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Characteristics of dynamic alignment for diatomic and linear triatomic molecules in intense femtosecond laser fields

Jianxin Chen [a,b,*], Xuefang Li [a]

[a] *School of Physics and OptoElectronics Technology, Fujian Normal University, Fuzhou 350007, PR China*
[b] *College of Science, Jiamusi University, Jiamusi 154007, PR China*

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Abstract

The process of dynamic alignment for diatomic and linear triatomic molecules is systematically investigated by solving numerically the rotation equation for angle θ between molecular axis and laser polarization direction in the high-frequency non-resonant regime. A two-step Coulomb explosion model of molecules in intense laser fields is used to determine the instant when molecular dynamic alignment is over. A counting approach and a fourth-order Runge–Kutta algorithm are used to calculate the angular distribution of molecules at a particular instant through solving numerically for a series of initial angle. Our computational results show that the majority of dynamic alignment for the light molecules N_2, H_2 and CO_2 takes place before the molecule ionizes and begins to dissociate. However, for the heavy molecules Br_2, I_2 and CS_2, the dynamic alignment occurs mainly during the process of molecular dissociation. The extent of molecular alignment tightly correlates not only with dynamic process of molecular Coulomb explosion, but also with molecular parameters and laser parameters. The effects of these factors to molecular dynamic alignment are extensively calculated and discussed.

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Keywords: Dynamic alignment; Diatomic and linear triatomic molecules; Intense femtosecond laser field

1. Introduction

Recently, the interactions of molecules and high intensity optical fields are investigated by using the method of a time-of-flight mass spectrometry and a chirped pulse amplifier laser. Many experiments performed on diatomic and triatomic molecules have observed that the angular distributions of ionic fragments arising from field ionization and Coulomb explosion of molecules are highly anisotropic: yields of ionic fragments are maximum along the laser polarization direction and minimum in the orthogonal direction [1–4]. Two alignment mechanisms have been proposed to explain such anisotropic angular distributions of ionic fragments. One is so-called dynamic alignment. The intense laser field induces dipole moments within the molecule. Strong torques on the molecular axis force the molecule to align along the laser polarization direction prior to or during the ionization and dissociation [5,6]. The other is geometric alignment. Molecules along the laser polarization direction are preferentially ionized and dissociated because of the dependence of ionization rate on the angle between the molecular axis and the direction of laser polarization vector [7,8]. Experimental investigation further indicated that alignment mechanism and alignment extent of molecules are strongly different for different pulse durations, laser intensity, especially for different molecules [3,4,8–13]. The spatial alignment of molecules has many practical applications, such as chemical reaction [14], trapping molecules [15], enhanced high-order harmonic generation [16,17] and especially control of photodissociation [18,19]. So, the study of molecular alignment process in theoretical simulations is necessary. At present, Baner-

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[*] Corresponding author. Tel.: +86 591 22686078; fax: +86 591 83456462.
E-mail address: chenjianxin@fjnu.edu.cn (J. Chen).

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jee et al. have studied the molecular reorientation using a classical model and accounted for both enhanced ionization and hyperpolarizability in the alignment of molecules [12]. Dion et al. simulated numerically the alignment dynamics of HCN by non-perturbative solutions of the time-dependent Schrödinger equation [20]. A two-dimensional filed-ionization Coulomb explosion model has been used to study the angular distribution of fragmental ions of diatomic molecules [21]. A time-dependent perturbation theory based on unitary transformations combined with averaging techniques is established to investigate molecular reorientation [22]. The dependences of the total ionization rates and yields on the orientation of the molecule are presented using the strong-field *S*-matrix approach [23].

In this paper, dynamic alignment of molecules $N_2, H_2, Br_2, I_2, CO_2$ and CS_2 is systematically investigated by solving numerically the rotation equation for θ between molecular axis and laser polarization direction in the high-frequency non-resonant regime. The effects of molecular parameters and laser parameters on the extent of molecular dynamic alignment are mainly calculated and discussed.

2. Description of the model

In the dynamic alignment mechanism, the intense laser filed induces dipole moments within the molecule, which, in turn, interacts with the field. Consequently, this exerts a torque that aligns the molecules along the direction of laser polarization vector. The dipole moment of molecule in the electric field can be expressed as [19,24]:

$$\vec{\mu} = \vec{\mu}_0 + \frac{1}{2}\alpha\vec{E} + \frac{1}{6}\beta\vec{E}\vec{E} + \frac{1}{24}\gamma\vec{E}\vec{E}\vec{E} + \cdots$$

where $\vec{\mu}_0$ is the permanent dipole moment, α the linear polarizability tensor and β and γ the first and second hyperpolarizability tensors. When the laser intensity exceeds 10^{16} W/cm^2, the reorientation of a molecule due to the hyperpolarizability will become comparable to that due to the linear polarizability [12]. In previous experiments, the laser intensity of about 10^{15} W/cm^2 was used to study molecular alignment [3,4,8–12]. So, the interaction potential between the laser electric field and molecules can be written as:

$$V = -\vec{\mu} \bullet \vec{E}(t) = -\mu_0 E(t)\cos(\theta) - \frac{[E(t)]^2}{2}[\alpha_{||}\cos^2(\theta) + \alpha_\perp \sin^2(\theta)]$$

in which, $\alpha_{||}$ and $\alpha_\perp$ are the parallel and perpendicular components of the linear polarizability tensor and θ the angle between the molecular axis and the laser polarization direction. In order to make our calculations realistic, a Gaussian laser pulse $E(t) = E\exp(-t^2/2\tau^2)\cos(\omega t) = E_0(t)\cos(\omega t)$ is used in our calculations. The Lagrangian function is written as:

$$L = \frac{M[r(t)]^2}{2}[\dot{\theta}^2 + \sin^2\theta\dot{\varphi}^2] + \mu_0 E_0(t)\cos(\omega t)\cos(\theta) + \frac{[E_0(t)]^2[\cos(\omega t)]^2}{2}[\alpha_{||}\cos^2(\theta) + \alpha_\perp \sin^2(\theta)]$$

where *r(t)* is the molecular internuclear distance, which is the function of time during the ionization and dissociation of molecules. M is the reduced mass of molecule and φ the azimuthal angle.

When the laser frequency is much larger than the rotational frequency of molecules, which is in the high-frequency non-resonant regime, the rotation equation for θ according to Lagrange equation can be deduced:

$$\ddot{\theta} = -\frac{\alpha_{\mathrm{eff}}[E_0(t)]^2}{4M[r(t)]^2}\sin(2\theta) - \frac{[\mu_0]^2[E_0(t)]^2\sin(2\theta)}{4\{M[r(t)]^2\}^2\omega^2} + \frac{\dot{\varphi}^2\sin(2\theta)}{2} - \frac{2\dot{\theta}\dot{r}(t)}{r(t)} - \frac{[\alpha_{\mathrm{eff}}]^2[E_0(t)]^4}{128\{M[r(t)]^2\}^2\omega^2}\sin(4\theta) \quad (1)$$

where $\alpha_{\mathrm{eff}} = \alpha_{||} - \alpha_\perp$ represents the anisotropy of the molecular linear polarizability. The molecule–laser interaction is independent of the azimuthal angle φ. That means $\dot{\varphi} = 0$. Our calculations show that contributions of the second-order field-induced dipole moment and the higher-order correction term to molecular rotation acceleration compared to the linear polarizability are negligible for 800 nm 10^{15} W/cm^2 intensity. Eq. (1) can be simplified to:

$$\ddot{\theta} = -\frac{\alpha_{\mathrm{eff}}[E_0(t)]^2}{4M[r(t)]^2}\sin(2\theta) - \frac{2\dot{\theta}\dot{r}(t)}{r(t)} \quad (2)$$

The process of molecular dynamic alignment is tightly correlated with that of molecular Coulomb explosion. The two-step Coulomb explosion model presents clearly the process of multielectronic dissociative ionization of molecules in intense laser fields [25]. The molecular Coulomb explosion occurs in two steps according to this model. 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 $R_c = 4.07/I_p$ (in atomic units), where I_p is the ionization potential of the molecule. In our calculations, the damping term in Eq. (2) 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 dynamic rotation of molecules 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. (1) by using a fourth-order Runge–Kutta algorithm. So using a counting approach, the angular distribution of molecules at a particular instant can also be obtained through solving numerically for a series of initial angles θ_0. The extent of the molecular

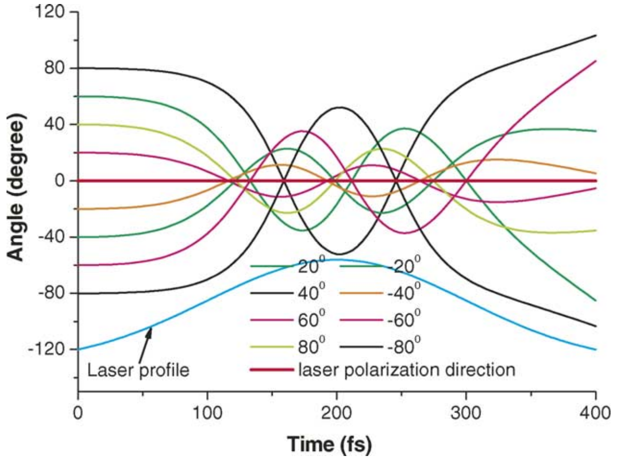

Fig. 1. Time evolution of the different initial angle for N_2 for a laser intensity of 10^{15} W/cm^2 and a pulse length of 100 fs without considering molecular ionization and dissociation in intense laser fields.

alignment can be estimated at the full width at half maximum (FWHM) of the angular distribution. In this paper, the method of a Gaussian fit was used to calculate the FWHM of the angular distribution.

3. Results and discussion

3.1. Molecular dynamic alignment without considering molecular ionization and dissociation in intense laser fields

When intense femtosecond laser pulses irradiate the molecule, the electric field of the intense laser will induce the dipole moment within the molecule. The induced dipole moments set up a torque on the molecular axis that can be large enough to spatially reorientate the molecule. If we only consider the effects of neutral molecule polarizability (without considering molecular ionization and dissociation), the damping term in Eq. (2) is zero and the molecule continues to rotate even after the laser field is switched off. Fig. 1 gives the time evolution of the different initial angle in the above case. The laser intensity of 10^{15} W/cm^2 and the pulse width of 100 fs for N_2 are used in our calculation. It can be seen that all the molecules firstly rotate to the direction of the laser polarization vector at the rising edge of the laser pulse. Then, they oscillate around the laser polarization vector with small amplitude. The longer the pulse length is, the quicker the oscillation is. All the molecules in intense laser fields align to the direction of the laser polarization vector. This result is consistent with the experimental observations that the fragment ions are predominantly ejected along the polarization direction of incident laser pulses [1–4]. This also shows that Eq. (2) can explain some experimental data even if we only consider the effects of neutral molecule polarizability.

The molecular oscillations about the laser polarization axis will cause the peak of the molecular angular distribution to be shifted away from the laser polarization direction at some instants. As shown in Fig. 2, the N_2 angular distribution varies significantly with several particular instants ($t = 100$, 200, 300 and 400 fs). At the instant of 100 fs, the molecules are aligned mostly in the direction of the laser polarization vector. At the instant of 200 fs, the maximum value of molecular counts is still in the laser polarization vector. But there exist a second peak at other direction. As for the instant of 300 fs, the peak of molecular counts is shifted away from the laser polarization direction. Finally, when the laser

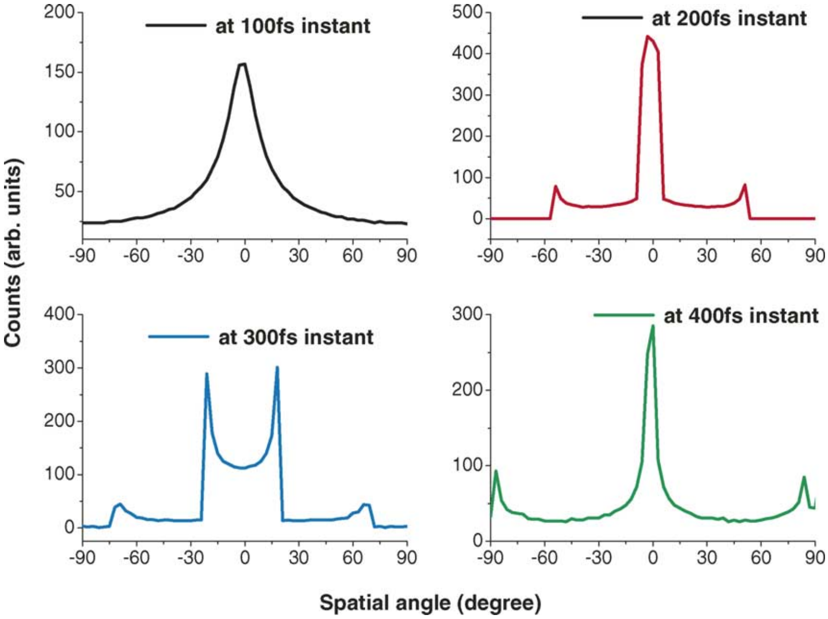

Fig. 2. Angular distributions for N_2 at several particular instants ($t = 100$, 200, 300 and 400 fs) for laser intensity of 10^{15} W/cm^2 and a pulse length of 100 fs.

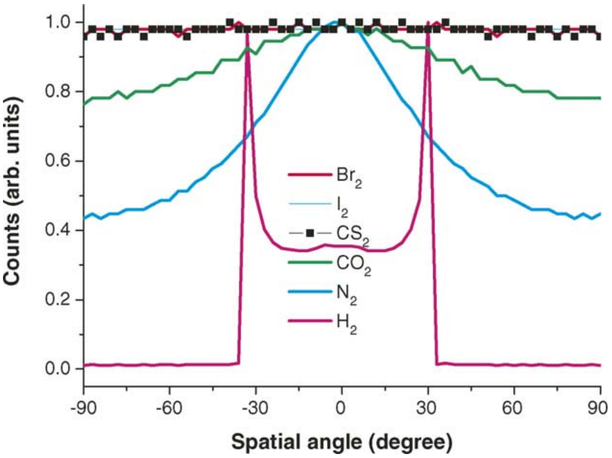

Fig. 3. Angular distributions for H_2, N_2, Br_2, I_2, CO_2 and CS_2 at the instant when the molecules ionized for a laser intensity of 10^{15} W/cm^2 and a pulse length of 100 fs.

pulse is over ($t = 400$ fs), the peak comes back to the laser polarization direction again. Similar calculations have been carried out for H_2, Br_2, I_2, CO_2 and CS_2. Similar results are obtained. These indicate that the instants at which the molecule is ionized and especially molecular dissociation is over are important factors in analyzing the extent of dynamic alignment.

3.2. Dependences of the extent of molecular dynamic alignment on molecular parameters

The molecular ionization and dissociation are closely related to molecular parameters including molecular reduced mass, ionization potential, the moment of inertia and linear polarizability. Figs. 3 and 4 show the counts of H_2, Br_2, I_2, CO_2 and CS_2 as a function of spatial angle at the instants at which the molecule ionized and molecular dissociation was over, respectively. The laser intensity of 10^{15} W/cm^2 and the pulse width of 100 fs are used in our calculations. At the instant of molecular ionization, there are no reorientations of molecular axis with the direction of laser polarization vector for Br_2, I_2 and CS_2. However, for N_2 and CO_2, the dynamic alignment of molecules is apparent and important. In the case of H_2, the peak of angular distribution is shifted away from zero, which occurs at the point where the polar angle between the molecular axis and the direction of the laser polarization vector is $\pm 30^\circ$. At the instant that the molecular dissociation was over (that means molecular dynamic rotation ended), the angular distributions of N_2 and CO_2 have no apparent change compared to that at the instant of molecular ionization. For H_2, the peak of molecular counts is still at the point that the angle is $\pm 30^\circ$. But some molecules have come back to zero. There is a second maximum peak of angular distribution along the laser polarization direction. In the case of Br_2, I_2 and CS_2, the extent of molecular reorientation caused by the dynamic alignment is not negligible. These show that the dynamic alignment of the light molecules H_2, N_2 and CO_2 occurs mainly before the molecule ionizes and begins to dissociate. However, for heavy molecules Br_2, I_2 and CS_2, most dynamic alignment occurs during the process of molecular dissociation.

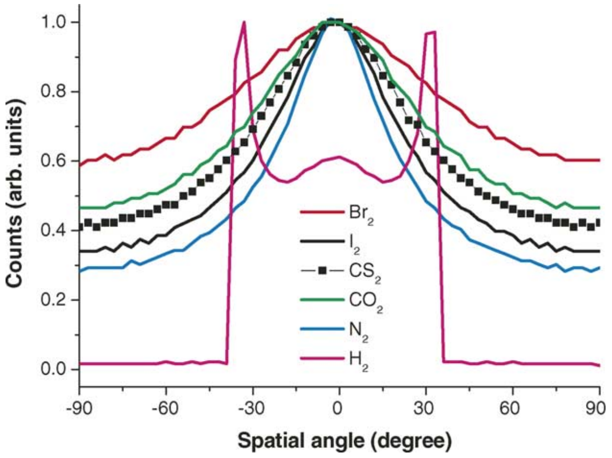

Fig. 4. Angular distributions for H_2, N_2, Br_2, I_2, CO_2 and CS_2 at the instant when the molecular dissociation was over for a laser intensity of 10^{15} W/cm^2 and a pulse length of 100 fs.

Three major molecular parameters are responsible for these. These are molecular reduced mass, molecular ionization potential and the ratio of the molecular linear polarizability to the moment of inertia. Table 1 presents the comparison of these parameters for H_2, Br_2, I_2, CO_2 and CS_2. R refers to the ratio of the molecular linear polarizability to the moment of inertia at the equilibrium internuclear distance of neutral molecule, R_e. The ionization potential of H_2, N_2 and CO_2 is larger than that of Br_2, I_2 and CS_2. That means the threshold intensities of ${H_2}^+$, ${N_2}^+$ and ${CO_2}^+$ ions are higher than that of ${Br_2}^+$, ${I_2}^+$ and ${CS_2}^+$ ions. When the molecule is exposed to the same laser field, the molecule with higher ionization potential requires longer time to align before the molecule ionizes and begins to dissociate. For example, the times of dynamic alignment for H_2, Br_2, I_2, CO_2 and CS_2 are 79.79, 78.18, 26.27, 13.8, 60.77, and 21.35 fs, respectively, at laser intensity of 10^{15} W/cm^2 and pulse duration of 100 fs (the peak instant of laser pulse is 200 fs).

On the other hand, for a larger ratio R a larger torque is induced by the dipole moments. For H_2, the ratio R is suf-

Table 1
Comparison of molecular parameters including the molecular reduced mass, the molecular ionization potential, and the ratio R of molecular linear polarizability to moment of inertia at the equilibrium internuclear distance of neutral molecule, R_e, for H_2, N_2, Br_2, I_2, CO_2 and CS_2

Molecules /Parameters	Molecular reduced mass (kg)	Molecular ionization energy (eV)	R (AS/V) (kg)
H_2	0.84×10^{-27}	15.43	7.64×10^{6}
N_2	11.62×10^{-27}	15.58	5.71×10^{5}
Br_2	66.32×10^{-27}	10.52	1.35×10^{5}
I_2	105.33×10^{-27}	9.40	9.99×10^{4}
CO_2	13.77×10^{-27}	13.77	3.80×10^{5}
CS_2	26.56×10^{-27}	10.07	4.50×10^{5}

ficiently large that the molecular axis has already aligned with the direction of laser electric vector before the molecule ionizes. And the molecular axis oscillates about the laser polarization vector with small amplitude. This leads to the peaks of molecular counts to divert from the laser polarization direction at the instant when the molecule is ionized. In the case of molecules Br_2, I_2 and CS_2, the low ionization energy and the relatively small value of R cause virtually no reorientation of the molecular axis with the laser polarization direction before the molecule ionizes and begins to dissociate. Once the molecule is ionized, the molecular dissociation will begin because of mutual repulsion of internuclear Coulomb force. The damping force will play an important role for the alignment due to the elongation of the internuclear distance. At this moment, the effects of the molecular reduced mass on the degree of alignment come to be important. The molecule with the low reduced mass will dissociate more rapidly and end quickly to its rotation and dynamic alignment, while the one with the large reduced mass undergoes a long time to align before the two nuclei separate completely. For example, the times of dynamic alignment for molecules N_2, H_2, Br_2, I_2 and CS_2 are 23.61, 3.69, 119.70, 181.60, 39.49 and 88.13 fs during the process of dissociation, respectively. So Br_2, I_2 and CS_2 will experience stronger alignment compared to H_2, N_2 and CO_2 after they are ionized. The extent of dynamic alignment for H_2, N_2 and CO_2 is larger than that for other molecules at the conditions of the same laser intensity and pulse length.

3.3. Dependences of the extent of molecular dynamic alignment on laser parameters

According to Eq. (2), the angular acceleration of molecular rotation is proportional to the square of the electric field. As the laser intensity increases, the torque exerted on the molecule will become strong. However, the time of molecular dynamic alignment becomes short with the increasing of laser intensity before the molecule is ionized and begins to dissociate. The effects of these two factors on molecular alignment compete with each other during the process of the molecular alignment. Figs. 5 and 6 present the angular distributions of N_2 and I_2 at different laser intensities. For N_2, the FWHM of angular distribution increases from $4.7 \pm 0.2^\circ$ with laser intensity of 3×10^{14} W/cm^2 to $73.0 \pm 0.9^\circ$ with laser intensity of 5×10^{15} W/cm^2. As for I_2, the angular distribution narrows with increasing of laser intensity. These suggested that the times of dynamic alignment before the molecule is ionized for N_2 play a main role for molecular alignment compared to the increasing torque. Conversely, the effect of the increasing torque is important for the alignment of I_2 when the laser intensity increases.

At a given laser intensity, the molecule will have enough time to align along the laser polarization direction and perhaps overshoot the laser polarization axis if the laser pulse duration is long enough. In our calculation, it has been found that the pulse length is longer than about 160 fs for N_2 and 2000 fs for I_2 at a laser intensity of 5×10^{14} W/cm^2, most molecules will overshoot the laser polarization axis and oscillate about the laser polarization axis with small amplitude. In these cases, the peak of angular distribution will be shifted away from the laser polarization direction. Figs. 7 and 8 present the dependence of FWHM of angular distribution on pulse length at a laser intensity of 5×10^{14} W/cm^2 for N_2 and I_2, respectively. For N_2, the FWHM of angular distribution becomes small when the laser pulse varies from 40 to 140 fs. But when the pulse length is increased further to 150 fs, the angular distribution is slightly broadened. This is because a little part of the molecules overshoots the laser polarization axis and cannot return to it before the dissociation is over. For I_2, when the width of the laser pulse is smaller than 300 fs, the torque makes the angular distribution narrow because molecular alignment happens near the peak of the laser pulse. As the pulse length is longer than 300 fs, the time of dynamic alignment plays a main role for the molecular alignment although the molecular alignment occurs far from the peak of laser pulse. This leads to narrowing of angular

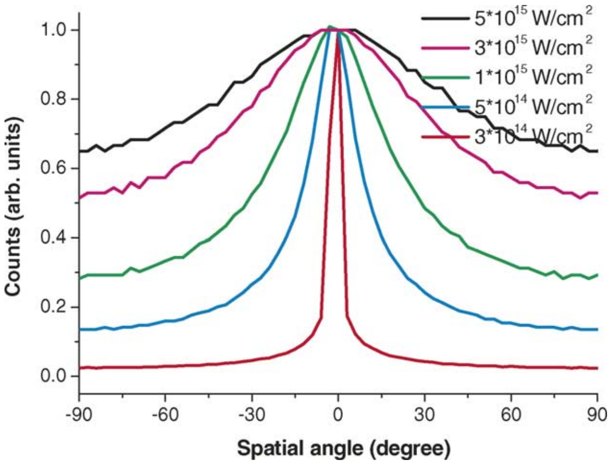

Fig. 5. Angular distributions of N_2 at pulse length of 100 fs when the laser intensity varies from 3×10^{14} to 5×10^{15} W/cm^2.

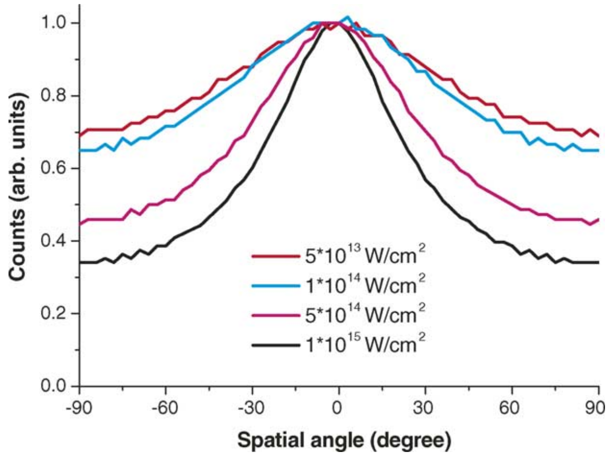

Fig. 6. Angular distributions of I_2 at pulse length of 100 fs when the laser intensity varies from 5×10^{13} to 1×10^{15} W/cm^2.

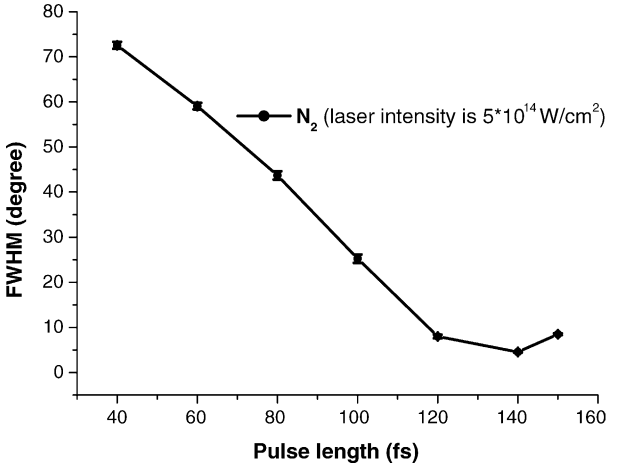

Fig. 7. Dependences of FWHM of angular distribution on the pulse length for laser intensity of 5 × 10^{14} W/cm^2 for N_2.

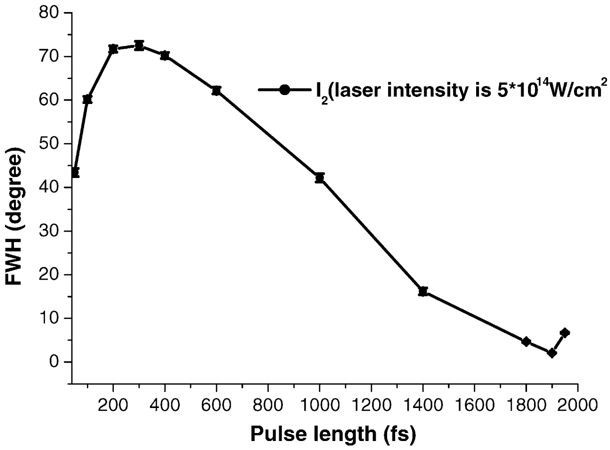

Fig. 8. Dependences of FWHM of angular distribution on the pulse length for laser intensity of 5 × 10^{14} W/cm^2 for I_2.

distributions. When the pulse length is increased further to 1950 fs, the similar situation with N_2 appears. The angular distribution begins to slowly broaden. Then the peak of angular distribution is shifted away from zero for a pulse length longer than 2000 fs.

4. Conclusions

The process of dynamic alignment for diatomic and linear triatomic molecules is clearly presented in this paper. The instants at which the molecule is ionized and molecular dissociation is over are important factors in analyzing the extent of dynamic alignment. The process of the dynamic alignment for the light molecules N_2, H_2 and CO_2 is significantly different from that for the heavy molecules Br_2, I_2 and CS_2. The dynamic alignment of molecules N_2, H_2 and CO_2 with high ionization potential, the low reduced mass and relatively, big value of *R* mainly occurs before the molecule ionizes and begins to dissociate. Conversely, molecules Br_2, I_2 and CS_2 will experience stronger alignment compared to molecules N_2, H_2 and CO_2 after they are ionized. The times of dynamic alignment before the molecule is ionized for N_2 play a main role for molecular alignment compared to the increasing torque when laser pulse length is constant. Conversely, the effect of increasing torque is important for the alignment of I_2 when the laser intensity increases.

At a given laser intensity, dependences of dynamic alignment for N_2 and I_2 on pulse duration are very different. It is worthy to note that we mainly investigate and discuss dynamic alignment mechanics of molecules in detail in our paper. In fact, dynamic and geometric alignments all contribute to the anisotropic angular distributions of ionic fragments when femtosecond laser pulses interact with molecules. It is very necessary to reassess which alignment plays an important role for the observed anisotropy of angular distributions of ionic fragments for a given condition from the theoretical view.

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