

## Controlling the molecular rotational wave packet accurately by femtosecond laser pulses

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Received 5 July 2010; Accepted (in revised version) 30 July 2010

Published online 28 February 2011

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**Abstract.** We investigate the rotational wave packet modified by femtosecond laser pulses. The calculations are performed by solving numerically the full time-dependent Schrödinger equation for the N<sub>2</sub> molecule at finite rotational temperature. It is demonstrated that the rotational wave packet induced by the first laser pulse can be controlled exactly just by selecting the optimal time at which the second laser pulse is introduced. Whether the pulse duration of the two lasers is equal or not, the molecular alignment induced by the first laser pulse can be enhanced or degraded by precisely inserting the peak of the second laser pulse at the maximum or minimum position of the slope curve for the alignment parameter by the first laser. Furthermore, the already enhanced alignment by the two lasers can be enhanced or degraded by precisely inserting the peak of the third laser at the maximum or minimum position of the slope curve for the alignment by the two lasers. The already degraded alignment by the two lasers can be increased again from the isotropic distributed ensemble by precisely inserting the peak of the third laser at the peak position of the slope curve by the two lasers.

**PACS:** 33.80.Rv, 42.50.Hz, 33.90.+h

**Key words:** rotational wave packets, femtosecond laser pulses, time delay, the slope curve for the alignment

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## 1 Introduction

When a laser pulse with both ultra short pulse width and ultrahigh laser intensity illuminates a molecule, the induced dipole moment will impart a torque. The molecule can then rotate to the laser polarization direction. As a result, molecules are partially aligned or oriented along the laser polarization direction [1–3], the field-free alignment and orientation of gas

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molecules have recently found many imperative applications. Molecular properties such as refractive index [4,5], ionization rate [6], high harmonic generation efficiency [7,8], diffraction [9,10], and molecular imaging [11] depend strongly on molecular alignment. What's more, molecular alignment and orientation also play an important part in molecular electronic stereo dynamics [12–14].

The molecular alignment can be achieved adiabatically and nonadiabatically [2], adiabatic alignment is induced from field-free rotational states to pendular states but it is lost at the end of the laser pulse. Nonadiabatic alignment of the molecules attracts much attention for being able to study the molecules in a field-free alignment condition. It is demonstrated that the excellent aligned molecules can be obtained by selecting the proper parameter of the aligning laser pulse [15–17] and reducing the rotational temperature [18,19]. However, the improvement is often limited by the saturation of molecular alignment and the maximum intensity that can be applied to the molecules without ionizing it. Spectral phase shaping method [20] provides a passive control schemes for improving the alignment degree beyond the limit. The alignment degree can be maximized by the accurate optimized laser pulse by evolutionary algorithm. However, this strategy is often time consuming and very hard to apply in experiment.

An intriguing and more efficient way for achieving an enhanced alignment degree is to employ a train of laser pulses [21]. Since the alignment is resulted from the in-phase overlapping of spherical harmonics in the time evolution of rotational wave packet, the odd and the even rotational states evolve out of phase around quarter revival times and in phase around full revival times. The alignment degree can be enhanced by applying the second laser pulse at the rising edge of the alignment parameter induced by the first laser pulse [22–24]. On the other hand, the rotational wave packet induced by the first laser pulse can be annihilated by the second laser pulse introduced at the half revival time [25,26]. What's more, a coherent molecular rotational wave packet can be manipulated selectively by strong femtosecond laser pulse [27,28].

Recently, a general method that relates the two-pulse alignment degree to their delay times has been developed, a new strategy to actively determine the optimized time delay between the two pulses with the same pulse duration is proposed according to the so called 'slope rule' [29]. In fact, the control for the rotational wave packet depends strongly on the pulse duration of two lasers. In this paper, the relation between the slope of alignment and the optimum time delay between the two pulses with both same pulse duration and different pulse duration is investigated, the enhancement and the annihilation of the molecular alignment are controlled exactly using two and three lasers with different peak intensity and pulse duration.

## 2 Theoretical method

We calculate the nonadiabatic alignment of linear molecules using the method presented in references [19,22]. For a molecule in a linearly polarized laser field, the Schrödinger equation

under the rigid rotor approximation can be written as

$$i \frac{\partial \Psi(\theta, \phi, t)}{\partial t} = \left( B \hat{J}^2 - \frac{E^2(t)}{2} (\alpha_{//} \cos^2 \theta + \alpha_{\perp} \sin^2 \theta) \right) \Psi(\theta, \phi, t), \quad (1)$$

where the angle  $\theta$  is between the laser polarization and the interatomic axis of the molecule,  $\phi$  is azimuthal angle of molecular axis in a fixed frame.  $\alpha_{//}$  and  $\alpha_{\perp}$  are the polarizabilities along and perpendicular to the molecular axis, respectively.  $B$  is the rotational constant,  $\hat{J}^2$  being the angular-momentum operator.  $E(t)$  with Gaussian temporal profile is written as

$$E(t) = \sum_k E_k e^{-2 \ln 2 ((t-t_k)/\tau_k)^2} \cos \omega t, \quad (2)$$

where  $E_k$ ,  $\tau_k$  and  $t_k$  are the field amplitude, full-width at half maximum (FWHM), center time of the  $k$ th laser pulse, respectively.  $\omega$  is the frequency of the laser.

Non-adiabatic field-free alignment of nitrogen molecules is achieved by the excitation of a rotational wave packet, which can be expressed by the wave function of field-free rotational states  $|JM\rangle$

$$\Psi(t) = \sum_J B_{JM} |JM\rangle. \quad (3)$$

The time evolution of the wave packet can be calculated by solving the time-dependent Schrödinger Eq. (1) with the pseudospectral method [30, 31]. Finally the time-dependent alignment parameter  $\langle \cos^2 \theta \rangle$  is obtained from the calculated rotational wave function of the molecules.

### 3 Results and discussion

Using the methods mentioned above, the dynamics of rotational wave packet for  $N_2$  is investigated firstly. The molecular ensemble is at the temperature of 300 K. Fig. 1 shows the alignment parameter  $\langle \cos^2 \theta \rangle$ , its slope  $d\langle \cos^2 \theta \rangle/dt$  by the first pulse, the maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  by the two pulses with the time delay in the shown time range, respectively. In our calculation, the laser intensity and pulse duration (FWHM) of the two pulses are fixed as  $2 \times 10^{13}$  W/cm<sup>2</sup> and 50 fs, respectively. the wavelength of the lasers is 800 nm, the center of the first laser is centered at 0 fs, we just adjust the center of the second laser to control the time delay between the two lasers. It is demonstrated that the curve of the maximum alignment by the two lasers matches with the slope of  $\langle \cos^2 \theta \rangle$  by the first laser exactly, and the minimum alignment by the two lasers matches with the slope of  $\langle \cos^2 \theta \rangle$  by the first laser reversely. When the second laser pulse is applied at the peak position of the slope curve around the full revival of the rotational wave packet by the first laser,  $\langle \cos^2 \theta \rangle_{max}$  reaches maximum and  $\langle \cos^2 \theta \rangle_{min}$  reaches minimum, which means a strongly enhanced alignment and antialignment. Reversely, when the second laser pulse is applied at the minimum position of the slope curve around the half revival of the rotational wave packet by the first laser, both the  $\langle \cos^2 \theta \rangle_{max}$  and  $\langle \cos^2 \theta \rangle_{min}$  get close to 1/3, which

means the rotational wave packet by the first pulse is strongly suppressed by the second laser and molecular ensemble exhibits an isotropic distribution, which is in good agreement with the result reported in reference [29] for  $O_2$  molecules. As the pulse duration plays an important role in the molecular alignment, in order to understand whether the 'slope rule' can be applicable in general when the pulse duration of the two lasers is different from each other, the following calculation is carried out.

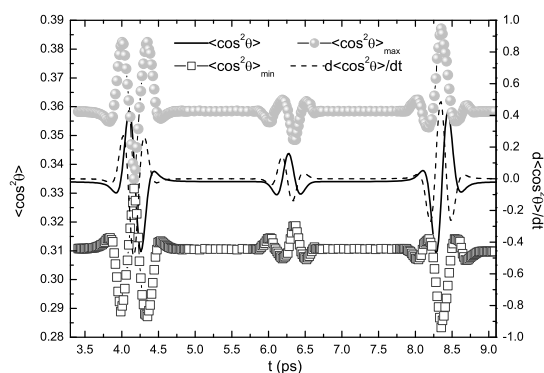


Figure 1: The alignment parameter  $\langle \cos^2 \theta \rangle$  (the left axis) and its slope  $d\langle \cos^2 \theta \rangle / dt$  (the left axis) induced by the first aligning pulse, when applying the second aligning pulse at the time delay in the shown time range, the maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  are shown respectively. the laser intensity and pulse duration (FWHM) of the two pulses are fixed as  $2 \times 10^{13} \text{W/cm}^2$  and 50 fs, respectively.

Fig. 2 shows the alignment parameter  $\langle \cos^2 \theta \rangle$ , its slope  $d\langle \cos^2 \theta \rangle / dt$  induced by the first pulse, the maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  by the two aligning pulse with the time delay in the shown time range, respectively. In our calculation, the first laser is the same used in Fig. 1, the second laser intensity is also fixed as  $2 \times 10^{13} \text{W/cm}^2$ , but the pulse duration of the second laser is 20 fs in Fig. 2(a) and 150 fs in Fig. 2(b), respectively. As can be seen, the curve of the maximum alignment also matches with the slope of alignment exactly, and the minimum alignment matches with the slope reversely both in the Fig. 2(a) and Fig. 2(b), which is the same as in Fig. 1. In a word, whether the pulse duration of the two lasers is equal or not, the 'slope rule' can be applicable in general. When the second laser pulse is introduced at the peak position of the slope curve, an obviously enhanced alignment and antialignment can be obtained. Reversely, when the second laser pulse is introduced at the minimum position of the slope curve, the revivals are greatly weakened, a decreased alignment and antialignment may be obtained.

Based on what mentioned above, the modified rotational wave packet by the two pulses with different pulse duration was investigated according to the slope rule. In the calculation, both the two lasers intensity is fixed as  $2 \times 10^{13} \text{W/cm}^2$ . Fig. 3 indicates the maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  by the two laser pulses, respectively. It is demonstrated that both the maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  varies as a function of the second pulse duration  $\tau_2$  when the pulse duration  $\tau_1$

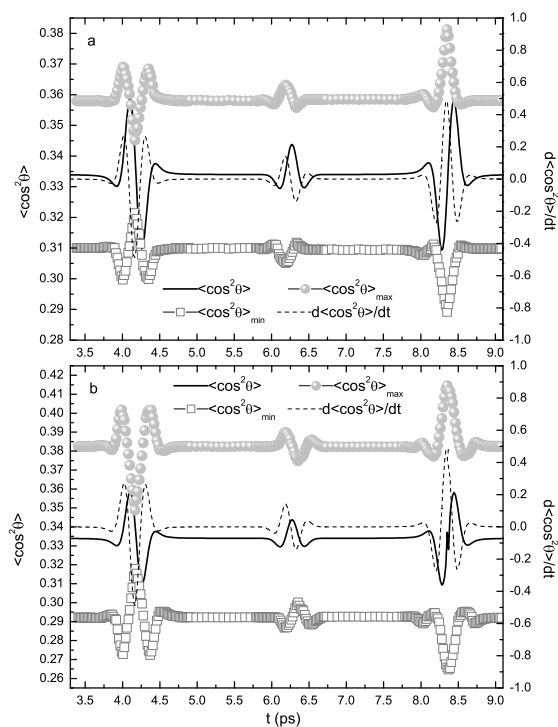


Figure 2: The alignment parameter  $\langle \cos^2 \theta \rangle$  (the left axis) and its slope  $d\langle \cos^2 \theta \rangle/dt$  (the left axis) induced by the first pulse with laser intensity  $2 \times 10^{13}$  W/cm<sup>2</sup> and pulse duration 50 fs, when applying the second aligning pulse with laser intensity  $2 \times 10^{13}$  W/cm<sup>2</sup> and pulse duration (a) 20 fs, (b) 150 fs at the time delay in the shown time range, the maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  are shown respectively.

of the first laser is fixed, the larger value the maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  reaches, the smaller the minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  becomes. In Fig. 3(a) and (b), the second laser pulse is introduced at the minimum position of the slope curve for the alignment by the first laser. As can be seen, for the certain pulse duration  $\tau_1$  of the first laser, the maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  can be decreased and increased by adjusting the pulse duration  $\tau_2$  of the second laser, respectively. Especially, the maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  get closed to 1/3 when the two lasers pulse duration is equal, the isotropic distributed molecular ensemble can be reproduced. In Fig. 3(c) and (d), the second laser pulse is introduced at the peak position of the slope curve for the alignment by the first laser. As can be seen, for the certain pulse duration  $\tau_1$  of the first laser, the maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  can be increased and decreased by adjusting the pulse duration  $\tau_2$  of the second laser, respectively. The  $\langle \cos^2 \theta \rangle_{max}$  reaches maximum and  $\langle \cos^2 \theta \rangle_{min}$  reaches minimum when the second

pulses duration is a certain value. For example, when the pulse duration of the two lasers is 150 fs, the molecular alignment and antialignment can be enhanced greatly.

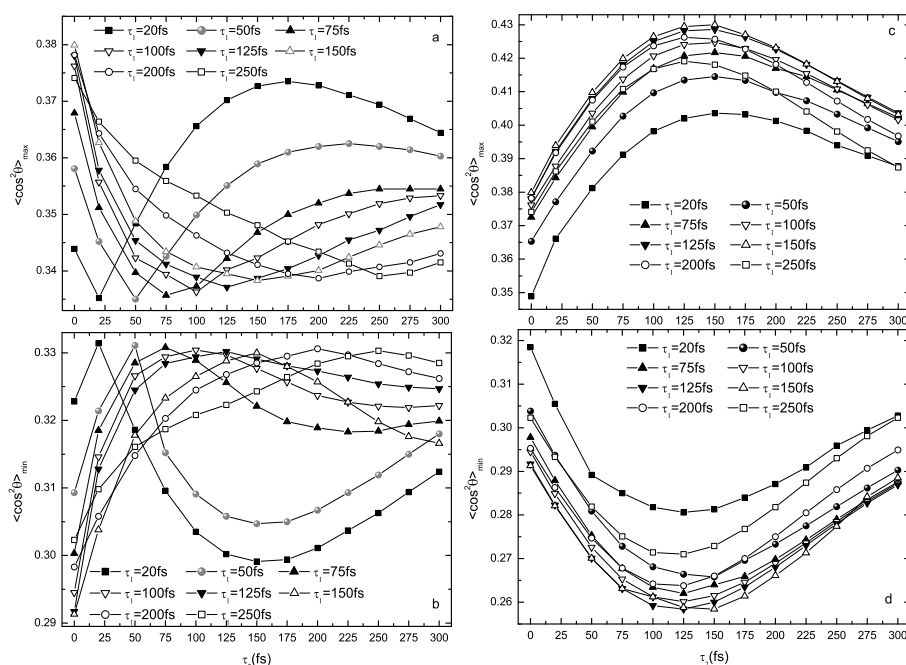


Figure 3: The maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  are shown respectively. The second laser pulse is introduced at the minimum position of the slope curve around the half revival of the rotational wave packet by the first laser in (a) and (b), but at the peak position of the slope curve around the first full revival of the rotational wave packet by the first laser in (c) and (d).

In order to investigate the alignment dependence on laser intensity, Fig. 4 shows the maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  by the two pulses. In the calculation, both the two pulse duration is fixed as 150 fs, the first laser intensity  $I_1$  is fixed as  $2 \times 10^{13}$  W/cm<sup>2</sup>, the second laser intensity  $I_2$  is changed limited by the saturation of molecular alignment and the maximum intensity that can be applied to the molecules without ionizing it. In the Fig. 4(a), the second laser is introduced at the maximum position of the slope curve for the alignment by the first laser, as can be seen, the maximum alignment increases monotonically as the second pulse intensity is increased from 0 to  $8 \times 10^{13}$  W/cm<sup>2</sup>, reflecting an increased degree of incoherent alignment. However, the minimum alignment decreases when the intensity of the second pulse is less than  $5 \times 10^{13}$  W/cm<sup>2</sup>, as the second intensities greater than  $5 \times 10^{13}$  W/cm<sup>2</sup>, it can not be decreased any more. In the Fig. 4(b), the second laser is introduced at the minimum position of the slope curve for the alignment

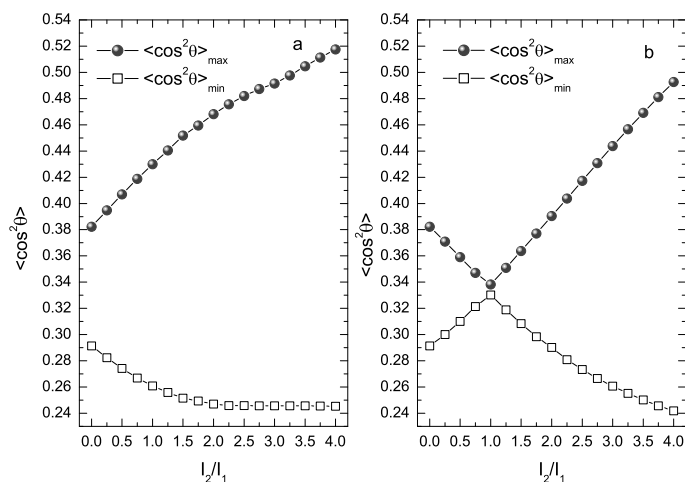


Figure 4: The maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  by the two pulses when the second laser is introduced (a) at the peak position of the slope curve, (b) at the minimum position of the slope curve for the alignment by the first laser. In the calculation, both the two pulse duration is 150 fs, the first laser intensity  $I_1$  is fixed as  $2 \times 10^{13}$  W/cm<sup>2</sup>, the second laser intensity  $I_2$  is changed.

by the first laser, the maximum alignment decreases and minimum alignment increases as the second pulse intensity is increased from 0 to  $2 \times 10^{13}$  W/cm<sup>2</sup>. As the second intensities greater than  $2 \times 10^{13}$  W/cm<sup>2</sup>, the maximum alignment increases and minimum alignment decreases, reversely. It can be concluded that the molecular alignment by the first laser can be perfectly degraded by the second pulse which has the same intensity and pulse duration with the first laser.

Fig. 5(a) demonstrates the alignment parameter  $\langle \cos^2 \theta \rangle$ , its slope by the first pulse, the maximum alignment and minimum alignment reaches 0.3799 and 0.2913, respectively. Fig. 5(b) shows the alignment parameter  $\langle \cos^2 \theta \rangle$  by the two laser pulses, the peak of the second laser is inserted at 4.187 ps when the slope for the alignment parameter by the first laser locates a local minimum. In the calculation, the laser intensity and pulse duration of the two lasers are  $2 \times 10^{13}$  W/cm<sup>2</sup> and 150 fs. As can be seen, the alignment is strongly degraded. The maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  reaches 0.3383 and 0.3303, respectively, both of which get close to 1/3. The second pulse annuls the effect of the first pulse, the wave packet is approximately returned to the isotropic distributed state if the two lasers had not been applied. Fig. 5(c) shows the alignment parameter  $\langle \cos^2 \theta \rangle$  by the two laser pulses as the peak of the second laser is inserted at 8.370 ps when the slope for the alignment parameter by the first laser locates a local maximum. As can be seen, the alignment is maximized. The maximum alignment  $\langle \cos^2 \theta \rangle_{max}$  and minimum alignment  $\langle \cos^2 \theta \rangle_{min}$  reaches 0.43 and 0.2584, which are increased about 13% and decreased about 11% respectively. An obviously enhanced alignment and antialignment is obtained.

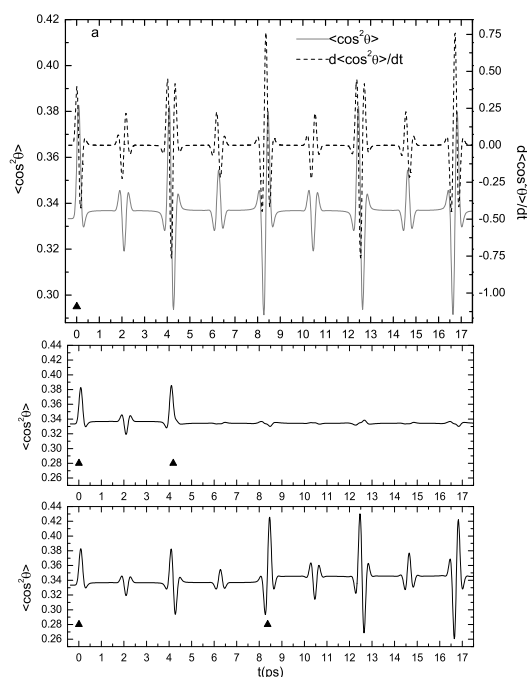


Figure 5: (a) The alignment  $\langle \cos^2\theta \rangle$  (the left axis) and its slope curve (the right axis) by the first aligning pulse. The peak of the first aligning pulse is centered at zero point. (b) and (c) show the alignment  $\langle \cos^2\theta \rangle$  by the two lasers, the peak of the second laser is inserted (b) at 4.187 ps when the slope for the alignment parameter by the first laser locates a local maximum, (c) at 8.370 ps when the slope for the alignment parameter by the first laser locates a local maximum. In the calculation, both the lasers intensity and pulse duration are fixed as  $2 \times 10^{13}$  W/cm<sup>2</sup> and 150 fs. The triangles indicate the peak position of the laser pulse.

To illustrate the applicability of ‘slope rule’ in molecular alignment by three laser pulses, the three-pulse alignment for N<sub>2</sub> molecules is calculated. Based on the above results, in our calculations, the first and second pulse intensities are fixed as  $2 \times 10^{13}$  W/cm<sup>2</sup> and the two lasers pulse duration is 150 fs. Fig. 6(a) shows the alignment  $\langle \cos^2\theta \rangle$  and its slope curve as the peak of the second laser is inserted at 8.370 ps when the slope for the alignment parameter by the first laser locates a local maximum. Fig. 6(b) indicates the alignment  $\langle \cos^2\theta \rangle$  by the three lasers as the peak of the third laser is inserted at 12.553 ps when the slope for the alignment parameter by the two lasers locates a local minimum, the third laser intensity is  $3.6 \times 10^{13}$  W/cm<sup>2</sup> and pulse duration 150 fs. It is obvious that the already enhanced alignment by the two lasers can be strongly degraded by introducing the third laser at the minimum position of the slope curve by the two lasers. The third pulse annuls the effect of the first and second pulses, the wave packet is approximately returned to the isotropic distributed state if the three lasers had not been applied. Fig. 6(c) indicates the alignment  $\langle \cos^2\theta \rangle$  by three lasers as the peak of the third laser is inserted at 16.736 ps when the slope for the alignment parameter by



the two lasers locates a local maximum, the third laser intensity is  $2 \times 10^{13} \text{ W/cm}^2$  and pulse duration 150 fs. It is obvious that the already enhanced alignment by the two lasers can be further enhanced just by introducing the third laser at the peak position of the slope curve by the two lasers.

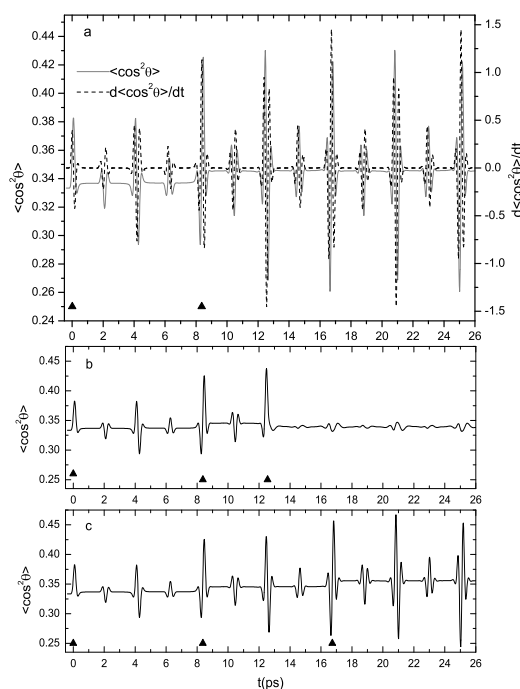


Figure 6: (a) The alignment  $\langle \cos^2 \theta \rangle$  (the left axis) and its slope curve (the right axis) by two pulses with equal intensity  $2 \times 10^{13} \text{ W/cm}^2$  and pulse duration 150 fs on the condition that the peak of the second laser is inserted at 8.370 ps when the slope for the alignment parameter by the first laser locates a local maximum. The peak of the first aligning pulse is centered at zero point. The alignment  $\langle \cos^2 \theta \rangle$  by the three lasers as the peak of the third laser is inserted (b) at 12.553 ps when the slope for the alignment parameter by the two lasers locates a local minimum, (c) at 16.736 ps when the slope for the alignment parameter by the two lasers locates a local maximum. In the calculation, the third laser intensity is  $3.6 \times 10^{13} \text{ W/cm}^2$  in (b) and  $2 \times 10^{13} \text{ W/cm}^2$  in (c), both the pulse duration is 150 fs. The triangles indicate the peak position of the laser pulse.

Fig. 7(a) shows the alignment  $\langle \cos^2 \theta \rangle$  and its slope curve as the peak of the second laser is inserted at 4.187 ps when the slope for the alignment parameter by the first laser locates a local minimum. Fig. 7(b) indicates the alignment  $\langle \cos^2 \theta \rangle$  by the three lasers as the peak of the third laser is inserted at 8.570 ps when the slope for the alignment parameter by the two lasers locates a local maximum. In the calculation, the three lasers intensity is all  $2 \times 10^{13} \text{ W/cm}^2$  and pulse duration is 150 fs. It is obvious that the degraded alignment even the isotropic distributed molecular ensemble by the second laser can be enhanced again just by

introducing the third laser at the peak position of the slope curve by the two lasers. Obviously, the alignment is the same as the one by a single pulse indicated in Fig. 7(c), the third pulse annuls the effect of the second pulse, and the wave packet is approximately returned to the state if only the first pulse is applied, which is in good agreement with the zero effect pulse pairs in Ref. [26].

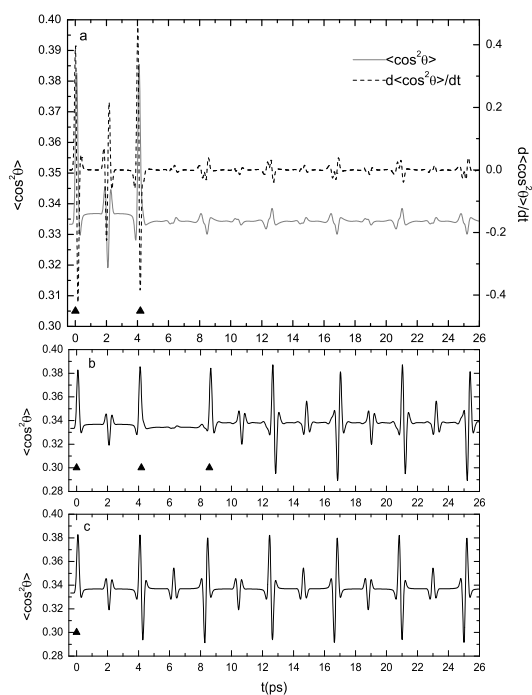


Figure 7: (a) The alignment  $\langle \cos^2 \theta \rangle$  (the left axis) and its slope curve (the right axis) by two pulses on the condition that the peak of the second laser is inserted at 4.187 ps when the slope for the alignment parameter by the first laser locates a local minimum. The peak of the first aligning pulse is centered at zero point. (b) The alignment  $\langle \cos^2 \theta \rangle$  by the three lasers as the peak of the third laser is inserted at 8.570 ps when the slope for the alignment parameter by the two lasers locates a local maximum, (c) The alignment  $\langle \cos^2 \theta \rangle$  by a single laser. In the calculation, all the lasers intensity is  $2 \times 10^{13}$  W/cm<sup>2</sup> and the pulse duration is 150 fs. The triangles indicate the peak position of the laser pulse.

## 4 Conclusions

In summary, the rotational wave packet modified by femtosecond laser pulses is investigated by solving numerically the full time-dependent Schrödinger equation. Whether the pulse duration of the two lasers is equal or not, the molecular alignment induced by the first laser pulse can be enhanced by precisely inserting the peak of the second laser at the peak position of the slope curve for the alignment parameter by the first laser, the alignment degree can be

further enhanced by adjusting the two pulse duration and increasing the second laser intensity without ionizing molecules. Reversely, the molecular alignment induced by the first laser can be degraded by precisely inserting the peak of the second laser pulse at the minimum position of the slope curve for the alignment parameter by the first laser; both the maximum alignment and minimum alignment get close to 1/3 when the rotational wave packet is modified by the two pulses with the same pulse duration and equal intensity, the second pulse annuls the effect of the first pulse, the wave packet is approximately returned to the isotropic distributed state if the two lasers had not been applied. Our further research indicates that the already enhanced alignment by the two lasers can be enhanced or annulled by introducing the third laser at the peak position or minimum position of the slope curve by the two lasers. The already annulled rotational wave packet by the two lasers can be enhanced again by introducing the third laser at the peak position of the slope curve by the two lasers, the third pulse annuls the effect of the second pulse, and the wave packet is approximately returned to the state if only the first pulse is applied. The scheme provides an exact way to create and annihilate the rotational wave packets using femtosecond lasers, which may pave an adaptive road for rotating molecules using laser pulses [23, 26], ultrafast refractive index modulation [4, 5], improving revival quality at long time scales, and handling mixtures of different species, isomers, or conformers [27, 28].

**Acknowledgments.** The authors thank the supports from the Initial Research Fund for High Level Talents of Shihezi University under Grant No. RCZX200743. We also acknowledge the support from the Opening Foundation of Xinjiang Laboratory of Phase Transitions and Microstructures of Condensed Matters under Grant No. XJDX0912-2010-08.

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