Selective excitation of the molecular rotational wave packet by two time-delayed laser pulses*

Xu Shu-Wu(徐淑武), Huang Yun-Xia(黄云霞), and Ji Xian-Ming(纪宪明)†

Xinglin College, Nantong University, Nantong 226007, *China*

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In this paper, we investigate the control of the molecular wave packet of a linear molecule by two femtosecond laser pulses. It is shown that the odd and the even rotational wave packets created by a single laser pulse can be selectively excited by accurately controlling the time delay of another laser pulse. By inserting the peak of the second laser pulse at the position of maximum or minimum value around quarter or three quarter rotational period of the slope curve with odd (or even) rotational wave packet contribution that is created by the first laser pulse, the odd rotational wave packet can be enhanced (or suppressed) while the even rotational wave packet is suppressed (or enhanced). As a result, the molecular alignments around quarter and three quarter rotational periods can be obtained. Moreover, it is also shown that by inserting the second laser pulse around the quarter or three quarter rotational periods, the changes in the maximum degree of the molecular alignment for the odd and the even rotational wave packet contributions are consistent with their corresponding slope curves at these positions.

Keywords: molecular rotational wave packet, selective excitation, slope curve

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1. Introduction

Subject to the action of a femtosecond laser field with a strong intensity, a molecule will be aligned along the polarization direction of the laser pulse by the torque generated by the interaction between the laser field and the induced dipole moment of the molecule.[1−3] Molecular alignment plays an important role in the study of chemical reaction dynamics,[4−7] multiphoton ionization[8,9] and high-order harmonic generation.[10−12] Generally, there are two ways to align the molecule. If the duration of the laser pulse is much longer than the rotational period of the molecule, the molecule is adiabatically aligned. Oppositely, if the width of the laser pulse is much shorter than the rotational period of the molecule, it is called nonadiabatic alignment. In the latter case, the field-free molecular alignment can be obtained due to periodic dephasing and rephasing of the rotational wave packet, and therefore it has attracted widespread attention for its further potential applications.[13]

During the evolution of the rotational wave packet in the field-free molecular alignment, the odd and the even components will show different dynamic behaviours. As a result, molecular alignment in one rotational period due to the interference of the wave packets is often distinct from the odd or even component contribution. The molecular rotational wave packet created by a laser pulse can be modified by applying the second laser pulse or trains of laser pulses.[14−23] Especially, Gong Q H *et al.* have demonstrated that by accurately controlling the delay time between the two femtosecond laser pulses, the alignment structures,[17] the coherent rotational wave packet,[18] wave packet interference induced rotational population and the revival structures[19−21] can be manipulated and many interesting results are obtained. In the present paper, we show that the odd and the even rotational wave packets created by a laser pulse can be accurately excited by applying another time-delayed laser pulse. Different from the case of the above-mentioned schemes, the delay time here is accurately determined by the slope curve of the molecular alignment produced by the first laser pulse. By inserting the peak of the second laser pulse at the position of maximum or the minimum value around quarter or three quarter rotational period of the slope curve with odd (or even) rotational wave packet contribution created by the first laser pulse, the odd rotational wave packet can be enhanced (or suppressed) while the even

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†Corresponding author. E-mail: jixm@ntu.edu.cn

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rotational wave packet is suppressed (or enhanced). The total result of the interference between the odd and the even rotational wave packets is that molecular alignment around quarter and three quarter rotational periods can be obtained. Furthermore, we also show that the maximum degrees of the molecular alignment for the odd and the even rotational wave packet contributions are consistent with their corresponding slope curves when the second pulse is inserted around the quarter or three quarter rotational periods.

2. Theoretical model

We consider a linearly polarized laser field with the Gaussian distribution, given by

$$E(t) = E_0[f(t)\cos\omega t + f(t-T_{\rm d})\cos\omega(t-T_{\rm d})], \quad (1)$$

where E_0 is the field amplitude, $f(t)$ is the pulse envelope temporal profile $f(t) = \exp(-2\ln 2t^2/\tau^2)$, τ is the pulse duration, ω is the carrier frequency of the pulse and $T_{\rm d}$ is the time delay between the first and second laser pulses. When a linear molecule is exposed to the laser field, the time-dependent Schrödinger equation can be expressed, based on the rigid rotor model, as

$$\mathrm{i}\hbar\frac{\partial|\psi(t)\rangle}{\partial t} = H(t)|\psi(t)\rangle \quad (2)$$

with

$$H(t) = BJ(J+1) - \mu E(t)\cos\theta - \frac{1}{2}\left[\left(\alpha_{\parallel}-\alpha_{\perp}\right)\cos^2\theta + \alpha_{\perp}\right]E^2(t), \quad (3)$$

where B is the rotational constant of the molecule, J is the angular momentum, μ is the permanent dipole moment of the molecule, θ is the angle between the molecular axis and the polarization direction of the laser field, and $\alpha_{||}$ and $\alpha_{\perp}$ are the polarizability components parallel and perpendicular to the molecular axis, respectively. The time evolution of the rotational wave packet of molecular alignment is usually characterized by the expectation value of $\cos^2\theta$ (i.e. $\langle\cos^2\theta\rangle$). Considering the thermal equilibrium of the molecular ensemble, $\langle\cos^2\theta\rangle$ should be averaged over the Boltzmann distribution of the initial rotational state and written as

$$\langle\cos^2\theta\rangle = \sum_J W_J \sum_{M=-J}^{J}\langle\Psi_{JM}\left|\cos^2\theta\right|\Psi_{JM}\rangle, \quad (4)$$

where W_J is Boltzmann weight factor with $W_J = Q^{-1}g_J\exp\left[-BJ(J+1)/kT\right]$, with Q being the partition function with $Q = \sum_J g_J(2J+1)\exp[-BJ(J+1)/kT]$ and g_J the spin degeneracy factor.

In our calculation, the time-dependent Schrödinger equation in Eq. (2) is numerically solved by a split-operator method.[24,25] The CO molecule is used as an example and the molecular parameters are $B = 1.93$ cm^{-1}, μ= 0.112 D, $\alpha_{||}$= 2.294 Å^3 (1 Å=0.1 nm), $\alpha_{\perp}$= 1.77 Å^3.[26–28] Thus, the rotational period of the CO molecule can be calculated as $T_{\rm rot} = 1/(2Bc) \approx 8.64$ ps, where c is the velocity of the light in vacuum. The central wavelengths of the two laser pulses are both 800 nm with the pulse duration of 100 fs and the peak intensity $I = 2\times10^{13}$ W/cm^2. The initial rotational temperature is set to be 50 K.

3. Results and discussion

We first present the time evolutions of the molecular alignment $\langle\cos^2\theta\rangle$ excited by a single laser pulse, together with the odd (dashed line) and the even (dotted line) rotational wave packet contributions, which are shown in Fig. 1. One can see that the odd and the even rotational wave packet contributions exhibit the same behaviours around half and full revival periods while they show the opposite behaviours around quarter and three quarter rotational periods. The molecular alignment is the result of the interference between the odd and the even rotational wave packets. Consequently, the odd and the even rotational wave packet contributions will cancel out each other around quarter and three quarter rotational periods and therefore no net molecular alignment can be obtained.

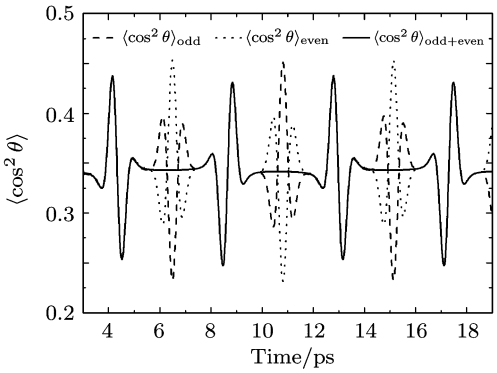


Fig. 1. Time evolutions of the molecular alignment $\langle\cos^2\theta\rangle$ created by a single laser pulse with the laser intensity $I = 2\times10^{13}$ W/cm^2 and the pulse duration $\tau = 100$ fs, together with the odd (dashed line) and even (dotted line) rotational wave packet contributions.

Figure 2 gives the time-dependent molecular alignment $\langle\cos^2\theta\rangle$ with the odd (a) and the even (b)

rotational wave packet contributions (solid lines, left axis) excited by a single laser pulse, together with their slope curves (the dashed lines, right axis). As can be seen, around half and full rotational periods, such as t=8.65 ps and 12.98 ps, the slope curves for both the odd and the even rotational wave packets show that they have the same behaviours and simultaneously have the maximum or minimum values. It has been demonstrated that by applying another laser pulse at these positions, the molecular alignment will be enhanced or suppressed.[23,29] However, around quarter and three quarter rotational periods, such as t = 6.48 ps and t = 10.8 ps, the slope curves for the odd and the even rotational wave packets exhibit opposite behaviours. The minimum (maximum) value is for the odd rotational wave packet while the maximum (minimum) value is for the even rotational wave packet. Therefore, if the second laser pulse is inserted at these positions, selective excitation between the odd and the even rotational wave packets can be achieved. Since the molecular alignment is the result of the interference between the odd and even rotational wave packets, it can be obtained around these positions.

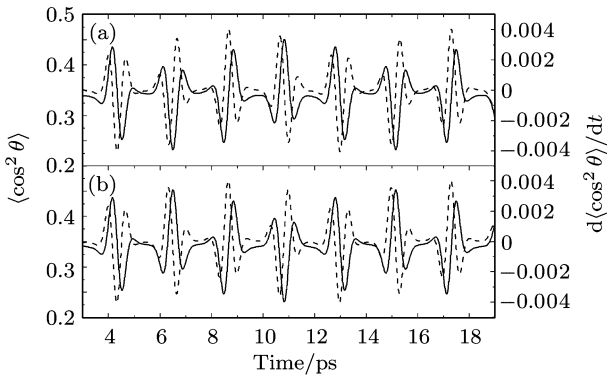


Fig. 2. Time evolutions of molecular alignment $\langle\cos^2\theta\rangle$ with the odd (a) and the even (b) rotational wave packet contributions (solid lines, left axis) excited by a single laser pulse, together with their slope curves (dashed lines, right axis).

Figure 3 shows the time evolutions of the molecular alignment $\langle\cos^2\theta\rangle$ with the odd (a), even (b), and total (c) rotational wave packet contributions (solid lines) by applying the second laser pulse at time delay T_d = 6.33 ps, together with the contributions by only the first laser pulse (dashed lines). Notice that the slope curves for the odd and the even rotational wave packet contributions have minimum and maximum values at T_d=6.33 ps, respectively (see Fig. 2). One can see that around quarter and three quarter rotational periods, the molecular alignment for the odd rotational wave packet contribution is suppressed (see Fig. 3(a)), while that for the even rotational wave packet contribution is enhanced (see Fig. 3(b)). That is to say, the selective excitation between the odd and the even rotational wave packets can be realized. Moreover, it can be found that the same behaviours can also be observed around the half and full rotational period. As shown in Fig. 3(c), the molecular alignment around quarter and three quarter rotational periods can be obtained due to the interference between the odd and the even rotational wave packets.

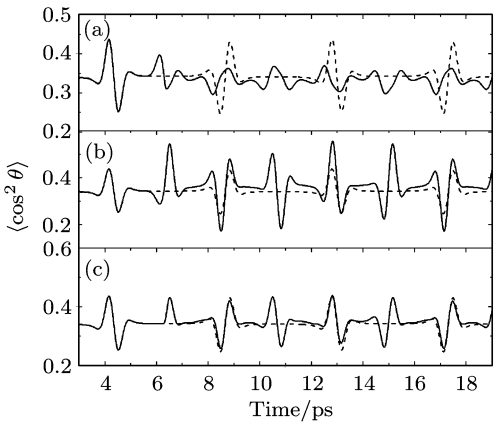


Fig. 3. Time evolutions of the molecular alignment $\langle\cos^2\theta\rangle$ with the odd (a), even (b), and total (c) rotational wave packet contributions (solid lines) by applying the second laser pulse at time delay T_d = 6.33 ps, together with the contributions by only the first laser pulse (dashed lines).

Similar results can be achieved by applying the second laser pulse at time delay T_d= 6.65 ps as shown in Fig. 4. Because the slope curves for the odd and the even rotational wave packet contributions at this time delay have the maximum and the minimum values, the molecular alignments for the two contributions are correspondingly enhanced and suppressed as shown in Figs. 4(a) and 4(b). Thus, the molecular alignment for the total rotational wave packet contribution around quarter and three quarter rotational periods can also be obtained, which is shown in Fig. 4(c), which is the same as that shown in Fig. 3(c). The same results can also be obtained by applying the second laser pulse at maximum (or minimum) value of the slope curves around three quarter rotational periods, and they are not shown here.

In order to demonstrate the effects of the second laser pulse on the molecular alignment for the odd and the even wave packet contributions, next we investigate the maximum degrees of the molecular align-

ment for the odd and the even wave packet contributions when the second laser pulse is applied at the time delay from 5.5 ps to 7.5 ps, and the calculated results are shown in Fig. 5, together with the time-dependent molecular alignment (solid lines, left axis) and its slope curve (dashed lines, right axis) induced by the first laser pulse. One can see that the change in the maximum degree of the molecular alignment shows a good agreement with the slope curve of alignment degree created by the first laser pulse for both the odd and even rotational wave packets. These observations further illustrate that the odd and the even rotational wave packets created by a laser pulse can be selectively excited by properly introducing another laser pulse.

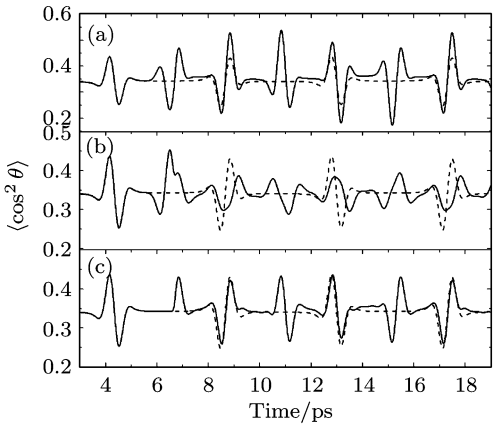


Fig. 4. The same as Fig. 3 but with $T_{\rm d} = 6.65$ ps.

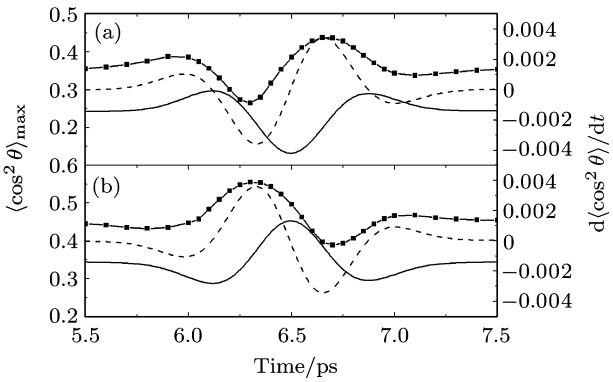


Fig. 5. Time evolutions of maximum degrees of the molecular alignment $\langle\cos^2\theta\rangle_{\rm max}$ for the odd (a) and even (b) wave packet contributions when the second laser pulse is applied with the time delay from 5.5 ps to 7.5 ps (squares), together with the time-dependent molecular alignment (solid lines, left axis) and its slope curve (dashed lines, right axis) induced by the first laser pulse.

4. Conclusion

We demonstrated that the molecular odd and even rotational wave packets created by a laser pulse can be selectively excited by accurately inserting another laser pulse. By applying the second pulse at the position of maximum or minimum value of the slope curve around quarter or three quarter rotational periods, the odd rotational wave packet is enhanced (or suppressed) while the even rotational wave packet can be suppressed (or enhanced). As a result, the molecular alignment around quarter and three quarter rotational periods can be obtained. Moreover, it is shown that when the second laser pulse is inserted around the quarter or three quarter rotational periods, the changes in the maximum degree of the molecular alignment for both the odd and the even rotational wave packets are consistent with their corresponding slope curves at these positions. We believe that these results can provide a theoretical basis for experimental study and have a potential application in the control of various related fields.

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