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Selective Alignment of D_2 Induced by Two Ultrashort Laser Pulses

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The dynamics of molecular rotational wave packets of D_2 induced by ultrashort laser pulses was investigated numerically by solving the time-dependent Schrödinger equation. Results show that an ultrashort pulse can manipulate a coherent rotational wave packet of D_2 selectively. In the calculation, a first laser pulse was used to create a coherent rotational wave packet from an initial thermal ensemble of D_2 at the temperature of 300 K. The second laser pulse was used to manipulate the rotational wave packet selectively around the first quarter and the three quarters revival. The alignment parameter and its Fourier transform amplitude both illustrate that the relative populations of even and odd rotational states in the final rotational wave packet of D_2 can be manipulated by precisely selecting the time delay between the first and the second ultrashort pulse.

Key words: Rotational wave packet, Ultrashort laser pulse, Time delay, Odd and even states

I. INTRODUCTION

The research of interaction between molecular and laser pulses with both ultrashort pulse width and ultrahigh laser intensity has been one intriguing area in the last decade. In particular, laser-induced spatial alignment [1,2] and orientation [3-5] is a central subject in molecular science. When the laser intensity reaches 10^{12} W/cm^2, molecules are aligned along the direction of the laser polarization through the interaction between nonresonant laser fields and the induced dipole moment of the molecule. A sample of aligned or oriented molecules is an excellent quantum logical system, which enables us to investigate quantum phenomena associated with molecular symmetry and correlations between a molecular axis and the laser polarization as well as stereodynamics in chemical reactions. Since some important physical phenomena such as high-order harmonic generation, ionization, and dissociation are related closely to the anisotropic properties of molecules [6-10], molecular alignment and orientation pave an effective road for electronic stereodynamics in molecules [11-13].

The molecular alignment can be divided into the adiabatic [14] and the nonadiabatic regimes [15] in general. For the majority of research, a femtosecond laser pulse (much less than the rotational period of molecules) is used to interact with a molecular ensemble, and a coherent rotational wave packet is therefore created. The coherence of the rotational wave packet is preserved after the laser is turned off and as a result, transient alignment is achieved periodically. For many research projects, a high degree of alignment is desired, so many researchers focus on the enhancement of the degree of alignment among various controls of nonadiabatic molecular alignment. The field-free alignment degree strongly depends on the parameter of the aligning laser pulse [16-19] as well as the temperature of the molecular ensemble [20,21]. Different rotational components exhibited different dynamical behaviors during the evolution of the rotational wave packet. For example, the odd and the even rotational states evolve out of phase around quarter revival times and in phase around full revival times. The coherent rotational wave packet induced by a femtosecond laser pulse can be modified by another femtosecond laser pulse or a train of laser pulses [22-25]. The final rotational wave packet can be manipulated by selecting the intensity ratio or the time delay between the laser pulses. Both the enhancement and the annihilation of the molecular alignment has been achieved in experiment by using two properly time-delayed short laser pulses. What is more, field-free three-dimensional alignment has been achieved experimentally by using two time-delayed, orthogonally polarized, nonresonant, femtosecond laser pulses [26].

Recently, some researches have demonstrated that a strong femtosecond pulse can manipulate a coherent molecular rotational wave packet of N_2 and N_2O selectively [27,28], which may find applications to analysis, enrichment, and actual physical separation of molecular spin modifications. Being the most fundamental molecule, the deuterium molecule has far-reaching implications in physics, including the precise time-domain

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spectroscopy [29] and the stability of physical constants and cosmology [30]. Dynamic alignment of much lighter D_2 molecules induced by ultrashort pulses has been realized in experiment [31,32] and enhanced alignment by two femotosecond lasers has also been proposed [33]. In this work, we also compared the rotational wave packets modified by two laser pulse with the one induced by a single laser pulse.

II. THEORETICAL METHOD

The theoretical method and calculations are similar to those described in [33]. First, we numerically solve the time-dependent Schrödinger equation which describes interaction of a neutral molecule in the ground vibrational state with a laser pulse.

$$i\frac{\partial\Psi(\theta,\varphi,t)}{\partial t}=\Big[\frac{1}{2I}\hat{J}^2+V(\theta)\Big]\Psi(\theta,\varphi,t) \tag{1}$$

where φ is azimuthal angle of molecular axis in a fixed frame, the angle θ is between the laser polarization and the interatomic axis of the molecule, and I is the moment of inertia of a molecule, and $\hat{J}^2$ is the angular-momentum operator.

$$V(\theta)=-\frac{E^2(t)}{2}(\Delta\alpha\cos^2\theta+\alpha_\perp) \tag{2}$$

where E is intensity profile of the aligning laser field, and $\Delta\alpha$ is the effective polarizability defined as the difference between $\alpha_{//}$ and $\alpha_\perp$, parallel and perpendicular to the molecular interatomic axis, respectively. The laser pulse excites a wave packet which is formed from many rotational states, and which may be written

$$\Psi(t)=\sum_J C_J|JM\rangle \tag{3}$$

where $|JM\rangle$ is the wavefunction of field-free rotational states. The time-dependent Schrödinger Eq.(1) can be solved using the pseudospectral method [34,35] numerically when the molecule interacts with laser. The second-order split-operator technique in spherical coordinates shall be extended for determining the time propagation of the Schrödinger equation:

$$\begin{aligned}\Psi(\theta,\varphi,t+\Delta t)\approx&\exp\frac{-i\hat{H}_0\Delta t}{2}\cdot\\&\exp\left[-i\hat{V}\left(\theta,\varphi,t+\frac{\Delta t}{2}\right)\Delta t\right]\cdot\\&\exp\frac{-i\hat{H}_0\Delta t}{2}\Psi(\theta,\varphi,t)+O(\Delta t^3)\end{aligned} \tag{4}$$

where $\hat{H}_0=\hat{J}^2/(2I)$, which is the kinetic energy operator, and $\hat{V}$ is the remaining Hamiltonian depending on the spherical coordinates only. Δt is the time step. Thus the propagation of the wavefunction is accomplished by repeating Eq.(5) until the end of the laser field, and time-dependent wavefunction of the rigid rotor of the molecule is obtained.

In general, the degree of molecular alignment of a whole molecular ensemble is described by alignment parameter $\langle\cos^2\theta\rangle$, which in the present case is the quantity $\langle\cos^2\theta\rangle_{JM}=\langle\Psi_{JM}|\cos^2\theta|\Psi_{JM}\rangle$. In the case of finite temperature [14], a statistical ensemble of initial wavefunctions distributed according to the Boltzmann weights ρ_J should be considered, so that

$$\langle\cos^2\theta\rangle=\sum_J\rho_J\sum_M\langle\cos^2\theta\rangle_{JM} \tag{5}$$

When $\langle\cos^2\theta\rangle>1/3$, the molecular axis is preferentially aligned along the laser polarization axis, while $\langle\cos^2\theta\rangle<1/3$ corresponds to a planar delocalization where the molecular axis is preferentially distributed into a plane perpendicular to the polarization axis. The value $\langle\cos^2\theta\rangle=1/3$ characterizes an isotropic distribution of the molecular axis in space. In general cases, $\langle\cos^2\theta\rangle$ is between 0 and 1.

III. RESULTS AND DISCUSSION

For the D_2 molecule, the rotational level spacing is much larger than any other molecule, and as well it has low polarizability anisotropy, so it is particularly hard to align. Therefore, laser with higher intensity and shorter pulse duration (or with a broader bandwidth) is needed in order to better align D_2 molecules [32]. In the present work, the laser intensity is fixed as 10^{14} W/cm^2 and pulse duration 10 fs. In this case, only a small fraction of molecules within the interaction region is ionized at the peak of the pulse; the majority of the molecules remains neutral and experience only rotational excitation [31,32]. In the following, the post-pulse alignment of D_2 molecules is calculated using the methods above. In our calculation, the laser pulse takes the Gaussian form:

$$E(t)=E\mathrm{e}^{-2\ln 2[(t-t_0)/t_w]^2}\cos\omega t \tag{6}$$

where E is peak intensity of the lasers, t_w is pulse duration (FWHM) of the laser, t_0 is the center of the laser pulse, and ω is the frequency of the laser. The wavelength of the laser interacting with molecules is 800 nm in our calculation.

Field-free alignment of D_2 induced by a single ultrashort pulse was calculated. In our calculation, the temperature of the molecular ensemble is 300 K, the FWHM is 10 fs and the pulse is centered at 0 fs. Figure 1 shows the behavior of alignment of D_2 during the first two revivals and its Fourier transform of the alignment parameter. From Fig.1(a), before the aligning pulse is reached, the molecular ensemble is isotropic and $\langle\cos^2\theta\rangle$ is 1/3. When the laser with intensity of

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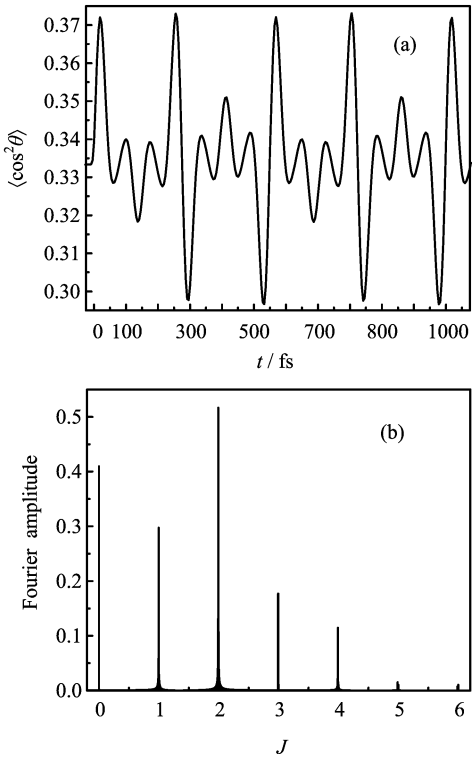

FIG. 1 (a) Calculated time-dependent alignment parameter $\langle\cos^2\theta\rangle$ for D_2 excited by 800 nm, 10 fs, 10^{14} W/cm^2 laser pulses. (b) The rotational states-amplitude from Fourier transform of the alignment.

10^{14} W/cm^2 irradiates the molecule, the electric field of the intense laser induces the dipole moment within the molecule. The induced dipole moments set up a torque on the molecular axis that can be large enough to rotate the molecule of D_2 to the laser polarization direction, so the arrival of the aligning pulse is followed by a prompt net alignment. The molecule continues to rotate even after the laser field is switched off, and then the periodical alignment appears. The revival period of D_2 is about 557 fs and the peak alignment is 0.3721. The Fourier transform of the alignment parameter gave the beat frequencies $\Delta\omega_{JM}$ between adjacent rotational states, where $\Delta\omega_{JM}=(4J+6)\omega_1/2$, and $\omega_1=4\pi Bc$ is the fundamental rotational frequency, B is the rotational constant of molecule by $B=1/(2I)$, I is the moment of inertia of a molecule, and c is speed of light. In the present work, the beat frequencies $\Delta\omega_{JM}$ are replaced by the rotational quantum number J directly. The Fourier amplitude is determined by the corresponding rotational state populations in the coherent rotational wave packet. Figure 1(b) describes the revival structure decomposing into different rotational states. The amplitudes of the Fourier transform of the alignment parameter demonstrated that the even and the odd states were populated differently in the rotational wave packet. Both the alignment and the Fourier

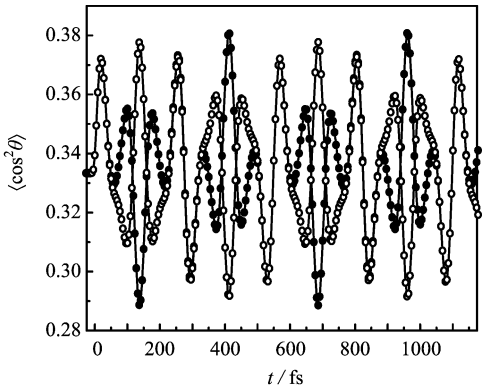

FIG. 2 Calculated time-dependent $\langle\cos^2\theta\rangle$ of D_2 for the odd (○) and even (●) rotational states excited by 800 nm, 10 fs, 10^{14} W/cm^2 laser pulses.

amplitude agree very well with those in Refs.[31,32].

In fact, the Hamiltonian of the laser-molecule interaction demonstrates that the even and the odd rotational states do not couple to each other. The rotational wave packets created by the first laser pulse could be divided into two components categorized as an even and an odd rotational wave packet respectively. Figure 2 shows the time-dependent $\langle\cos^2\theta\rangle$ of D_2 for the odd and even rotational states separately in the first two revivals. Compared with alignment for all rotational states in Fig.1(a), the odd rotational wave packet and the even rotational wave packet exhibit opposite behaviors around the first quarter revival and the three quarters revival. The odd rotational wave packet has the maximum alignment parameter when the even wave packet has the minimum alignment parameter and vice versa. The odd rotational states are different from the evens in the statistics of nuclei spin, so the opposite localizations of the odd and the even wave packets cancel each other out partially, and result in partial alignment for the total rotational wave packet of D_2 at the time of the first quarter revival and the three quarters revival after a single laser pulse excitation. Therefore, when a second ultrashort pulse is turned on around the first quarter revival or the three quarters revival, the even and odd components in the rotational wave packet can be excited selectively. In order to verify this, the following calculation is carried out.

Figure 3 shows the peak alignment $\langle\cos^2\theta\rangle_{\max}$ of D_2 for the odd and even components after excited by the second laser pulse with intensity of 10^{14} W/cm^2 and pulse duration of 10 fs around the first quarter revival of the rotational wave packets created by the first laser pulse, Δt stands for the time delay of the first laser pulse and the second laser pulse. The $\langle\cos^2\theta\rangle_{\max}$ is very sensitive to the time delay Δt and exhibits different dynamics for the odd and even components around the quarter-revival. The peak alignment varies as a function of Δt, which varies dramatically when the time de-

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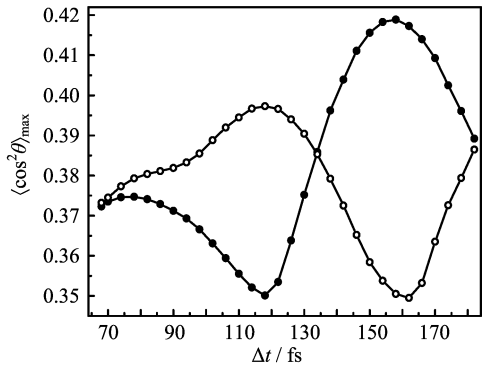

FIG. 3 $\langle\cos^2\theta\rangle_{\max}$ of D_2 for the odd (○) and even (●) components after being excited by the second laser pulse with intensity of 10^{14} W/cm^2 and pulse duration of 10 fs around the first quarter revival of the rotational wave packets created by the first laser pulse.

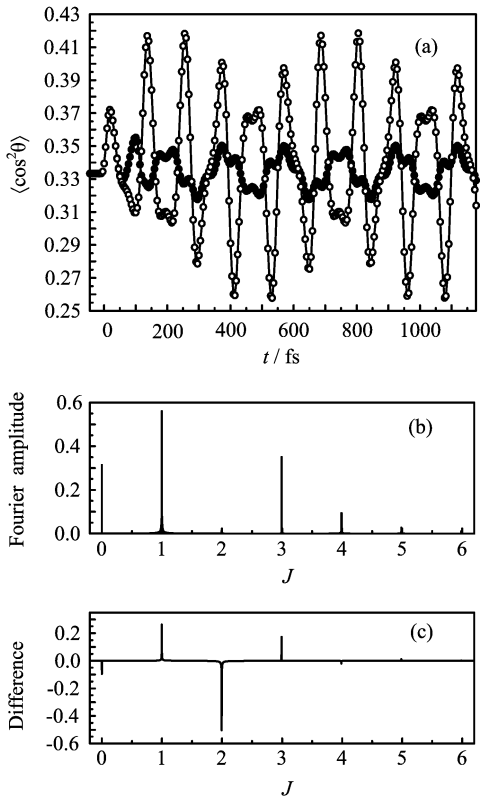

FIG. 4 (a) Calculated time-dependent $\langle\cos^2\theta\rangle$ for D_2 excited by two laser pulses with time delay Δt=118 fs in two revivals (● even, ○ odd). (b) The rotational states amplitude from Fourier transform of the alignment. (c) The difference between the Fourier transforms of the alignment signals excited by two laser pulses and a single laser pulse.

lay Δt changes from 102 fs to 178 fs. When the second laser pulse interacts with the rotational wave packet just "before" and "after" the first quarter revival, the peak alignment for the different components exhibits different behaviors. Just "before" the first quarter revival, the odd wave packet of D_2 rotates from antialignment to alignment, but the even wave packet of D_2 rotates from alignment to antialignment. The second laser pulse increases odd's average rotational energy and favors its motion, but it decreases even's average rotational energy and stops its motion, so the alignment for the odds is enhanced but evens decreased, and the odds prevail on the evens in the rotational wave packet. Just "after" the first quarter revival, the even wave packet rotates from antialignment to alignment, but the odd wave packet rotates from alignment to antialignment, the second laser pulse increases evens average rotational energy and favors its motion, while it decreases odds average rotational energy and stops its motion [27,28], so the alignment for the evens is enhanced but odds decreased, the evens prevail on the odds in the rotational wave packet. The peak alignment for the odd components achieves maximum but the evens reaches minimum when Δt=118 fs. On the contrast, the peak alignment for the even components achieves maximum but the odds reaches minimum when Δt=158 fs. The difference of time delay Δt is 40 fs when the peak alignment for the odd and the even components achieves maximum respectively.

Figure 4(a) shows time evolution of rotational wave packet D_2 irradiated by two laser pulses separated by Δt=118 fs with both pulse duration 10 fs and intensity 10^{14} W/cm^2. The $\langle\cos^2\theta\rangle$ varies between 0.2967 and 0.3721 (see Fig.1). After the rotational wave packet is modified by the second laser pulse, the opposite effects on the odd and the even wave packets created by the first laser pulse are concealed. The $\langle\cos^2\theta\rangle$ varies from 0.2591 to 0.3973 for the odd wave packet and exhibits a subsequently enhanced alignment and antialignment. The $\langle\cos^2\theta\rangle$ varies from 0.3200 to 0.3501 for the even wave packet and exhibits a subsequently decreased alignment and antialignment. From the above, we can conclude that the second laser pulse has opposite effects on the odd and the even rotational wave packets created by the first laser pulse. When the delay Δt=118 fs, the odd rotational wave packet is promoted by the second laser pulse, and so the peak alignment is increased. In contrast, the even rotational wave packet is degraded by the second laser pulse, with the result that the peak alignment is decreased. Figure 4(b) shows the amplitude of rotational states from the Fourier transforms of the alignment parameter. Obviously, after the rotational wave packet is excited by the second laser pulses, the amplitude of odd rotational states increases but that of the evens decreases compared with the one excited by a single laser pulse (see Fig.1). The second laser pulse with time delay Δt=118 fs favors the motion of the odd rotational states. Meanwhile, the second laser pulse stops the motion of the even rotational states. Figure 4(c) shows the difference of the Fourier transforms for the alignment parameter excited by two

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laser pulses and a single laser pulse. The positive values mean that the second laser pulse has a positive effect on the rotational state and increases its population. In contrast, the negative values mean that the second laser pulse has a negative effect on the rotational state and decreases its population. The results clearly demonstrate the effect of the second laser pulse on the rotational wave packet created by the first ultrashort pulse. When the second laser pulse was delayed by Δt=118 fs, the odd rotational wave packet was enhanced and the even rotational wave packet was degraded. Thus the odd states are increased and the evens decreased in the final rotational wave packet created by the two pulses.

Figure 5(a) shows time evolution of rotational wave packet irradiated by two laser pulses separated by Δt=158 fs with both pulse duration 10 fs and intensity 10^{14} W/cm^2. The $\langle\cos^2\theta\rangle$ varies from 0.3168 to 0.3479 for the odd wave packet and exhibits a subsequently decreased alignment and antialignment. In contrast, the $\langle\cos^2\theta\rangle$ varies from 0.2795 to 0.4189 for the even wave packet and exhibits a subsequently enhanced alignment and antialignment. Figure 5(b) shows that the amplitude of rotational states from the Fourier transforms of the alignment parameter. Obviously, after the rotational wave packet is excited by the second laser pulses, the amplitude of even rotational states is increased but the odd is decreased compared with the one excited by a single laser pulse (see Fig.1). Figure 5(c) shows the difference of the Fourier transforms of the alignment parameter excited by two laser pulses and a single laser pulse. The even rotational wave packet was enhanced and the odd rotational wave packet was degraded. The second laser pulse with time delay Δt=158 fs favors the motion of the even rotational states and enhances their peak alignment. Meanwhile, the second laser pulse stops the motion of the odd rotational states and degrades their peak alignment. Thus the final rotational wave packet excited by the two laser pulses was mainly composed of the even rotational states.

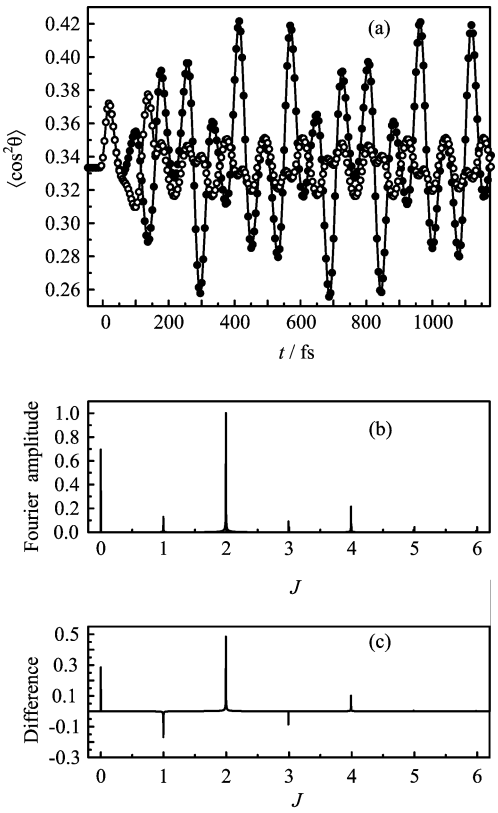

FIG. 5 (a) Calculated time-dependent $\langle\cos^2\theta\rangle$ for D_2 excited by two laser pulses with time delay Δt=158 fs in two revivals (• even, ◦ odd). (b) The rotational states amplitude from Fourier transform of the alignment. (c) The difference between the Fourier transforms of the alignment signals excited by two laser pulses and a single laser pulse.

In order to investigate alignment character irradiated by the second ultrashort pulse around the three quarters revival of the rotational wave packet, the following calculation was carried out. Figure 6 shows the $\langle\cos^2\theta\rangle_{\max}$ of D_2 for the odd and even components after excited by the second laser pulse with intensity of 10^{14} W/cm^2 and pulse duration of 10 fs around the three quarters revival of the rotational wave packet created by the first laser pulse, Just as in Fig.3, the $\langle\cos^2\theta\rangle_{\max}$ for the odd and even components varies dramatically when the time delay Δt changes from 375 fs to 450 fs. When the second laser pulse interact with the rotational wave packet just "before" and "after" the three quarters revival, the peak alignment for the odds and evens exhibits different behaviors. Just "before" the three quarters revival, the even wave packet is enhanced but the odd wave packet decreased. Just "after" the three quarters revival, the even wave packet is decreased but the odd wave packet enhanced. The peak alignment for the evens achieves maximum but that of the odds reaches minimum when Δt=390 fs. On the contrary, the peak alignment for the

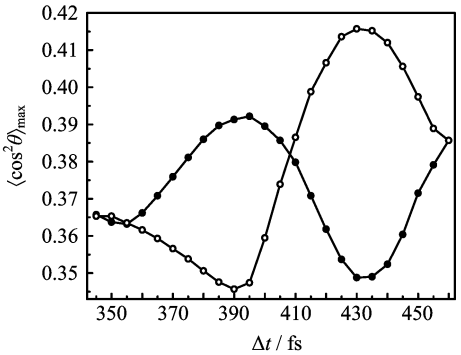

FIG. 6 The $\langle\cos^2\theta\rangle_{\max}$ of D_2 for the odd (◦) and even (•) components after being excited by the second laser pulse with intensity of 10^{14} W/cm^2 and pulse duration of 10 fs around the three quarters revival of the rotational wave packets created by the first laser pulse.

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odds achieves maximum but that of the evens reaches minimum when Δt=430 fs. Similarly, the difference of time delay Δt is 40 fs when the peak alignment for the even and odd rotational wave packet achieves maximum. This is similar to the second laser pulse interaction with the rotational wave packets induced by the first laser pulse around the first quarter revival (see Fig.3), but with the odds and evens reversed.

Figure 7(a) shows the time evolution of rotational wave packet of D_2 irradiated by two laser pulses separated by Δt=390 fs with both pulse duration 10 fs and intensity 10^{14} W/cm^2. The $\langle\cos^2\theta\rangle$ exhibits a subsequently decreased alignment and antialignment for the odd wave packet, but a subsequently enhanced alignment and antialignment for the even wave packet. Figure 7 (b) and (c) shows the even rotational wave packet was enhanced and the odd rotational wave packet was degraded just as time delay Δt=158 fs. As shown in Figs.5 (b) and (c), the final rotational wave packets excited by the two laser pulses were mainly composed of the even rotational states. Figure 8(a) shows the time evolution of rotational wave packet irradiated by two laser pulses separated by Δt=430 fs. The alignment parameter exhibits a subsequently enhanced alignment and antialignment for the odd wave packet, but a subsequently decreased alignment and antialignment for the even wave packet. Figure 8 (b) and (c) shows the odd rotational wave packet was enhanced and the even rotational wave packet was degraded exactly when Δt is 118 fs. As Figs.4 (b) and (c) show, the odds prevail on the evens in the final rotational wave packet created by the two pulses. Different rotational components exhibit different dynamical behaviors during the evolution of the rotational wave packet. The odd and the even rotational states evolve in phase around full revival times but out of phase around the fractional revival times. When the second laser interacts with the already rotating wave packets around the other fractional revival, the dynamics should be the same as what was discussed above. Rotational revivals can also be applied to most linear molecules even nonlinear molecules. The current selectivity should be further pronounced as long as further optimization and better selectivity ratio can be achieved by optimizing the modifying aligning laser pulse, or using multipulse alignment [18,25,36].

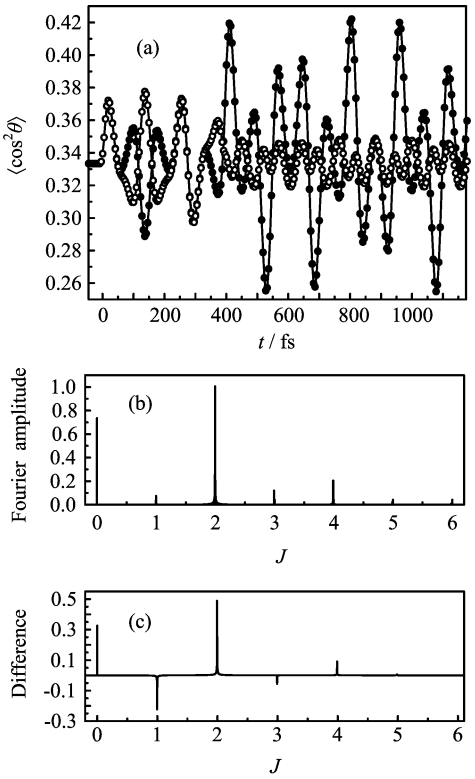

FIG. 7 (a) Calculated time-dependent $\langle\cos^2\theta\rangle$ for D_2 excited by two laser pulses with time delay Δt=390 fs in two revivals (• even, ◦ odd). (b) The rotational states amplitude from Fourier transform of the alignment. (c) The difference between the Fourier transforms of the alignment signals respectively excited by two laser pulses and a single laser pulse.

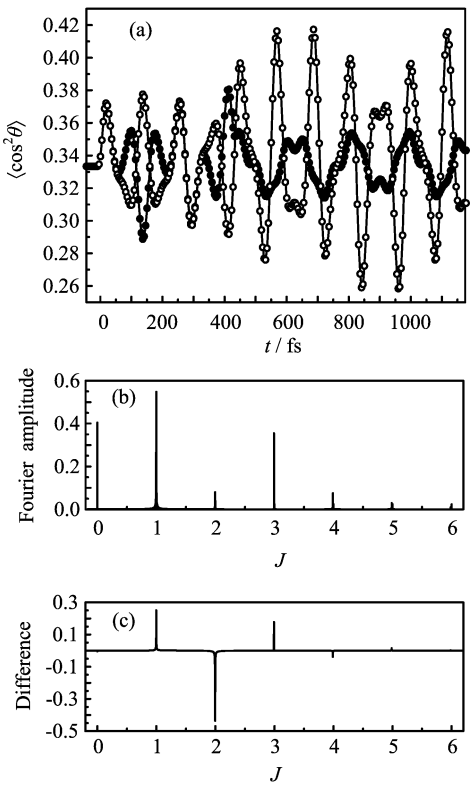

FIG. 8 (a) Calculated time-dependent $\langle\cos^2\theta\rangle$ for D_2 excited by two laser pulses with time delay Δt=430 fs in two revivals (• even, ◦ odd). (b) The rotational states amplitude from Fourier transform of the alignment. (c) The difference between the Fourier transforms of the alignment signals respectively excited by two laser pulses and a single laser pulse.

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IV. CONCLUSION

The relative populations between even and odd rotational states can be effectively controlled in a coherent rotational wave packet for D_2 by adaptively selecting the time delay between the two laser pulses. When the time delay is 118 and 430 fs, the second laser pulse stops the motion of the even rotational states and degrades their peak alignment, so the odds states are increased and the evens are decreased in the final rotational wave packet created by the two pulses. When the time delay is 158 and 390 fs, the second laser pulse stops the motion of the odd rotational states and degrades their peak alignment, so the even states are increased and the odds are decreased in the final rotational wave packet created by the two pulses, resulting in a final coherent rotational wave packet mainly composed of even rotational states. It can be concluded that rotational fractional revivals offer an excellent opportunity to discriminate between the odd and even rotational states, which can be applied to most other linear molecules even top molecules.

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