Dynamic Alignment of C_2H_4 Investigated by Using Two Linearly Polarized Femtosecond Laser Pulses

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We have studied multielectron ionization and Coulomb explosion of C_2H_4 irradiated by 110 fs, 800 nm laser pulses at an intensity of $\sim 10^{15}$ W/cm^2. Strong anisotropic angular distributions were observed for the atomic ions $C^{n+}(n = 1$–$3)$. Based on the results of two crossed linearly polarized laser pulses, we conclude that such anisotropic angular distributions result from dynamic alignment, in which the rising edge of the laser pulses aligns the neutral C_2H_4 molecules along the laser polarization direction. The angular distribution of the exploding fragments, therefore, reflects the degree of the alignment of molecules before ionization. Using the same femtosecond laser with intensity below the ionization threshold, the alignment of C_2H_4 molecules was also observed.   (J Am Soc Mass Spectrom 2006, 17, 1717–1724) © 2006 American Society for Mass Spectrometry

Understanding the behavior of molecules in strong laser fields is crucial to manipulate the internal and external degrees of freedom of molecules using femtosecond lasers [1]. When the laser intensity is below the ionization threshold of molecules, the laser–molecule interaction tends to align the molecules with the most polarizable axis along the laser polarization direction [2]. Because the laser pulse duration is less than the molecular rotational period, such transient alignment can be revived periodically after extinction of the exciting laser. When the laser intensity is beyond the ionization threshold, the external field asymmetrically distorts the potential and forms a potential barrier on one side of the molecules. The electron in the highest occupied molecular orbit tunnels through the barrier, which leads to the field ionization of the molecules [3]. Various models have been proposed to deal with the ionization of molecules in intense laser fields [3–11]. One of the most important predications of these models is that the strong field ionization rate for neutral molecules depends on the molecular orientation with respect to the laser polarization. The angle-dependent ionization rate is, therefore, a key parameter to test the models.

When the laser intensity reaches 10^{14} W/cm^2, the electric field of the laser is comparable to the Coulombic field felt by the valence electrons in molecules. Several electrons may be stripped away if the mole-

cules are subjected to such an intense laser field. Due to Coulomb repulsion, the highly charged molecular ions will explode into multicharged atomic ions with large kinetic energies [12–17]. The angular distributions of the exploding fragments have been observed to be highly anisotropic relative to the laser polarization, which is attributed to alignment before ionization or angle-dependent ionization. The former is also called dynamic alignment and the latter geometric alignment [18, 19]. In the case of dynamic alignment, the interaction between the laser field and the induced dipole moment sets up a torque to align the molecule along the laser polarization [18]. The aligned molecules are ionized and Coulomb-exploded at the peak of the laser intensity. Because the Coulomb explosion process is rapid, the initial velocities of the exploding fragments are parallel to the molecular axis. Thus, the angular distribution of the exploding fragments reflects the degree of alignment of molecules before ionization. In the case of geometric alignment, the ionization rate strongly depends on the angle between the molecular axis and the laser polarization direction [7, 9, 19]. The angular distribution of the exploding fragments, therefore, reflects the angle-dependent ionization probability of molecules. Recently, Alnaser et al. [20] studied the angular distributions of the exploding fragments of N_2 and O_2 irradiated by an 8 fs laser at an intensity of 2×10^{14} W/cm^2. They observed that atomic ions peaked at $\theta = 0°$ for N_2 and $\theta = 40°$ for O_2, where θ is the angle between the molecular axis and the laser polarization direction. Because dynamic alignment could be neglected under the experimental condi-

tions, the angular distribution of the exploding fragments represented the angle-dependent ionization probability. The results agreed with theoretical predictions of molecular ADK theory, the maximum ionization occurs at $\theta = 0°$ for N_2 and $\theta = 40°$ for O_2 [9].

However, dynamic alignment and geometric alignment coexist for the commonly used femtosecond laser with tens of fs duration. Due to the inapplicability of perturbation theory, theoretical studies on the alignment mechanism are difficult. Many methods were therefore proposed to experimentally evaluate the contribution of dynamic alignment and geometric alignment to the anisotropic angular distribution of the exploding fragments [21–25]. Among these methods, double pulses with crossed polarization can be used to directly assess the contribution of geometric and dynamic alignment [25]. If geometric alignment dominates, molecules with axes perpendicular to the polarization of the first laser pulse will survive and interact with the second laser pulse whose polarization is perpendicular to that of the first one. Therefore, the first laser pulse will not reduce the ion signal generated by the second laser pulse. On the other hand, if dynamic alignment dominates, the first laser pulse will align all molecules within the laser focus, irrespective of their initial orientation. Thus, all molecules will be ionized and depleted by the first laser pulse. Therefore, the ion signal generated by the second laser pulse will be reduced. The amount of depletion is a measure of the laser-induced alignment. Using this method, Posthumus et al. [26, 27] concluded that dynamic alignment dominated for H_2 and geometric alignment dominated for I_2 under a 50 fs laser field.

The alignment mechanism depends strongly on the laser pulse duration and laser intensity. For example, Rosca-Pruna et al. [28] studied the laser-induced alignment of I_2 for different pulse durations and revealed that geometric alignment dominated for 80 fs pulses and dynamic alignment began to play a significant role for pulse durations longer than 500 fs. However, for CO, dynamic alignment dominated for both 35 ps [25] and 110 fs [29] laser pulses at an intensity of 10^{15} W/cm^2.

Many groups have studied ionization of C_2H_4 irradiated by an intense femtosecond laser field. Talebpour et al. [30] observed strong molecular ions and abundant fragment ions when C_2H_4 was irradiated by an 800 nm, 200 fs pulse at an intensity of 10^{14} W/cm^2. The abundance of the fragment ions was attributed to the multiphoton ionization of inner-valence electrons followed by radiationless transitions to various dissociation channels of the molecular ions. Prall et al. [31] concluded that field ionization dominated when C_2H_4 was irradiated by a 780 nm, 125 fs pulse at an intensity of 6×10^{13} W/cm^2 and predicted that molecules were more easily ionized than the atom with the same ionization potential.

Muth-Bohm et al. [32] calculated the ionization yield of C_2H_4 using the intense-field many-body S-matrix theory and concluded that the ionization rate does not exceed that of the atom with comparable ionization potential. Kjeldsen et al. [33] studied the influence of molecular symmetry on strong field ionization using the molecular strong-field approximation. Their investigations show that the orientational dependence of ionization yields is primarily determined by the nodal surface structure of the molecular orbital. Coulomb explosion of C_2H_4 was also studied and the explosion pathways were identified [34, 35].

In this article, we focus on the origin of the highly anisotropic angular distribution of the exploding fragments of C_2H_4. Using two linearly polarized laser pulses with crossed polarization, we found that the first laser pulse completely depleted the ion signal generated by the second laser pulse. This result demonstrated clearly that dynamic alignment dominated, in which the neutral C_2H_4 was aligned along the laser polarization direction before ionization by the rising edge of the laser pulse. The angular distribution of exploding fragments reflected the alignment degree of molecules before ionization. Using the same laser with intensity below the ionization threshold, the field-induced alignment was also observed using the weak-field polarization technique.

Experimental

The experimental setup is shown in our previous paper [29, 36]. A Ti:sapphire femtosecond laser system with a regenerative chirp-pulsed amplifier (TSA-10, Spectra-Physics Inc., Mountain View, CA) delivered laser pulses with a central wavelength of 800 nm and a pulse duration of 110 fs at a repetition rate of 10 Hz. The maximum pulse energy was 6 mJ and the laser polarization was horizontal. A beamsplitter was used to divide the laser into two beams with equal intensity and a half-wave plate was inserted into one of the laser beams to rotate its linear polarization. The relative optical paths of the two laser beams were precisely adjusted using an optical translational stage controlled by a stepping motor. Then, the two laser beams were recombined by a beamsplitter. The spatial and temporal overlap of the two laser beams could be detected by observing the interference patterns after recombination. The recombined laser beams were focused into the chamber of a time-of-flight spectrometer by a lens with a focal length of 150 mm. A gaseous sample was introduced into the chamber via a pulsed valve (Park Inc., Cleveland, OH) with a 0.2 mm orifice. The ions produced in the laser beam were accelerated by a two-stage electric field and detected by a micro-channel plate (MCP). The signals were typically averaged over 256 laser pulses and recorded using a 1 GHz data acquisition card (DP110, Acqiris Digitizers, Geneva, Switzerland).
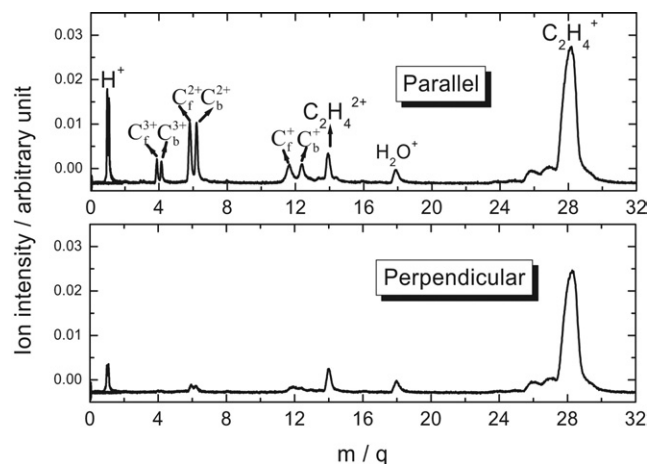
Figure 1. TOF mass spectra of C_2H_4 irradiated by 800 nm, 110 fs laser pulses at an intensity of 2×10^{15} W/cm² with laser polarization (upper graph) parallel and (lower graph) perpendicular to the TOF axis.

Results and Discussion

Single-Pulse Experiment

Mass spectra. Figure 1 shows typical TOF mass spectra of C_2H_4 irradiated by 110 fs, 800 nm at an intensity of 2×10^{15} W/cm². The laser polarization was parallel (upper graph) and perpendicular (lower graph) to the TOF axis. The mass spectra were consistent with previous reports [34, 35]. Singly and doubly charged parent ions had a similar shape and intensity for both parallel and perpendicular laser polarizations. However, the atomic ions $C^{n+}(n = 1–3)$ exhibited different behaviors. When the laser polarization was parallel to the TOF axis, each atomic ion was split into double peaks. However, when the laser polarization was perpendicular to the TOF axis, the C^{n+}ions almost completely disappeared. The splitting double peaks of the atomic ions indicated that Coulomb explosion occurred under the experimental conditions. The difference of mass spectra for parallel and perpendicular polarizations could be attributed to the large kinetic energy of the fragmental ions produced in the Coulomb explosion. When these ions were ejected in the direction parallel to the TOF axis, they would produce two peaks in the mass spectrum corresponding to the ions with initial velocities toward and away from the detector. When these ions were ejected in the direction perpendicular to the TOF axis, they missed the microchannel plate and could not be collected by the detector.

Angular distributions. The difference of mass spectra between the perpendicular and the parallel polarizations implied that the exploding fragments of C_2H_4 were highly anisotropic relative to the laser polarization direction. By rotating the half-wave plate, we changed the linear polarization direction of the laser beam relative to the TOF axis and measured the ion intensity at different laser polarization angles. Thus, we obtained the angular distribution of the ions. The results are shown in Figure 2. Singly and doubly charged parent ions were characteristically isotropic (not shown), while the atomic ions were highly anisotropic, with a maximum along the laser polarization and a minimum perpendicular to it. The full widths at half maxima (FWHM) were 52.8°, 63.6°, and 63.6° for C^+, C^{2+}, and C^{3+}, respectively. The anisotropic angular distributions were equal for C^{2+} and C^{3+} and became narrower for C^+. In addition, the C^+ contained a certain isotropic component. It is unclear why the angular distribution of C^+ was narrower than those of C^{2+} and C^{3+}.

Double-Pulse Experiment

The anisotropic angular distributions of the exploding fragments have been attributed to dynamic alignment or geometric alignment mechanisms. We utilized a double-pulse experiment with crossed polarization to assess the contribution of geometric and dynamic alignment. The key for double-pulse experiments is the control of the complete overlap of the spacing and the timing [25]. In the experiment, two pulses with the same polarization were used to
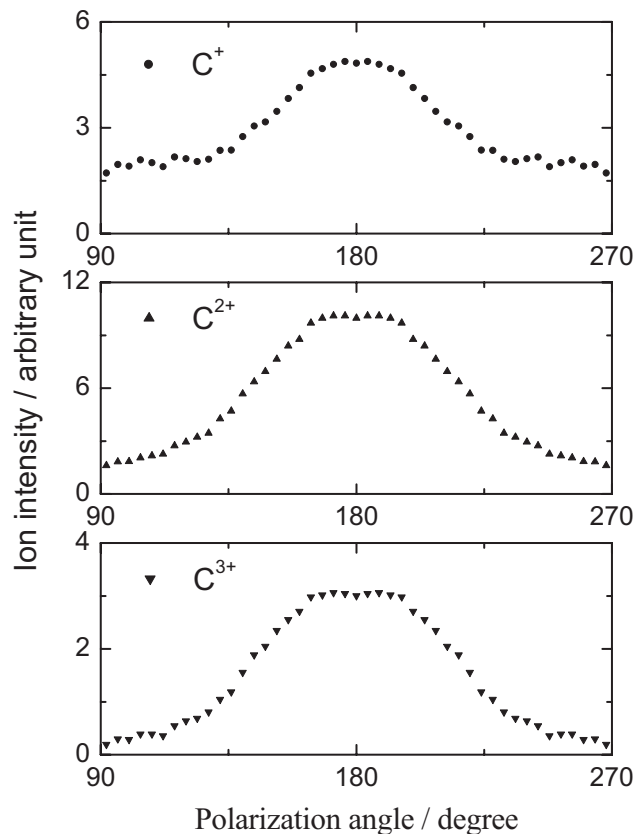

Figure 2. Angular distributions of the ions C^+, C^{2+}, and C^{3+} produced in the Coulomb explosion of C_2H_4 at a laser intensity of 2×10^{15} W/cm². These exploding ions were highly anisotropic with a maximum along the laser polarization and minimum normal to the laser polarization.
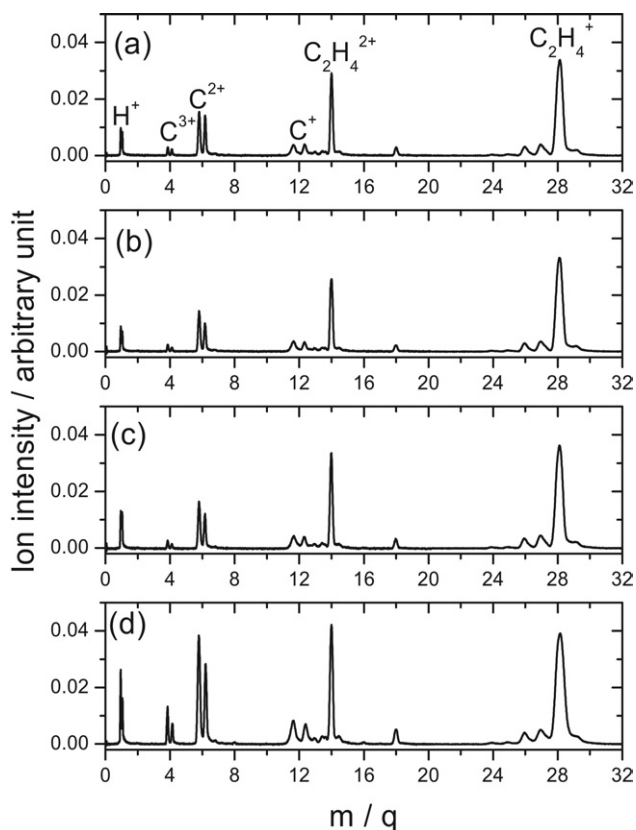
Figure 3. TOF mass spectra of C_2H_4 irradiated by (**a**) the first laser pulse alone, (**b**) the second laser pulse alone, (**c**) both of the laser pulses with a time delay of 1 ps, and (**d**) both of the laser pulses with timing overlap. The intensity was 6.2×10^{14} W/cm^2 for the first laser pulse and 5.0×10^{14} W/cm^2 for the second laser pulse. Both laser polarizations were parallel to the TOF axis.

test the spatial and timing overlap for the two pulses, and two pulses with crossed polarization were used to determine the alignment mechanism.

Two pulses with the same polarization. In the experiment, we first rotated the half wave plate in the first laser beam path and made the two laser beams have the same linear polarization. Then, we adjusted the time delay between the two laser beams. When they were over-lapped in timing, some interference stripes could be observed. Finally, we precisely adjusted the collinearity of the two laser beams. When they were completely spatially collinear, a flickering interference disk was observed. A lens was used to focus the combined laser beams into the TOF vacuum chamber and the laser focus was, therefore, completely overlapped both in spacing and timing. The overlap was tested by the experimental signals of the two pulses with the same polarization.

Figure 3 shows the mass spectra of the double-pulse laser with the polarizations parallel to the TOF axis. The laser intensity was 6.2×10^{14} W/cm^2 for the first laser pulse and 5.0×10^{14} W/cm^2 for the second laser pulse. Figure 3a and b show the single-pulse

mass spectra of C_2H_4 irradiated by the first laser pulse and the second laser pulse, respectively. Figure 3c and d show the double-pulse mass spectra of C_2H_4 irradiated by both the first laser pulse and the second laser pulse. However, the first laser pulse preceded the second laser pulse by 1 ps for Figure 3c and the first laser pulse overlapped the second laser pulse in timing for Figure 3d. By comparing the mass spectra in Figure 3a and c, we conclude that the two laser beams were indeed focused at the same point of the interaction volume. Otherwise, the signals produced by the two pulses should be the total of the signals produced by the two pulses separately, i.e., the signal in Figure 3c should be the sum of Figure 3a and b. The difference between Figure 3c and d resulted from different delay times between the first laser pulse and the second laser pulse. The maximum laser intensity would be greatly enhanced when the two laser pulses completely overlapped in timing. The ion signal was, therefore, greatly enhanced in Figure 3d, which was consistent with our experimental observation that the signals under zero delay were larger than those with
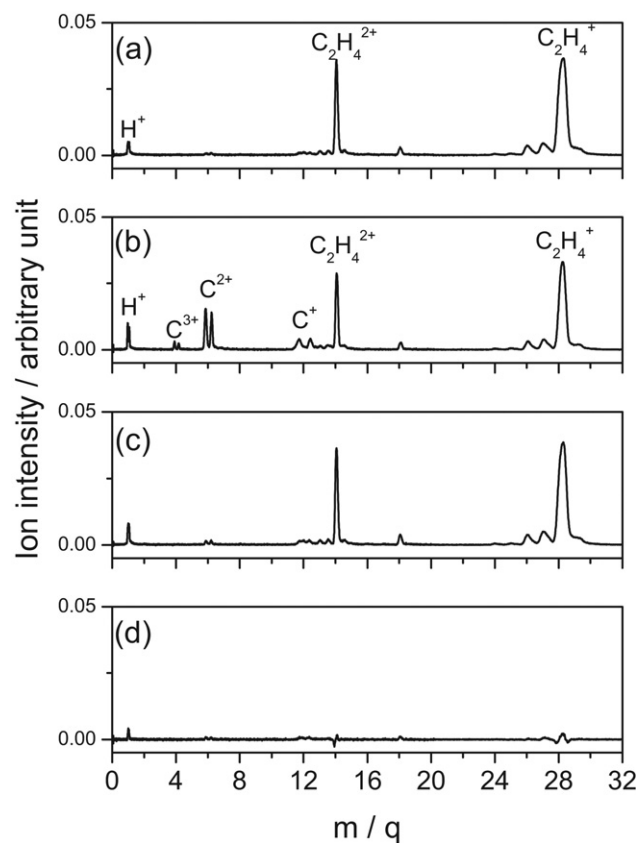

Figure 4. TOF mass spectra of C_2H_4 irradiated by (**a**) the per-pendicular polarized laser pulse alone, (**b**) the parallel polarized laser pulse alone, (**c**) both of the lasers with the perpendicular polarized laser pulse preceding the parallel polarized laser pulse by 1 ps, and (**d**) is the difference between (**a**) and (**c**), which represents the ion signal generated by the parallel polarized laser pulse in the double-pulse mass spectra. The intensity was 6.2×10^{14} W/cm^2 for the perpendicular polarized laser pulse and 5.0×10^{14} W/cm^2 for the parallel polarized laser pulse.

a 1-ps delay. Thus, we fully confirmed the spatial and timing overlap of the two laser pulses. By controlling the difference in the optical paths of the two laser beams, we could change the delay times between the two laser beams.

Two pulses with crossed polarization.　By rotating the half waveplate, we obtained two laser beams with crossed polarization. Figure 4 shows typical mass spectra during the double-pulse experiment with crossed polarizations. Figure 4a shows the single-pulse mass spectrum of C_2H_4 irradiated by the perpendicular polarized laser pulse alone. The laser intensity was 6.2×10^{14} W/cm². Figure 4b shows the single-pulse mass spectrum of C_2H_4 irradiated by the parallel polarized laser pulse alone. The laser intensity was 5.0×10^{14} W/cm². Figure 4c shows the double-pulse mass spectra of C_2H_4 irradiated by both the perpendicular polarized laser pulse and the parallel polarized laser pulse with a 1-ps delay time. Subtracting Figure 4a from c, we obtained Figure 4d, which represents the ion signal generated by the parallel polarized laser pulse in the presence of the perpendicular polarized laser pulse. The difference between Figure 4b and d reflects the extent of the depletion. It can be seen that the perpendicular polarized laser completely reduced the signal generated by the parallel polarized laser pulse.

The results of the above double-pulse experiment implied that the anisotropic angular distribution of the exploding fragmental ions resulted from dynamic alignment, in which the rising edge of the laser aligned the molecules along the laser polarization before ionization. Then multielectron ionization and Coulomb explosion occurred from the aligned neutral molecules at the peak of the laser intensity, which resulted in the anisotropic angular distribution of the exploding fragments. The angular distribution of the exploding fragments reflected the degree of alignment of neutral molecules before ionization.

Field-Induced Alignment with Laser Intensity Below the Ionization Threshold

Based on the above discussion, we draw the conclusion that we can align C_2H_4 molecules without ionizing them if the femtosecond laser intensity is below the ionization threshold. Recent theoretical calculations support this conclusion. Underwood et al. [37] calculated the interaction between C_2H_4 and strong femtosecond laser fields with intensity below the ionization threshold. Their calculation indicated that the laser-molecule interaction gave molecules a rapid "kick" to reposition the molecular axis in the laser polarization direction. Following the laser pulse, the molecules move through a moment of collective alignment. Then they continue to rotate and pass through the transient alignment periodically. Such laser–molecule interaction can, therefore, produce macroscopic ensembles of highly aligned molecules under field-free conditions.

Here, we experimentally studied the field-induced alignment of C_2H_4 by a strong femtosecond laser field with intensity below the ionization threshold using a typical weak-field polarization technique [38]. The 110 fs output pulse was split into two parts to provide a strong energy pump beam and a weak energy probe beam, both linearly polarized at 45° with respect to each other. The relative separation times between the two pulses were precisely adjusted using an optical translation stage controlled by a stepping motor. Both the pump beam and the probe beam were focused with a 300 mm focal length lens into a 200 mm long gas cell at a small angle. The gas cell was filled with C_2H_4 at room temperature and one atmospheric pressure. The field-free aligned molecules induced by the short pump laser will cause birefringence and depolarize the probe laser. After the cell, the depolarization of the probe laser, which represents the alignment degree, was analyzed with a polarizer set at 90° with respect to its initial polarization direction.

The degree of alignment of a rotational wave packet is given by the average of $\langle \cos^2\theta \rangle$, where θ is the angle between the molecular axis and the polarization vector of the laser field. $\langle \cos^2\theta \rangle = 1/3$ represents an isotropic angular distribution evenly distributed across all θ. If $\langle \cos^2\theta \rangle > 1/3$, the molecule is predominantly aligned along the laser polarization. If $\langle \cos^2\theta \rangle < 1/3$, the molecule is delocalized in the plane orthogonal to the laser polarization. The experimental measured alignment signal is proportional to $(\langle \cos^2\theta \rangle - 1/3)^2$ for commonly used homodyne detection techniques. In the present work, we used heterodyne detection technique, in which an in-phase local oscillator field was added by slightly rotating the polarizer in the probe beam path. Thus, the measured alignment signal is proportional to $\langle \cos^2\theta \rangle$ and the alignment revival structure can be obtained directly.

C_2H_4 is an asymmetric top molecule and the temporal structure of the alignment signal is more complicated than that of a linear molecule. Following spectroscopic convention, we define the molecular frame coordinates with principal axes (a, b, c) for C_2H_4 as $z = a$, $x = b$, and $y = c$. The a axis is along the C=C bond, the b axis is orthogonal to the C=C bond in the plane of the molecule and the c axis is perpendicular to the plane of the molecule. The rotational constants are A = 4.828 cm⁻¹, B = 1.001 cm⁻¹, and C = 0.828 cm⁻¹. The polarizabilities are $\alpha_{xx} = 3.682$ Å³, $\alpha_{yy} = 3.251$ Å³, and $\alpha_{zz} = 5.022$ Å³ [39]. Irradiated by a short linearly polarized laser, the most polarizable C=C bond axis of C_2H_4 is kicked towards alignment along the laser polarization direction. Compared with the linear molecule, the temporal structure of the alignment signal is more complicated for the asymmetric top molecule C_2H_4. There are two sets of alignment signals, which originate from J type transient and C type transient, respectively [39]. For a linear molecule, the J transient is the only type observed in pure rotational spectroscopy and characterized by a pseudoperiod $T_J = 1/2(B + C) =$
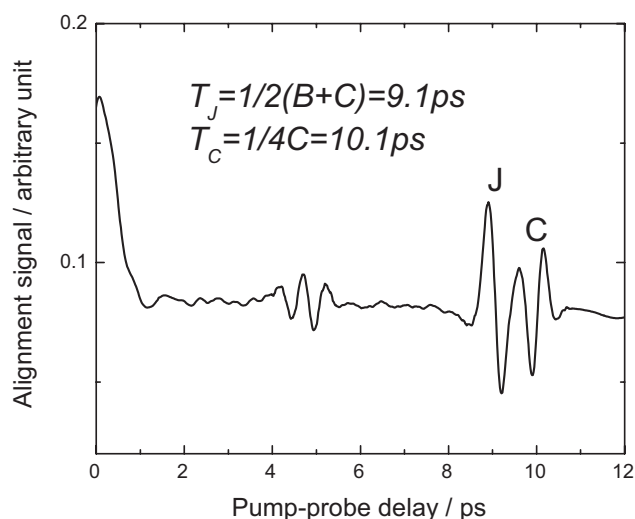
Figure 5. Field-free alignment of C_2H_4 induced by 110 fs, 800 nm at an intensity of 6×10^{13} W/cm^2.

9.1 ps. The C type transient results from the asymmetry of the molecules and describes a rotation of the molecule about the molecular c axis with $T_c = 1/4$ C = 10.1 ps.

Figure 5 shows the measured alignment signal versus the pump-probe delay. The intensity of the aligned laser was 6.0×10^{13} W/cm^2. We did not observe ion signal in our TOF mass spectrometer below this laser intensity. It can be seen that the aforementioned two types of alignment signals, J type transient and C type transient, were observed. Both transients result in alignment and planar delocalization of the C=C bond axis of C_2H_4. The characteristic pseudoperiods are indeed 9.1 ps and 10.1 ps for J type and C type transient, respectively. The alignment signal in Figure 5 further indicates that the C=C bond axis is aligned before delocalization for the J type transient, and aligned after delocalization for the C type transient. These observations agree with the theoretical calculation carried out by Rouzée et al. [39].

These observations demonstrated that a 110 fs laser pulse can indeed align C_2H_4 molecules; moreover, the C=C bond axis is aligned along the laser polarization. The above results support the explanation about the anisotropic angular distribution of C^{n+} ($n = 1$–3) produced by Coulomb explosion of C_2H_4 in an intense femtosecond laser field. The interaction between the induced dipole moment and the rising edge of the laser sets up a torque to align the C=C bond axis along the laser polarization. The aligned molecules are ionized and Coulomb exploded at the peak of the laser intensity. Because the Coulomb explosion process is rapid, the initial velocities of the exploding fragments C^{n+} ($n = 1$–3) are parallel to the C=C bond axis. The alignment of the C=C bond axis along the laser polarization direction before ionization resulted in the anisotropic angular distribution of the C^{n+} ($n = 1$–3) relative to the laser polarization. In addition, the transient alignment can be revived

after the exciting laser is extinguished. Aligned molecules can thus be achieved under field-free conditions. The possibility of preserving the alignment of a large number of molecules under field-free conditions demonstrates that a strong femtosecond laser is a potentially powerful tool to manipulate molecules and provides a novel practical approach to prepare aligned molecules in the laboratory.

Discussion

Strong field ionization is an important topic because it is related to high harmonic, X-ray, and attosecond pulse generation processes [40, 41]. Various models have been proposed to deal with the ionization of molecules in intense laser fields. The angle-dependent ionization rate is an important parameter to test these models. However, for our commonly used laser with tens of fs duration, the rising edge of the laser pulse aligns the molecules along the laser polarization before ionization. The coexistence of alignment and ionization makes it difficult to experimentally measure the angle-dependent ionization rates of molecule. Recently, Alnaser et al. [20] studied the angular distributions of the exploding fragments of N_2 and O_2 irradiated by an 8 fs laser at an intensity of 2×10^{14} W/cm^2. They observed that atomic ions peaked at $\theta = 0°$ for N_2 and $\theta = 40°$ for O_2. Because the laser pulse is so short, alignment before ionization can be neglected. The angular distribution of the exploding fragments represents the angle-dependent ionization probability of molecules. The results agreed with theoretical prediction of molecular ADK theory [9]; the maximum ionization occurs at $\theta = 0°$ for N_2 and $\theta = 40°$ for O_2. This is the first time that the angle-dependent ionization probabilities were experimentally measured for molecules in an intense femtosecond laser field even though some assumptions were assumed [20].

Litvinyuk et al. [42] provided another experimental approach to measure the angle-dependent ionization probability of N_2 molecules by intense femtosecond laser fields. They utilized a 40 fs linearly polarized laser pulse to prepare a macroscopic alignment of N_2 molecules under field-free conditions and another 40 fs linearly polarized laser pulse to ionize the aligned N_2 molecules. They discovered that at an ionizing laser intensity of 2×10^{14} W/cm^2, N_2 molecules aligned parallel to the laser field are ~four times more likely to ionize than molecules oriented perpendicular to it. However, they did not obtain the angle-dependent ionization probability for O_2 using a similar technique. The possible reason is that the rising edge of the ionization laser destroys the alignment degree of O_2 formed by the aligned laser. The ionization laser will realign the aligned molecules before ionization.

Our experiment indicated that the rising edge of the 110 fs laser would align C_2H_4 before ionization. Therefore, for our commonly used laser with tens of fs duration, the alignment before ionization should be taken into account when quantitatively measuring the

angle-dependent ionization probability of molecules in intense laser fields. Here, we propose another approach to measure the angle-dependent ionization of molecules. We can use a femtosecond laser with tens of fs duration to produce field-free aligned molecules. The aligned laser intensity is controlled to be below the ionization threshold of molecules. Then another short-pulse laser (<8 fs) is used to ionize the field-free aligned molecules. Because such a short laser pulse does not realign the aligned molecules before ionization, the angle-dependent ionization probability for C_2H_4 can be precisely measured. This type of measurement is currently being carried out in our laboratory.

Conclusions

Using two linearly polarized laser pulses with crossed polarization, we have studied the alignment mechanism of C_2H_4 irradiated by a 110 fs, 800 nm laser at an intensity of $\sim 10^{15}$ W/cm^2. The fact that the first laser pulse completely depleted the ion signal generated by the second laser pulse demonstrated clearly that dynamic alignment dominated under the experimental conditions. The neutral C_2H_4 was aligned along the laser polarization direction before ionization by the rising edge of the laser pulse. At the peak of the laser intensity, multielectron ionization and Coulomb explosion occurred from these aligned neutral molecules, which resulted in anisotropic angular distribution of the exploding fragments. This conclusion was supported by the field-induced alignment of C_2H_4 induced by the femtosecond laser with laser intensity below the ionization threshold. Coexistence of alignment and ionization suggests the degree of alignment should be taken into account to qualitatively measure the angle-dependent ionization probability of C_2H_4 in intense femtosecond lasers with tens of fs duration.

Acknowledgments

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