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2011 J. Phys. B: At. Mol. Opt. Phys. 44 055403

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Field-free alignment in linear molecules by a square laser pulse

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Received 12 November 2010, in final form 22 January 2011

Published 15 February 2011

Online at stacks.iop.org/JPhysB/44/055403

Abstract

In this paper, we propose a square laser pulse to realize the field-free alignment in linear molecules. We show that the molecular alignment after the laser pulse can be obtained in both non-adiabatic and adiabatic regimes. Comparing with the conventional Gaussian laser pulse, the maximum degree of the molecular alignment can be enhanced with the same laser energy in the short laser pulse and the same laser intensity in the long laser pulse. We also show that, by observing the time evolution of the molecular alignment during the interaction with a long laser pulse, the optimum pulse duration and its corresponding alignment degree at the full revival time for a given laser intensity and rotational temperature can be directly obtained.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Molecular alignment has attracted considerable attention due to its extensive application on chemical reaction dynamics, ultrafast optics, high-harmonic generation and surface processing [1]. An intense nonresonant laser field has been proven the most versatile tool to create the field-free molecular alignment along the laser polarization direction, and the field-free molecular alignment can be realized in both non-adiabatic and adiabatic regimes [2–10]. In the non-adiabatic regime, i.e. the laser pulse duration is much smaller than the rotational period of the molecule, the maximum degree of the molecular alignment is obtained after the laser pulse and repeatedly constructed at multiple revival periods by a periodic dephasing and rephasing of the rotational wave packet. In the adiabatic regime, i.e. the laser is slowly turned on and off as compared to the rotational period of the molecule, the maximum degree of the molecular alignment is reached during the interaction with the laser pulse, but whether the molecular alignment after the laser pulse occurs depends on the descending edge of the laser pulse; it can be obtained for the slow turn-on and rapid turn-off laser pulse [11], while it will disappear for the Gaussian laser pulse [7].

For further application of the aligned molecules, it is also crucial to obtain as high a degree of the molecular

alignment as possible under the free-field condition. This requires the lowest rotational temperature of the aligned molecules and the optimal intensity and duration of the laser pulse. Furthermore, some complicated laser fields are usually suggested to further enhance the molecular alignment without increasing its intrinsic saturation and ionization rates, such as the laser pulse trains with the pulse separations that are approximated to the rotational revival period [12–18] or the shaped laser pulses by the spectral phase modulation [19–21].

In this work, we utilize a square laser pulse to achieve the field-free molecular alignment. Now it is possible to obtain the square laser pulse by tailoring the ultrashort laser pulse in the frequency domain [22]. It is shown that the molecular alignment after the laser pulse can be obtained in both non-adiabatic and adiabatic regimes. As compared to the conventional Gaussian laser pulse, the maximum degree of the molecular alignment created by the square laser pulse can be enhanced with the same laser energy in the short laser pulse and the same laser intensity in the long laser pulse. It is also shown that the time evolution of the molecular alignment during the interaction with a long laser pulse can directly reflect the optimum pulse duration and its corresponding alignment degree at the full revival time for a given laser intensity and rotational temperature.

2. Theoretical model

We consider a linearly polarized laser field with a square pulse envelope, given by

$$E(t) = A_{\text{am}} \text{rect}(t/\tau) \cos \omega t, \quad \text{rect}(t/\tau) = \begin{cases} 0 & |t| > \tau/2 \\ 1/2 & |t| = \tau/2, \\ 1 & |t| < \tau/2 \end{cases} \quad (1)$$

where A_{am} is the field amplitude, τ is the pulse duration of the square laser pulse and ω is the laser centre frequency. When a linear molecule is subjected to the laser field $E(t)$ in equation (1), the time-dependent Schrödinger equation can be expressed based on a rigid-rotor model as

$$i\hbar \frac{\partial |\Psi(\theta, t)\rangle}{\partial t} = H(t) |\Psi(\theta, t)\rangle. \quad (2)$$

Here, $H(t)$ is the total Hamiltonian of the molecule interacting with the laser field $E(t)$, and is written as

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

where B is the rotational constant of the molecule, J_i is the angular momentum, μ is the permanent dipole moment, θ is the angle between the molecular axis and the polarization direction of the laser field and α_{\parallel} and α_{\perp} are the polarizability components parallel and perpendicular to the molecular axis, respectively. In equation (3), the first term is the rotational energy of the molecule, and the second and third terms respectively represent the interaction potentials with the permanent dipole moment and the polarizability. To eliminate the artificial orienting effect, the pulse duration τ of the square pulse is much larger than the reciprocal of its frequencies $\omega/2\pi$ (i.e. $\tau \gg 2\pi/\omega$); thus, the permanent dipole interaction integrated over τ becomes zero. Finally, the degree of the molecular alignment is defined by the expectation value of $\cos^2 \theta$, and is given by

$$\langle \cos^2 \theta \rangle = \sum_{J_i} \frac{g_{J_i} \exp[-B J_i(J_i + 1)/kT]}{Q} \times \sum_{M_i=-J_i}^{J_i} \langle \Psi_{J_i M_i} | \cos^2 \theta | \Psi_{J_i M_i} \rangle, \quad (4)$$

where Q is the rotational partition function and g_{J_i} is the spin degeneracy factor.

In our simulation, the time-dependent Schrödinger equation in equation (2) is numerically solved by a split-operator method [23, 24]. We take the N_2 molecule as an example, and the molecular parameters are $B = 1.989 \text{ cm}^{-1}$, $\alpha_{\parallel} = 2.38 \text{ \AA}^3$ and $\alpha_{\perp} = 1.45 \text{ \AA}^3$ [7]. Thus, the rotational period of the N_2 molecule can be calculated as $T_{\text{rot}} = 1/(2BC) \approx 8.38 \text{ ps}$, where C is the speed of light. The centre frequency ω of the laser field is $12\,500 \text{ cm}^{-1}$, corresponding to the centre wavelength of 800 nm .

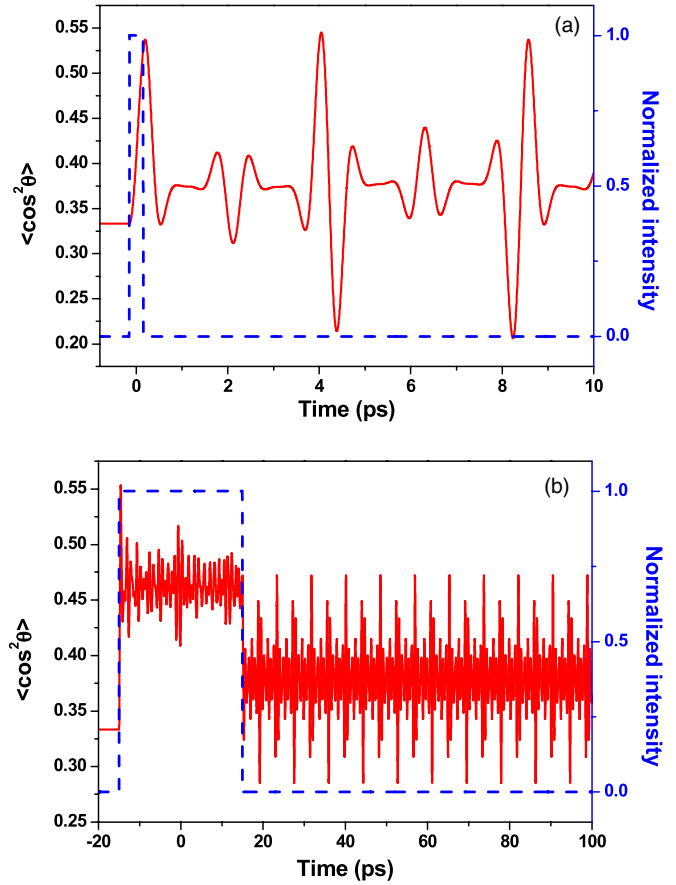


Figure 1. Time evolution of the molecular alignment for the rotational temperature $T = 50 \text{ K}$ and the laser intensity $I = 1 \times 10^{13} \text{ W cm}^{-2}$ with the pulse duration $\tau = 300 \text{ fs}$ (a) and 30 ps (b).

3. Results and discussion

Figure 1 shows the time evolution of the molecular alignment created by the square laser pulse for the rotational temperature $T = 50 \text{ K}$ and the laser intensity $I = 1 \times 10^{13} \text{ W cm}^{-2}$ with the pulse duration $\tau = 300 \text{ fs}$ (a) and 30 ps (b), corresponding to the non-adiabatic and adiabatic alignments. As can be seen, the molecular alignment after the laser pulse can be obtained in both the non-adiabatic and adiabatic regimes. This observation is different from the molecular alignment created by the conventional Gaussian laser pulse, where the molecular alignment after the laser pulse will vanish in the adiabatic regime [7]. Furthermore, it can be found from figure 1(b) that the revival structure of the molecular alignment in the adiabatic regime becomes more complicated and is sensitive to the pulse duration. This result indicates that the laser pulse duration in the adiabatic regime can tremendously change the time evolution of the rotational wave packet after the laser pulse.

To further apply the aligned molecules, it is important to ensure that the degree of the molecular alignment obtained under the field-free laser field is as high as possible. The maximum degree of the molecular alignment depends on the laser energy rather than the laser intensity in a sufficiently short laser pulse (i.e. $\tau^2 \ll 8B/[(\alpha_{\parallel} - \alpha_{\perp})(\pi A_{\text{am}})^2]$), while it only relies on the laser intensity and is independent on the

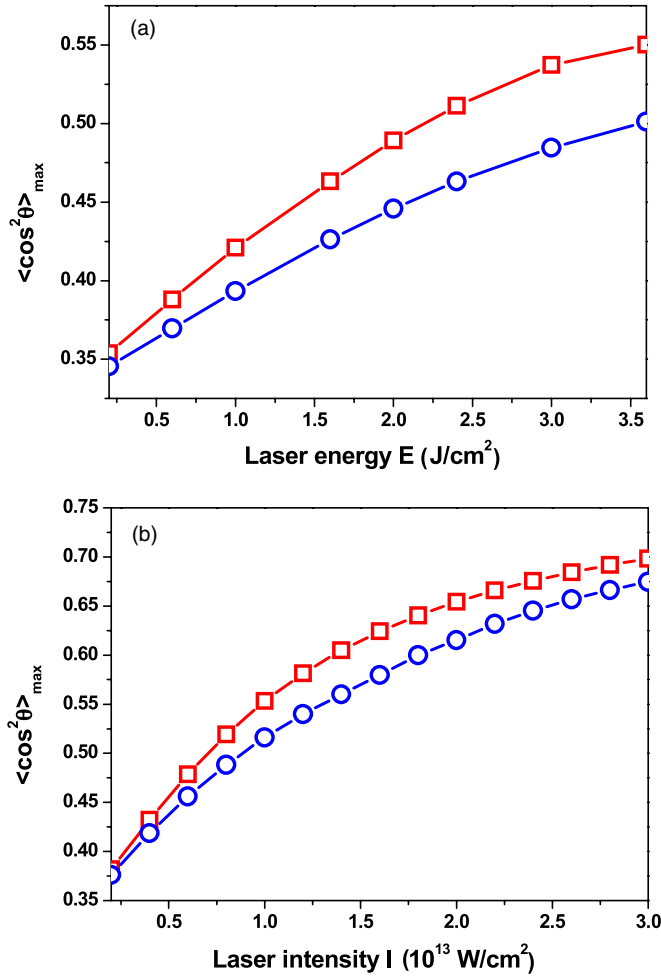


Figure 2. Maximum degree of the molecular alignment at the rotational temperature $T = 50 \text{ K}$ as the function of the laser energy E for the laser intensity $I = 1 \times 10^{13} \text{ W}/\text{cm}^2$ (a) and the laser intensity I for the pulse duration $\tau = 2 \text{ ps}$ (b) with the square laser pulse (red squares) and the conventional Gaussian laser pulse (blue circles).

pulse duration in the long laser pulse (i.e. $\tau^2 \gg 8B/[(\alpha_{\parallel} - \alpha_{\perp})(\pi A_{\text{am}})^2]$) [7, 25]. Next we compare with the maximum degree of the molecular alignment created by the square laser pulse and conventional Gaussian laser pulse in both the short and long laser pulses. Figure 2 shows the maximum degree of the molecular alignment at the rotational temperature $T = 50 \text{ K}$ as the function of the laser energy E for the laser intensity $I = 1 \times 10^{13} \text{ W}/\text{cm}^2$ (a) and the laser intensity I for the pulse duration $\tau = 2 \text{ ps}$ (b) with the square laser pulse (red squares) and the Gaussian laser pulse (blue circles); here the pulse duration of the Gaussian laser pulse refers to the full width at half maximum (FWHM). It can be seen that, comparing with the Gaussian laser pulse, the maximum degree of the molecular alignment created by the square laser pulse can be enhanced in both short and long laser pulses. This means that the temporal intensity profile of the laser pulse has a significant effect on the molecular alignment in the non-adiabatic and adiabatic regimes, and therefore it is an important parameter to improve the molecular alignment. However, the enhancement degree is correlated with the laser energy E and laser intensity I . In the short laser pulse, the enhancement degree increases

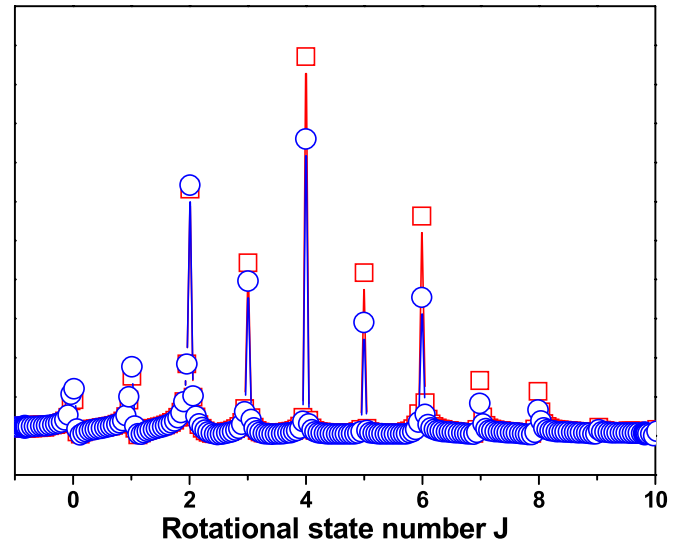


Figure 3. Fourier transform of the molecular alignment signal for the laser intensity $I = 1 \times 10^{13} \text{ W}/\text{cm}^2$, the pulse duration $\tau = 300 \text{ fs}$ and the rotational temperature $T = 50 \text{ K}$ (red squares), together with that obtained by the Gaussian laser pulse with the same laser intensity, laser energy and rotational temperature (blue circles).

with the increase of the laser energy E . In the long laser pulse, it is optimal for the intermediate laser intensities I ($\sim 1.5 \times 10^{13} \text{ W}/\text{cm}^2$), and will be reduced for high laser intensities due to its intrinsic saturation.

To illustrate the physical origin of the alignment enhancement created by the square laser pulse, we further observe the population in each rotational state after the laser pulse, which can be obtained by the Fourier transform of the alignment signal. Figure 3 shows the Fourier transform of the alignment signal with the laser intensity $I = 1 \times 10^{13} \text{ W}/\text{cm}^2$, the pulse duration $\tau = 300 \text{ fs}$ and the rotational temperature $T = 50 \text{ K}$ (red squares), together with that obtained by the Gaussian laser pulse with the same laser intensity, laser energy and rotational temperature (blue circles). It can be found that, comparing with the Gaussian laser pulse, the population in low rotational states ($J \leq 2$) induced by the square laser pulse is slightly decreased, but that in higher rotational states ($J \geq 3$) is greatly increased. The phenomenon reflects the fact that the square laser field is easier to promote the population in the higher rotational states.

It is crucial to find the optimum pulse duration that corresponds to the maximally attainable degree of the molecular alignment. However, the optimum pulse duration increases with the decrease of the laser intensity and rotational temperature [7]. In previous studies employing the Gaussian laser pulse, the optimum pulse duration was usually obtained by studying the maximum degree of the molecular alignment versus the pulse duration [7, 25]. Here, we provide a more convenient way to directly obtain the optimum pulse duration for the square laser pulse. Figure 4(a) shows the maximum degree of the molecular alignment at the full revival time T_{rot} as the function of the pulse duration τ from 0.02 to 2 ps for the laser intensity $I = 0.5 \times 10^{13} \text{ W}/\text{cm}^2$ and the rotational temperature $T = 10 \text{ K}$. It can be seen that the optimum pulse duration is 0.642 ps and its corresponding alignment degree is

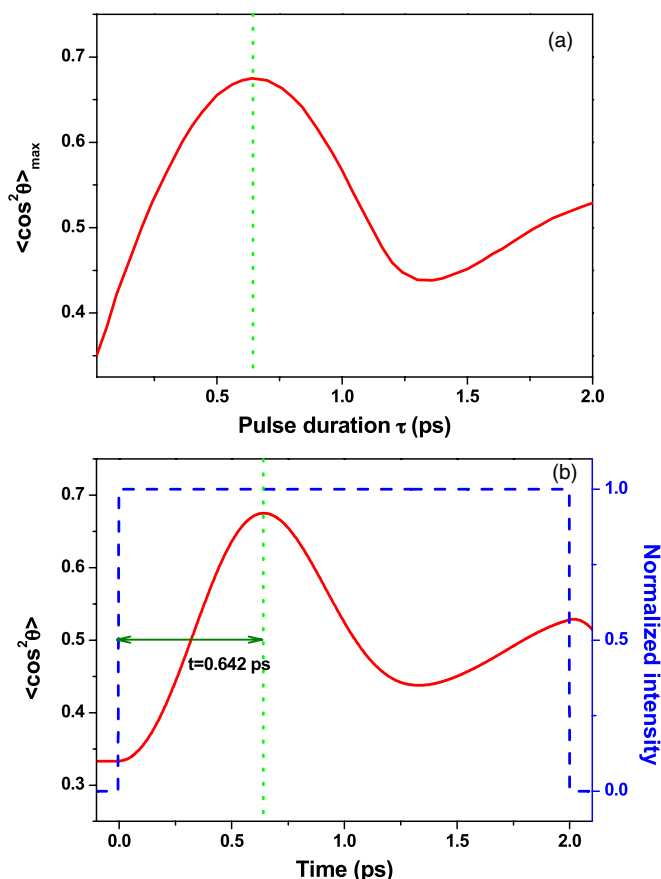


Figure 4. (a) Maximum degree of the molecular alignment at the full revival time T_{rot} as the function of the pulse duration τ from 0.02 to 2 ps for the laser intensity $I = 0.5 \times 10^{13} \text{ W cm}^{-2}$ and the rotational temperature $T = 10 \text{ K}$. (b) Time evolution of the molecular alignment during the interaction with the long laser pulse for the pulse duration $\tau = 2 \text{ ps}$ with the same laser intensity and rotational temperature in figure 4(a).

0.668. Figure 4(b) shows the time evolution of the molecular alignment during the interaction with the long laser pulse for the pulse duration $\tau = 2 \text{ ps}$ with the same laser intensity and rotational temperature in figure 4(a). The maximum degree of the molecular alignment is 0.668 at the time $t = 0.642 \text{ ps}$, which is consistent with that obtained in figure 4(a). That is to say, the optimum pulse duration and its corresponding alignment degree at the full revival time T_{rot} can be directly obtained by observing the time evolution of the molecular alignment within a long laser pulse.

Furthermore, it can be seen from figure 4 that the maximum degree of the molecular alignment at the full revival time T_{rot} is slightly larger than the alignment degree at the end of the laser pulse for the short laser pulse, while they are absolutely equal for the long laser pulse. In other words, the state of the rotational wave packet at the end of the pulse in the adiabatic regime determines the maximum degree of the molecular alignment after the laser pulse, which is similar to that created by the slow turn-on and rapid turn-off laser pulse [9]. Consequently, it can provide an excellent method to directly judge the maximum degree of the adiabatic alignment after the laser pulse.

4. Conclusion

In summary, we have theoretically studied the field-free alignment created in linear molecules by a square laser pulse. Our results showed that, comparing with the conventional Gaussian laser pulse, there are three advantages: (1) the molecular alignment after the laser pulse can be obtained in both the non-adiabatic and adiabatic regimes; (2) the maximum degree of the molecular alignment can be enhanced with the same laser energy in the short laser pulse and the same laser intensity in the long laser pulse and (3) the optimum pulse duration and its corresponding alignment degree at the full revival time for a given laser intensity and rotational temperature can be directly obtained by observing the time evolution of the molecular alignment during the interaction with a long laser pulse. Since the square laser pulse can be experimentally realized by the ultra-fast pulse shaping method, we believe that the present technique is feasible and can apply on various linear molecules.

Acknowledgments

This work was partly supported by Shanghai Leading Academic Discipline Project (B408), Ministry of Education of China (30800), National Natural Science Fund (11004060) and Shanghai Municipal Science and Technology Commission ((10XD1401800, 09142200501, 09ZR1409300, 09JC1404700 and 10JC1404500).

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