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Qi-Yuan Cheng(程起元)[1,2], Yu-Zhi Song(宋玉志)[2], Qing-Tian Meng(孟庆田)[2]
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Field-free molecular orientation steered by combination of super-Gaussian and THz half-cycle laser pulses*

Qi-Yuan Cheng(程起元)[1,2], Yu-Zhi Song(宋玉志)[2], and Qing-Tian Meng(孟庆田)[2,†]

[1] *Medical Engineering Department, Shandong Provincial Hospital Affiliated to Shandong University, Jinan* 250021, *China*
[2] *School of Physics and Electronics, Shandong Normal University, Jinan* 250358, *China*

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The molecular orientation created by laser fields is important for steering chemical reactions. In this paper, we propose a theoretical scheme to manipulate field-free molecular orientation by using an intense super-Gaussian laser pulse and a time-delayed terahertz half-cycle pulse (THz HCP). It is shown that the degree of field-free orientation can be doubled by the combined pulse with respect to the super-Gaussian pulse or THz HCP alone. Moreover, different laser intensities, carrier envelop phases, shape parameters, and time delays have great influence on the positive and negative orientations, with other conditions unchanged. Furthermore, it is indicated that the maximum degree and direction of molecular orientation can be precisely controlled by half of the duration of the super-Gaussian pulse. Finally, by adjusting the laser parameters of the super-Gaussian laser pulse and THz HCP, the optimal results of negative orientation and corresponding rotational populations are obtained at different temperatures of the molecular system.

Keywords: molecular orientation, terahertz half-cycle pulse, super-Gaussian pulse

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1. Introduction

Controlling the rotational freedom of molecules by the laser fields has been an important research subject in recent decades. Generally, there are two cases for the controlling: alignment and orientation.[1] Molecular alignment requires the principal axis of a molecule to be along a laboratory-fixed axis, however the technique for orientation means that the confinement of molecular rotation includes not only a particular direction but also a head-tail order as good as possible. Obviously, the molecular orientation is more difficult to realize than the alignment. The field-free molecular orientation can be achieved by various kinds of modulated laser pulses when the pulse duration is shorter than the rotational period of the molecule. In this case, the result of orientation can be repeated periodically after the laser has been turned off, which make it more possible to be practically used in photoelectron angular distribution,[2,3] high-harmonic generation,[4–6] and ultra-fast molecular switch.[7]

Initially, the single intense laser field was used to realize the molecular alignment/orientation based on the anisotropic polarization and hyperpolarization interaction.[8–10] However, the interaction between the field and the molecular permanent dipole moment is relatively weak, which limits the improvement of orientation. To make up the deficiency of a single field, researchers designed the combination of multiple fields to enhance the molecular orientation. Friedrich *et al.*[11,12] utilized the respective characteristics of the static field and intense nonresonant laser pulse to control the orientation of the ICl molecule. Then Jin *et al.*[13] took the NO molecule as an object to study the orientation by using several linearly polarized resonant pulses based on rearranging the distribution of the $M = 0$ rotational states. Next, Goban *et al.*[14] realized the field-free molecular orientation by the combination of an electrostatic field and an intense rapidly turned-off laser field, which is shaped by the plasma shutter technique. They all obtained better results in orientating the molecules by the combined field than by the electrostatic field or the intense laser pulse alone. Although a certain degree of the molecular orientation has been achieved by the above strategies, the easiness of the static fields to induce the Stark effects and post-field disorientation has limited their further improvement in the orientation of the molecules.

With the development of laser technology, various kinds of terahertz (THz) laser pulses are used to study the molecular spatial effect by interacting with the permanent dipole moment of the molecule instead of the static fields. It is shown that in the range of THz, a half-cycle laser pulse (HCP) has a certain advantage over a few-cycle pulse (FCP) in enhancing the alignment/orientation of linearly polar molecules.[15,16] Recently, Shalaby *et al.*[17] have implemented preliminarily a THz HCP by optical rectification in the organic crystal experimentally. By solving the time-dependent Schrödinger equation, Machholm analyzed the orientation of LiH and NaI molecules excited by a plane polarized electromagnetic THz

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†Corresponding author. E-mail: qtmeng@sdnu.edu.cn

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HCP.[18] Then, Cong *et al.*[19] proposed a theoretical scenario of using a few THz HCP pulses to enhance the field-free molecular orientation, and found that three HCPs can jointly excite more rovibrational transitions and reach a higher degree of the orientation than a single HCP. Recently, we investigated the alignment of the FCN molecules by the THz HCP and found that a THz HCP has a certain advantage over an FCP in improving the FCN molecular alignment under the same conditions.[16]

In addition to the above, other techniques, such as two dual-color ultrashort laser pulses,[20] femtosecond two-color laser fields,[21] phase-locked four-color laser fields,[22] combining femtosecond, and THz laser pulses,[23,24] combining an infrared laser pulse with an HCP,[25] *etc*., were also successively used to study the molecular orientation. However, most of them are based on the conventional Gaussian envelope, and the obtained molecular alignment and orientation are relatively low in these cases. Meng *et al.*[26,27] have found that the degree of alignment and orientation induced by a super-Gaussian pulse is about 0.1 higher than that by a Gaussian pulse at the same pulse energy or intensity. Although Liu *et al.*[28] studied the molecular orientation by combining an STRT (slow turn-on and rapid turn-off) with THz laser pulse, the maximum degrees of molecular orientation by adding THz pulse has not been improved effectively, and the negative orientation has not been discussed in their work either. Dang *et al.*[29] orientated the molecules by combining a nonresonant shaped laser pulse with a THz laser pulse train, but they paid the most attention to the effect of pulse number on orientation. Here in this work, on the basis of these recent research results and technological advances, we propose a scheme to achieve a high degree of the molecular orientation by a combination of super-Gaussian and THz half-cycle laser pulses. It is shown that the molecular orientation can be significantly improved after the THz HCP has been added, even the maximum degrees of orientation can be doubled. And by tuning the laser parameters to a certain range, the positive and negative orientations of molecules can have large changes, particularly, the difference value $\Delta\langle\cos\theta\rangle_{\max} = 0.51$ when the variation of the pulse half-width duration $\Delta\sigma = 500$ fs.

The remaining part of the paper is arranged as follows. First, we give the time evolution of field-free molecular orientation induced by a THz HCP, super-Gaussian pulse and combination of THz half-cycle and super-Gaussian pulses, respectively. And the relationship between the time-varying population of $J = 0$ to 5 states and the degree of orientation is discussed. Then, we analyze how the parameters of laser pulses affect the maximum degrees of positive and negative orientations. Finally, an optimized result of field-free molecular orientation is provided and the populations of the rotational states at different temperatures are discussed in detail.

2. Theoretical method

Here, a linearly polarized femtosecond laser pulse with a super-Gaussian envelope expressed as

$$E_{\rm fs}(t) = E_0 \exp(-t^N/\sigma^N)\cos\omega_0 t \tag{1}$$

is used to orientate the linear molecule, where E_0, σ, ω_0, and N denote the amplitude of laser pulse, the pulse half-width duration, the laser central frequency, and the laser shape parameter, respectively. The THz HCP is given by

$$E_{\rm THz}(t) = E_1 \exp\left[-2\ln 2\frac{(t-t_{\rm d})^2}{\tau^2}\right]\cos\left[\omega_1(t-t_{\rm d})+\varphi\right], \tag{2}$$

where E_1 is the amplitude of THz HCP, $t_{\rm d}$ is the time delay between a super-Gaussian and THz pulse, τ is the full width at half maximum (FWHM), ω_1 is the central frequency, and φ is the carrier envelope phase (CEP). τ can be tuned to match the optical period $t_{\rm p} = 2\pi/\omega_1$. For a THz HCP, $\tau = t_{\rm p}/2$. Based on the rigid rotor approximation, the linear molecular Hamiltonian in the laser field is given by[30]

$$\begin{aligned}\hat{H}(t) \;=\; & B_{\rm e}\hat{J}^2 - \mu E_{\rm THz}(t)\cos\theta \\ & -\frac{1}{4}[(\alpha_{\parallel}-\alpha_{\perp})\cos^2\theta+\alpha_{\perp}]E_{\rm fs}^2(t) \\ & -\frac{1}{8}[(\beta_{\parallel}-3\beta_{\perp})\cos^3\theta+3\beta_{\perp}\cos\theta]E_{\rm fs}^3(t),\end{aligned} \tag{3}$$

where the first term on the right-hand side is the molecular rotation energy, the second term is the dipole interaction between the THz HCP and the molecule, the last two terms represent the interaction potentials of the super-Gaussian laser pulse with polarization and hyperpolarization, respectively, $B_{\rm e}$ denotes the molecular rotational constant, $\hat{J}$ is the angular momentum operator and θ is the angle between the laser polarization and the molecular axis, $\alpha_{\parallel}$ and $\alpha_{\perp}$ denote the polarizability parallel and perpendicular to the molecular axis, respectively, $\beta_{\parallel}$ and $\beta_{\perp}$ are the parallel and orthogonal components of the hyperpolarizability with respect to the axis of molecule, respectively. The degree of molecular orientation can be calculated from

$$\langle\cos\theta\rangle = {\rm Tr}\{\cos\theta\hat{\rho}(t)\}, \tag{4}$$

where Tr and $\hat{\rho}$ are the trace and the density operator respectively. The relation between the density operator and the Hamiltonian obeys the Liouville equation

$$\frac{{\rm d}\hat{\rho}(t)}{{\rm d}t} = -\frac{\rm i}{\hbar}[\hat{H},\hat{\rho}(t)]. \tag{5}$$

The density operator is expressed in the eigenstates of the rigid rotor Hamiltonian as

$$\hat{\rho}(t) = \sum_{J,M,J',M'}\rho_{J,M,J',M'}(t)\left|JM\right\rangle\left\langle J'M'\right|, \tag{6}$$

where $\rho_{JMJ'M'}(t)$ are obtained by solving the coupling differential equations[31]

$$\begin{aligned}
&\frac{\mathrm{d}\rho_{JMJ'M'}(t)}{\mathrm{d}t} \\
&= -\frac{\mathrm{i}}{\hbar}\Big\{(\varepsilon_J-\varepsilon_{J'})\rho_{JMJ'M'}(t) \\
&\quad -\mu E_{\mathrm{THz}}(t)\sum_{J_1M_1}\big[V^{(1)}_{JMJ_1M_1}\rho_{J_1M_1J'M'}(t) \\
&\quad -V^{(1)}_{J_1M_1J'M'}\rho_{JMJ_1M_1}(t)\big] \\
&\quad -\frac{1}{4}\Delta\alpha E^2_{\mathrm{fs}}(t)\sum_{J_1M_1}\big[V^{(2)}_{JMJ_1M_1}\rho_{J_1M_1J'M'}(t) \\
&\quad -V^{(2)}_{J_1M_1J'M'}\rho_{JMJ_1M_1}(t)\big] \\
&\quad -\frac{1}{8}{E_{\mathrm{fs}}}^3(t)\sum_{J_1M_1}\big[(\beta_{\parallel}-3\beta_{\perp})\big(V^{(3)}_{JMJ_1M_1}\rho_{J_1M_1J'M'}(t) \\
&\quad -V^{(3)}_{J_1M_1J'M'}\rho_{JMJ_1M_1}(t)\big)+3\beta_{\perp}\big(V^{(1)}_{JMJ_1M_1}\rho_{J_1M_1J'M'}(t) \\
&\quad -V^{(1)}_{J_1M_1J'M'}\rho_{JMJ_1M_1}(t)\big)\big]\Big\} \qquad (7)
\end{aligned}$$

with $V^{(i)}_{JMJ'M'} = \langle JM|\cos^i\theta\,|J'M'\rangle$ $(i = 1, 2, 3)$. The initial density operator obeys the temperature-dependent Boltzmann distribution

$$\rho_0(T) = \frac{1}{Z}\sum_{J=0}^{\infty}\sum_{M=-J}^{M=J}|JM\rangle\langle JM|\exp\left(\frac{-B_{\mathrm{e}}J(J+1)}{k_{\mathrm{B}}T}\right), \qquad (8)$$

where

$$Z = \sum_{J=0}^{\infty}\sum_{M=-J}^{M=J}\exp\left(-\frac{B_{\mathrm{e}}J(J+1)}{k_{\mathrm{B}}T}\right)$$

is the partition function with the Boltzmann constant k_{B} at temperature T. In this work, the fourth-order Runge–Kutta method is used to solve coupling differential equations, and the CO molecule is taken as a target to investigate its field-free orientation with molecular parameters being as follows:[32] $B_{\mathrm{e}} = 1.93\ \mathrm{cm}^{-1}$, $\mu = 0.112$ D, $\alpha_{\parallel} = 2.294\ \text{Å}^3$, $\alpha_{\perp} = 1.77\ \text{Å}^3$, $\beta_{\parallel} = 2.748\times10^9\ \text{Å}^5$, and $\beta_{\perp} = 4.994\times10^8\ \text{Å}^5$. The rotational period of the CO molecule $T_{\mathrm{rot}} = h/2B_{\mathrm{e}} = 8.64$ ps, and the rotational temperature T is set to be 0 K. The parameters of the super-Gaussian laser pulse are taken to be $E_0 = 65$ MV/cm, $\sigma = 500$ fs, $N = 4$, and $\omega_0 = 12500\ \mathrm{cm}^{-1}$, corresponding to the central wavelength of 800 nm. For the THz HCP, $E_1 = 10$ MV/cm, $\varphi = 0.5\pi$, $t_{\mathrm{d}} = 2.5$ ps, and $\tau = 463$ fs.

3. Results and discussion

Figure 1(a) shows the time evolution of molecular orientation induced by a super-Gaussian (red dash), THz HCP (black dot), and combination of super-Gaussian and THz half-cycle pulses (olive solid). Evidently, a higher orientation degree can be obtained by the combined laser field than by a super-Gaussian pulse or THz HCP alone. Especially, the negative orientation of molecules, which corresponds to the orientation arranged in the negative direction, can be improved greatly. The maximum degree of molecular orientation $\langle\cos\theta\rangle_{\max}$ increases from 0.23 to 0.5 after the THz HCP has been used. The increase is much larger than that of combination of STRT and THz pulses.[28] In other words, the super-Gaussian pulse has more advantages in enhancing molecular orientation degree than the shaped pulse when combined with the THz pulse. The time-varying populations of rotational states at $t_{\mathrm{d}} = 2.5$ ps are depicted in Fig. 1(b). As can be seen, most of molecules are excited to $|2,0\rangle$ and $|4,0\rangle$ states and few to $|1,0\rangle$, $|3,0\rangle$, and $|5,0\rangle$ states after the super-Gaussian pulse has been over. When the THz HCP is added, the populations of $J = 2$ and 4 states begin to decrease and those of $J = 1$, 3, and 5 begin to increase to some degree. Then the rotational populations of molecules remain unchanged when $t > 3.35$ ps. For the molecular orientation steered by the super-Gaussian pulse and THz pulse, the selection rules are $\Delta J = \pm1$ and $\Delta J = 0$, ±2, respectively. We can deduce that the molecules excited to odd states by the THz HCP play a vital role in this case.

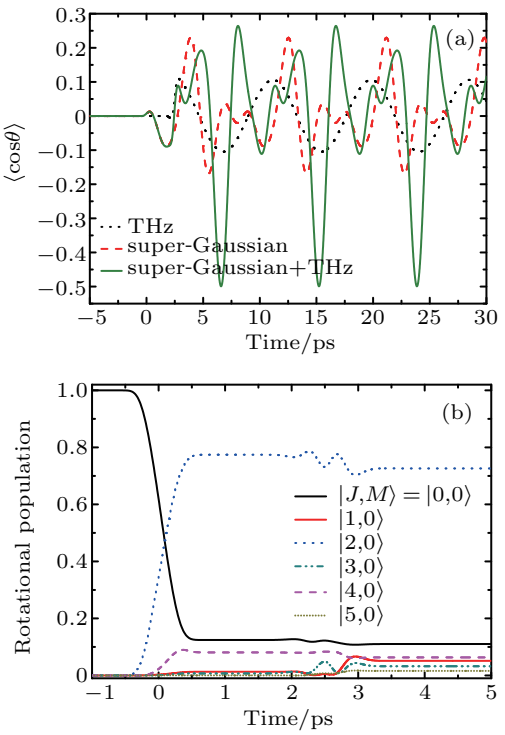


Fig. 1. (a) Time evolution of molecular orientation induced by THz HCP (black dot), super-Gaussian pulse (red dash), and combination of THz half-cycle and super-Gaussian pulses (olive solid). (b) Time-dependent population of $J = 0$ to 5 states in panel (a).

In order to examine how the laser intensity affects the molecular orientation, we calculate the maximum degrees of positive and negative orientations with the increase of the amplitude of super-Gaussian pulse and THz HCP, and the results are shown in Fig. 2. From Fig. 2(a), it is found that the positive and negative maximum orientations of molecules have almost the same changing tendency when the field amplitude E_0 is less than 20 MV/cm. With amplitude increasing continuously,

the positive orientation intends to increase gradually, while the negative orientation obviously presents a minimum value of 0.3 at the amplitude of the super-Gaussian $E_0 = 80$ MV/cm and a maximum value of 0.68 at the amplitude of the super-Gaussian $E_0 = 100$ MV/cm. For the THz HCP in Fig. 2(b), the maximum degrees of positive and negative orientations increase first and then decrease. It can be found that almost each negative maximum is greater than the positive maximum at the same pulse intensity. Meanwhile the negative orientation takes a maximum value when $E_1 = 10.5$ MV/cm, which is less than $E_1 = 15.5$ MV/cm at which the positive orientation takes a maximum value. The contribution of the THz HCP to the negative orientation is a little larger when the field amplitude is relatively low, which can prevent the excessive intensity caused nonlinear effect from occurring.

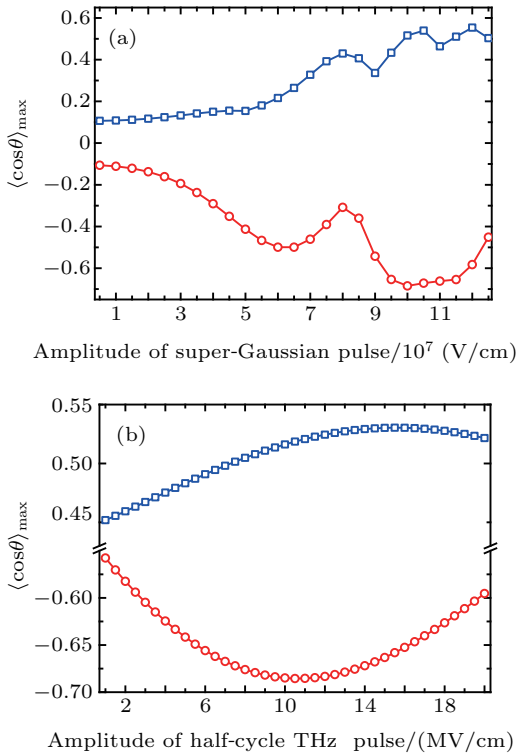


Fig. 2. Maximum degrees of the positive (blue square) and negative (red circle) orientations $\langle\cos\theta\rangle_{\max}$ *versus* (a) amplitude of super-Gaussian pulse and (b) THz HCP, respectively.

To illustrate the effect of the CEP on field-free molecular orientation created by a combination of super-Gaussian and THz HCP pulses, we give the maximum degrees of the positive and negative orientations through tuning the CEP of THz HCP from 0 to 4π in steps of 0.1π as indicated in Fig. 3. It can be found that the maximum orientation of molecules presents an irregular variation in a 2π period. The positive and negative maximum degree are 0.58 and 0.59, corresponding to the CEP $\varphi = 1.7\pi$ and 0.7π, respectively. This phenomenon is different from previous results that the maximum orientation increases initially and decreases afterwards, *i.e.*, the optimal degree of orientation can be obtained when the CEP $\varphi = \pi$.[20,22] And by adjusting the CEP, we are able to achieve a change of 0.4 for $\langle\cos\theta\rangle_{\max}$, which is larger than 0.2 obtained in the study by Liu *et al.*[28] From the above we can conclude that the CEP of THz HCP is an important parameter influencing the molecular orientation under the combined pulse, and a specific CEP is beneficial to the improvement of molecular orientation.

Figure 4 shows the maximum degrees of the positive and negative orientation $\langle\cos\theta\rangle_{\max}$ *versus* delay time t_d between the super-Gaussian pulse and THz HCP. For comparison, we give the molecular orientation (green dash curve) induced only by a super-Gaussian pulse, where the maximum values of positive and negative orientation are, respectively, 0.23 and 0.17. Here, the delay time t_d is taken in a range from $0.05T_{rot}$ to $2T_{rot}$ in steps of $0.05T_{rot}$. It can be seen that the maximum degrees of positive and negative orientations revive at a period of nT_{rot} ($n = 1, 2, \ldots$) and can be obviously enhanced at the delay time $t_d = 1.93$ and 2.16 respectively. The optimal degrees of positive and negative orientation are 0.58 and 0.56, respectively. Compared with the maximum degrees in the case of the super-Gaussian pulse, the maximum degrees of positive and negative orientations have increased by 0.35 and 0.39 in the case of THz HCP, respectively. Based on the preexcitation by the super-Gaussian pulse, applying the THz HCP at different time delays can make different transitions and change the rotational population of molecules created by the first super-Gaussian pulse. Therefore, the delay time also has a crucial effect on the molecular orientation.

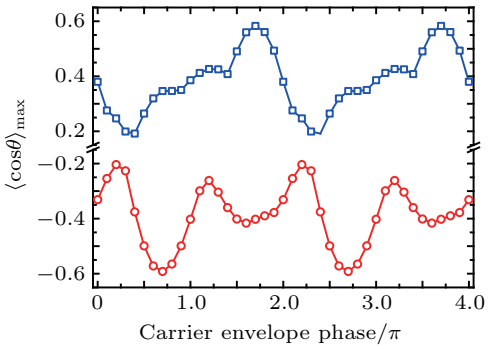


Fig. 3. Maximum degrees of positive (blue square) and negative (red circle) orientation $\langle\cos\theta\rangle_{\max}$ *versus* CEP of THz HCP.

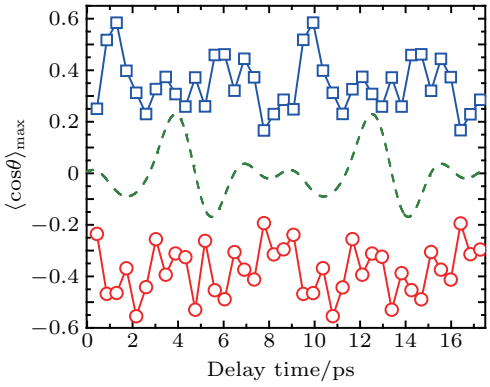


Fig. 4. Maximum degrees of the positive (blue square) and negative (red circle) orientation $\langle\cos\theta\rangle_{\max}$ *versus* time delay t_d. The green dashed curve refers to the molecular orientation varying with time by a super-Gaussian pulse only.

Owing to the fact that the tuning of laser shape parameter can change the on and off ramp of the super-Gaussian laser pulse, we study the maximum degrees of molecular orientation by changing the laser shape parameter N from 1 to 21 as shown in Fig. 5. As the laser shape parameter N increases, the maximum degrees of both positive and negative orientations end up with the saturation values 0.37 and 0.45, respectively. Evidently, the positive maximum orientation by combining the super-Gaussian pulse with THz HCP is always greater than by combining the traditional Gaussian or $N = 1$ pulse with THz HCP. However, it is slightly different from the negative maximum orientation. The maximum degrees by the super-Gaussian pulses of $N = 3$, 4, and 5 are a little higher but the maximum degrees by $N > 5$ pulses are lower than by the Gaussian pulse. In this case, the maximum degree of negative orientation $\langle\cos\theta\rangle_{\max} = 0.5$ at $N = 3$. Meanwhile, the result also reflects the fact that compared with the negative orientation, the positive orientation is very sensitive to the laser shape parameter.

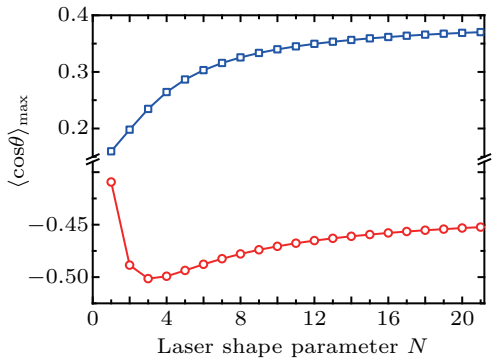


Fig. 5. Maximum degrees of the positive (blue square) and negative (red circle) orientation $\langle\cos\theta\rangle_{\max}$ *versus* laser shape parameter N.

Figures 6(a) and 6(b) shows the maximum degrees of positive (blue squares) and negative (red circles) orientation varying with pulse half-duration of the super-Gaussian pulse and the FWHM of THz HCP, respectively. It can be seen from Fig. 6(a) that the maximum degrees of positive and negative orientations have roughly the same trend and fluctuate greatly with the change of the pulse half-duration. Specifically, the maximum degree of positive orientation $\langle\cos\theta\rangle_{\max}$ decreases from 0.77 to 0.16 as the pulse half-duration σ varies from 5.0 ps to 5.5 ps, *i.e.*, the variation of the pulse half-duration $\Delta\sigma = 0.5$ ps can create $\Delta\langle\cos\theta\rangle_{\max} = 0.51$. And for the negative orientation, it has a maximum increment of 0.72 when the variation of the pulse half-duration $\Delta\sigma = 4$ ps. From Fig. 6(b), we can obtain that the maximum variation of the positive orientation $\Delta\langle\cos\theta\rangle_{\max} = 0.3$ as the FWHM τ changes from 0.01 ps to 1.6 ps, and the maximum value $\langle\cos\theta\rangle_{\max} = -0.52$ when the FWHM $\tau = 1.45$ ps for the negative orientation. Although the FWHM of THz HCP can improve the positive and negative orientations to some extent, compared with the pulse half-duration, its ability to improve the molecular orientation is relatively weak. From the above we can know that in a non-adiabatic regime, the positive and negative orientations can be obviously enhanced or suppressed by the pulse half-duration. Since the energy of the super-Gaussian pulse is determined by the pulse half-duration, with other parameters unchanged, *i.e.*, the bigger the pulse half-duration, the higher the pulse energy will be, different pulse energy can make the molecule transit to different rotational energy levels, thus leading the molecular orientation to change. It should be noted that the dependence of field-free positive and negative orientation on the pulse half-duration may have an application in controlling the maximum degrees and direction of molecular orientation in accordance with specific requirements.

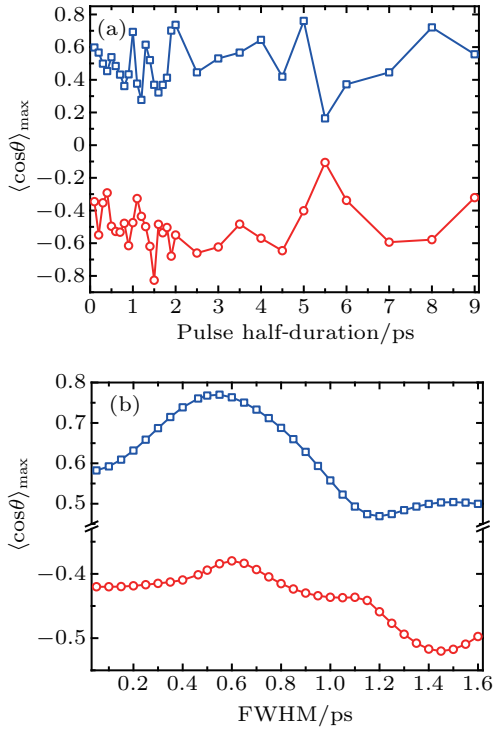


Fig. 6. Maximum degrees of the positive (blue square) and negative (red circle) orientation $\langle\cos\theta\rangle_{\max}$ *versus* (a) pulse half-duration σ at $\tau = 463$ fs and (b) FWHM τ at $\sigma = 5$ ps, where $E_0 = 120$ MV/cm, $E_1 = 15.5$ MV/cm, CEP $= 0.5\pi$, $t_d = 1.296$ ps, and $N = 20$.

The above discussion mainly focuses on the influence of laser pulse parameters. For further investigating the features of molecular orientation, we change the temperature of the molecular system to examine the orientation steered by the combined laser field. The curves of time-dependent molecular orientation and the population of rotational quantum states at different temperatures are plotted in Fig. 7. We can find from Fig. 7(a) that the negative orientation is greatly improved by setting the appropriate laser parameters, and the maximum orientation is −0.852, −0.753, and −0.396 at the temperature $T = 0$, 3, and 5 K, respectively. Obviously, the rotational temperature only affects the peak value of the orientation revival. The orientation achieved in Fig. 7(a) at $T = 0$ K has a duration of 1.2 ps for $\langle\cos\theta\rangle > 0.5$. This duration is long enough

to probe the electron dynamics with femtosecond or picosecond laser pulses.[33] Figure 7(b) shows the population of the energy states at different rotational temperatures. An obvious population transfer from $J = 1$ to 4, 5, and 6 is found when the temperature T increases from 0 K to 5 K. This phenomenon is different from the population transfer caused by increasing the laser intensity or adding another laser pulse, where the molecular alignment can be enhanced when the rotational population is transferred from lower to higher energy level.[16]

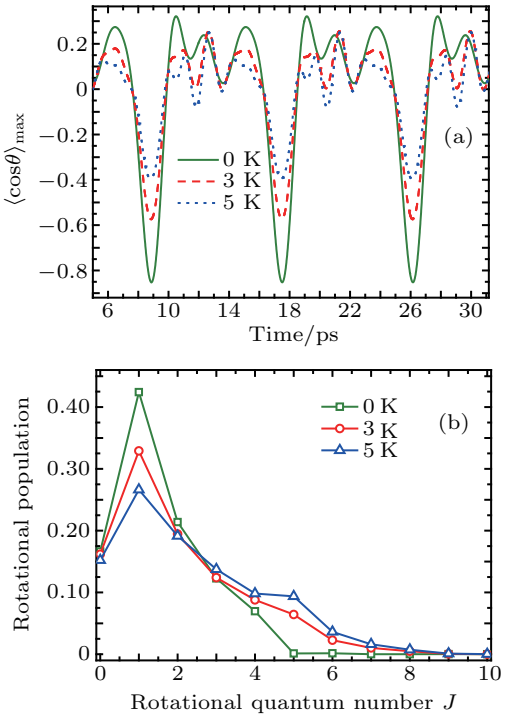


Fig. 7. Plots of (a) orientation *versus* time and (b) population of rotational states *versus* rotational quantum number at molecular temperature $T = 0$ K (olive line), 3 K (red line), and 5 (blue line), for $E_0 = 120$ MV/cm, $E_1 = 15.5$ MV/cm, CEP $= 0.5\pi$, $t_d = 1.296$ ps, $\sigma = 1513$ fs, and $N = 20$.

4. Conclusions

In this work, we theoretically study the molecular orientation induced by a super-Gaussian laser pulse and a time-delayed THz HCP. The maximum degrees of both the positive and negative orientations have a much higher value by two-step excitation than by the super-Gaussian or THz pulse excitation alone. By analyzing the time evolution of rotational population $J = 0$ to 5 states, we find that the molecular transition between odd states by the THz HCP is a crucial factor to improve the molecular orientation. The effects of laser parameters including the pulse intensity, CEP, time delay, shape parameter and pulse half-duration on field-free molecular orientation are discussed in detail. Finally, we obtain an optimal negative orientation by adjusting the laser parameters. To elucidate the mechanism of molecular orientation, the populations of rotational states $J \leq 10$ are given at different temperatures. Certainly the above conclusions need further supporting from experiments.

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