PHYSICAL REVIEW A **83**, 043410 (2011)

Controlling field-free molecular orientation with combined single- and dual-color laser pulses

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(Received 6 January 2011; published 19 April 2011)

We propose a scheme to achieve the field-free molecular orientation around the half rotational periods with the combination of single- and dual-color laser pulses. We show that the molecular orientation can be obtained and controlled by precisely controlling the time delay between the two laser pulses. Furthermore, we discuss the effect of the laser intensity and pulse duration of the single-color laser pulse on the molecular orientation created by the dual-color laser pulse, and show that the molecular orientation depends on the change in the alignment degree between the odd and even rotational wave-packet contributions created by the single-color laser pulse.

DOI: 10.1103/PhysRevA.83.043410 PACS number(s): 33.80.Wz, 33.15.Bh

Molecular alignment and orientation have attracted both physicists' and chemists' interest because of their extensive applications in chemical reaction dynamics [1–3], high-harmonic generation [4,5], surface processing [6], and attosecond science [7]. An intense linearly polarized laser field has been shown to be the most effective method to obtain the molecular alignment and orientation [8–28]. For example, the molecular alignment can be realized with the single-color laser pulse [8–17], and the molecular orientation can be achieved with the dual-color laser pulse [18–21], the half-cycle laser pulse [22–25], or the single-color laser pulse combined with a weak dc field [26–28]. The laser-induced molecular alignment and orientation can be obtained in both nonadiabatic and adiabatic regimes. Especially, the nonadiabatic alignment and orientation can be used to produce aligned and oriented molecules under field-free field conditions, and so are desirable for further applications in various related fields. Usually, the degrees of the molecular alignment and orientation are limited by the maximally applicable laser intensity before its intrinsic saturation and ionization. Recently, various techniques have been proposed to further enhance the molecular alignment and orientation, such as the laser-pulse trains, with the pulse separations being commensurate with the rotational period [12–15,19], the slow turn-on and rapid turn-off laser pulse [20,27,28], the shaped laser pulse [16,17], or the hybrid laser pulse [29].

The molecular orientation can be repeatedly obtained around the full rotational periods by applying the short two-color laser pulse [18–21]. However, the odd and even wave-packet contributions around the half rotational periods cancel each other out, and so no molecular orientation for the total wave-packet contribution at these time delays is observed. In this paper, we present a scheme to obtain the molecular orientation around the half rotational periods that even exceeds that obtained around the full rotational periods. We employ two time-delayed laser pulses with the combination of the single- and dual-color laser pulses, and show that the molecular orientation around the half rotational periods can be obtained and controlled by varying the time delay between the two laser pulses. We discuss the dependence of the molecular orientation around the half rotational periods created by the dual-color laser pulse on the laser intensity and pulse duration of the single-color laser pulse, and illustrate that the molecular orientation depends on the change in the alignment degree between the odd and even rotational wave-packet contributions created by the single-color laser pulse.

We consider a hybrid laser pulse with the combination of single- and dual-color laser pulses $E(t) = A_s f_s(t)\cos(\omega_s t) + A_d f_d(t - T_d)\{\cos[\omega_d(t - T_d)] + \cos[2\omega_d(t - T_d)]\}$, where A_s and A_d are the field amplitude, $f_s(t)$ and $f_d(t - T_d)$ are the pulse envelope with $f(t) = \exp(-2\ln 2t^2/\tau^2)$, τ is the pulse duration, and T_d is the time delay between the single- and dual-color laser pulses. When a linear molecule is exposed to the hybrid laser pulse, the time-dependent Schrödinger equation can be expressed based on the rigid-rotor model as [20] $i\hbar\partial\psi(\theta,t)/\partial t = [H_0 + V_\mu(\theta)E(t) + V_{\mathrm{pol}}(\theta)E^2(t) + V_{\mathrm{hyp}}(\theta)E^3(t)]\psi(\theta,t)$ with $H_0 = BJ_i(J_i+1), V_\mu(\theta) = -\mu\cos\theta, V_{\mathrm{pol}}(\theta) = -[(\alpha_{||} - \alpha_\perp)\cos^2\theta + \alpha_\perp]/2$, and $V_{\mathrm{hyp}}(\theta) = -[(\beta_{||} - 3\beta_\perp)\cos^3\theta + 3\beta_\perp]/6$, where B is the rotational constant, J_i is the angular momentum, μ is the permanent dipole moment, θ is the angle between the molecular axis and the laser polarization, $\alpha_{||}$ and $\alpha_\perp$ are the polarizability components parallel and perpendicular to the molecular axis, and $\beta_{||}$ and $\beta_\perp$ are hyperpolarizability components parallel and perpendicular to the molecular axis. Finally, the degrees of the molecular alignment and orientation are, respectively, given with the expectation value of $\langle\cos^2\theta\rangle$ and $\langle\cos\theta\rangle$ by considering the temperature-dependent Boltzmann distribution of initial rotational states. We use the CO molecule as an example, and the molecular parameters are $B = 1.93\ \mathrm{cm}^{-1}$, $\mu = 0.112$ D, $\alpha_{||} = 2.294\ \text{Å}^3$, $\alpha_\perp = 1.77\ \text{Å}^3$, $\beta_{||} = 2.748 \times 10^9\ \text{Å}^5$, and $\beta_\perp = 4.994 \times 10^8\ \text{Å}^5$ [20,30,31]. Thus, the rotational period of the CO molecule can be calculated as $T_{\mathrm{rot}} = 1/(2Bc) \approx 8.64$ ps, where c is the speed of light. The initial rotational temperature of the CO molecule is set to be 30 K. The center frequencies of the single- and dual-color laser pulse ω_s and ω_d are both set to be 12 500 cm^{-1}, corresponding to the laser center wavelength of 800 nm.

We first show the field-free molecular orientation $\langle\cos\theta\rangle$ only created by the dual-color laser pulse, and the calculated result is depicted in Fig. 1 with the laser intensity of $1 \times 10^{13}\ \mathrm{W/cm}^2$ and the pulse duration of 200 fs, together with the odd (red dashed line) and even (blue dotted line) rotational

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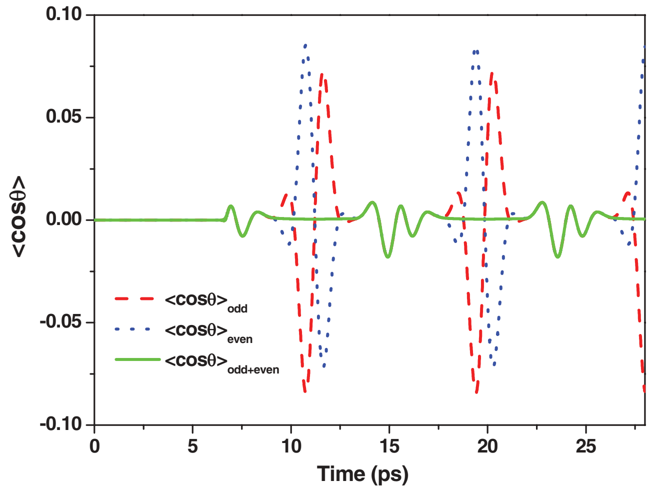

FIG. 1. (Color online) Time-dependent molecular orientation $\langle\cos\theta\rangle$ created by the two-color laser pulse with the laser intensity of 1×10^{13} W/cm^2 and the pulse duration of 200 fs, together with the odd (red dashed line) and even (blue dotted line) rotational wave-packet contributions.

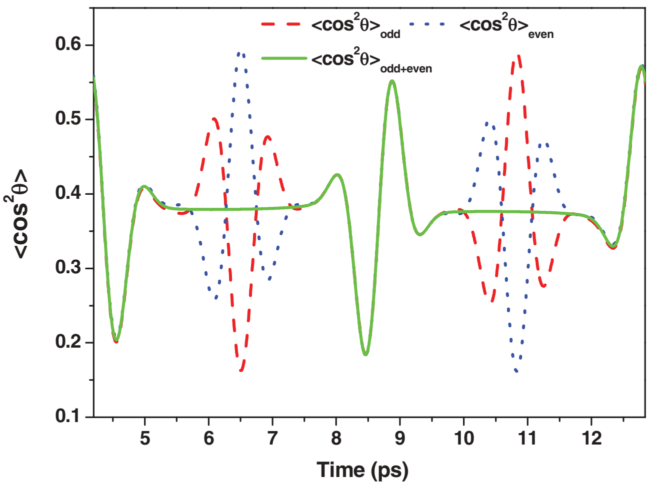

FIG. 2. (Color online) Time-dependent molecular alignment $\langle\cos^2\theta\rangle$ created by the single-color laser pulse with the laser intensity of 2×10^{13} W/cm^2 and the pulse duration of 200 fs, together with the odd (red dashed line) and even (blue dotted line) rotational wave-packet contributions.

wave-packet contributions. The odd and even rotational wave-packet contributions exhibit opposite behavior around the half rotational periods (i.e., $t = nT_{\rm rot}/2$, where n is integer), while showing the same behavior around the full rotational periods (i.e., $t = nT_{\rm rot}$). Since the field-free molecular orientation is the result of the interference between the odd and even rotational wave-packet contributions, the two contributions will cancel each other out around the half rotational periods and interfere instructively around the full rotational periods; therefore, the molecular orientation for the total contribution is only observed around the full rotational periods. However, the orientation degrees for both the odd and even rotational wave-packet contributions around the half rotational periods are much larger than those around the full rotational periods. If the relative excitation between the two contributions around the half rotational periods can be manipulated, the molecular orientation for the total contribution at these time delays can be obtained, and even exceeds that obtained around the full rotational periods. Our primary goal is to obtain the molecular orientation around the half rotational periods by controlling the relative excitation between the odd and even rotation wave-packet contributions.

An intense nonresonant single-color laser pulse has been proven to be a well-established method to create the molecular alignment, but it can not create the molecular orientation. Here, we employ the single-color laser pulse to create the molecular alignment, which is used to control the relative excitation between the odd and even rotational wave-packet contributions when the time-delayed dual-color laser pulse is applied, and finally obtain the molecular orientation around the half rotational periods. Figure 2 shows the time-dependent molecular alignment $\langle\cos^2\theta\rangle$ only created by the single-color laser pulse with the laser intensity of 2×10^{13} W/cm^2 and the pulse duration of 200 fs, together with the odd (red dashed line) and even (blue dotted line) rotational wave-packet contributions. The odd and even rotational wave-packet contributions exhibit opposite behavior around ($2n$-1)/4 rotational periods (i.e., $t = (2n-1)T_{\rm rot}/4$) and, therefore, no alignment for the total contribution is observed. However, both the odd and even rotational wave-packet contributions have the same behavior around $n/2$ rotational periods (i.e., $t = nT_{\rm rot}/2$), and, thus, the molecular alignment for the total contribution can be obtained. When the aligned molecules are subjected to the dual-color laser pulse, the alignment behavior for the odd and even rotational wave-packet contributions will produce a significant effect on the molecular orientation. If the dual-color laser pulse is set at the time delay $T_d = 6.5$ ps (or 10.8 ps), the odd rotational wave-packet contribution has a minimum (or maximum) alignment that suppresses (or promotes) the molecular orientation, while the even rotational wave-packet contribution has a maximum (or minimum) alignment that promotes (or suppresses) the molecular orientation, and the molecular orientation around the half rotational periods can be obtained. If the two-color laser pulse is set at $T_d = 8.4$ ps (or 8.8 ps), both the odd and even rotational wave packets have a minimum (or maximum) alignment that simultaneously suppresses (or promotes) the molecular orientation, and the molecular orientation around the half rotational periods still does not occur.

To validate the above predications, we calculate the time-dependent molecular orientation $\langle\cos\theta\rangle$ when the dual-color laser pulse is set at $T_d = 6.5$, 10.8, 8.4, and 8.8 ps, and the calculated results are presented in Fig. 3, together with the odd (red dashed line) and even (blue dotted line) rotational wave-packet contributions. When the dual-color laser pulse is applied at $T_d = 6.5$ ps (or 10.8 ps) [see Figs. 3(a) and 3(b)], the orientation degree for the odd rotational wave-packet contribution decreases (or increases), while that for the even rotational wave-packet contribution increases (or decreases), and therefore the molecular orientation around the half rotational periods is obtained. When the dual-color laser pulse is applied at $T_d = 8.4$ ps (or 8.8 ps) [see Figs. 3(c) and 3(d)], the orientation degrees for both the odd and even rotational wave-packet contributions decrease (or increase)

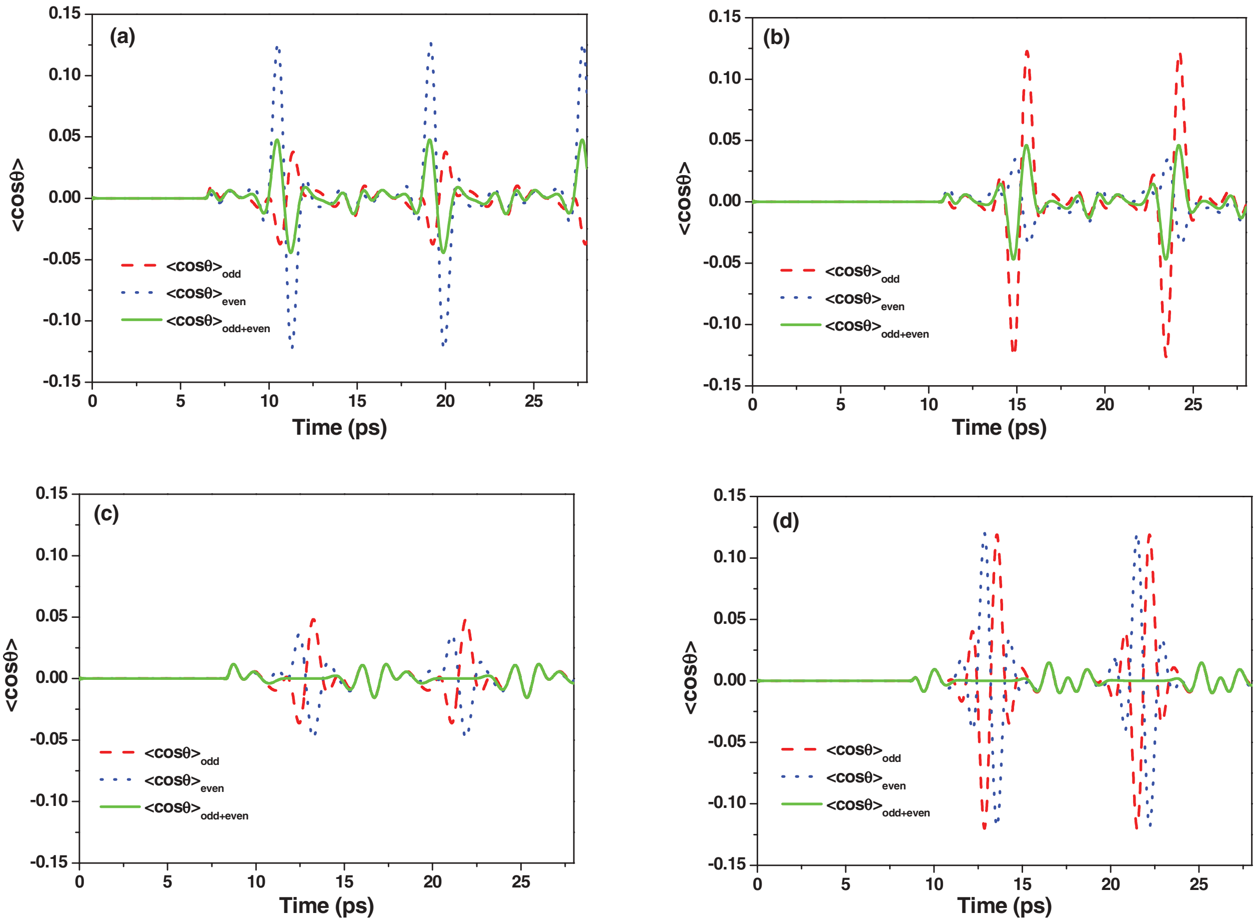

FIG. 3. (Color online) Time-dependent molecular orientation $\langle\cos\theta\rangle$ created by the combined single- and dual-color laser pulses with the pulse separation of (a) $Td = 6.5$, (b) 10.8, (c) 8.4, and (d) 8.8 ps, together with odd (red dashed line) and even (blue dotted line) rotational wave-packet contributions.

and, thus, the molecular orientation around the half rotational periods is still not obtained. It is obvious that these observations are consistent with our above expectations.

As compared with the single dual-color laser pulse (see Fig. 1), the degree of the molecular orientation induced by the combined single- and dual-color laser pulses can be significantly enhanced, reaching by a factor of ∼2.9 [see Figs. 3(a) and 3(b)]. Therefore, it can provide an effective method to further enhance the field-free molecular orientation. To demonstrate the advantage of our scheme, we compare it with a previous scheme using the two dual-color laser pulses [19] and find that a higher degree of the molecular orientation is obtained by our scheme under the same laser intensity (not shown here). Note that the maximal degree of the molecular orientation does not occur at the same time for the two schemes. This result indicates that our scheme is more effective in enhancing the molecular orientation.

As shown in Fig. 3, the molecular orientation around the half rotational periods can be obtained by the combined single- and dual-color laser pulses. Next, we study the effect of the laser intensity and the pulse duration of the single-color laser pulse on the molecular orientation created by the dual-color laser pulse. Figure 4 shows the maximum degrees of the positive (red squares) and negative (blue circles) orientation $\langle\cos\theta\rangle_{\max}$ around the half rotational periods by varying (a) the laser intensity and (b) the pulse duration of the single-color laser pulse; here the dual-color laser pulse is set at $T_d = 6.5$ ps. As can be seen, the molecular orientation increases with the increase of the laser intensity before its intrinsic saturation and ionization. However, with the increase of the pulse duration, the molecular orientation first increases and then decreases, and the optimal pulse duration is about 250 fs. The optimal pulse duration is correlated with the initial rotational temperature, which will increase with the increase of the rotational temperature. Obviously, by optimizing the laser intensity and pulse duration of the single-color laser pulse, the molecular orientation around the half rotational periods created by the dual-color laser pulse can be significantly improved.

To explore the physical origin that the molecular orientation is obtained around the half rotational periods by the combined single- and dual-color laser pulses, we calculate the alignment degrees for both the odd and even rotational wave-packet contributions at the time delay $T_d = 6.5$ ps and their difference by varying the laser intensity and pulse duration of the single-color laser pulse (the calculated

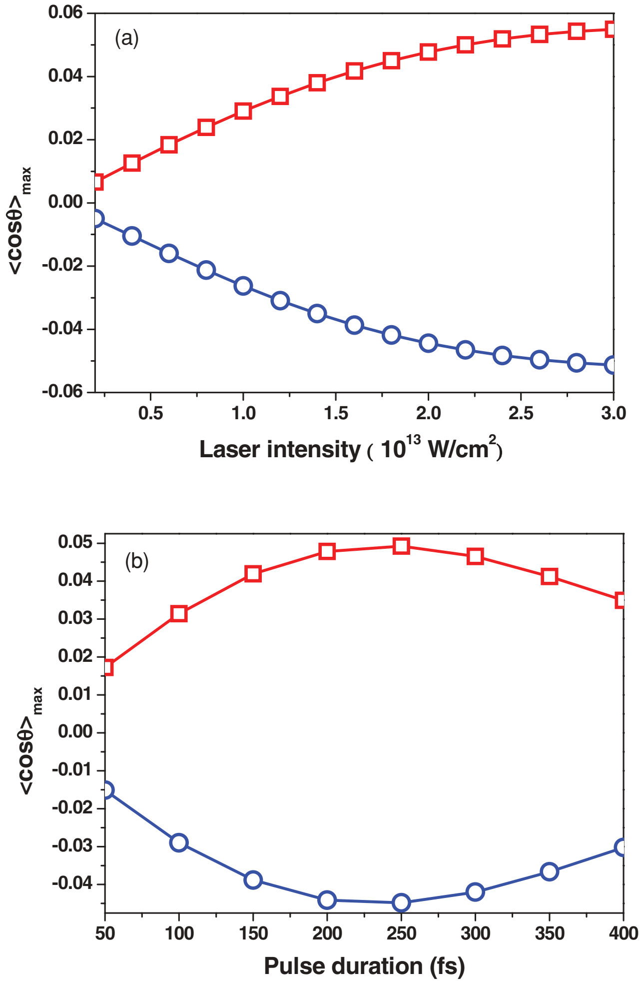

FIG. 4. (Color online) Maximum degrees of the positive (red squares) and negative (blue circles) orientation $\langle \cos\theta \rangle_{\max}$ around the half rotational periods by varying (a) the laser intensity and (b) the pulse duration of the single-color laser pulse; here the dual-color laser pulse is set at the time delay $T_d = 6.5$ ps.

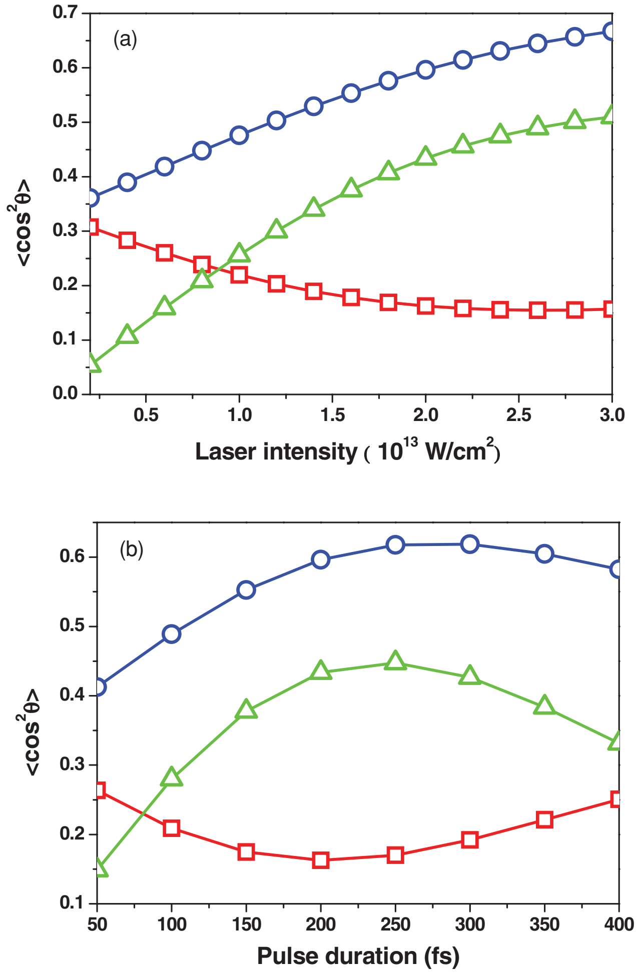

FIG. 5. (Color online) Alignment degrees $\langle \cos^2\theta \rangle$ for the odd (red squares) and even (blue circles) wave-packet contributions at the time delay $T_d = 6.5$ ps by varying (a) the laser intensity and (b) the pulse duration of the single-color laser pulse. The green triangles are used to show the change in the alignment degree between the odd and even rotational wave-packet contributions.

results are shown in Fig. 5). Comparing with Fig. 4, it can be found that the molecular orientation around the half rotational periods should depend on the change in the alignment degree between the odd and even rotational wave-packet contributions but not the alignment degree for odd or even rotational wave-packet contribution. With the increase of the change in the alignment degree between the odd and even wave-packet contributions, the molecular orientation around the half rotational periods will increase. This conclusion can also be further confirmed in Fig. 3, where the change in the alignment degree is zero and the molecular orientation around the half rotational periods will disappear.

In conclusion, we have theoretically shown that the molecular orientation around the half rotational periods can be obtained with the combined single- and dual-color laser pulses. The molecular alignment created by the first single-color laser pulse was used to control the relative excitation between the odd and even wave-packet contributions when the second dual-color laser pulse was applied, and, thus, the molecular orientation around the half rotational periods can be obtained and controlled by precisely controlling the time delay between the two laser pulses. Moreover, the effect of the laser intensity and pulse duration of the single-color laser pulse on the molecular orientation created by the dual-color laser pulse was discussed, and the change in the alignment degree between the odd and even rotational wave-packet contributions created by the single-color laser pulse was considered as the decisive factor for the achievement of the molecular orientation around the half rotational periods.

This work was partly supported by Ministry of Education of China (Grant No. 30800), National Natural Science Fund (Grants No. 11004060 and No. 11027403), and Shanghai Municipal Science and Technology Commission (Grants No. 10XD1401800, No. 09142200501, No. 09ZR1409300, No. 09JC1404700, and No. 10JC1404500).

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