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Manipulating field-free molecular alignment by V-shaped femtosecond laser pulses

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We demonstrate theoretically that the shaped femtosecond laser pulse with a V-style spectral phase modulation can provide a well-established tool to control the maximal degree and temporal structure of the field-free molecular alignment. By properly designing these controlling parameters characterizing the V-style spectral phase modulation, we can effectively suppress the maximum degree of molecular alignment or reconstruct as that induced by the transform-limited laser pulse while the temporal structure of the molecular alignment keeps unchanged; we also can control the temporal structure of the molecular alignment with a desired shape and switch the molecular alignment and antialignment at any time delay for any temporal structure.

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I. INTRODUCTION

When a molecule with anisotropic polarizability, such as a linear molecule, is exposed to a strong laser field, an induced dipole moment can be created, which is usually not parallel to the electric field of the laser. The interaction between the induced dipole and the laser field will produce a torque on the molecular axis and tends to align the molecule with the laser polarization [1,2]. Recently, the laser-induced molecular alignment has attracted considerable attention because of its wide applications in chemical reaction dynamics [3–5], high-order-harmonic generation [6,7], surface processing [8], and attosecond science [9,10]. By employing the strong laser pulse, there are two alternative ways to align the molecules that depend on the laser pulse duration and the molecular rotational period. If the laser field is turned on and off slowly as compared to the molecular rotational period, which is termed adiabatic alignment, the molecular alignment proceeds adiabatically and the molecular ensemble will return smoothly to the isotropic angular distribution when the field fades away. If the strong laser pulse is much shorter than the molecular rotational period, which is termed nonadiabatic alignment, a freely evolved rotational wave packet will suffer from a periodic dephasing and rephrasing after the light-molecule interaction and the molecular alignment can be obtained under the free laser field. Compared with the adiabatic alignment, the nonadiabatic alignment has more advantages because of its further applications under field-free conditions.

The ability to control the field-free molecular alignment degree and its dynamical behavior is always an important issue for those who focus on the research of molecular dynamics. With the advent of the ultrafast pulse shaping technique, the shaped laser pulse with the spectral phase modulation has shown to be an effective method to manipulating the molecular alignment. Various shaping schemes, such as cubic phase modulation or periodic phase step modulation, have been utilized to control the maximum degree of the molecular alignment and its temporal structure [11–20]. In this paper we propose a V-style spectral phase modulation to manipulate the maximum degree and temporal structure of the field-free molecular alignment. Our theoretical results show that, by rationally designing the modulation parameters characterizing the V-style phase modulation, the maximum degree of molecular alignment can be effectively suppressed or reconstructed as that induced by the transform-limited laser pulse and the temporal structure of the molecular alignment can be artificially controlled and the molecular alignment and antialignment can be switched at any time delay for any temporal structure.

II. THEORETICAL MODEL

We consider that a linear molecule is subjected to a linearly polarized laser field with the Gaussian distribution $E(t) = E_0 f(t)\cos(\omega_0 t)$, where E_0 is the laser field amplitude, ω_0 is the laser central frequency, and $f(t)$ is the pulse envelope temporal profile with $f(t) = \exp[(-2\ln 2)t^2/\delta_t^2]$, where δ_t is the pulse duration. Thus, the time-dependent Schrödinger equation can be approximated based on the rigid rotor model as [18–20]

$$i\hbar\frac{\partial|\psi(\theta,t)\rangle}{\partial t} = H(t)|\psi(\theta,t)\rangle, \tag{1}$$

with

$$H(t) = BJ(J+1) - \mu E(t)\cos\theta \\ - \tfrac{1}{2}[(\alpha_{||}-\alpha_{\perp})\cos^2\theta + \alpha_{\perp}]E^2(t), \tag{2}$$

where B, J, and μ are, respectively, the rotational constant, the angular momentum, and the permanent dipole moment, θ is the angle between the molecular axis and the laser polarization direction, and $\alpha_{||}$ and $\alpha_{\perp}$ are the polarizability components parallel and perpendicular to the molecular axis, respectively. The first term in Eq. (2) is the molecular rotational energy and the second and third terms are the interaction potentials with the permanent dipole moment and the polarizability, respectively. The degree of the molecular alignment is characterized by the expectation value of $\cos^2\theta$ (i.e., $\langle\cos^2\theta\rangle$). Considering the thermal equilibrium of the molecular ensemble, $\langle\cos^2\theta\rangle$ is averaged over the Boltzmann

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distribution and is written as

$$\langle\cos^2\theta\rangle = \sum_J Q^{-1} g_J \exp[-BJ(J+1)/kT] \times \sum_{M=-J}^{J} \langle\Psi_{JM}|\cos^2\theta|\Psi_{JM}\rangle, \quad (3)$$

where Q is the partition function, g_J is the spin degeneracy factor, and Ψ_{JM} is the rotational wave function of the time-evolved molecules.

III. RESULTS AND DISCUSSION

In our theoretical simulation, the time-dependent Schrödinger equation in Eq. (1) is numerically solved by a split-operator method [21,22]. The CO molecule is used as our study example and the molecular parameters are set as $B = 1.93\ \text{cm}^{-1}$, $\mu = 0.112$ D, $\alpha_{\|} = 2.294\ \text{Å}^3$, and $\alpha_{\perp} = 1.77\ \text{Å}^3$ [23–25]. Thus, the rotational period of the CO molecule can be calculated as $T_{\text{rot}} = 1/2Bc \approx 8.64$ ps, where c is the speed of the light in vacuum. The molecular rotational temperature is set to be 30 K. The laser parameters are set as follows: the laser central frequency is $\omega_0 = 12\,500\ \text{cm}^{-1}$, the pulse duration (full width at half maximum) is $\delta_t = 200$ fs, and the laser intensity is $I = 1 \times 10^{13}\ \text{W/cm}^2$.

The femtosecond laser pulse shaped by modulating the laser spectral phase has been widely employed to control the molecular alignment, such as cubic phase modulation or periodic phase step modulation [11–20]. Here we utilize a V-style phase modulation to control the molecular alignment. The V-style phase modulation can be defined by the function of $\Phi(\omega) = \tau|\omega - \omega_0 - \delta\omega|$, where τ and $\delta\omega$ represent the modulation depth and modulation position, respectively. Thus, the laser pulse shaped by V-style phase modulation in the frequency domain $E_{\text{V}}(\omega)$ can be written as $E_{\text{V}}(\omega) = E(\omega)\exp[i\Phi(\omega)]$, where $E(\omega)$ is the Fourier transform of the unshaped laser field $E(t)$ and the shaped laser pulse in time domain $E_{\text{V}}(t)$ is given by the reverse Fourier transform of $E_{\text{V}}(\omega)$. The V-style spectral phase modulation in the frequency domain is shown in Fig. 1(a) and the temporal intensity profiles in the time domain with different modulation parameters are shown in Fig. 1(b), together with the unshaped femtosecond laser pulse. One can see from Fig. 1(b) that the V-style phase modulation will lead to the formation of two time-delayed subpulses, where the modulation depth τ determines the time separation Δt of the two subpulses with $\Delta t = 2\tau$ and the modulation position $\delta\omega$ determines the relative intensity ratio between the two subpulses. By the V-style spectral phase modulation, the single laser pulse can be divided into two subpulses with controllable time delay and relative intensity ratio while the total laser energy remains unchanged, which is useful to control the maximal degree and temporal structure of the molecular alignment and can avoid the intrinsic saturation and ionization in the single laser pulse with high intensity.

Figure 2(a) shows the maximal degree of the field-free molecular alignment $\langle\cos^2\theta\rangle_{\max}$ as a function of the modulation depth τ for the modulation position $\delta\omega = 0$. As can be seen, the molecular alignment degree is completely suppressed at the modulation depth $\tau = 2.16$ ps, corresponding the time separation of the two subpulses of $\Delta t = 4.32$ ps, which is equal to the half of the molecular rotational period. This result is the same as that obtained by two time-delayed laser pulses [26,27]. However, when the modulation depth is $\tau = 4.32$ ps, corresponding to a time separation of the two subpulses of $\Delta t = 8.64$ ps, which is equal to the whole the molecular rotational period, the molecular alignment degree does not recover to that induced by the transform-limited laser pulse, which is different from that induced by the two time-delayed laser pulses [28]. The fundamental reason is that two subpulses by the V-style phase modulation are broadened and therefore their laser intensities will decrease [see Fig. 1(b)]. Figure 2(b)

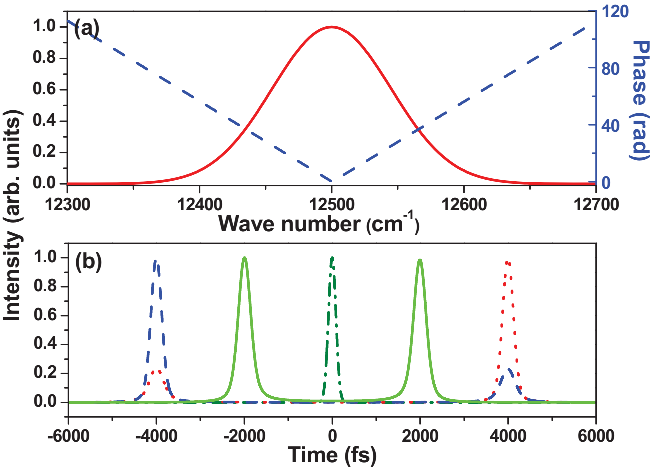

FIG. 1. (Color online) (a) Schematic diagram of the 200-fs laser spectrum (red solid line) and the V-style spectral phase modulation $\Phi(\omega) = \tau|\omega - \omega_0 - \delta\omega|$ with $\tau = 2000$ fs and $\delta\omega = 0$ (blue dashed line). (b) Temporal intensity profile of the unshaped laser pulse (olive dash-dotted line) and the shaped laser pulse by the V-style spectral phase modulation with $\tau = 2000$ fs and $\delta\omega = 0$ (green solid line), together with $\tau = 4000$ fs and $\delta\omega = 20$ (red dotted line) and $-20\ \text{cm}^{-1}$ (blue dashed line).

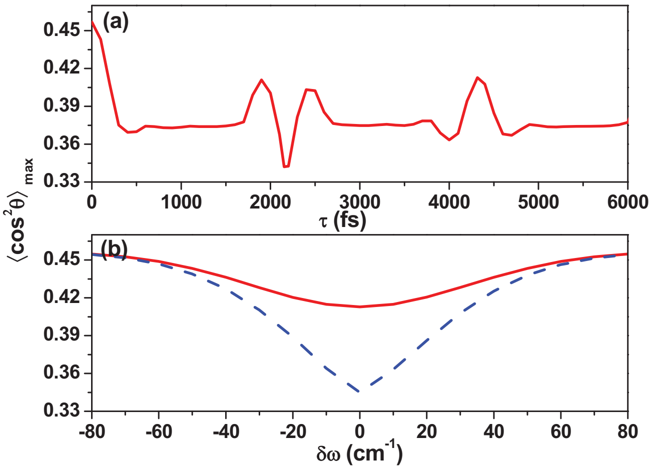

FIG. 2. (Color online) Maximal degree of the field-free molecular alignment $\langle\cos^2\theta\rangle_{\max}$ as a function of (a) the modulation depth τ with the modulation position $\delta\omega = 0\ \text{cm}^{-1}$ and (b) the modulation position $\delta\omega$ with the modulation depths $\tau = 4.32$ (red solid line) and 2.16 ps (blue dashed line).

shows the maximal degree of the field-free molecular alignment $\langle\cos^2\theta\rangle_{\max}$ as a function of the modulation position $\delta\omega$ for the modulation depths $\tau = 2.16$ (red solid line) and 4.32 ps (blue dashed line). One can see that, by varying the modulation position $\delta\omega$, the molecular alignment degree can recover to that induced by the transform-limited laser pulse, which is due to the fact that the large modulation position $\delta\omega$ will make one of the two time-delayed subpulses disappear [see Fig. 1(b)]. Furthermore, it is noticed that the positive or negative modulation position $\delta\omega$ with the same absolute value has the same control effect on the molecular alignment degree, that is to say, the same relative intensity ratio between the two subpulses does not affect the molecular alignment degree.

The control of the maximal degree of the field-free molecular alignment by the V-style spectral phase modulation can be intuitively understood by a train of kicks; this method has been successfully utilized to interpret the enhancement or suppression of the molecular alignment or orientation excited by the multiple laser pulses [29–33]. Two time-delayed subpulses are formed by the V-style spectral phase modulation. The first subpulse creates a coherent rotational wave packet and the field-free evolution of the rotational wave packet results in periodic revivals of molecular alignment and antialignment. When the second subpulse is applied at full of the molecular rotational period, the rotational wave packet induced by the first subpulse evolves from the molecular antialignment to alignment, thus the second subpulse produces a kick force to the rotational wave packet and therefore the molecular alignment is enhanced. However, if the second subpulse is sent at the half of the molecular rotational period, it provides an antikick force since the rotational wave packet created by the first subpulse evolves from the molecular alignment to antialignment and consequently suppresses the molecular alignment.

Furthermore, the physical control mechanism of the maximal degree of molecular alignment by the V-style spectral phase modulation can also be explained by the impulsive Raman transition probability [34]. The Raman transition probability from the molecular rotational states $|J\rangle$ to $|J+2\rangle$ can be approximated by the second-order perturbation theory as [12,35]

$$P_{J-J+2} \propto \left| \int_{-\infty}^{+\infty} E_{\mathrm{V}}(\omega) E_{\mathrm{V}}^{*}(\omega-\omega_J)\, d\omega \right|^2, \qquad (4)$$

where ω_J is the Raman frequency with $\omega_J = 2\pi(E_{J+2} - E_J)/h = (4J+6)\pi/T_{\mathrm{rot}}$. Considering the molecular ensembles in thermal equilibrium, the total Raman transition probability P is given by

$$P \propto \sum_J \{Q^{-1} g_J \exp[-BJ(J+1)/kT]\}^2$$
$$\times \left| \int_{-\infty}^{+\infty} E_{\mathrm{V}}(\omega) E_{\mathrm{V}}^{*}(\omega-\omega_J)\, d\omega \right|^2. \qquad (5)$$

Figure 3 presents the Raman transition probability P (green solid lines) as a function of the modulation depth τ with the modulation position $\delta\omega = 0$ cm^{-1} [Fig. 3(a)] and the modulation position $\delta\omega$ with the modulation depth $\tau = 2.16$ ps [Fig. 3(b)]. By comparing Figs. 2 and 3, one can see that the Raman transition probability P has the same evolution behavior as the maximal degree of the molecular alignment. The Raman transition probability P is completely suppressed at the modulation depth $\tau = 2.16$ ps and reconstructed as that by the transform-limited laser pulse for the larger modulation position $\delta\omega$. Consequently, it can be concluded that the change of the Raman transition probability in these molecular rotational states result in the control of the molecular alignment degree.

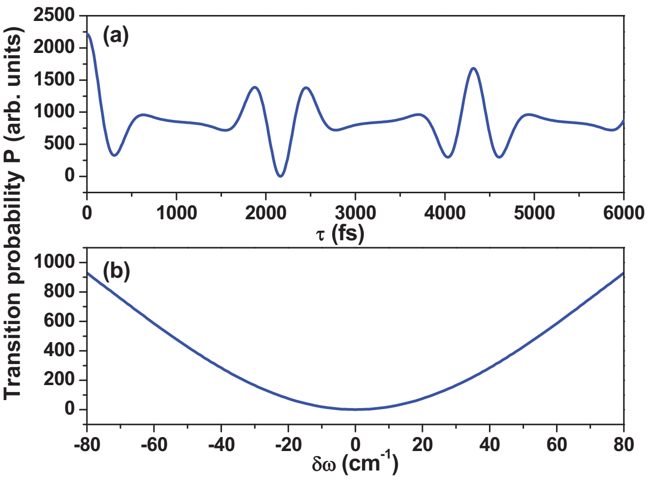

FIG. 3. (Color online) Raman transition probability P as a function of (a) the modulation depth τ with the modulation position $\delta\omega = 0$ cm^{-1} and (b) the modulation position $\delta\omega$ with the modulation depth $\tau = 2.16$ ps.

As shown in Fig. 2, by controlling the modulation depth τ and the modulation position $\delta\omega$, the maximum degree of the molecular alignment can be effectively controlled. Next, we further demonstrate the control effect of these modulation parameters on the temporal structure of the molecular alignment. Figure 4 shows the time-dependent molecular alignment $\langle\cos^2\theta\rangle$ by the V-style spectral phase modulation with the

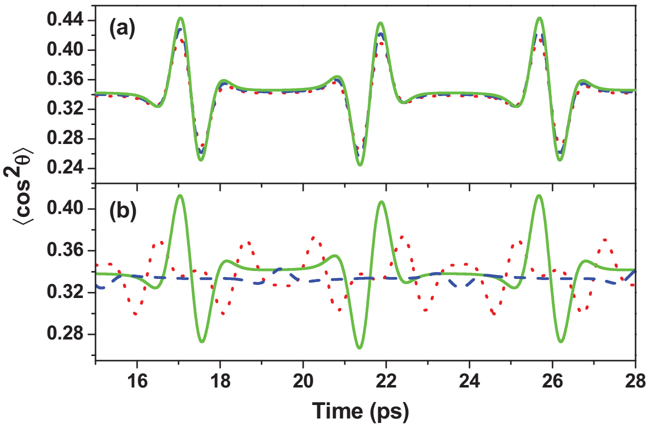

FIG. 4. (Color online) Time-dependent molecular alignment $\langle\cos^2\theta\rangle$ by the V-style spectral phase modulation with (a) the modulation depth $\tau = 4.32$ ps for the modulation positions $\delta\omega =$ 10 (red dotted line), 30 (blue dashed line), and 50 cm^{-1} (green solid line) and (b) the modulation position $\delta\omega = 0$ cm^{-1} for the modulation depths $\tau = 1.08$ (red dotted line), 2.16 (blue dashed line), and 4.32 ps (green solid line).

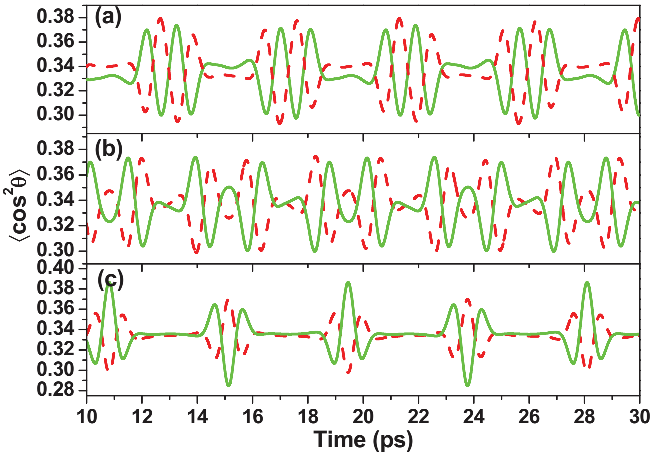

FIG. 5. (Color online) Time-dependent molecular alignment $\langle\cos^2\theta\rangle$ by the V-style spectral phase modulation with the modulation position $\delta\omega = 0$ cm^{-1} for the modulation depths (a) $\tau = 0.568$, (b) $\tau = 1.238$, and (c) $\tau = 2.050$ ps (green solid lines), together with (a) $\tau = 3.749$, (b) $\tau = 3.079$, and (c) $\tau = 2.267$ ps (red dashed lines).

modulation depth $\tau = 4.32$ ps for the modulation positions $\delta\omega = 10$ (red dotted line), 30 (blue dashed line), and 50 cm^{-1} (green solid line) [Fig. 4(a)] and the modulation position $\delta\omega = 0$ cm^{-1} for the modulation depths $\tau = 1.08$ (red dotted line), 2.16 (blue dashed line), and 4.32 ps (green solid line) [Fig. 4(b)]. It can be seen from Fig. 4(a) that the modulation position $\delta\omega$ will vary the maximal degree of the molecular alignment degree but does not affect its temporal structure and the larger absolute value of the modulation position $|\delta\omega|$ yields the higher molecular alignment degree, which indicates that one can continuously adjust the molecular alignment degree while keeping its temporal structure unchanged. On the other hand, as can be seen in Fig. 4(b), by varying the modulation depth τ, both the maximal degree and temporal structure of the molecular alignment can be controlled, which is different from that obtained by varying the modulation position $\delta\omega$, where only the maximal degree of the molecular alignment is varied. Since the modulation depth τ is to control the time separation of the two subpulses, the variation of the temporal structure of the molecular alignment can be well illustrated by analyzing the two time-delayed subpulses. These results indicate that, by properly designing the modulation depth τ and the modulation position $\delta\omega$ of the V-style spectral phase modulation, one can control both the maximal degree and temporal structure of the molecular alignment [see Fig. 4(b)] or only the maximal degree while the temporal structure remains unchanged [see Fig. 4(a)].

Since the temporal structure of the molecular alignment can be controlled by varying the modulation depth τ, as shown in Fig. 4(b), we further analyze these temporal structures of the molecular alignment and their relationship. Figure 5 presents the time-dependent molecular alignment $\langle\cos^2\theta\rangle$ by the V-style spectral phase modulation with the modulation position $\delta\omega = 0$ for the modulation depths $\tau = 0.568$ [Fig. 5(a)], 1.238 [Fig. 5(b)], and 2.050 ps [Fig. 5(c)] (green solid lines), together with $\tau = 3.749$ [Fig. 5(a)], 3.079 [Fig. 5(b)], and 2.267 ps [Fig. 5(c)] (red dashed lines). By varying the modulation depth τ, one can obtain the temporal structure of the molecular alignment with a desired shape, such as one alignment (or antialignment) and two antialignments (or alignments) [see Fig. 5(c)] or two alignments (or antialignments) and two antialignments (or alignments) [see Figs. 5(a) and 5(b)]. Furthermore, by comparing the solid and dashed lines in Fig. 5, it is noteworthy that the molecular alignments and antialignments can be switched at any time delay for any temporal structure by precisely designing the modulation depths τ_1 and τ_2. Our calculated results find that the two modulation depths τ_1 and τ_2 satisfy the relation of $\tau_1 + \tau_2 = T_{\mathrm{rot}}/2$. Considering the relation of the modulation depth τ and the time separation Δt of two subpulses with $\Delta t = 2\tau$, the two subpulse separations Δt_1 and Δt_2 exist in the relation of $\Delta t_1 + \Delta t_2 = T_{\mathrm{rot}}$. That is to say, the molecular dynamical behavior induced by the two subpulses with the time separation Δt_1 is always opposite to that induced by the two subpulses with the time separation $T_{\mathrm{rot}} - \Delta t_1$.

Finally, we simply analyze the feasibility of our proposed scheme in the experiment. In our scheme, the crucial question is how to get the shaped femtosecond laser pulse by the V-style phase modulation. With the advent of the ultrafast pulse shaping technique, the shaped femtosecond laser pulse with almost arbitrary temporal pulse profile can be obtained by controlling the laser spectral phase and/or amplitude [36]. The programmable $4f$-configuration zero-dispersion pulse shaper combined with a one-dimensional liquid-crystal spatial light modulator has been proven to be a well-established setup to manipulate the femtosecond laser pulse. Recently, the shaped femtosecond laser pulse by the V-style phase modulation has been successfully achieved and applied in the control of the molecular absorption and laser manufacturing [37–39].

IV. CONCLUSION

We have presented a scheme to control the field-free molecular alignment by shaping the femtosecond laser pulse with a V-style spectral phase modulation, involving the maximum degree and the temporal structure of the molecular alignment. Our results showed that, by controlling the modulation depth and modulation position, i.e., the time separation and relative intensity ratio of the two subpulses, the maximum degree of the molecular alignment can be effectively suppressed and reconstructed as that by the transform-limited laser pulse, the temporal structure of the molecular can be controlled with a desired shape, and the molecular alignment or antialignment can be switched at any time delay for any temporal structure. These theoretical results provide an ideal method to understand and control the maximal degree and temporal structure of the molecular alignment and will have potential applications in various related fields.

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