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International Journal of Mass Spectrometry 254 (2006) 37–43

International Journal of Mass Spectrometry

www.elsevier.com/locate/ijms

Effects of polarizability and hyperpolarizability on molecular spatial alignment in intense light fields

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Received 22 March 2006; received in revised form 26 April 2006; accepted 26 April 2006

Abstract

Considering the influences of polarizability and hyperpolarizability on molecular spatial alignment, we newly deduce the classical equation of molecular motion for the internuclear distance r and for the angle θ between molecular axis and laser polarization direction in intense light fields. Numerical simulations of molecular dynamic alignment are performed and discussed at intensities of 10^{15}–10^{17} W/cm^2 and pulse durations of 100 fs and shorter than 100 fs. In our calculation, N_2 and Cl_2 molecules are selected as sample because they can represent the light molecule and heavy molecule, respectively. Our computational results show that the contributions of the molecular polarizability and hyperpolarizability to the degree of dynamic alignment all become strong when the laser intensity increases and the pulse duration narrows. And the role of hyperpolarizability on molecular dynamic alignment becomes comparable to that of linear polarizability term, even exceeds it in the conditions of high laser intensity and short pulse length. The characteristics of molecular angular distribution with only considering the effect of polarizability, second hyperpolarizability, and higher-order correction term of the hyperpolarizability, respectively, are very different. Their contributions to molecular dynamic alignment are systematically calculated and discussed in different laser intensities and pulse durations.

PACS: 33.80.Rv; 42.50.Hz; 33.90. +h

Keywords: Polarizability; Hyperpolarizability; N_2 and Cl_2 molecules; Intense light fields

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1. Introduction

The interaction of molecules with strong, non-resonant femtosecond laser pulses has been investigated in the last decade. Many fundamental physical processes have been understood by means of a variety of experimental techniques and theoretical methods [1–5]. Particularly, laser-induced spatial alignment is a great interesting project because of its potential applications [6,7]. The molecular spatial alignment in intense femtosecond laser fields has alternatively been explained in terms of geometric alignment and dynamic alignment mechanisms [8,9]. The contributions of dynamic alignment to the observed anisotropic angular distribution of fragmental ions base on molecular rotation and reorientation because the laser-induced dipole moments exert sufficiently strong torques on the molecular axis. For the majority of theoretical models, a description by solving numerically the classical equations of molecular motion for the internuclear distance r and for the angle θ between molecular axis and laser polarization direction is usually used to understand molecular dynamic alignment. At present, this model only considers the role of the molecular polarizability on the laser-induced alignment dynamics but neglects the effect of molecular hyperpolarizability [10–16]. Recently, the infra-red femtosecond lasers at intensities of above 5×10^{15} W/cm^2 or shorter than 100 fs pulse duration are used to experimentally study laser-induced alignment in molecular Coulomb explosion [12,17–21]. So, accounting for the influence of molecular hyperpolarizability on spatial alignment is necessary.

In this paper, we simultaneously consider the effects of polarizability and hyperpolarizability on molecular spatial alignment

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1387-3806/$ – see front matter © 2006 Published by Elsevier B.V.
doi:10.1016/j.ijms.2006.04.013

and newly deduce the equation of motion for θ between the molecular axis and electric field vector. Numerical simulations of molecular dynamic alignment are performed and discussed at intensities of 10^{15}–10^{17} W/cm^2 and pulse durations of 100 fs and shorter than 100 fs. Our studies focus on N_2 and Cl_2 molecules because they can represent the light molecule and heavy molecule, respectively.

2. Numerical method

For N_2 and Cl_2 molecules, their permanent dipole moment and the first hyperpolarizability are zero because of the property of the centrosymmetrical structure. The field-molecule interaction potential is given by:

$$V(\theta) = -\vec{\mu}\cdot\vec{E}(t) = -\frac{[E(t)]^2}{2}[\alpha_\parallel \cos^2(\theta) + \alpha_\perp \sin^2(\theta)] - \frac{[E(t)]^4}{24}[\gamma_\parallel \cos^4(\theta) + \gamma_\perp \sin^4(\theta)] \quad (1)$$

where θ is the angle between the molecular axis and the laser field direction, $E(t)$ is laser field strength, $\alpha_\parallel$ and $\alpha_\perp$ are the polarizability components parallel to and perpendicular to the molecular bond, and $\gamma_\parallel$ and $\gamma_\perp$ are the parallel and perpendicular components of the second hyperpolarizability tensor. In this paper, we use the ground-state polarizability and hyperpolarizability of a neutral molecule but neglect other uncertainties of polarizability and hyperpolarizability.

In our deductions and calculations, the laser pulses are defined as Gaussian distribution: $E(t) = E\exp(-t^2/2\tau^2)\cos(\omega t) = E_0(t)\cos(\omega t)$. Than the Lagrangian function $L = T - V$ is written as

$$L = T - V = \frac{1}{2}M[\dot{r}(t)]^2 + \frac{M[r(t)]^2}{2}[\dot{\theta}^2 + \sin^2\theta\dot{\varphi}^2] + \frac{[E_0(t)]^2[\cos(\omega t)]^2}{2}[\alpha_\parallel \cos^2(\theta) + \alpha_\perp \sin^2(\theta)] + \frac{[E_0(t)]^4[\cos(\omega t)]^4}{24}[\gamma_\parallel \cos^4(\theta) + \gamma_\perp \sin^4(\theta)]$$

with M the reduced mass of molecule and φ the azimuthal angle. So we can obtain the expression of $(\mathrm{d}/\mathrm{d}t)(\partial L/\partial\dot{\theta})$ and $\partial L/\partial\theta$.

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{\partial L}{\partial\dot{\theta}}\right) = 2Mr(t)\dot{r}(t)\dot{\theta} + M[r(t)]^2\ddot{\theta},$$

$$\frac{\partial L}{\partial\theta} = M[r(t)]^2\dot{\varphi}^2\sin(\theta)\cos(\theta) - \frac{[E_0(t)]^2}{2}\alpha_{\mathrm{eff}}\sin(2\theta) \times \left[\frac{1+\cos(2\omega t)}{2}\right] - \frac{[E_0(t)]^4}{24}[\gamma_\parallel \cos^2(\theta) - \gamma_\perp \sin^2(\theta)]\sin(2\theta)\left[\frac{3}{4} + \cos(2\omega t) + \frac{1}{4}\cos(4\omega t)\right]$$

$\alpha_{\mathrm{eff}} = \alpha_\parallel - \alpha_\perp$ represents the anisotropy of the polarizability. According to Lagrange equation, the motion equation for θ can be obtained:

$$\ddot{\theta} = \frac{\dot{\varphi}^2\sin(2\theta)}{2} - \frac{2\dot{r}(t)\dot{\theta}}{r(t)} - \frac{\alpha_{\mathrm{eff}}[E_0(t)]^2\sin(2\theta)}{4M[r(t)]^2} - \frac{[E_0(t)]^4\sin(2\theta)}{32M[r(t)]^2}[\gamma_\parallel \cos^2(\theta) - \gamma_\perp \sin^2(\theta)] - \frac{1}{M[r(t)]^2}\left\{\frac{\alpha_{\mathrm{eff}}[E_0(t)]^2}{4} - \frac{[E_0(t)]^4}{24}[\gamma_\perp \sin^2(\theta) - \gamma_\parallel \cos^2(\theta)]\right\}\sin(2\theta)\cos(2\omega t) - \frac{1}{M[r(t)]^2}\frac{[E_0(t)]^4}{96} \times [\gamma_\parallel \cos^2(\theta) - \gamma_\perp \sin^2(\theta)]\sin(2\theta)\cos(4\omega t) \quad (2)$$

When the laser frequency is much larger than the rotational frequency of molecules, which is in the high-frequency off-resonance regime, the right hand side of Eq. (2) can be classified into two terms: the time-dependent term $\ddot{\theta}_1$ and the other rapidly oscillating time-dependent term $\ddot{\theta}_2$.

$$\ddot{\theta}_1 = \frac{\dot{\varphi}^2\sin(2\theta)}{2} - \frac{2\dot{r}(t)\dot{\theta}}{r(t)} - \frac{\alpha_{\mathrm{eff}}[E_0(t)]^2\sin(2\theta)}{4M[r(t)]^2} - \frac{[E_0(t)]^4\sin(2\theta)}{32M[r(t)]^2}[\gamma_\parallel \cos^2(\theta) - \gamma_\perp \sin^2(\theta)],$$

$$\ddot{\theta}_2 = -\frac{1}{M[r(t)]^2}\left\{\frac{\alpha_{\mathrm{eff}}[E_0(t)]^2}{4} - \frac{[E_0(t)]^4}{24}[\gamma_\perp \sin^2(\theta) - \gamma_\parallel \cos^2(\theta)]\right\}\sin(2\theta)\cos(2\omega t) - \frac{1}{M[r(t)]^2}\frac{[E_0(t)]^4}{96} \times [\gamma_\parallel \cos^2(\theta) - \gamma_\perp \sin^2(\theta)]\sin(2\theta)\cos(4\omega t) \quad (3)$$

According to Eq. (3), the average kinetic energy of the oscillatory motion over optical cycle is

$$\frac{M[r(t)]^2}{2}\overline{\dot{\theta}_2^2} = \frac{1}{16M[r(t)]^2\omega^2}\left\{\frac{\alpha_{\mathrm{eff}}^2[E_0(t)]^4\sin^2(2\theta)}{16} + \frac{[E_0(t)]^8\sin^2(2\theta)}{24^2}[\gamma_\perp \sin^2(\theta) - \gamma_\parallel \cos^2(\theta)]^2 - \frac{\alpha_{\mathrm{eff}}[E_0(t)]^6\sin^2(2\theta)}{48}[\gamma_\perp \sin^2(\theta) - \gamma_\parallel \cos^2(\theta)]\right\} + \frac{[E_0(t)]^8\sin^2(2\theta)}{4\times 384^2M[r(t)]^2\omega^2}[\gamma_\perp \sin^2(\theta) - \gamma_\parallel \cos^2(\theta)]^2$$

Then the motion equation for θ can be obtained in terms of an effective force field derived from the sum of a time-dependent potential $V(\theta)$ (due to the time-dependent term $\ddot{\theta}_1$) and the time-averaged kinetic energy (due to the rapidly oscillating time-dependent term $\ddot{\theta}_2$)

$$I\ddot{\theta} = -\frac{\mathrm{d}(V(\theta) + (I/2)\overline{\dot{\theta}_2^2})}{\mathrm{d}\theta} = I\ddot{\theta}_1 - \frac{I}{2}\frac{\mathrm{d}\overline{\dot{\theta}_2^2}}{\mathrm{d}\theta}$$

We finally obtain the motion equation for θ

$$\begin{aligned}\ddot{\theta} = & -\frac{2\dot{r}(t)\dot{\theta}}{r(t)} - \frac{\alpha_{\text{eff}}}{4M[r(t)]^2} \times \sin(2\theta)[E_0(t)] \\ & - \frac{1}{32M[r(t)]^2}[\gamma_{\|}\cos^2(\theta) - \gamma_{\perp}\sin^2(\theta)] \times \sin(2\theta)[E_0(t)]^4 \\ & - \left\{\frac{[\alpha_{\text{eff}}]^2\cos(2\theta)}{64\{M[r(t)]^2\omega\}^2}\right\}\sin(2\theta)[E_0(t)]^4 \\ & - \frac{\alpha_{\text{eff}}}{768\{M[r(t)]^2\omega\}^2}[(4\cos 2\theta\cos^2\theta - \sin^2 2\theta)\gamma_{\|} \\ & - (4\cos 2\theta\sin^2\theta + \sin^2 2\theta)\gamma_{\perp}]\}\sin(2\theta)[E_0(t)]^6 \\ & - \Bigg\{\frac{1}{2} \times \left(\frac{1}{\{48M[r(t)]^2\omega\}^2} + \frac{1}{\{384M[r(t)]^2\omega\}^2}\right) \\ & \times\ [\gamma_{\perp}\sin^2(\theta) - \gamma_{||}\cos^2(\theta)] \times [(2\cos 2\theta\sin^2\theta \\ & + \sin^2 2\theta)\gamma_{\perp} - (\sin^2 2\theta - 2\cos 2\theta\cos^2\theta)\gamma_{\|}]\Bigg\} \\ & \times \sin(2\theta)[E_0(t)]^8 \equiv -\frac{2\dot{r}(t)\dot{\theta}}{r(t)} - A - B - C - D - E \end{aligned} \tag{4}$$

In Eq. (4), we have taken $\dot{\varphi} = 0$ because the molecule-laser interaction is independent of the azimuthal angle φ. And the parameters A–E in the right hand side of Eq. (4) are introduced for the future convenience. These terms can cause molecular reorientation. They based on molecular polarizability and hyperpolarizability. In this paper, we mainly investigate the contributions of these terms to the molecular alignment at different laser intensities and pulse lengths. The parameters used, which comprise the polarizability, the second hyperpolarizability of N_2 and Cl_2 molecules, are originated from the computational results of Maroulis and Archibong [22,23]. $(2\dot{r}(t)\dot{\theta}/r(t))$ is the damping term that blocks molecular rotation, which derived from the increasing of internuclear distance during the process of molecular Coulomb explosion and multielectron dissociative ionization. The motion equation for internuclear distance $r(t)$ can be written as:

$$\ddot{r}(t) = \frac{Q_1 Q_2}{M[r(t)]^2}$$

Q_1 and Q_2 are the charges of the two ions, respectively. In the following calculations, we consider the situation of $Q_1 = Q_2 = 1$.

Experiments have proved that the ratio of measured kinetic energy of fragmental ions and Coulomb explosion energy is about 45% for N_2, CO, and O_2 molecules but about 70% for Cl_2 and I_2 molecules [24–26] (this is also why we select N_2 and Cl_2 molecules as sample in our calculations). The two-step Coulomb explosion model and the field ionization Coulomb explosion model described clearly this process from theoretical point of view. These two models indicated that the Coulomb explosion and multielectron dissociative ionization of molecules in intense femtosecond laser fields occurs at a critical internuclear distance larger than the equilibrium internuclear distance. [27,28]. The molecular Coulomb explosion undergoes two steps according to these two models. The first step is that the neutral molecules are ionized and the resulting molecular ions align along the laser polarization direction. Once the molecule is ionized beyond the first stage, the two nuclei mutually repel due to the Coulomb force. Then additional electrons are lost rapidly when the internuclear distance elongates to the critical distance. In our calculations, the damping term $2\dot{r}(t)\dot{\theta}/r(t)$in Eq. (4) begins to play a role for the molecular alignment when the neutral molecule is ionized. As the internuclear distance extends to the critical distance, we think that the repulsion between the two nuclei vanishes. That means that the molecule does not survive and the dynamic rotation of molecules arose from the interaction of induced dipole moments and laser field is over at this moment.

The time evolution of the angle for a given initial angle θ_0 can be described by solving numerically Eq. (4) using a fourth-order Runge–Kutta algorithm. So we develop a counting approach to describe molecular spatial angular distribution. A corresponding series of final angles are calculated through solving numerically for a series of initial angles θ_0. Thus, the angular distribution of molecules at a particular instant is obtained through counting a series of final angles.

3. Results and discussion

Fig. 1 presents the angular distribution of N_2 molecule at intensities of 10^{15}–10^{17} W/cm^2 and pulse durations of 100 and 20 fs. The laser wavelength is 800 nm. The black, red green, blue and magenta curves of each figure represent the degree of molecular alignment when only considering A–E terms in the right hand side of Eq. (4), respectively. It can be seen that no significant reorientation of N_2 molecule occurs with only considering C–E terms when laser intensity varies from 10^{15} to 10^{17} W/cm^2 with 100 and 20 fs pulse durations. These three terms have the same characteristics, which are inversely proportional to the square of laser frequency. In our calculations, laser frequency is 2.356×10^{15}/s (corresponding to laser wavelength of 800 nm). This makes the value of C–E terms become small. And C–E terms are also tightly related to laser intensity, the angle θ, molecular polarizability and hyperpolarizability. In fact, these three terms can be as the higher-order correction terms of contributions of the polarizability and hyperpolarizability to molecular alignment according to the deduced process of Eq. (4). For N_2 molecule, our computational results indicate that the contributions of higher-order correction terms to molecular alignment can be neglected in conditions of 800 nm laser wavelength and 20 fs (or 100 fs) pulse width for the laser intensity range of 10^{15}–10^{17} W/cm^2.

From Fig. 1, we can obtain that molecular rotation and reorientation due to the second hyperpolarizability (corresponding to the B term) will become comparable to that due to the A term (corresponding to the polarizability) at an intensity of 10^{17} W/cm^2 with 100 fs pulse length. As for 20 fs pulse duration, the contribution of the second hyperpolarizability (corresponding to the B term) to molecular alignment has exceeded

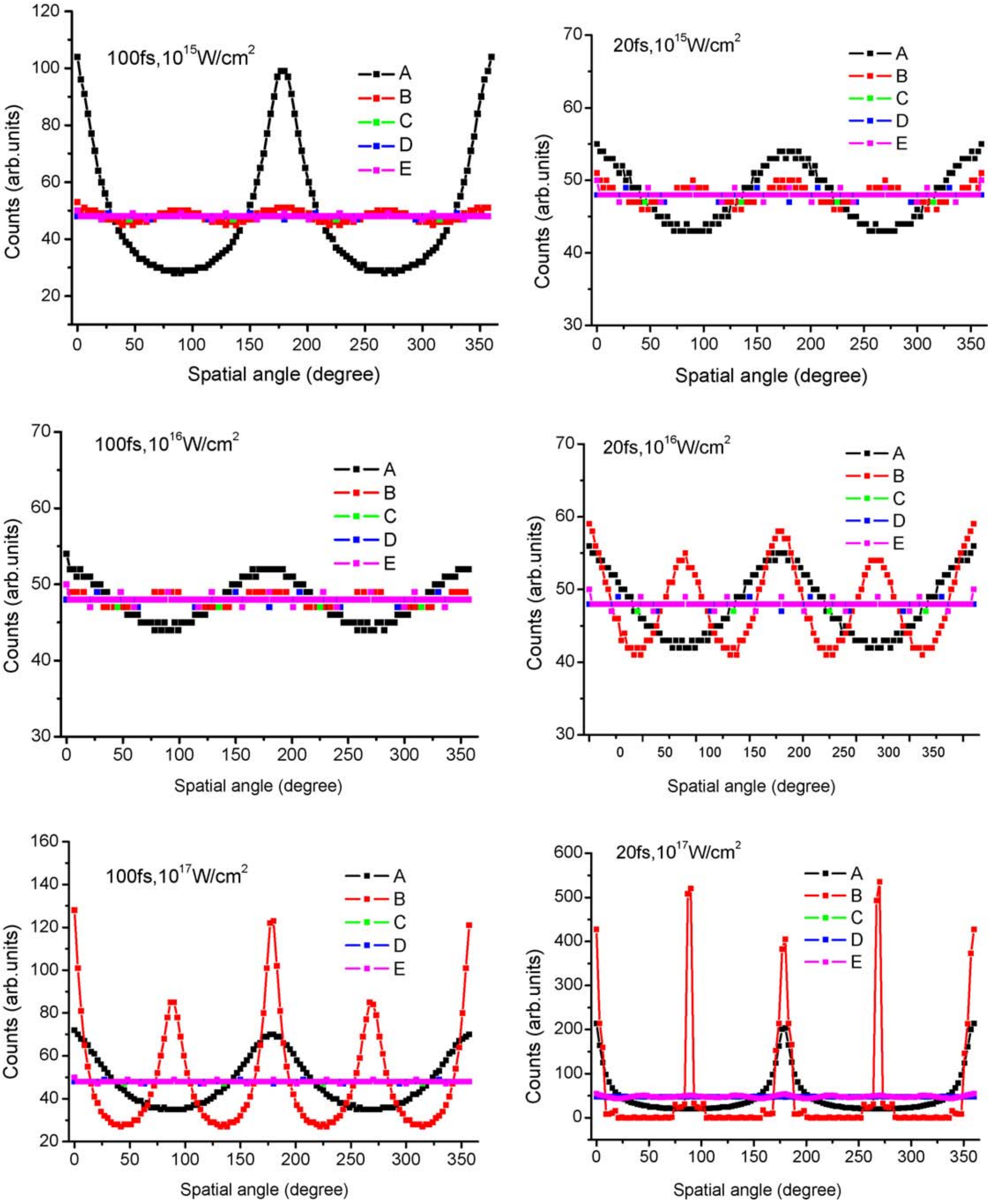

Fig. 1. Angular distributions of N_2 molecule at different intensities: 10^{15}, 10^{16}, and 10^{17} W/cm^2 with 20 and 100 fs pulse durations. The laser wavelength is 800 nm. The black, red green, blue and magenta curves of each figure represent the degree of molecular alignment when only considering A–E terms in the right hand side of Eq. (4), respectively (for interpretation of the references to colour in this figure legend, the reader is referred to the web version of the article).

that of the polarizability term at an intensity of 10^{16} W/cm^2. This indicates that the second hyperpolarizability term plays an important role for the degree of alignment when the femtosecond laser with short pulse length and high laser intensity irradiates the molecule. In the conditions of different laser pulses, the dependences of molecular dynamic alignment due to the polarizability term on laser intensities show apparently different characteristics. For 100 fs pulse duration, the full-width at half maxima (FWHMs) of angular distribution of N_2 molecule are $44.8^\circ \pm 0.9^\circ$, $87.9^\circ \pm 2.4^\circ$, $63.5^\circ \pm 0.7^\circ$ at intensities of 10^{15}, 10^{16} and 10^{17} W/cm^2, respectively. As for 20 fs pulse length, the angular distribution narrows with the increasing of laser intensity. The FWHMs of angular distribution decrease from $84.3^\circ \pm 2.6^\circ$ with laser intensity of 10^{15} W/cm^2 to $22.4^\circ \pm 0.8^\circ$ with laser intensity of 10^{17} W/cm^2. According to the expression of polarizability term in Eq. (4), we can find answer. The angular acceleration of molecular rotation with only considering the polarizability depends quadratically on laser intensity. The increasing of laser intensity results in the strong torque exerted on the molecule. However, the time of molecular alignment becomes short. The effects of these two factors on dynamic alignment compete with each other during the process of the

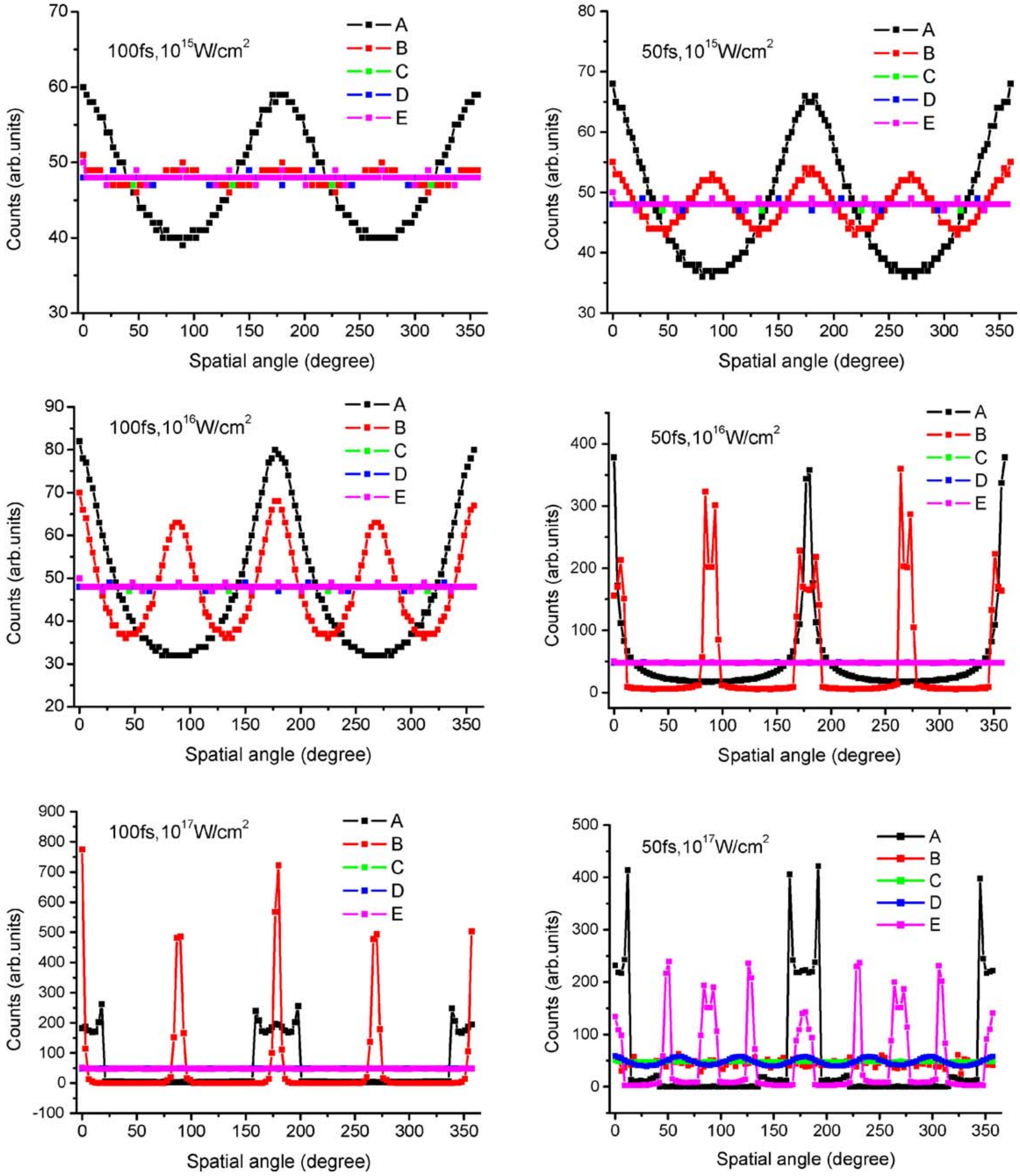

Fig. 2. Angular distribution of Cl_2 molecule at different intensities: 10^{15}, 10^{16}, and 10^{17} W/cm^2 with 50 and 100 fs pulse durations. The laser wavelength is 800 nm. The black, red green, blue and magenta curves of each figure represent the degree of molecular alignment when only considering A–E terms in the right hand side of Eq. (4), respectively (for interpretation of the references to colour in this figure legend, the reader is referred to the web version of the article).

molecular alignment. When the effect of the increasing torque on molecular alignment exceeds that of the shorten time of dynamic alignment, the angular distribution narrows with the increasing of laser intensity. From Fig. 1, we can obtain that the contribution of the molecular polarizability to the degree of dynamic alignment becomes strong in intense laser field with short pulse duration.

In numerical calculations of Cl_2 molecule, we choose the pulse widths of 50 and 100 fs instead of 20 and 100 fs because the molecule with the large reduced mass undergoes a long time to align before the two nuclei separate completely [14,16]. The angular distributions of Cl_2 molecule for different values of laser intensities and different pulse durations are presented in Fig. 2. It can be seen that contributions of the higher-order correction terms (C–E terms) of the polarizability and hyperpolarizability to molecular alignment may be negligible except for the condition of 50 fs pulse length and 10^{17} W/cm^2 laser intensity. For either 100 or 50 fs laser pulses, the FWHMs of angular distribution only due to the polarizability term become narrow with the increasing of laser intensity. When laser intensity reaches 10^{17} W/cm^2, the peak of molecular counts shifted away from the laser polarization direction but is near the laser polarization direction. The minimal value of molecular counts reaches zero, which is different from that case of N_2 molecule. The strong torque exerted on molecular axis is responsible for these variations. The torque generated by the laser field is sufficient to make molecules firstly rotate to the laser polarization direction. Then molecules oscillate around the laser polarization vector with small amplitude. A part of the molecules overshoots the laser polarization axis and cannot return to it before the two nuclei completely separate. As a result, the peak of angular distribution will shift away from the laser polarization direction. For the conditions of 100 fs and 50 fs pulse durations at the intensity larger than 10^{16} W/cm^2, molecular reorientation due to the hyperpolarizability terms will become comparable to that due to the polarizability. Especially in the condition of 50 fs pulse length and 10^{17} W/cm^2 laser intensity, although the contribution of the second hyperpolarizability becomes little, the effect of its higher-order correction terms (E term) on molecular alignment becomes comparable to that of linear polarizability term.

From Figs. 1 and 2, we note that the characteristics of molecular angular distribution with only considering the effect of polarizability (A term), second hyperpolarizability (B term), and higher-order correction term of the second hyperpolarizability (E term), respectively, are very different. The polarizability-field interaction makes the angular distributions of molecule have a maximum along the polarization axis and a minimum perpendicular to it. The role of the second hperpolarizability keeps the molecular counts a maximum along the laser polarization direction but a minimum at the position of angle of 45° between the molecular axis and the polarization direction. And there is the second maximum of molecular counts perpendicular to the polarization axis. When only considering the higher-order correction term of the second hyperpolarizability (see Fig. 2 in the condition of 50 fs pulse length and 10^{17} W/cm^2 laser intensity), the property of molecular dynamic alignment becomes very complex. Eq. (4) has given the dependences of these three terms on the angle θ. The time evolution of the different initial angle in these three cases can well explain the results of numerical simulation (as show as Fig. 3a–c). About 50 fs pulse length and 10^{17} W/cm^2 laser intensity are used in the calculations. It can be seen that all the molecules firstly rotate to the direction of the laser polarization vector when only considering the polarizability term. Then they oscillate around the polarization axis with small amplitude. This makes molecules predominantly align along the polarization direction of incident laser pulses.

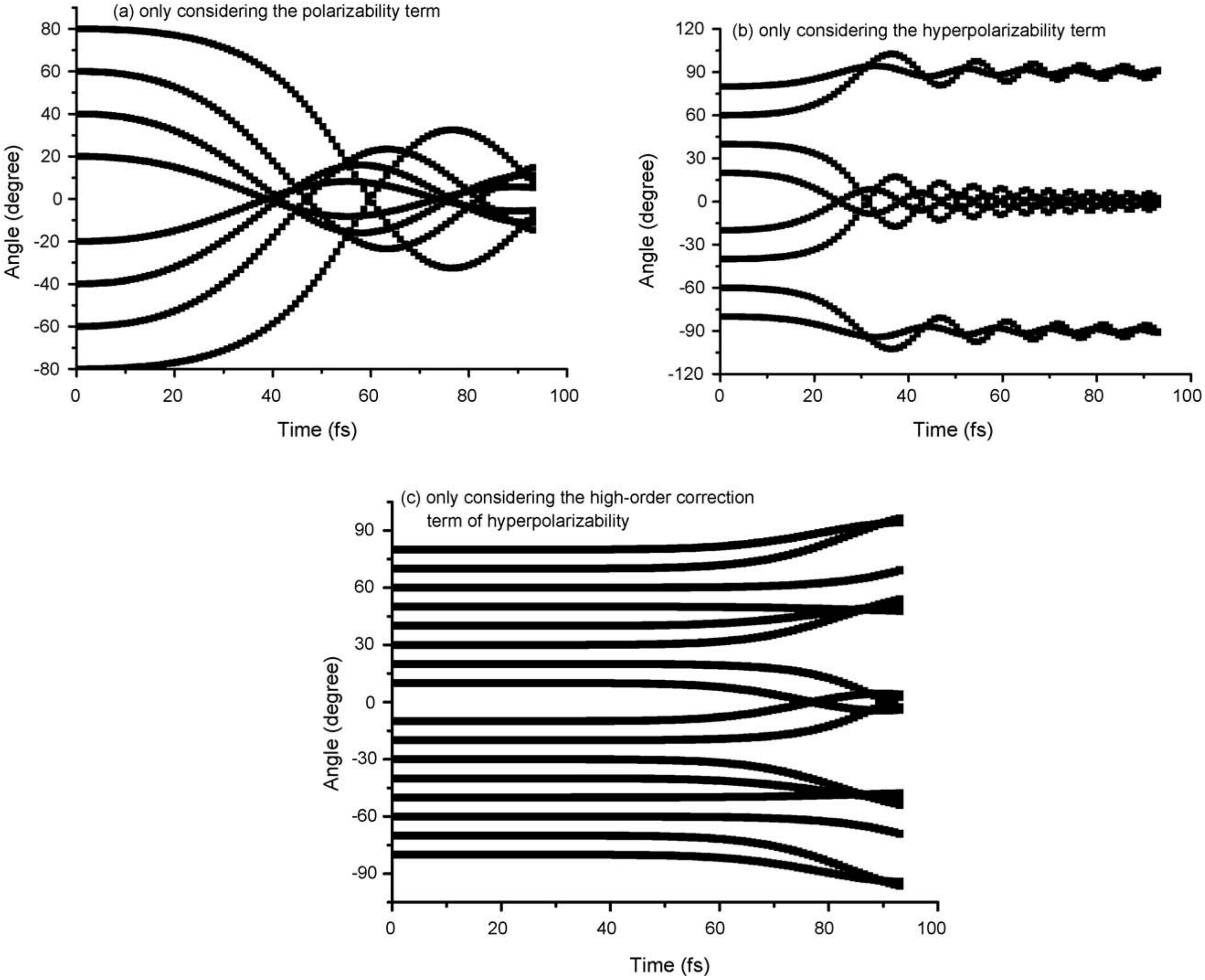

Fig. 3. Time evolution of the different initial angles for Cl_2 molecule with only considering the polarizability, the second hyperpolarizability terms and the higher-order correction term of the second hyperpolarizability, respectively. 50 fs pulse length and 10^{17} W/cm^2 laser intensity are used in the calculations.

While only taking into account the second hyperpolarizability term, a part of molecules rotate to the laser polarization direction and the other part rotate to the vertical direction of the laser field. And they also oscillate around the laser polarization direction or perpendicular to it with small amplitude. Fig. 3c presents the molecules rotate the position of angle of near 0°, 45°and 90° due to the higher-order correction term of the second hyperpolarizability, respectively. This is consistent with the result that the peaks of molecular counts located at the angle of near 0°, 45°and 90° while only taking into account the E term.

4. Conclusion

We have systematically calculated and discussed the influence of polarizability, hyperpolarizability and their high-order correction terms on molecular spatial alignment at intensities of 10^{15}–10^{17} W/cm^2 and pulse durations of 100 fs and shorter than 100 fs. The polarizability-field interaction makes the angular distributions of molecule have a maximum along the polarization axis and a minimum perpendicular to it. The role of the second hyperpolarizability keeps the molecular counts a maximum along the laser polarization direction but a minimum at the position of angle of 45° between the molecular axis and the polarization direction. And there is the second maximum of molecular counts perpendicular to the polarization axis. The peaks of molecular counts located at the angle of near 0°, 45° and 90° while only taking into account the higher-order correction term of the second hyperpolarizability. For N_2 and Cl_2 molecules, the dependences of molecular degree of dynamic alignment on the polarizability and hyperpolarizability present different property in the similar conditions. Usually, the role of hyperpolarizability on molecular dynamic alignment becomes comparable to that of linear polarizability term, even exceeds it in the conditions of high laser intensity and short pulse length. The comparison of different experimental results of molecular angular distribution between the conditions of high and low laser intensity or long and short pulse length may manifest the calculation results.

Acknowledgements

The project was supported by National Natural Science Foundation of China (No. 60508017), Heilongjiang Province Natural Science Foundation of China (A2004-04), and Natural Science Foundation of Fujian Province of China (No. A0510015), and Scientific Program of the Educational Hall of Fujian Province (NO.JA05215). We also appreciate the support of Professor Maroulis of Department of Chemistry in University of Patras and Fujian Provincial Key Laboratory of Photonic Technology.

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