

## ARTICLE

**Dynamic Alignment of D<sub>2</sub> Enhanced by Two Few-cycle Pulses**

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Dynamic alignment of D<sub>2</sub> induced by two few-cycle pulses was investigated by solving the time-dependent Schrödinger equation numerically based on a rigid rotor model. The results show that alignment of D<sub>2</sub> can be enhanced by two few-cycle pulses compared with the level achievable by a single few-cycle pulse as long as the time delay between two pulses is chosen properly, and the pulse duration of two lasers plays an important role in the aligning process of D<sub>2</sub> molecules.

**Key words:** Dynamic alignment, Few-cycle pulses, Pulse duration**I. INTRODUCTION**

The interaction of molecules with non-resonant femtosecond laser pulses has been investigated in the last decade. Particularly, laser-induced spatial alignment is a greatly intriguing area because of its potential applications. Certain key properties of molecules, such as ionization and dissociation [1,2], high-order harmonic generation [3,4], quantum information [5], and chemical-reaction dynamics [6-8], are affected greatly by the molecular alignment and orientation, so molecular alignment and orientation play an important part in all these fields. Although there are many ways to manipulating molecular alignment and orientation at present, one effective way for manipulating molecular alignment is by using a laser field [9]. In recent years, many fundamental physical processes have been understood by means of a variety of experimental techniques [10-13] and theoretical methods [14,15]. For the majority of these studies, a femtosecond laser pulse (much less than the rotational period of molecules) is used to interact with a molecular ensemble, which results in a rotational wavepacket. The wavepacket will not disappear completely; instead, a periodical revival is achieved after the laser is turned off. As a result, dynamic alignment of molecules is formed which provides a convenient means for the research of relationships between molecular alignment and properties of molecules in laser fields. In general,  $\langle \cos^2\theta \rangle$  is used to describe the extent of alignment of a molecular ensemble, where  $\theta$  is the angle between the molecular axis and polarization direction of laser and the angle brackets stand for the statistical thermal average over the whole ensemble.

Many studies show that the degree of alignment is closely related to the parameters of the laser, so excellent molecular alignment can be realized through se-

lecting the proper parameters of laser, such as increasing laser intensity [16], selecting proper pulse duration [17], and reducing temperature of the molecular ensemble [18]. But all of these means are limited in certain conditions: molecules may be ionized and dissociated if laser intensity is too strong, pulse duration may be too long to achieve the periodical revival, and reducing temperature may be limited by the experimental environment. Therefore, many methods have been suggested to achieve excellent molecular alignment. Some researchers presented optimal control theory [19,20] to optimize the laser pulse and enhanced the degree of alignment, but it is very difficult to realize in experiment. There have recently been proposals to modify the normal evolution of rotational wave packets by applying additional laser pulses to the rotating molecules at selected times after the first pump pulse [21,22]. One predicted result was that these extra pulses could be designed to successively increase the peak alignment beyond the level achievable by a single aligning pulse. In what follows, enhanced molecular alignment induced by two time-delayed laser pulses has been observed in experiment [23-26]. Both the time delays and laser intensity ratio of the two pulses play a significant role in achieving enhanced alignment, but pulse duration of the two pulses is not investigated.

Recently, dynamic alignment of much lighter D<sub>2</sub> molecules induced by a few-cycle pulses has been observed [27]. Being the most fundamental molecule, the deuterium molecule has far-reaching implications in physics, including the precise time-domain spectroscopy [28] and the stability of physical constants and cosmology [29]. But because of the low polarizability and anisotropy of deuterium molecule, it is very difficult to achieve excellent alignment by only one aligning pulse. In this work, in order to study the behavior of dynamic alignment of D<sub>2</sub> induced by two few-cycle femtosecond lasers, the alignment of D<sub>2</sub> was investigated by solving the time-dependent Schrödinger equation numerically based on a rigid rotor model. Different pulse durations were used in order to enhance the extent of molecular

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alignment on the condition that the intensity of the two lasers was conserved.

## II. THEORETICAL METHOD

The molecule can be modeled as rigid rotors based on the fact that the rotational motion of molecules occurs in a weak laser field, provided that the molecules do not exhibit ionization, electronic and vibrational excitation. For homonuclear diatomic molecule, the permanent dipole moment is zero because of the property of the centrosymmetrical structure. The field-molecule interaction potential is given by

$$V(\theta) = -\frac{E^2(t)}{2}(\alpha_{//} \cos^2 \theta + \alpha_{\perp} \sin^2 \theta) \quad (1)$$

where  $\alpha_{//}$  and  $\alpha_{\perp}$  are parallel and perpendicular to the molecular axis, respectively. For diatomic molecules [30],  $\alpha_{//} = \alpha_{zz}$ ,  $\alpha_{\perp} = \alpha_{xx} = \alpha_{yy}$  ( $x$ - $y$ - $z$  is fixed in the molecule,  $z$  axis is the direction of molecular axis).  $E(t)$  is the laser intensity strength and  $\theta$  is the angle between the molecular axis and laser field direction.

According to quantum dynamics, the Hamiltonian of rigid rotors in laser field is given by

$$\hat{H}(t) = BJ^2 + V(\theta) \quad (2)$$

where  $B$  is the rotational constant of molecule by  $B=1/(2I)$ , and  $I$  is the moment of inertia of a molecule.

The time-dependent Schrödinger equation of a rigid rotor in laser field is given by

$$i \frac{\partial \Psi(\theta, \varphi, t)}{\partial t} = \hat{H}(t) \Psi(\theta, \varphi, t) \quad (3)$$

where  $\varphi$  is azimuthal angle of molecular axis in a fixed frame.

The time-dependent Schrödinger Eq.(3) can be solved using the pseudospectral method [31,32] 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:

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

where  $\hat{H}_0 = B\hat{J}^2$ , which is the kinetic energy operator, and  $\hat{V}$  is the remaining Hamiltonian depending on the spherical coordinates only. Eq.(4) shows that the propagation of the wavefunction from  $t$  to  $(t+\Delta t)$  is accomplished by three steps: (i) First the wavefunction

$\Psi(\theta, \varphi, t)$  is propagated for a half-time step  $\Delta t/2$  in the energy space spanned by  $\hat{H}_0$ :

$$\exp\left(\frac{-i\hat{H}_0\Delta t}{2}\right)\Psi(\theta, \varphi, t) = \Psi_1(\theta, \varphi, t) \quad (5)$$

(ii) Then the wavefunction  $\Psi_1(\theta, \varphi, t)$  is transformed back to the coordinate space and propagated for a time step  $\Delta t$ :

$$\exp\left[-i\hat{V}\left(\theta, \varphi, t + \frac{\Delta t}{2}\right)\Delta t\right]\Psi_1(\theta, \varphi, t) = \Psi_2(\theta, \varphi, t) \quad (6)$$

(iii) Finally the wavefunction  $\Psi_2(\theta, \varphi, t)$  is transformed back to the energy space again spanned by  $\hat{H}_0$  and propagated for another half-time step  $\Delta t/2$ :

$$\exp\left(\frac{-i\hat{H}_0\Delta t}{2}\right)\Psi_2(\theta, \varphi, t) = \Psi(\theta, \varphi, t + \Delta t) \quad (7)$$

The propagation of the wavefunction is accomplished by repeating Eq.(4) until the end of laser field, and time-dependent wavefunction of the rigid rotor of molecule is obtained.

$$\Psi(\tau_p) = \sum_J C_J |JM\rangle \quad (8)$$

where  $|JM\rangle$  is the wavefunction field-free rotational states and  $\tau_p$  is the time when laser is turned off.

After the pulse is turned off, the molecular wavefunction continues to propagate in the free space

$$\Psi(t + \tau_p) = \sum_J C_J e^{-iBJ(J+1)t} |JM\rangle \quad (9)$$

For homonuclear diatomic molecules, the degree of molecular alignment of a whole ensemble can be described by  $\langle \cos^2 \theta \rangle$ . For different states,  $\langle \cos^2 \theta \rangle_{JM}$  averaging thermally over the  $|JM\rangle$  is given by

$$\begin{aligned} \langle \cos^2 \theta \rangle_{JM} = & \int_0^\pi \cos^2 \theta \sin \theta d\theta \cdot \\ & \int_0^{2\pi} |\Psi_{JM}(\theta, \varphi, t)|^2 d\varphi \end{aligned} \quad (10)$$

Assuming the Boltzman distribution of the rotational states at the initial time, the alignment parameter at a given temperature is defined by

$$\langle \cos^2 \theta \rangle = Q^{-1} \sum_J \exp\left[\frac{-BJ(J+1)}{k_B T}\right] \sum_{M=-J}^J \langle \cos^2 \theta \rangle_{JM} \quad (11)$$

$$Q = \sum_J (2J+1) \exp\left[\frac{-BJ(J+1)}{k_B T}\right] \quad (12)$$

here,  $Q$  is the partition function,  $k_B$  and  $T$  are the Boltzmann constant and ensemble temperature, respectively. For the alignment parameter,  $\langle \cos^2\theta \rangle = 1/3$  reflects an isotropic distribution of molecular alignment, whereas  $\langle \cos^2\theta \rangle = 1$  indicates complete alignment. The limiting case in which all molecules are perpendicular to the alignment axis, called antialignment, is denoted by  $\langle \cos^2\theta \rangle = 0$ . In general cases,  $\langle \cos^2\theta \rangle$  is between 0 and 1.

### III. RESULTS AND DISCUSSION

The technique of chirped pulse amplifier can be used to obtain different pulse duration in experiment. In this paper, the alignment of D<sub>2</sub> molecules is calculated using the methods above. In our calculation, the laser pulse takes the Gaussian form:

$$E(t) = E_1 e^{-2 \ln 2 [(t-t_{01})/t_{w1}]^2} \cos \omega t + E_2 e^{-2 \ln 2 [(t-t_{02})/t_{w2}]^2} \cos \omega t \quad (13)$$

where  $E_1$  and  $E_2$  are the peak intensity of the two lasers,  $t_{w1}$  and  $t_{w2}$  are pulse duration (FWHM) of the two lasers, and  $t_{01}$  and  $t_{02}$  are the centers of the pulses, respectively.  $\omega$  is the frequency of the laser and the wavelength is 800 nm.

Firstly, dynamic alignment of D<sub>2</sub> induced by a single few-cycle pulse was calculated. In the calculation, the pulse duration (FWHM) is 10 fs and the pulse is centered at 0 fs. Figure 1 shows the behavior of alignment of D<sub>2</sub> during the two revival periods. Before the aligning pulse is reached, the molecular ensemble is isotropic and alignment parameter  $\langle \cos^2\theta \rangle$  is 1/3. The arrival of the aligning pulse is followed by a prompt net alignment and then the periodical alignment appears. The revival period of D<sub>2</sub> is about 0.55 ps. When the laser with intensity  $10^{14}$  W/cm<sup>2</sup> interacts with molecules, the maximum alignment is 0.3758. When the laser intensity is increased to  $2 \times 10^{14}$  W/cm<sup>2</sup>, the peak alignment is 0.4189 which agrees very well with Ref.[27]. Obviously, the degree of alignment is enhanced through increasing

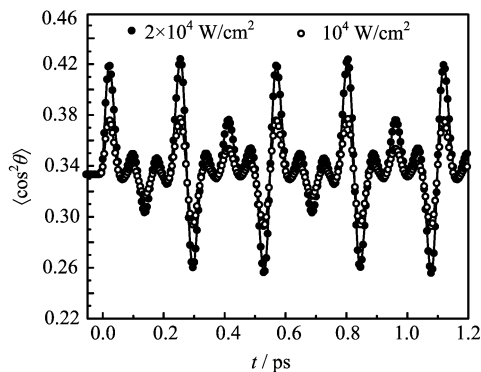


FIG. 1 Alignment of D<sub>2</sub> induced by a single few-cycle pulses with FWHM=10 fs.

laser intensity, but the aligning extent is not excellent because of the low polarizability and anisotropy of D<sub>2</sub>. If we increase laser intensity further, the molecules may be ionized and even dissociated. Therefore, two few-cycle pulses were used in order to enhance the degree of alignment.

In the calculation for two few-cycle pulses, the peak intensity of the first laser is  $10^{14}$  W/cm<sup>2</sup> and the second  $2 \times 10^{14}$  W/cm<sup>2</sup>, and both the pulse durations are 10 fs. We apply the second aligning pulse around the first peak alignment and investigate the alignment in the second aligning period; where the first pulse is centered at 0 fs. Figure 2(a) shows the maximum alignment of D<sub>2</sub> induced by two aligning pulses as a function of time  $t$  and time delays  $\Delta t$  between the two aligning pulses. From Fig.2(a), we can see that the peak alignment has the same tendency with time for certain time delays between the two aligning pulses. Each set of revivals differs by the location in time of the second aligning pulse  $\Delta t$ , which came from 0.464 ps to 0.587 ps after the first pulse. As can be seen by the peak in Fig.2(a), alignment was only enhanced for certain time delays between the two aligning pulses, while other time delays

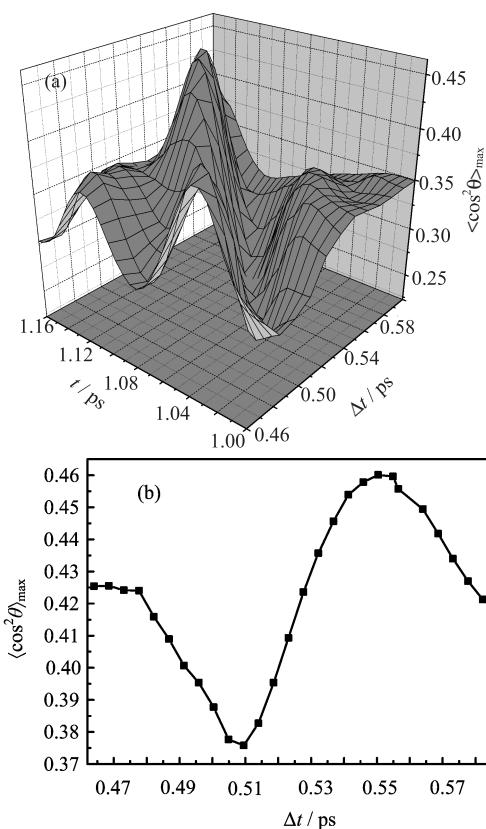


FIG. 2 (a) Peak alignment of D<sub>2</sub> as a function of time  $t$  and time delays  $\Delta t$  induced by two few-cycle pulses. (b) Peak alignment of D<sub>2</sub> as a function of time delays  $\Delta t$  highlighted from (a).

did not enhance the alignment. All of these are the same as those of  $N_2$  referred to in Ref.[23]. Different aspects of Fig.2(a) are highlighted in Fig.2(b). From Fig.2(b), we can see that the peak alignment reaches maximum when  $\Delta t$  is 0.5504 ps, but the peak alignment comes to minimum when  $\Delta t$  is 0.5095 ps.  $\langle \cos^2\theta \rangle_{\max}$  is merely 0.3758 ( $\Delta t=0.5095$  ps) which is less than the level induced by a single pulse with intensity of  $2 \times 10^{14}$  W/cm<sup>2</sup> and we can not enhance the degree of alignment effectively. However,  $\langle \cos^2\theta \rangle_{\max}$  is 0.4601 ( $\Delta t=0.5504$  ps) which are increased by about 22% and 10% compared with the level by a single pulse with intensity of  $10^{14}$  and  $2 \times 10^{14}$  W/cm<sup>2</sup>, respectively. As a result, molecular alignment is enhanced. When the time delay lies between 0.5413 and 0.5640 ps,  $\langle \cos^2\theta \rangle_{\max}$  is more than 0.45, which means the molecular alignment is increased effectively by applying the second pulse in this range of time delays. This is because a rotational wavepacket produced after the first aligning pulse interacted with the molecular ensemble of  $D_2$ . When the time delay is from 0.5413 ps to 0.5640 ps, the wavepackets begin to rephase, and the majority of molecules is aligned at the direction of laser polarization with a small angle  $\theta$ . According to the classic rigid rotor model [22], the first pulse prealigns the molecules in the harmonic region of the angular interaction potential while the second pulse completes the angular focusing. As a result, molecules rotate to the laser polarization direction at the same time and molecular alignment is enhanced. However, if the second laser is applied at other time delays when the angle between the axis of molecules and the laser polarization direction lies in a large region, the second pulse can not realize the angular squeezing effectively and alignment will not be enhanced perfectly.

Figure 3 shows the alignment behavior of  $D_2$  induced by two pulses both with pulse duration 10 fs near the second revival. The two-pulse revivals which had the strongest alignment ( $\Delta t=0.5504$  ps) and antialignment ( $\Delta t=0.5095$  ps). The revivals that was induced by applying only one pulse are also shown in Fig.3. The corresponding revival from only the first pulse was similar in shape and amplitude to the one by using two lasers. This plot is shown to illustrate the degree of increase in the quality of the revivals. The time when alignment parameter  $\langle \cos^2\theta \rangle$  comes to maximum is different, which is mainly due to the difference of time when the second pulse interacts with molecules and strong torques on the molecular axis force the molecule to align along the laser polarization direction at different time.

From the above, we can see that molecular alignment of  $D_2$  can be increased by applying two pulses with the same pulse duration as a single pulse, as long as the time delays between the two lasers is selected properly. We should note, however, that the enhanced alignment is not ideal because of the low polarizability and anisotropy of deuterium molecules. In fact, pulse duration plays an important role in the aligning process [33], and for sufficiently short pulses the degree of ro-

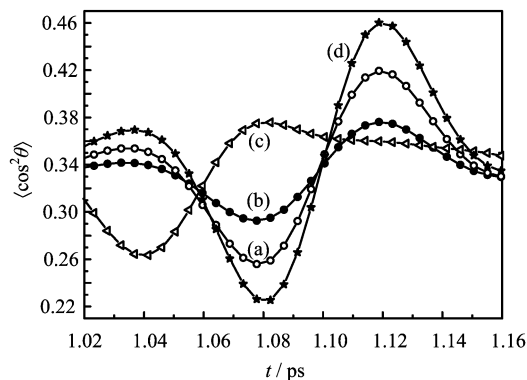


FIG. 3 Alignment parameter of  $D_2$  as a function of time  $t$  near the second revival. (a) and (b) is revival by a single pulse with laser intensity of  $2 \times 10^{14}$  and  $10^{14}$  W/cm<sup>2</sup>, respectively. (c) and (d) is revival by two pulse with laser intensity of  $2 \times 10^{14}$  and  $10^{14}$  W/cm<sup>2</sup> with  $\Delta t=0.5095$  and 0.5504 ps, respectively. FWHM=10fs.

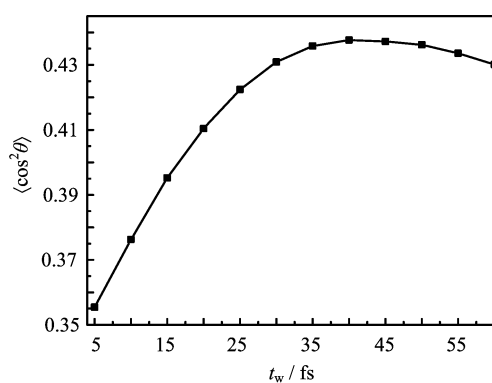


FIG. 4 Peak alignment parameter of  $D_2$  as a function of pulse duration on the condition that the laser intensity is fixed as  $10^{14}$  W/cm<sup>2</sup>.

tational excitation depends on the total energy rather than upon the peak intensity of the laser pulse. Thus, for a given intensity, the degree of alignment is limited by the pulse duration.

Figure 4 shows peak alignment of  $D_2$  induced by one laser with intensity of  $10^{14}$  W/cm<sup>2</sup> as a function of the pulse duration. The peak alignment varies with pulse duration and reaches maximum when the pulse duration is 40 fs (about 0.07 h/2B). In order to understand the role of the pulse duration in two-pulse alignment, the alignment of  $D_2$  by two lasers with different pulse duration is investigated, and in our calculations, the pulse duration of the first laser is fixed as 10 fs.

Figure 5 shows peak alignment of  $D_2$  induced by two lasers with different pulse durations, with the pulse duration of the first laser fixed as 10 fs, the intensity of the two lasers intensity fixed as  $10^{14}$  and  $2 \times 10^{14}$  W/cm<sup>2</sup>, respectively. From Fig.5, we can see that peak alignment  $\langle \cos^2\theta \rangle_{\max}$  increases when the pulse duration of the second laser ( $t_{w2}$ ) varies from 6 fs to 35 fs but de-

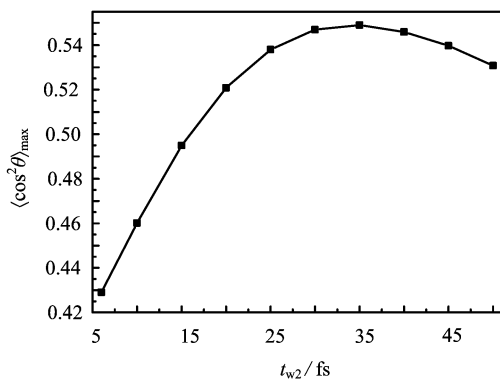


FIG. 5 Peak alignment parameter of D<sub>2</sub> as a function of pulse duration of the second laser on the condition that the pulse duration of the first laser is fixed as 10 fs.

increases from 35 fs to 50 fs, and the alignment is enhanced compared with the level only by one pulse as long as time delays is selected correctly. The alignment parameter achieves a maximum when  $t_{w2}=35$  fs (about  $0.06 h/2B$ ). In this case, the pulse duration of the second laser may be not too long or too short in order to enhance the extent of alignment. In general, the pulse duration of the first laser should be shorter compared with the second laser. As a result, stronger torques on the molecular axis forces the molecule to align along the laser polarization direction and excellent alignment is achieved.

Figure 6 shows the aligning behavior of D<sub>2</sub> induced by two pulses with different pulse duration near the second revival. While the duration of the two pulses is 10 and 35 fs, alignment of D<sub>2</sub> is increased perfectly,  $\langle \cos^2 \theta \rangle_{\max}$  is 0.5490 which is increased by about 46% and 31% compared with the level by a single pulse with intensity  $10^{14}$  and  $2 \times 10^{14}$  W/cm<sup>2</sup>, respectively. When  $t_{w2}=6$  fs, alignment can not be enhanced perfectly just as the case with  $t_{w2}=35$  fs. We can also see that the time when alignment parameter  $\langle \cos^2 \theta \rangle$  comes to maximum is different, which is also due to the difference of time when the second pulse interacts with molecules and torques on the molecular axis force the molecule to align along the laser polarization direction at different times. If the lasers' intensity is optimized for D<sub>2</sub>, more excellent alignment can be obtained and D<sub>2</sub> molecules can be even better aligned. For other molecules interacting with two lasers, the dynamic alignment should exhibit the same characteristic, but since different molecules have different polarizability anisotropy and rotational constants, the optimum pulse duration of the two lasers is different. The enhanced alignment has immediate practical implications for improved experiments using aligned molecules, but enhanced alignment is only part of a more general result that rotational revival structures can be modified by using a second laser pulse. Enhanced alignment is one possible effect, but there are infinitely many possible pulse combinations avail-

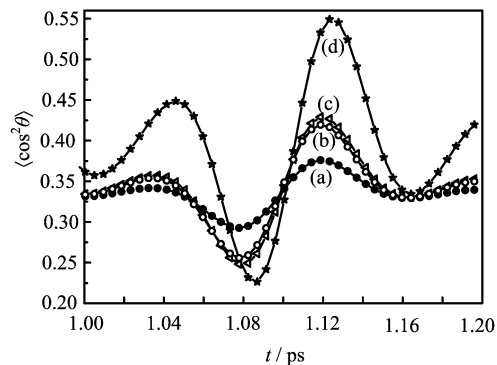


FIG. 6 Alignment parameter of D<sub>2</sub> as a function of time  $t$  near the second revival. (a) and (b) is revival by a single pulse with laser intensity  $10^{14}$  and  $2 \times 10^{14}$  W/cm<sup>2</sup>, respectively. (c) and (d) is revival by two pulses with laser intensity  $2 \times 10^{14}$  and  $10^{14}$  W/cm<sup>2</sup>. The first pulse duration (FWHM) is fixed as 10 fs, and the second pulse duration of (d) and (c) are 35 and 6 fs, respectively.

able. In the next study, we will apply the second pulse at a fractional revival, where components of the rotational wavepacket can be addressed independently [34], which may let us switch revivals on and off coherently. Such modified revivals have implications not only for alignment, but also for using revivals as a new type of quantum logic system [35]. Because the recollision between the electron and its parent ion play an important part in correlated multielectron ionization in strong laser field [36,37], the entanglement approach can be applied to exploit the correlation between the electronic and nuclear wave packets within the attosecond time scale.

#### IV. CONCLUSION

Alignment of D<sub>2</sub> was investigated by two few-cycle pulses. The investigation verifies that molecular alignment of D<sub>2</sub> can be enhanced by applying two time delay-selected few-cycle pulses, and also the pulse duration play an important role in the aligning process. Alignment of D<sub>2</sub> can be enhanced perfectly by adjusting the pulse duration as 10 and 35 fs on the condition that the intensity of the two lasers are  $10^{14}$  and  $2 \times 10^{14}$  W/cm<sup>2</sup>, respectively. We will continue to investigate modified revivals by the second pulse at a fractional revival where components of the rotational wavepacket can be addressed independently and the revivals may be switched on and off coherently.

#### V. ACKNOWLEDGMENTS

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