. Based on a transformed Schrödinger equation, we discuss the coherence generation under the one- and two-photon excitations, respectively. In the one-photon excitation, our analysis shows that the envelope of the pump pulse gets modulated in presence of the PDMs. The modulation becomes stronger while the difference between the PDMs of the two levels becomes larger. Gradually, the adiabatic process of the D-STIRAP fails to complete and the robustness of the technique disappears. Therefore, parameters have to be chosen with care if maximum coherence is desired in systems with large or giant difference between the PDMs. Similar results can also be found in the two-photon excitation, although the influence of the PDMs on the D-STIRAP seems weaker compared with that in the one-photon excitation case. Finally, we point out that the negative role of the PDMs can be effectively avoided by simply choosing longer pulses instead and the D-STIRAP can be safely applied in two-level systems with PDMs.

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I. INTRODUCTION

Coherent quantum systems have shown applications in many areas such as in nonlinear optics [1–3]. They are also known as the basis in quantum information [4]. For decades, many protocols have been proposed to create different kinds of coherent quantum systems [5–8]. Among these schemes, the adiabatic technique is of large interest since it allows engineering quantum systems with high fidelity. Especially, the adiabatic technique shows the potential in quantum information to meet the requirement that quantum systems should be prepared and manipulated with minimal errors.

Recently, growing interest was shown to an adiabatic technique that can prepare maximum coherence in a two-level system [9]. The technique is an analog to the well-known stimulated Raman adiabatic passage (STIRAP) in a three-level system and it is illustrated in Fig. 1. It is found that the Bloch equation for a two-level system is mathematically similar to the Schrödinger equation for a three-level Λ system. Therefore, a counterintuitive sequence of the time-dependent detuning (which we call the"detuning pulse") and the pump pulse leads that the value of the population inversion $w = -1$ can be completely transferred to the real part of the coherence term u, which is defined as $2\rho_{12} = u + iv$, while the imaginary part v is still kept at zero. Finally, a stable maximum coherence is achieved. Similar to the STIRAP, the adiabatic condition should also be satisfied during the process, which is

$$\int_{-\infty}^{\infty} \sqrt{\Delta^2 + \Omega^2} dt \gg \pi/2. \qquad (1)$$

Note Δ and Ω are the time-dependent detuning and Rabi frequency of the pump pulse, respectively. The integration in Eq. (1) requires that the amplitudes of the detuning pulse and Rabi frequency should be strong with pulse durations sufficiently long. Especially, the overlapped decreasing side of the detuning pulse and the rising side of the pump pulse are critical and they should be long enough for the adiabatic process to complete, as can be seen in Fig. 1(b). Then, the technique is robust against parameters such as the relative time delay between the two pulses, and the amplitudes of the two pulses. Since the process is accomplished under the assistance of a "detuning pulse", we call the technique the detuning-induced STIRAP (D-STIRAP).

Such elegant extension from the STIRAP to the D-STIRAP has drawn a lot of attention. The D-STIRAP was soon carried out in experiment [10]. Based on the technique, coherence preparation in complicated systems such as in dense two-level systems [11] or two-level systems with hyperfine structure [12] was discussed. Transparency of pulses in two-level systems with or without hyperfine structure [13–15] was also found. Besides, it was suggested that the technique can be applied to generate the extreme ultraviolet (XUV) light with high efficiency [16]. Moreover, the technique was found quite useful in the Bose-Einstein condensate [17]. Lately, even the classical simulation of the D-STIRAP was realized in optical waveguides [18].

For an ideal quantum control scheme, it is always desired to make it applicable in materials as many as possible. Among these materials, a big portion of them are proven to be quantum systems with broken inversion symmetry. In these systems, the diagonal matrix elements of the dipole moment operator, which are called the permanent dipole moments (PDMs), are nonequivalent. Generally, quantum control schemes behave differently in systems with PDMs and they should be investigated carefully with necessity. During the

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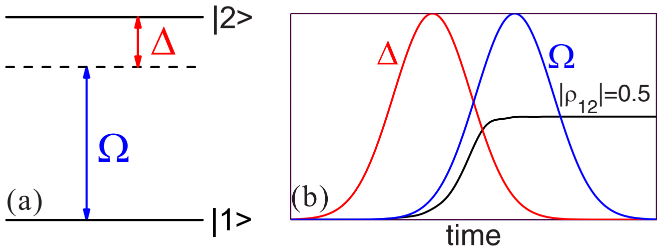

FIG. 1. (a) Prototype scheme for the D-STIRAP in a two-level system. (b) With a counterintuitive sequence of the time-dependent detuning and the pump pulse, stationary maximum coherence is obtained.

past decades, there have been many literatures concentrating on the topic, including the investigation of the optical bistability [19,20], the four-wave mixing [21], the harmonic generation [22], the STIRAP [23–25], and the electromagnetically induced transparency (EIT) [26,27] in systems with PDMs.

For the D-STIRAP, it also faces the same situation as described above. Therefore, it is our purpose here to discuss the behavior of the D-STIRAP in two-level systems with PDMs. In this paper, we carry out the study based on a transformed Schrödinger equation and we focus on the one- and two-photon excitations, respectively. In the one-photon excitation, it is found that systems with large or giant difference between the PDMs of the two levels can obviously prevent the adiabatic process of the D-STIRAP from being completed. This is due to the reason that the presence of the PDMs modulates the envelope of the pump pulse. The modulation is stronger when the difference between the PDMs becomes larger. Hence, satisfaction of the adiabatic condition of the D-STIRAP meets more difficulty and the robustness of the technique gradually vanishes. Consequently, maximum coherence has to be generated by controlling parameters carefully. Similar results are also found in the two-photon excitation, although it looks like the D-STIRAP under the two-photon excitation is less influenced by the PDMs. As to the negative effect of the PDMs (especially those lead to the giant difference between the PDMs) on the D-STIRAP, the following discussion shows that we can simply use longer pulses instead to avoid it. Therefore, the D-STIRAP is still available in such systems.

II. THEORY

We start by considering a two-level system with nonzero PDMs as shown in Fig. 1(a). When interacting with an off-resonant pump pulse, the evolution of the system can be described by the Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\Psi = H\Psi. \tag{2}$$

Under the semiclassical dipole approximation, it can be written as

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix}a_1\\a_2\end{pmatrix} = \begin{pmatrix}\mathscr{E}_1-\mu_{11}E(t) & -\mu_{12}E(t)\\ -\mu_{21}E(t) & \mathscr{E}_2-\mu_{22}E(t)\end{pmatrix}\begin{pmatrix}a_1\\a_2\end{pmatrix}. \tag{3}$$

In the above equation, a_j and $\mathscr{E}_j$ represent the probability amplitude and the energy of level $|j\rangle$, respectively. μ_{12} and μ_{21} are the nondiagonal dipole moments. Here, we assume they are equal with each other, which is $\mu_{21}=\mu_{12}$. μ_{jj} is the diagonal PDM defined with respect to level $|j\rangle$, and $E(t)$ is the time-dependent electric field associated with the pump pulse.

Generally, it is difficult to carry out the theoretical analysis by directly using Eq. (3) since the equation can only be solved numerically. In order to clarify the underlying physics induced by the PDMs, it would be very helpful to obtain a transformed Schrödinger equation following the method introduced in Ref. [28], which has been successfully applied in many other systems with PDMs [20,23,26]. The method first treats Eq. (3) in the interaction representation b, which is defined as

$$a_j = b_j\exp\left\{-\frac{i}{\hbar}\left[\mathscr{E}_j(t-t_0)-\mu_{jj}\int_{t_0}^{t}E(t')dt'\right]\right\}. \tag{4}$$

In the above equation, t_0 is the initial time when the interaction begins. Equation (3) then becomes

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix}b_1\\b_2\end{pmatrix} = \begin{pmatrix}0 & H_{12}\\ H_{21} & 0\end{pmatrix}\begin{pmatrix}b_1\\b_2\end{pmatrix}, \tag{5}$$

with the Hamiltonian having the following form:

$$H_{12} = H_{21}^{*} = -\frac{\mu_{12}E(t)}{\hbar}\exp\left\{-i\omega_{21}(t-t_0) + i\frac{d}{\hbar}\int_{t_0}^{t}E(t')dt'\right\}. \tag{6}$$

In Eq. (6), ω_{21} is the transition frequency of the two-level system and d is the difference between the PDMs of the two levels, which are

$$\omega_{21} = (\mathscr{E}_2-\mathscr{E}_1)/\hbar > 0, \quad d = \mu_{22}-\mu_{11}. \tag{7}$$

To proceed, the electric field of the pump pulse is assumed as

$$E(t) = \hat{e}E_0 f_{\mathrm{p}}(t)\cos(\omega_0 t+\delta), \tag{8}$$

where $\hat{e}$, E_0, $f_{\mathrm{p}}(t)$, ω_0, and δ are the polarization vector, the amplitude of the electric field, the envelope of the pump pulse, the central frequency, and the initial phase, respectively. Since the D-STIRAP can be carried out experimentally by using a single linearly chirped pump pulse [10] and the chirp rate can be positive or negative, we point out that the central frequency of the pump pulse, ω_0, is initially far detuned (red or blue detuned) in such a case. Then, the central frequency will increase or decrease with time and finally the pump pulse is resonant with the two-level system.

In order to simplify the Hamiltonian in Eq. (6), the authors in Ref. [28] suggested using the rotating wave approximation (RWA) and the approximation that the pump pulse duration τ_{p} is much longer than $1/\omega_0$, which is $\omega_0\tau_{\mathrm{p}} \gg 1$. However, if we also want to use the two approximations before carrying out the analysis, we should be careful about the approximation $\omega_0\tau_{\mathrm{p}} \gg 1$. Its validity should be discussed since the central frequency of the pump pulse, ω_0, is now time dependent, which is different from that in Ref. [28]. Especially, we should pay attention to the situation when a red-detuned pump pulse with positive chirp rate is employed to realize the D-STIRAP. In this situation, the minimal value of the central frequency can be $\omega_0^{\min} = \omega_{21}-\Delta_0$, where Δ_0 is the amplitude of the detuning pulse. Generally, Δ_0 should be large enough to satisfy the adiabatic condition for the D-STIRAP. But, still, it is found to be much smaller than the transition frequency, which is $\Delta_0 \ll \omega_{21}$, as has been indicated in previous literatures [12,16] and will be confirmed in the following calculations. Therefore, we can say that the approximation $\omega_0\tau_p \gg 1$ can be kept valid when the central frequency changes with time.

Now, we can continue to use the two approximations mentioned above. The Hamiltonian H_{12} is transformed to be

$$H_{12} = -\frac{\mu_{12}\cdot\hat{e}E_0}{\hbar}\frac{N}{z}J_N(zf_p(t))\exp(i\omega_{21}t_0 + iN\delta)\exp[-i(\omega_{21}-N\omega_0)t], \quad (9)$$

where $J_k(x)$ is the Bessel function of the first kind and N corresponds to the N-photon excitation case. z is defined as

$$z = \frac{d\cdot\hat{e}E_0}{\hbar\omega_0}. \quad (10)$$

If the amplitude of the Rabi frequency has the form

$$\Omega_0 = \frac{\mu_{12}\cdot\hat{e}E_0}{\hbar}, \quad (11)$$

then z can be rewritten as

$$z = \frac{d\Omega_0}{\mu_{12}\omega_0}. \quad (12)$$

We can see from Eq. (10) or (12) that the parameter z is a function of the difference between the PDMs, d. Since we focus on studying the effect solely induced by the PDMs on the D-STIRAP in this paper, we will change the value of d to investigate different evolution of the system under the D-STIRAP in the following section. We will do this by simply varying the value of z while keeping the other three parameters in Eq. (10) or (12) unchanged. Moreover, we should point out that one of the most important properties of the D-STIRAP is that it is quite robust against the fluctuation of the pump pulse intensity. Therefore, when discussing the effect of the PDM on the sensitivity of the D-STIRAP to the pump pulse intensity, the value of z should be changed proportional to the amplitude of the Rabi frequency Ω_0. As will be shown in the following section, z is a very important characteristic parameter to influence the adiabatic process of the D-STIRAP.

It is also interesting to see from Eq. (9) that both the odd- and even-photon excitations are allowed in two-level systems with the PDMs, which are generally mutually exclusive because of the inversion symmetry. In this case, the even-photon excitation such as the two-photon excitation can happen without the assistance of the intermediate or virtual level. This is due to the reason that the presence of the PDMs now breaks the inversion symmetry [29].

If we assume the interaction starts at $t_0 = 0$ and the initial phase of the pump pulse be set as $\delta = 0$ in Eq. (9), we can define the time-dependent detuning $\Delta(t)$ and the Rabi frequency of the pump pulse $\Omega(t)$ under the N-photon excitation as

$$\Delta(t) = \omega_{21} - N\omega_0 \quad (13)$$

and

$$\Omega(t) = \frac{1}{2}\Omega_0\frac{2NJ_N(zf_p(t))}{z}, \quad (14)$$

respectively. Then, Eq. (9) can be further simplified as

$$H_{12} = -\Omega(t)\exp[-i\Delta(t)t]. \quad (15)$$

Therefore, the transformed Schrödinger equation [Eq. (5)] combined with the simplified Hamiltonian expressed in Eq. (15) describe the interaction of a pump pulse with a two-level system with PDMs under the N-photon excitation. We can see from Eq. (14) that the only modification in Eq. (5) compared with the Schrödinger equation without considering the PDMs is that the envelope of the pump pulse $f_p(t)$ is now modulated by the parameter z (or the PDMs). Obviously, the modification will make the evolution of the D-STIRAP in two-level systems with PDMs different from that without PDMs. Besides, we note that the population of the two levels, $|\rho_{jj}| = |b_j b_j^*|$, and the value of the coherence, $|\rho_{12}| = |b_1 b_2^*|$, still keep unchanged in the interaction representation b, as can be easily seen from the definition in Eq. (4).

Based on the transformed Schrödinger equation, we will carry out the numerical calculations in the next section. In order to make the underlying physics more clear, the decaying rates from the upper level are ignored. By defining $b_1 = c_1 + ic_2$ and $b_2 = c_3 + ic_4$, the transformed Schrödinger equation can be expended as

$$\begin{aligned}
\frac{\partial}{\partial t}c_1 &= \Omega(t)\sin[\Delta(t)t]c_3 - \Omega(t)\cos[\Delta(t)t]c_4,\\
\frac{\partial}{\partial t}c_2 &= \Omega(t)\cos[\Delta(t)t]c_3 + \Omega(t)\sin[\Delta(t)t]c_4,\\
\frac{\partial}{\partial t}c_3 &= -\Omega(t)\cos[\Delta(t)t]c_2 - \Omega(t)\sin[\Delta(t)t)]c_1,\\
\frac{\partial}{\partial t}c_4 &= \Omega(t)\cos[\Delta(t)t]c_1 - \Omega(t)\sin[\Delta(t)t]c_2. \quad (16)
\end{aligned}$$

The above equation can be solved directly by using the fourth-order Runge-Kutta method. In the calculations, we assume that both the detuning and pump pulse have the Gaussian shape, which are

$$\Delta(t) = \Delta_0 f_\Delta(t) = \Delta_0\exp\left[-\ln 4\frac{(t-t_\Delta)^2}{\tau_\Delta^2}\right] \quad (17)$$

and

$$f_p(t) = \exp\left[-\ln 4\frac{(t-t_\Delta-t_p)^2}{\tau_p^2}\right], \quad (18)$$

respectively. In the above two equations, τ_Δ and τ_p are the durations of the detuning and pump pulse. t_Δ is the time delay of the detuning pulse, while t_p is the relative time delay between the detuning and pump pulse.

III. CALCULATION RESULTS AND DISCUSSION

In this section, we will give a general investigation of the D-STIRAP in two-level systems with PDMs under one- and two-photon excitations, respectively. As has been mentioned above, the presence of the PDMs can modulate the envelope of the pump pulse. Since the decreasing side of the detuning

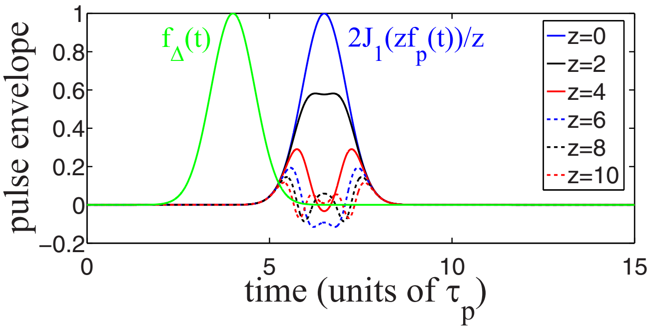

FIG. 2. Modulation on the pump pulse envelope by the PDMs, $2J_1(zf_p(t))/z$, in the one-photon excitation. To demonstrate the effect of the difference between the PDMs, d, the amplitude of the Rabi frequency is fixed at $\Omega_0 = 60/\tau_p$ while the parameter z increases from 0 to 10. The envelope of the detuning pulse $f_\Delta(t)$ is also given for reference and the relative time delay between the two pulses is fixed at $t_p = 2.5\tau_p$.

pulse and the rising side of the pump pulse are critical for the adiabatic process of the D-STIRAP to complete, we can predict that the modification on the pump pulse envelope will make the behavior of the D-STIRAP different from that in two-level systems without PDMs. Therefore, we will first study the variation of the pump pulse envelope under different value of the PDMs. Continuously, we will discuss how the variation influences the adiabatic process and the robustness of the D-STIRAP in both one- and two-photon excitation cases.

In the following calculations, the transition dipole moment, the PDMs, and the polarization vector of the laser are assumed to be aligned with one another, which is $\mu_{12} \parallel d \parallel \hat{e}$. All the parameters are chosen in units of the duration of the pump pulse τ_p throughout this paper. Besides, the duration of the detuning pulse is set equal to the pump pulse duration, which is $\tau_\Delta = \tau_p$, and the delay of the detuning pulse is fixed at $t_\Delta = 4\tau_p$. Note that the characteristic parameter z is unitless according to its definition.

A. One-photon excitation

The calculation of one-photon excitation can be carried out by simply choosing $N = 1$ in Eq. (16). First, the envelope of the pump pulse under different value of the characteristic parameter z is given in Fig. 2 based on Eq. (14). Since the parameter z is a function of the difference between the PDMs, d, and the amplitude of the Rabi frequency, Ω_0, according to Eq. (12), the amplitude of the Rabi frequency is fixed at $\Omega_0 = 60/\tau_p$ to clarify the effect of the PDMs in this figure. For reference, the detuning pulse is also given with the relative time delay between the two pulses fixed at $t_p = 2.5\tau_p$.

We can roughly see from Fig. 2 that the parameter z increases from 0 to 10, the modulation on the envelope of the pump pulse gradually gets stronger. To be more detailed, if there are no PDMs in the two-level system, which means $z = 0$, the Rabi frequency $\Omega(t)$ in Eq. (14) becomes the familiar form

$$\Omega(t) = \frac{\mu_{12} \cdot \hat{e} E_0}{2\hbar} f_p(t). \tag{19}$$

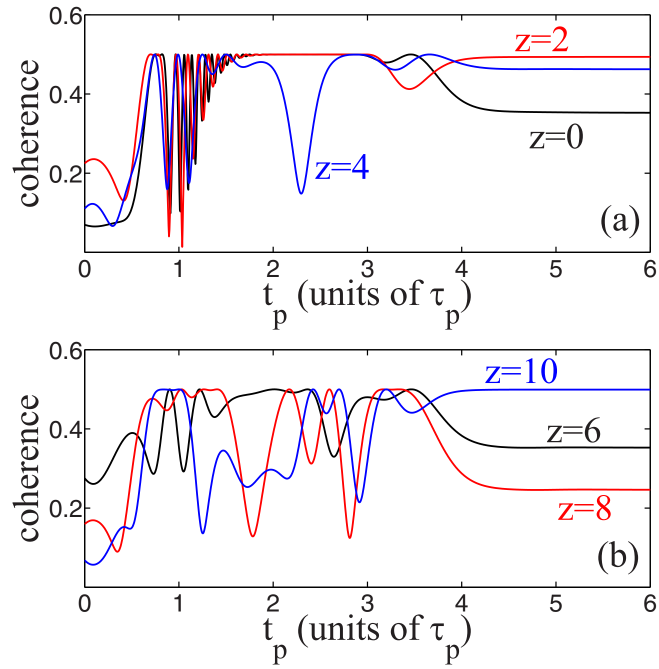

FIG. 3. Generated coherence at the time $t = 15\tau_p$ as a function of the relative time delay t_p, with the parameter z chosen in (a) as $z = 0$; $z = 2$; $z = 4$; and in (b) as $z = 6$; $z = 8$; $z = 10$. The amplitudes of the detuning pulse and the Rabi frequency are equal with each other, $\Omega_0 = \Delta_0 = 60/\tau_p$. All the other parameters are kept the same as those in Fig. 2.

Clearly, the modulation of the PDMs on the pump pulse envelope disappears, as shown by the blue line in Fig. 2. Therefore, the counterintuitive time sequence of the detuning and pump pulse leads to the standard D-STIRAP and maximum coherence generated with robustness can be predicted. When the parameter z is nonzero and increases from 2 to 10, we can find that the peak value of the pump pulse envelope decreases. Meanwhile, the peak splitting shows up. As has been mentioned in the Introduction section, the amplitude of the pump pulse should be strong and the rising side of the pump pule, which is overlapped with the decreasing side of the detuning pulse, should be sufficiently long for the adiabatic process to complete. Obviously, we can predict here that the modulation on the pump pulse envelope in presence of the PDMs may destroy the adiabatic condition of the D-STIRAP and the robustness of the technique may vanish. Consequently, maximum coherence may have to be generated with higher requirement.

To further verify our prediction, we will discuss the robustness of the D-STIRAP in presence of the PDMs in the following. First, generated coherence as a function of the relative time delay t_p under different value of the parameter z is given in Fig. 3. To be accordant with Fig. 2, the amplitude of the Rabi frequency is still chosen as $\Omega_0 = 60/\tau_p$ to demonstrate the effect of the difference between the PDMs, d. For simplicity, the amplitude of the detuning pulse is set equal to that of the Rabi frequency, which is $\Delta_0 = 60/\tau_p$. Generated coherence at the time $t = 15\tau_p$ after the two pulses have passed is chosen. All the other parameters are kept the same as those in Fig. 2. The detuning and pump pulse is

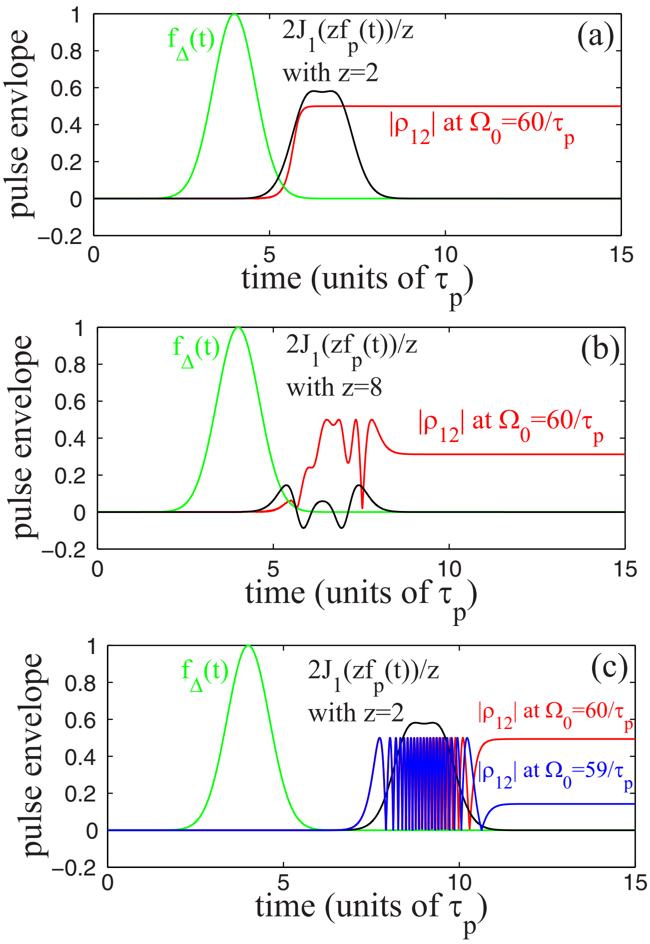

FIG. 4. Time evolution of the coherence with parameters chosen in (a) as $t_p = 2.5\tau_p$, $z = 2$, $\Omega_0 = 60/\tau_p$, and in (b) as $t_p = 2.4\tau_p$, $z = 8$, $\Omega_0 = 60/\tau_p$, and in (c) as $t_p = 5\tau_p$, $z = 2$, while the amplitude of the Rabi frequency slightly varies from $\Omega_0 = 60/\tau_p$ to $59/\tau_p$. In all the subfigures, the amplitude of the detuning pulse is fixed at $\Delta_0 = 60/\tau_p$.

partially overlapped when the relative time delay t_p is smaller than $4\tau_p$, which is around $t_p < 4\tau_p$.

When there are no PDMs in the two-level system with $z = 0$, which corresponds to the standard D-STIRAP case, the generated maximum coherence is quite insensitive to the relative time delay t_p as shown by the black line in Fig. 3(a). With the parameter z increases to 2, we can see that the robustness of the D-STIRAP is still well preserved, which is shown by the red line in Fig. 3(a). This implies that the existence of the PDMs has not yet affected the adiabatic process of the D-STIRAP. The phenomenon can be explained by the envelope of the pump pulse displayed in Fig. 2. Although the peak value of the envelope starts to decrease when $z = 2$, the rising side of the pump pulse is still long enough for the adiabatic process to complete under the choice of the amplitudes of the two pulses. It would be more clear if we give the time evolution of the coherence in this case, as can be seen in Fig. 4(a). In this figure, the relative time delay is fixed at $t_p = 2.5\tau_p$ while the other parameters are the same as those in Fig. 3. We can see from this figure that the adiabatic process is fully completed due to the partially overlapped detuning and modulated pump pulse. After that, the generated maximum coherence will not be affected by the rest of the pump pulse any longer.

If the parameter z keeps increasing from 4 to 10, we can see from Fig. 3 that generation of the maximum coherence becomes sensitive to the relative time delay, which means the robustness of the D-STIRAP disappears. The phenomena can also be explained by the variation of the pump pulse envelope. In these cases, the modulation on the envelope is stronger and the rising side of the pump pulse is not long enough for the adiabatic process to complete. Therefore, the adiabatic condition in Eq. (1) is actually destroyed. For instance, we show in Fig. 4(b) the time evolution of the coherence when the characteristic parameter $z = 8$ and the relative time delay $t_p = 2.4\tau_p$. All the other parameters are kept unchanged as those in Fig. 3. As can be seen in Fig. 4(b), the rising side of the pump pulse is not sufficiently long for the adiabatic process to accomplish and maximum coherence is unable to achieve. The generated coherence is finally determined by the rest part of the pump pulse.

We should point out that we can also observe the generation of maximum coherence in Fig. 3 when the parameter $z = 2$ or 10 with $t_p > 4\tau_p$. This phenomenon does not result from the adiabatic process, but simply the Rabi oscillation by the pump pulse. In this case, there is no overlapping between the detuning and pump pulse to form the D-STIRAP. A slight variation of the amplitude of the Rabi frequency can easily change the generated coherence, as can be seen in Fig. 4(c).

To continue, we investigate the generated coherence under the D-STIRAP as functions of the amplitudes of the detuning pulse Δ_0 and the Rabi frequency Ω_0 in presence of the PDMs. The sole effect of the difference between the PDMs, d, with different value on the robustness of the D-STIRAP is emphasized in Fig. 5. In order to achieve this target, the parameter z has to change proportional to the amplitude of the Rabi frequency Ω_0 in the calculations. Since the amplitude of the Rabi frequency is always fixed at $60/\tau_p$ in Figs. 2 and 3, here the parameter z is replaced by z', which is defined as

$$z' = z\frac{\Omega_0}{60} \tag{20}$$

for consistency. In Figs. 5(a) and 5(b) where $z = 0$ and 2, the relative time delay is fixed at $t_p = 2.5\tau_p$, while in Figs. 5(c)–5(f) where z increases from 4 to 10, the relative time delay is optimized to ensure that maximum coherence is generated based on Fig. 3. Therefore, the relative time delay $t_p = 2.9\tau_p$, $2\tau_p$, $3.2\tau_p$, and $3.2\tau_p$, while the parameter $z = 4$, 6, 8, and 10, respectively.

The calculation results in Fig. 5 turn out that the influence of the PDMs on the sensitivity of the D-STIRAP to the amplitudes of the detuning pulse Δ_0 and the Rabi frequency Ω_0 is quite similar to that in Fig. 3. The robustness of the technique disappears gradually while the value of the parameter z increases. In two-level systems without the PDMs, we can obtain the typical property of the D-STIRAP, which is dispalyed in Fig. 5(a) for reference. In this figure, maximum coherence can be generated as long as the amplitudes of the detuning pulse and the Rabi frequency are strong enough to satisfy the adiabatic condition. In presence of small difference between

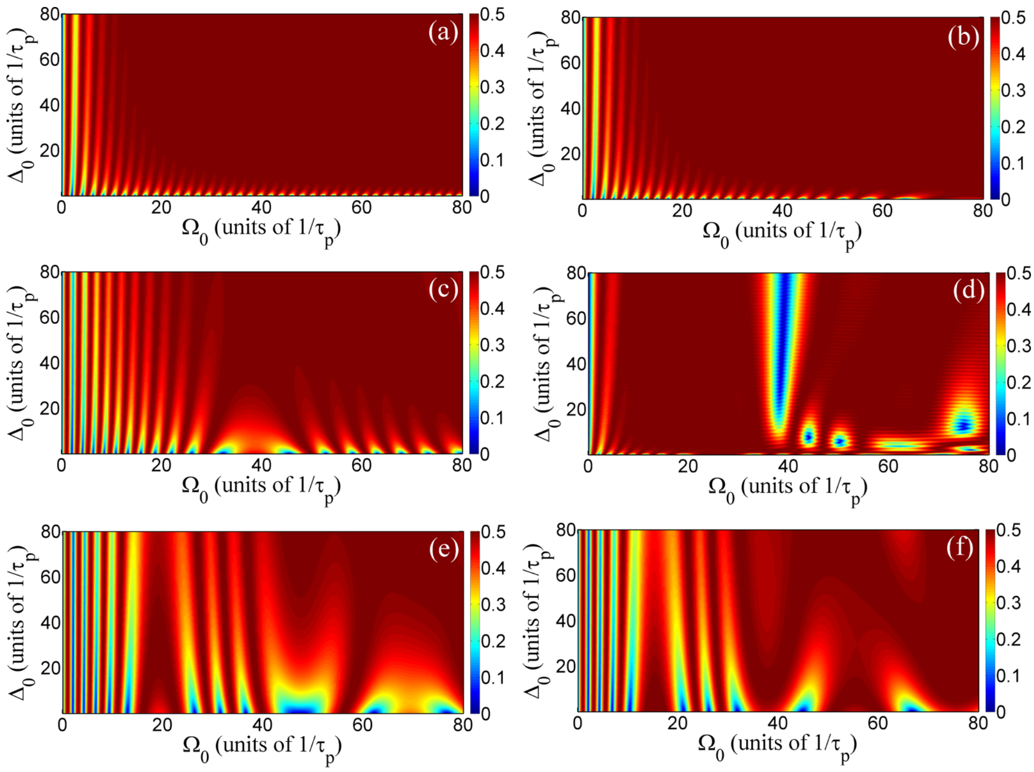

FIG. 5. Generated coherence at the time $t = 15\tau_p$ as functions of the amplitudes of the detuning pulse Δ_0 and the Rabi frequency Ω_0 is shown in (a) with $z = 0$, $t_p = 2.5\tau_p$, (b) with $z = 2$, $t_p = 2.5\tau_p$, (c) with $z = 4$, $t_p = 2.9\tau_p$, (d) with $z = 6$, $t_p = 2\tau_p$, (e) with $z = 8$, $t_p = 3.2\tau_p$, and (f) with $z = 10$, $t_p = 3.2\tau_p$. The relative time delay t_p in Figs. 5(c)–5(f) is optimized according to Fig. 3. The parameter z is replaced by z', which is $z' = z\Omega_0/60$, to emphasize the effect of d.

the PDMs with $z = 2$, the robustness of the D-STIRAP is still preserved, as can be found in Fig. 5(b). When z increases from 4 to 10, we find from Figs. 5(c)–5(f) that the region where maximum coherence can be generated is narrowed. At the mean time, there is discontinuity in the region, which means we should choose the amplitudes of the detuning pulse and the Rabi frequency carefully for maximum coherence. This phenomenon can also be explained by the variation of the pump pulse envelope in Fig. 2. The modulation on the envelope is weak when $z = 2$ and the D-STIRAP still applies well in this case. While the modulation becomes stronger when z increases from 4 to 10, the adiabatic process is getting harder to complete and the negative role played by the PDMs on the D-STIRAP is getting more obvious.

B. Two-photon excitation

Now, we consider the D-STIRAP in two-level systems with PDMs under the two-photon excitation. The two-photon excitation without the help of the intermediate or virtual level is special because it can happen when the two-level system has broken inversion symmetry due to the nonzero PDMs. Moreover, the "direct" two-photon excitation is found to be more effective than that through virtue levels [30]. Besides, the D-STIRAP carried out under the two-photon excitation has important applications such as generating pulses with ultrashort wavelength with high efficiency [16]. By setting $N = 2$ in Eq. (14), we will investigate the D-STIRAP following the same procedure as that in Sec. III A. We will first show the modulation on the pump pulse envelope by the PDMs under the two-photon excitation. Then, we will discuss the influence of the PDMs on the robustness of the D-STIRAP.

The modulation on the pump pulse envelope by the PDMs, $4J_2(zf_p(t))/z$, is shown in Fig. 6. The envelope of the detuning pulse is also given for reference. All the parameters are kept the same as those in Fig. 2. We can see from this figure that when there are no PDMs in the two-level system with $z = 0$, which corresponds to $4J_2(zf_p(t))/z = 0$, the envelope of the pump pulse vanishes. This implies clearly that the two-photon excitation is forbidden in two-level systems without the PDMs. When the parameter z increases from 2 to 10, similar evolution of the pump pulse envelope to that in Fig. 2

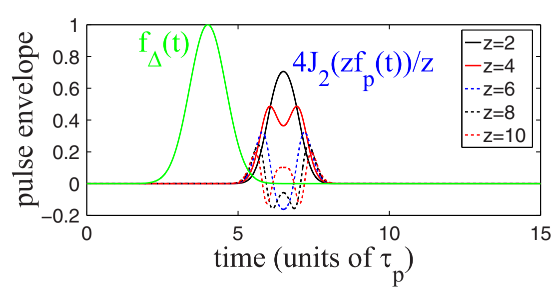

FIG. 6. Modulation on the pump pulse envelope by the PDMs, $4J_2(zf_p(t))/z$, under the two-photon excitation. For reference, the envelope of the detuning pulse is also given. All the parameters are the same as those in Fig. 2.

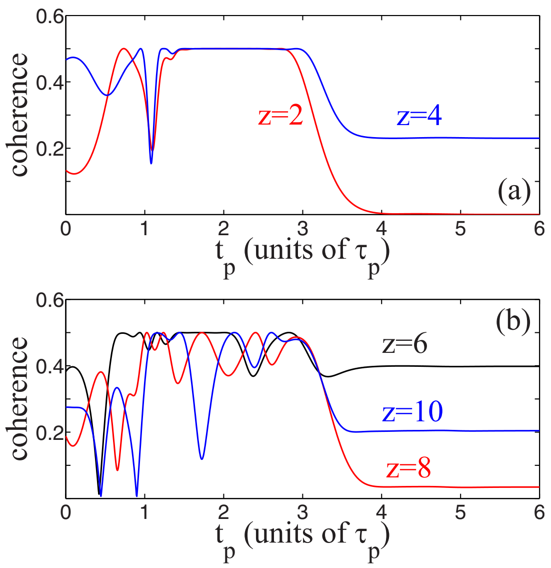

FIG. 7. Generated coherence at the time $t = 15\tau_p$ as a function of the relative time delay t_p under the two-photon excitation with the parameter z chosen in (a) as $z = 2$; $z = 4$; and in (b) as $z = 6$; $z = 8$; $z = 10$. All the parameters are kept the same as those in Fig. 3.

is found. The peak value of the envelope decreases gradually and the peak splitting appears at the same time, which makes the rising side of the pump pulse overlapped with the detuning pulse shorter. Nevertheless, we can still observe the difference between Figs. 6 and 2. The modulation on the pump pulse envelope by the PDMs is relatively weaker in Fig. 6 than that in Fig. 2. Therefore, we can predict that the robustness of the D-STIRAP under the two-photon excitation may be less influenced by the PDMs.

Based on Fig. 6, we continue to investigate the sensitivity of the D-STIRAP to the relative time delay, the amplitudes of the detuning pulse, and the Rabi frequency under different value of the PDMs. First, generated coherence with respect to the relative time delay t_p is presented in Fig. 7. In this figure, all the parameters are kept unchanged as those in Fig. 3. We can see from Fig. 7 that the sensitivity to the relative time delay under the two-photon excitation is similar to that under the one-photon excitation. While the parameter z increases, the robustness of the D-STIRAP against the relative time delay disappears. Moreover, we can observe from this figure that the impact of the PDMs on the robustness is weaker in the two-photon excitation. When the parameter z is small with $z = 2$, the robustness of the D-STIRAP is almost unchanged. While the parameter z becomes larger, which is $z = 4$, we find that the D-STIRAP is still robust against the relative time delay, which is different from that shown in Fig. 3(a). Even in the case when $z = 6$, fluctuation of the relative time delay is still allowed to generate the maximum coherence. Only when the value of the parameter z is very large, such as $z = 8$ or 10, the D-STIRAP becomes quite sensitive to the relative time delay. The phenomena are accordant with our prediction and the explanation is the same as that in Fig. 3. Since the modulation on the pump pulse envelope is weaker under the two-photon excitation, the D-STIRAP is more robust against the relative time delay in this case.

Moreover, calculation results on the sensitivity of the D-STIRAP to the amplitudes of the detuning pulse and the Rabi frequency are displayed in Fig. 8, which are found similar to those in Fig. 5. In the calculations, the parameter z is replaced by z', which is $z' = z\Omega_0/60$, due to the same reason as that in Fig. 5. The relative time delay t_p is also optimized in Figs. 8(d) and 8(e) to ensure that maximum coherence is generated according to Fig. 7. If the parameter z has small value, which corresponds to $z = 2$, there is slight modulation on the pump pulse envelope and maximum coherence can be generated as long as the amplitudes of the detuning pulse and the Rabi frequency are strong to satisfy the adiabatic condition. When the parameter z increases, the modulation on the envelope becomes stronger and the adiabatic condition is getting harder to be satisfied. Therefore, the region where maximum coherence can be obtained becomes smaller. Meanwhile, the discontinuity in the region appears. In these cases, we should choose the amplitudes of the detuning pulse and the Rabi frequency carefully for the maximum coherence.

In both the one- and two-photon excitations, the above general discussion implies that materials with small value of the characteristic parameter z do not apparently influence the adiabatic process of the D-STIRAP, while materials with large value of z are going to influence the adiabatic process with high probability. Therefore, it is of importance to discuss the feasibility of the D-STIRAP by looking for methods to avoid the negative role played by the PDMs, especially when materials with large value of z (which may be induced by the giant difference between the PDMs) are chosen in experiments. In this case, we can see from Eq. (12) that the difference between the PDMs, d, is fixed and the parameter z is proportional to the amplitude of the Rabi frequency Ω_0. Hence, in order to minimize the negative effect of the parameter z, it is natural to consider that we can reduce the value of z by using weaker pump pulse. However, this may simultaneously lead to the problem that reducing the amplitude of the electric field may destroy the adiabatic condition in Eq. (1) if the durations of the detuning and pump pulse are kept unchanged. To avoid this problem, we point out that we can use longer pulses instead to satisfy the adiabatic condition while maintaining the value of the parameter z small. Consequently, the negative influence of the giant difference between the PDMs, d, is effectively reduced.

We can take an example to demonstrate the above analysis. There are a lot of materials known to be two-level systems with large or giant difference between the PDMs [19,23,28,31–33], such as polar gases, organic compounds, and quantum dots. Here, we consider the frequently used HCN → HNC isomerization, whose parameters are given in Ref. [23]. The energy gap of the rovibrational transition is $\Delta\mathscr{E}_{12} = 0.02$ a.u. The difference of the PDMs is $d = -1$ a.u., which is giant compared with the transition dipole moment with $\mu_{12} = 0.01$ a.u. Now, we consider the D-STIRAP under the one-photon excitation. If the duration of the pump pulse is fixed at $\tau_p = 500$ fs, all the other parameters will change correspondingly. We know the amplitudes of the detuning pulse and the Rabi frequency with moderate value of $\Delta_0 = \Omega_0 = 60/\tau_p = 0.12 \times 10^{15}$/s can easily satisfy the adiabatic condition. However, the characteristic parameter z currently is $z = d\Omega_0/\mu_{12}\omega_0 \approx 14.5$, which is quite large. Obviously,

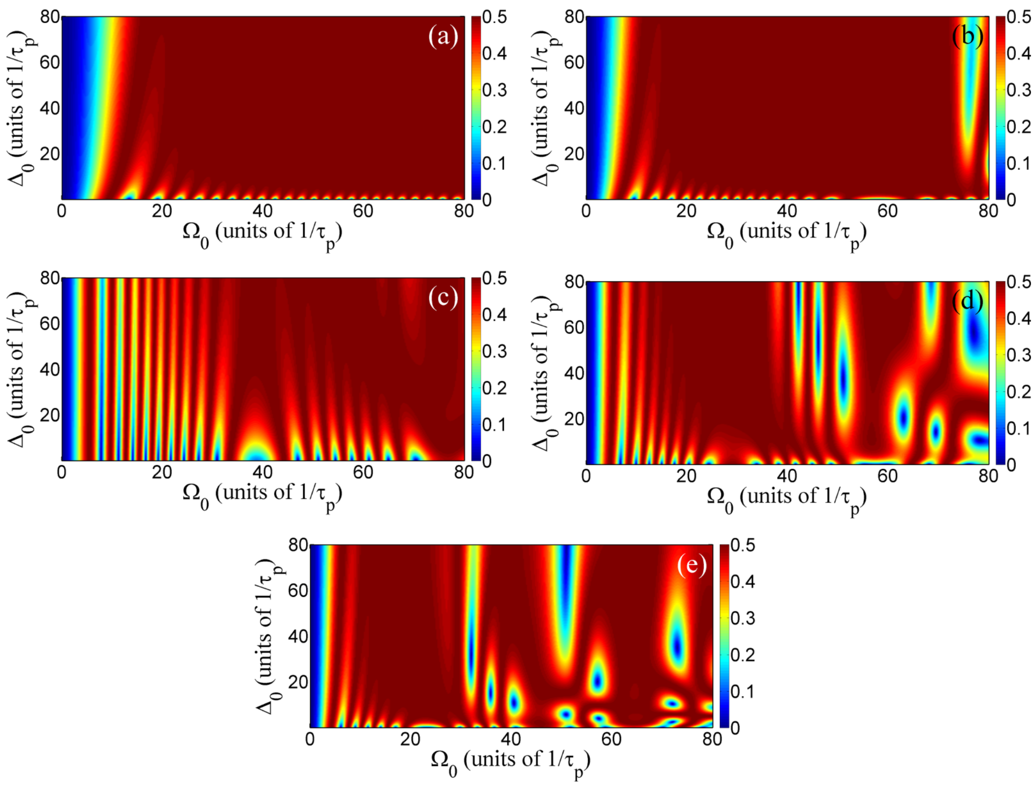

FIG. 8. Effect of the PDMs on the sensitivity of the D-STIRAP to the amplitudes of the detuning pulse and the Rabi frequency under the two-photon excitation. Same as that in Fig. 5, the parameter z also changes proportional to Ω_0, which is $z' = z\Omega_0/60$. The relative time delay in Figs. 8(d) and 8(e) is optimized according to Fig. 7. Parameters in each subfigure are chosen as (a) $z = 2$, $t_p = 2\tau_p$; (b) $z = 4$; $t_p = 2\tau_p$; (c) $z = 6$, $t_p = 2.8\tau_p$; (d) $z = 8$, $t_p = 2.4\tau_p$; and (e) $z = 10$, $t_p = 2.14\tau_p$, respectively.

the parameter z with such high value will destroy the adiabatic process of the D-STIRAP and the robustness of the technique will disappear. Therefore, parameters have to be chosen carefully for the maximum coherence. In order to avoid this problem, we can use pulses with longer durations to reduce the value of z based on the above analysis. If we choose the pump pulse with duration 10 times longer, which is $\tau_p = 5$ ps, we find that the amplitudes of the detuning pulse and the Rabi frequency are reduced to $\Delta_0 = \Omega_0 = 60/\tau_p = 0.012 \times 10^{15}$/s. According to Eq. (1), the adiabatic condition is still satisfied. However, the parameter z is now very small, which is $z \approx 1.45$. Clearly, the parameter z with such small value will not influence the adiabatic process of the D-STIRAP too much and the robustness will be well preserved according to Figs. 3 and 5. Finally, the negative role of the PDMs with giant value of d is avoided successfully. In this example, we should also note that the product of the minimal value of the central frequency and the pump pulse duration is $\tau_p\omega_0^{\min} \approx 354$ and $\tau_p\omega_0^{\min} \approx 4075$ while the pump pulse duration is $\tau_p = 500$ fs and $\tau_p = 5$ ps, respectively. Clearly, the approximation $\tau_p\omega_0 \gg 1$ is satisfied and the transformed Schrödinger equation is valid.

IV. CONCLUSION

In this paper, we have investigated the D-STIRAP in two-level systems possessing PDMs. Our analysis has shown that the D-STIRAP can still be applied safely in this kind of system under the one- and two-photon excitations. For a pump pulse with fixed duration, the PDMs (especially those with giant difference between them) in the two-level system play a negative role in the D-STIRAP by modulating the envelope of the pump pulse, which destroys the adiabatic condition of the D-STIRAP. The robustness of the technique is also influenced simultaneously and pulse parameters need to be chosen with care to obtain the maximum coherence. Nevertheless, we have shown that the negative influence of the PDMs can be effectively avoided by simply elongating the used pulses.

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