Enhancing molecular orientation by combining electrostatic and four-color laser fields

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We propose a scheme to enhance molecular orientation by combing an intense electrostatic field and a four-color laser field. We show that molecular orientation by the combined field can be obtained under a laser-field-free condition, and the maximal orientation degree can be enhanced by comparing with the sum of that individually created by the electrostatic field and the four-color laser field. Our results show that the orientation enhancement results from the larger asymmetry of the four-color laser field because of the existence of the electrostatic field. Furthermore, we also discuss the dependence of the orientation enhancement on the carrier-envelope phase, laser intensity, and pulse duration of the four-color laser field and the molecular rotational temperature.

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I. INTRODUCTION

Aligned or oriented molecules have attracted considerable attention in the last decade because of their widespread applications in various related fields [1,2], such as high-order-harmonic generation [3,4], chemical reaction dynamics [5,6], ultrafast molecular imaging [7–9], and attosecond science [10]. Compared with the molecular alignment that an order of the molecular geometry is defined with respect to a space fixed axis, the molecular orientation with a head-versus-tail order has more advantages in various related applications, but there are higher requirements for its experimental technique. By now, several techniques have been utilized to realize molecular orientation, such as a strong dc electrostatic field [11,12], the two-color laser field [13–15], or a half-cycle or Terahertz laser field [16,17]. Furthermore, various schemes have also been proposed to further enhance molecular orientation, such as those involving an intense laser field combined with a weak electrostatic field [18–20], multiple two-color laser pulses [13–15,21], use of a shaped Terahertz laser pulse [22], and so on.

Molecular orientation achieved by an electrostatic field is based on the effect of the permanent dipole interaction, whereas a molecular orientation by a two-color or multi-color laser field is based on the combined effects of the anisotropic polarizability and hyperpolarizability interaction. In this paper, we show that a hybrid field achieved by combing an electrostatic field and a four-color laser field can further enhance molecular orientation. In our previous study, we showed that a four-color laser field can induce the maximum molecular orientation in these multicolor laser fields under the same laser intensity [23]. Furthermore, with the development of the ultrafast laser technique, a four-color laser field is easily obtained experimentally via second, third, and fourth harmonic generation using beta barium borate (BBO) crystal. Our results show that, compared with the sum of the maximal orientation degree individually created by the electrostatic field and the four-color laser field, the orientation degree by a combined field can be enhanced and obtained under a laser-field-free condition. Our analysis demonstrates that because of the existence of the electrostatic field, a higher asymmetry of the four-color laser field is generated, which results in the orientation enhancement. In order to illustrate the maximally attainable enhancement efficiency of the molecular orientation, the effects of the carrier-envelope phase, laser intensity, and pulse duration of the four-color laser field and the molecular rotational temperature on the orientation enhancement are discussed.

II. THEORETICAL MODEL

We consider a hybrid field with the combination of an electrostatic field and a four-color laser field, where the four-color laser field is superimposed by a fundamental field and its harmonics. We assume that the polarization direction of the four-color laser field is the same as the propagation direction of the dc electrostatic field, and thus the combined field can be written as

$$\begin{aligned} E(t) &= E_S(t) + E_L(t) \\ &= E_0 + E_L f(t)[\exp(i\omega t) + \exp(i2\omega t) \\ &\quad + \exp(i3\omega t) + \exp(i4\omega t)]\exp(i\Phi_{\mathrm{CEP}}), \end{aligned} \tag{1}$$

where E_0 and E_L are, respectively, the amplitude of the electrostatic field and the four-color laser field, Φ_{CEP} is the carrier-envelope phase of the four-color laser field, $f(t)$ is the laser pulse envelope with a Gaussian shape of $f(t) = \exp[(-2\ln 2)t^2/\tau^2]$, and τ is the laser pulse duration. When a linear polar molecule is subjected to the combined electrostatic and four-color laser field, the time-dependent Schrödinger equation can be approximated based on the rigid rotor model as

$$i\hbar\frac{\partial|\psi(t)\rangle}{\partial t} = H(t)|\psi(t)\rangle, \tag{2}$$

with

$$\begin{aligned} H(t) &= BJ(J+1) - \mu_0 E(t)\cos\theta \\ &\quad - \tfrac{1}{2}[(\alpha_{||} - \alpha_\perp)\cos^2\theta + \alpha_\perp]E^2(t) \\ &\quad - \tfrac{1}{6}[(\beta_{||} - 3\beta_\perp)\cos^3\theta + 3\beta_\perp\cos\theta]E^3(t), \end{aligned} \tag{3}$$

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where B and μ_0 are, respectively, the rotational constant and the permanent dipole moment of the molecule, J is the angular momentum, θ is the angle between the molecular axis and the direction of the combined field, $\alpha_{||}$ and $\alpha_{\perp}$ are, respectively, the polarizability components that are parallel and perpendicular to the molecular axis, and $\beta_{||}$ and $\beta_{\perp}$ are, respectively, the hyperpolarizability components that are parallel and perpendicular to the molecular axis. The degree of the molecular orientation is usually given by the expectation value of $\cos\theta$, i.e., $\langle\cos\theta\rangle$. When the thermal equilibrium of the molecular ensemble is considered, the molecular orientation degree $\langle\cos\theta\rangle$ should be averaged over the Boltzmann distribution and is written as

$$\begin{aligned}\langle\cos\theta\rangle &= \sum_J W_J \sum_{M=-J}^{J} \langle\Psi_{JM}|\cos\theta|\Psi_{JM}\rangle \\ &= \sum_J \frac{g_J \exp[-BJ(J+1)/kT]}{Q} \\ &\quad \times \sum_{M=-J}^{J} \langle\Psi_{JM}|\cos\theta|\Psi_{JM}\rangle, \end{aligned} \tag{4}$$

where g_J is the spin degeneracy factor, Q is the rotational partition function, k is the Boltzmann constant, and T is the molecular rotational temperature.

III. RESULTS AND DISCUSSION

In our theoretical simulation, the time-dependent Schrödinger equation in Eq. (2) is numerically solved by the split-operator method [24,25]. The linear CO molecule is used as our model example, and the molecular parameters are set as $B = 1.93\,\mathrm{cm}^{-1}$, $\mu = 0.112$D, $\alpha_{||} = 2.294$ Å^3, $\alpha_{\perp} = 1.77$ Å^3, $\beta_{||} = 2.748 \times 10^9$ Å^5, and $\beta_{\perp} = 4.994 \times 10^8$ Å^5 [26,27]. The amplitude of the electrostatic field is set to be $E_0 = 200$ kV/cm, and the central frequency of the fundamental laser field is $\omega = 12\,500\,\mathrm{cm}^{-1}$, corresponding to a laser wavelength of 800 nm. In the following discussion, the other parameters are set as follows unless a special explanation is given. The laser intensity of the four-color laser field is $I = 1 \times 10^{13}$ W/cm^2, the laser pulse duration is $\tau = 100$ fs, the carrier-envelope phase is $\Phi_{\mathrm{CEP}} = 0$, and the molecular rotational temperature is $T = 1$ K.

Figure 1 shows the time-dependent molecular orientation $\langle\cos\theta\rangle$ excited by the combined field with a molecular rotational temperature of $T = 30$ K. As can be seen, the molecular orientation periodically evolves after the four-color laser field with the rotational period of $T_{\mathrm{rot}} = 1/(2Bc) \approx 8.64$ ps. Before interaction with the four-color laser field, i.e., negative time delay, an orientation degree $\langle\cos\theta\rangle = 0.006$ is observed which is created by the electrostatic field. It can be found that the molecular orientation can be obtained under the laser field-free condition, which is very useful for further applications in various related fields. Furthermore, the amplitude of the electrostatic field E_0 is set as 200 kV/cm, corresponding to a field intensity of 5.31×10^7 W/cm^2, which is relatively low compared with the laser intensity. In some special cases, the electrostatic field does not affect further applications of the oriented molecules, such as the photoionization and photodissociation of the oriented molecules.

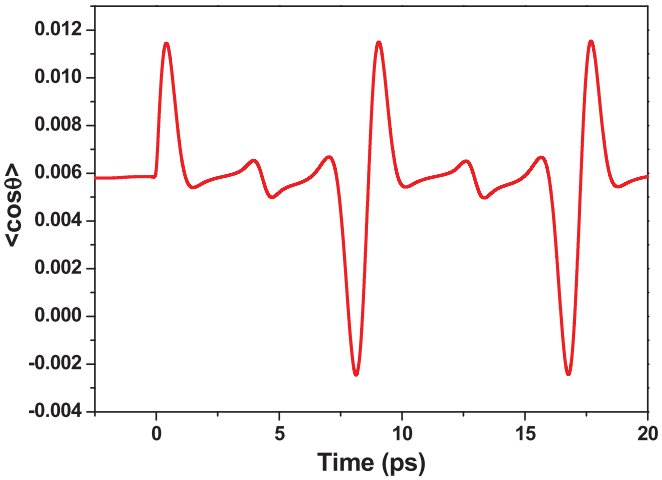

FIG. 1. (Color online) Time evolution of the molecular orientation $\langle\cos\theta\rangle$ excited by the combined electrostatic and four-color laser field with a molecular rotational temperature of $T = 30$ K.

Figure 2 shows the maximum orientation degree $\langle\cos\rangle_{\mathrm{max}}$ induced by the four-color laser field (pink open-circle line), the electrostatic field (olive open-square line), and their combined field (red solid-circle line) as a function of the carrier-envelope phase Φ_{CEP} of the four-color laser field (see left coordinate), together with the sum of the maximum orientation degree individually induced by the four-color laser field and the electrostatic field (green solid-square line). One can see that compared with the sum of the maximal orientation degree

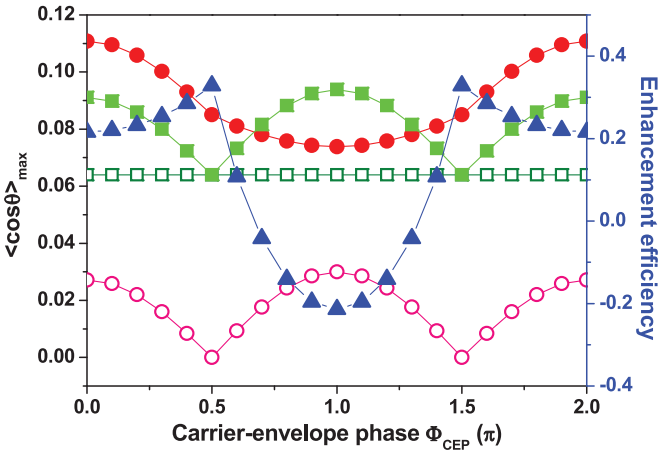

FIG. 2. (Color online) Left coordinate: the maximum orientation degree $\langle\cos\rangle_{\mathrm{max}}$ induced by the four-color laser field (pink open-circle line), the electrostatic field (olive open-square line), and their combined field (red solid-circle line) as a function of the carrier-envelope phase Φ_{CEP} of the four-color laser field, together with the sum of the maximum orientation degree individually induced by the four-color laser field and the electrostatic field (green solid-square line). Right coordinate: the enhancement efficiency of the maximum orientation degree induced by the combined field with respect to the sum of that induced by the two individual fields (blue solid-triangle line).

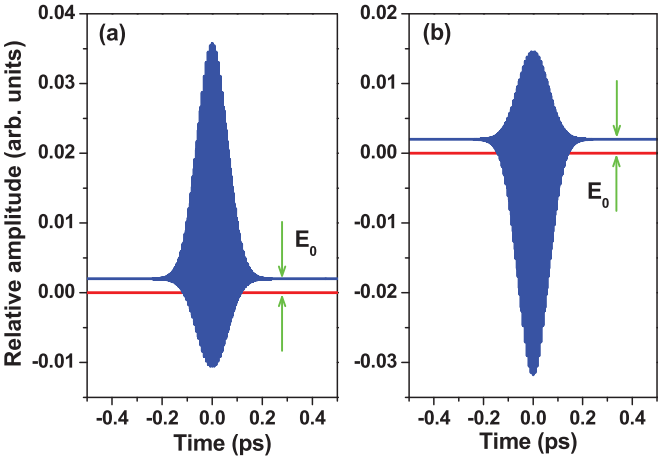

FIG. 3. (Color online) Schematic diagram of the combined electrostatic and four-color laser field with the carrier-envelope phase $\Phi_{CEP} = 0$ (a) and π (b), where E_0 is the amplitude of the electrostatic field.

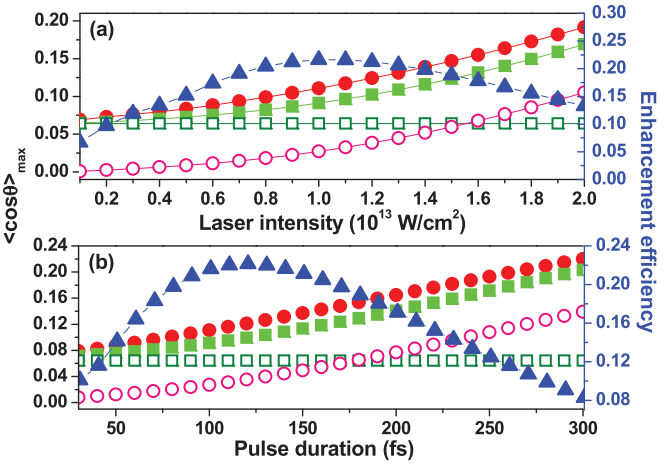

FIG. 4. (Color online) The maximum orientation degree $\langle\cos\rangle_{max}$ and enhancement efficiency as a function of the laser intensity (a) and pulse duration (b) of the four-color laser field. All lines are labeled the same as those shown in Fig. 2.

created by the two individual fields, the maximal orientation degree by the combined field can be enhanced around $\Phi_{CEP} = 0$ and 2π, while is efficiently suppressed at around $\Phi_{CEP} = \pi$. In other words, the carrier-envelope phase Φ_{CEP} can be used to control the molecular orientation enhancement or suppression. Figure 2 also presents the enhancement efficiency with a change of the carrier-envelope phase Φ_{CEP} (see right coordinate). It can be seen that the enhancement efficiency can increase by ~20%. Consequently, we can conclude that the four-color laser field combined with an intense electrostatic field can provide a feasible scheme to further enhance molecular orientation.

Since the degree of the molecular orientation depends on the asymmetry of the four-color laser field and the larger asymmetry yields the higher orientation degree under the same laser intensity, the modulation of the maximal orientation degree in Fig. 2 can be explained by observing the structure of the combined electrostatic and four-color laser fields. Figure 3 shows a schematic diagram of the combined electrostatic and four-color laser field with the carrier-envelope phase $\Phi_{CEP} = 0$ (a) and π (b). The carrier-envelope phase Φ_{CEP} determines the structure of the four-color laser field, which is maximal asymmetry for $\Phi_{CEP} = 0$ and antiasymmetry for $\Phi_{CEP} = \pi$. Thus, the existence of the electrostatic field will enhance the asymmetry of the four-color laser field for $\Phi_{CEP} = 0$ while suppressing its antiasymmetry for $\Phi_{CEP} = \pi$, and therefore the molecular orientation can be enhanced for $\Phi_{CEP} = 0$ and suppressed for $\Phi_{CEP} = \pi$.

The laser intensity and pulse duration of the four-color laser field are two important parameters that affect molecular orientation, and next we discuss the dependence of the orientation enhancement on the laser intensity and pulse duration. Figure 4 shows the maximal orientation degree $\langle\cos\rangle_{max}$ and enhancement efficiency as a function of the laser intensity (a) and pulse duration (b), where all lines are labeled the same as those of Fig. 2. With the increase of the laser intensity and pulse duration, the molecular orientation degree by the combined field is increased, which is the same as that of the four-color laser field. However, the enhancement efficiency is first increased and then decreased, and is maximally obtained around a laser intensity of $I = 1 \times 10^{13}$ W/cm^2 and a pulse duration of $\tau = 120$ fs. That is to say, in the nonadiabatic region, a higher degree of orientation can be obtained for a higher laser intensity or larger pulse duration, whereas the enhancement efficiency is not increased, which indicates there is an optimal value.

Finally, we demonstrate the effect of the molecular rotational temperature on the maximal orientation degree and enhancement efficiency, and the calculated results are shown in Fig. 5. Again all lines are labeled the same as those of Fig. 2. With a decrease of the molecular rotational temperature, both the maximal orientation degree and enhancement efficiency are increased. So, in order to simultaneously obtain the higher orientation degree and enhancement efficiency, reducing the molecular rotational temperature is a good method.

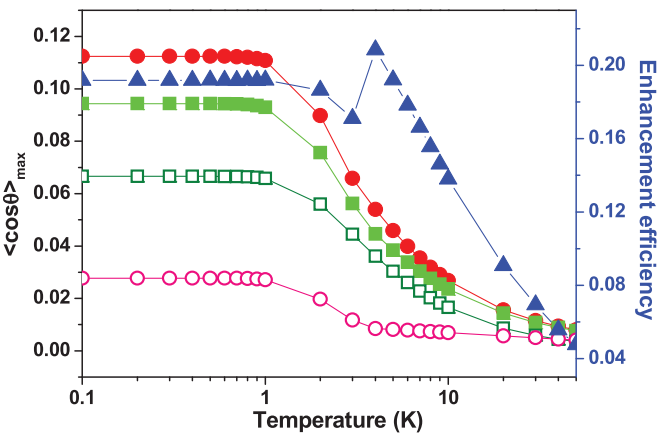

FIG. 5. (Color online) The maximum orientation degree $\langle\cos\rangle_{max}$ and enhancement efficiency as a function of the molecular rotational temperature. Here all lines are labeled the same as those shown in Fig. 2.

IV. CONCLUSIONS

In conclusion, we have demonstrated that by combining an electrostatic field and a four-color laser field, a hybrid field can be an effective tool to enhance molecular orientation. It was shown that the molecular orientation by the combined field can be obtained under a laser-field-free condition, and the maximal orientation degree can be enhanced by ∼20% compared with the sum of the orientation degree individually created by the electrostatic field and the four-color laser field. It was also shown that the orientation enhancement can be attributed to the larger asymmetry of the four-color laser field due to the existence of the electrostatic field. Furthermore, the effects of the carrier-envelope phase, laser intensity, and pulse duration of the four-color laser field and the molecular rotational temperature on the orientation enhancement were discussed. Both the electrostatic field and four-color laser field can be obtained based on the existing experimental conditions, so we believe that our proposed scheme is feasible in real experimentation.

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