Field-Induced Alignment of Oxygen and Nitrogen by Intense Femtosecond Laser Pulses

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Field-induced alignment of O_2 and N_2 was experimentally studied with laser intensities varying from 10^{13} to 10^{15} W/cm^2. When the laser intensity was below the ionization threshold for these molecules, the interaction between the induced dipole moment of molecules and the laser electric field aligned the molecules along the laser polarization direction. After extinction of the exciting laser, the transient alignment revived periodically. Thus macroscopic ensembles of highly aligned O_2 and N_2 molecules were obtained under field-free conditions. When the laser intensity exceeded the ionization threshold for these molecules, multielectron ionization and Coulomb explosion occurred. Using two linearly polarized laser pulses with crossed polarization, we demonstrated that the rising edge of the laser pulse aligned the molecules along the laser polarization direction prior to ionization, which resulted in strong anisotropic angular distributions of exploding fragments. These results suggest that the degree of alignment should be taken into account when qualitatively comparing the ion yield of these molecules with their companion atoms.

Introduction

The femtosecond laser is becoming a powerful tool to manipulate the behaviors of molecules.[1] When the laser intensity is below the ionization threshold, the interaction between molecules and strong nonresonant linearly polarized laser fields 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, the laser−molecule interaction gives the molecules a rapid "kick" to move the molecular axis toward 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 produce macroscopic ensembles of highly aligned molecules under field-free conditions and will not interfere with subsequent applications. A variety of new and exciting applications of field-free aligned molecules are currently emerging.[3−5]

When the laser intensity is further increased and passes the ionization threshold for these molecules, 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.[6,7] Various models have been proposed to deal with the ionization of molecules in intense laser fields.[8−11] One of the most important predications for 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. However, the rising edge of the laser pulse may align the molecules prior to ionization depending on the laser properties. Thus, coexistence between alignment and ionization makes it very difficult to

precisely measure the angle-dependent strong field ionization probabilities of molecules.

When the laser intensity exceeds 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 molecules are subjected to such an intense laser field and highly charged molecular ions are therefore formed. Due to Coulomb repulsion, the highly charged molecular ions explode into multicharged atomic ions with large kinetic energies. The angular distributions of the exploding fragments have been observed to be highly anisotropic, which is attributed to alignment prior to ionization or angle-dependent ionization. The former is also called dynamic alignment and the latter geometric alignment.[12,13] In the case of dynamic alignment, the induced dipole moment interacts with the rising edge of the laser pulse and sets up a torque to align the molecule 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 are parallel to the molecular axis. Therefore, the angular distribution of the exploding fragments represents the alignment degree of molecules prior to ionization. In the case of geometric alignment, the ionization rate depends on the angle between the laser polarization direction and the molecular axis. Therefore, the angular distribution of the exploding fragments represents the angle-dependent ionization probabilities of molecules.

Different methods have been used to determine the origin of the anisotropic angular distribution of the exploding fragments, that is, the alignment mechanism.[14−18] Among these methods, double pulses with crossed polarization can be used to directly assess the contribution of geometric and dynamic alignment.[18] If geometric alignment dominates, molecules with their 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

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laser pulse will not reduce the 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.[19] concluded that dynamic alignment dominated for H_2 and geometric alignment dominated for I_2 under a 50-fs laser field.

O_2 and N_2 are the major ingredients of the atmosphere, and their interaction with intense femtosecond laser fields has been extensively studied. Dooley et al.[20] used a 45-fs linearly polarized laser to produce macroscopic field-free aligned molecules at well-defined time delays and observed the time evolution of the alignment degree using an intense circularly polarized laser pulse to explode the aligned molecules. Thus they directly measured the alignment degree of molecules at various times after the extinction of the aligned laser. They also demonstrated that the degree of alignment could be improved by two laser pulses at separate times.[21] When the laser intensity exceeds the ionization threshold, these molecules will be ionized. The most remarkable finding is that suppressed ionization occurs for O_2 by an order of magnitude relative to its companion atom with nearly the same ionization potential.[22] Several mechanisms, such as electron rescattering,[23] charge-screening correction,[8] electron interference,[9] and destructive quantum interference,[24] were proposed to explain the suppressed ionization of O_2. According to the molecular ADK theory,[10,25] the strong field ionization rate depends on θ, which is defined as the angle between the molecular axis and the laser polarization direction. The maximum ionization occurs at $\theta = 0°$ for N_2 and $\theta = 40°$ for O_2. Recently, Alnaser and Voss et al.[26,27] 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, in which dynamic alignment could be neglected. Thus the angular distribution of the exploding fragments represents the angle-dependent ionization. The results agreed with theoretical prediction by the molecular ADK theory; the exploding fragments peaked at $\theta = 0°$ for N_2 and $\theta = 40°$ for O_2. It has been widely recognized that the alignment mechanism was determined by the laser properties. Voss et al.[27] demonstrated that geometric alignment dominated for both O_2 and N_2 by an 8-fs laser at an intensity of 2×10^{14} W/cm^2. When the laser pulse duration was lengthened or the laser intensity was increased, dynamic alignment took effect. Miyazaki et al.[28] concluded that no dynamic alignment happened for N_2 under 40-fs pulses at an intensity less than 10^{15} W/cm^2. However, Beylerian and Cornaggia[29] concluded that both dynamic and geometric alignment occurred for N_2 by a 40-fs laser pulse at an intensity of 10^{14} W/cm^2. Posthumus et al.[19] concluded that dynamic alignment dominated for N_2 in a 110-fs intense laser field.

In this article, we have systemically studied field-induced alignment of O_2 and N_2 molecules by 800-nm, 110-fs laser pulses with intensities varying from 10^{13} to 10^{15} W/cm^2. When the laser intensity was below the ionization threshold of the molecules, we demonstrated that field-free alignment could be achieved using the weak field polarization technique. When the laser intensity was beyond the ionization threshold of the molecules, we demonstrated that the rising edge of the laser pulse aligned the molecules along the laser polarization direction prior to ionization using two linearly polarized laser pulses with crossed polarization. The results indicated that alignment and ionization coexisted for laser pulses with tens of femtoseconds

duration. Therefore, the degree of alignment should be taken into account when qualitatively comparing the ion yield of these molecules with their companion atoms in commonly used tens of femtoseconds laser fields.

Experimental Section

The laser system consisted of a chirped-pulse-amplified Ti: sapphire system operating at 800 nm and at a repetition rate of 10 Hz. To study the interaction between molecules and the laser field with intensity below the ionization threshold, a typical weak field polarization technique was used.[30] 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 time separation between the two pulses could be 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 different gases at room temperature and at one atmospheric pressure. The field-free aligned molecules induced by the short pump laser caused birefringence and depolarized 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. To eliminate the laser fluctuation, a reference laser was introduced. The alignment signals and the reference laser signals were detected by two photoelectric cells and transferred to a computer via a four-channel A/D converter for analysis.

To study the interaction between molecules and laser fields with intensity above the ionization threshold, a typical time-of-flight mass spectrometer was used.[31] A beam splitter 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. Then the two laser beams were recombined by a beam splitter and focused into the chamber of a time-of-flight mass 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., USA) with a 0.2-mm orifice. The chamber pressure was maintained below 4×10^{-5} Pa in order to avoid space charge effects. The ions produced in the laser beam were accelerated by a two-stage electric field and detected by a microchannel plate (MCP). The signals were typically averaged over 256 laser pulses and recorded using a 1-GHz data acquisition card (DP110, Acqiris Digitizers, Switzerland).

Results

1. **Field-Free Alignment of N_2 and O_2 Molecules with Laser Intensity below the Ionization Threshold.**

Field-free alignment of molecules by strong femtosecond laser pulses has been the topic of rapidly growing experimental and theoretical interest in recent years. Now, researchers have developed two typical methods to measure experimentally the alignment degree of molecules. The first one is realized by breaking the aligned molecule through multielectron dissociative ionization or dissociation followed by ionization of the fragments.[21] The alignment degree $\langle \cos^2 \theta \rangle$ is thus deduced from the angular distribution of the ionized fragments. The disadvantage of this method is that the probe laser is so strong that it destroys the aligned molecules. The second method is the weak field polarization spectroscopy technique based on the birefringence caused by aligned molecules.[32] The advantage of this method is that the probe laser is so weak that it neither affects the alignment degree nor destroys the aligned molecules.
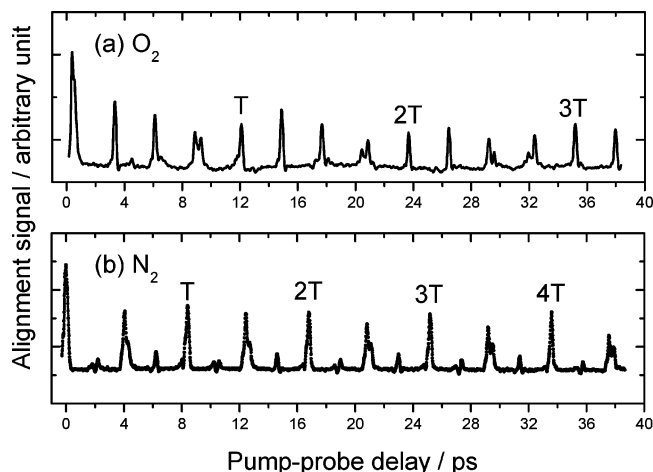
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Figure 1. Field-free alignment signals versus pump−probe delay for (a) O_2 and (b) N_2 at room-temperature irradiated by 800-nm, 110-fs laser pulses at an intensity of 6×10^{13} W/cm^2.

In the present measurement, we used the weak field polarization spectroscopy technique to measure the alignment signal of N_2 and O_2 molecules by strong femtosecond laser pulses.

Figure 1 shows the alignment signal for diatomic molecules (a) O_2 and (b) N_2 at room temperature irradiated by an 800-nm, 110-fs laser at an intensity of 6×10^{13} W/cm^2. The classical rotational period T of molecules is determined by the equation $T = 1/(2 B_0 c)$, where B_0 is the rotational constant in the ground vibronic state and c is the speed of the light. For N_2 and O_2, B_0 is 2.010 and 1.4456 cm^{-1}, respectively.[33] The corresponding rotational period T is therefore 8.3 ps for N_2 and 11.6 ps for O_2. Figure 1 clearly shows that the alignment signal was repeated every molecular rotational period. There were also moments of strong alignment that occurred at smaller intervals. The difference at quarter full revival for O_2 and N_2 can be explained by the different nuclear spin weights of the even and odd J states in the initial distribution. At $T/4$, $3T/4$, $5T/4$ ... revivals, the odd J wave packet has maxima (minima) whereas the even J wave packet has minima (maxima). For homonuclear diatomic molecules, the nuclear spin statistics control the relative weights between even and odd J states. In the case of O_2, only odd J states are populated. Since only a single localized wave packet existed, strong net alignment and antialignment were observed near the time of a quarter revival. In the case of N_2, the relative weights of the even and odd J were 2:1. As a result, the temporary localization of the even J wave packet at $T/4$ was partially canceled by its odd counterpart. Thus, small net N_2 alignment and antialignment were observed near $t = nT/4$, where n is an odd number. This work indicated that a femtosecond laser field can control the alignment of molecules and that macroscopic ensembles of highly field-free aligned N_2 and O_2 molecules can be achieved for practical application in the laboratory.

2. Field-Induced Alignment of N_2 and O_2 Molecules with Laser Intensity Exceeding the Ionization Threshold.

Chu et al.[11] calculated the ionization probabilities of N_2 and O_2 using an all-electron ab-initio theory. They reported that the ionization probability was 0.0285 for N_2 and 0.0601 for O_2 at an intensity of 1×10^{14} W/cm^2. Many researchers reported that multielectron ionization and Coulomb explosion occurred when the laser intensity exceeded 10^{14} W/cm^2.[34−36] The angular distributions of the exploding fragments were observed to be highly anisotropic, which was attributed to dynamic alignment or geometric alignment. If dynamic alignment dominates, the angular distribution of the exploding fragments represents the

alignment degree of molecules prior to ionization. On the contrary, if geometric alignment dominates, the angular distribution of the exploding fragments represents the angle-dependent ionization probabilities of molecules. Recently, Alnaser and Voss et al.[26,27] 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 the atomic ions peaked at $\theta = 0°$ for N_2 and $\theta = 40°$ for O_2. They concluded that dynamic alignment could be neglected under their experimental condition and the angular distribution of the exploding fragments resulted from the angle-dependent ionization of N_2 and O_2 by an 8-fs laser field. However, we demonstrated that a 110-fs laser could align N_2 and O_2 molecules along the laser polarization direction with intensity below the ionization threshold of these molecules. In addition, we observed that the atomic ions peaked at $\theta = 0°$ for both N_2 and O_2 when these molecules were irradiated by our intense 110-fs laser field. Therefore, we doubted that our measured anisotropic angular distribution of the exploding fragments resulted from alignment prior to ionization, that is, dynamic alignment. To investigate such a possibility, we used a double-pulse method to evaluate the contribution of dynamic alignment.

Two laser pulses with crossed polarizations were fired with separate times. Instead of observing the depletion of the ion signal generated by the second laser pulse, we observed the difference between the double-pulse mass spectra with the parallel laser preceding the perpendicular laser and that with the parallel laser following the perpendicular laser. When dynamic alignment dominates, all molecules at the laser focus are aligned and Coulomb-exploded by the first laser pulse, causing the atomic ion signal generated by the second laser pulse to be greatly reduced. Therefore, the double-pulse mass spectrum depends on the relative timing of the parallel and perpendicular polarized laser pulses. If the parallel polarized laser precedes the perpendicular polarized laser pulse, then the double-pulse mass spectrum will be similar to the single-pulse mass spectrum for a parallel polarized laser pulse alone. However, if the perpendicular polarized laser precedes the parallel polarized laser pulse, then the double-pulse mass spectrum will be similar to the single-pulse mass spectrum for a perpendicular polarized laser pulse alone. When geometric alignment dominates, the first laser pulse only Coulomb-explodes the molecules with axes parallel to the laser polarization, and the remaining molecules with axes perpendicular to the first laser polarization will be ionized and Coulomb-exploded by the second laser pulse. Therefore, there is no apparent difference between the double-pulse mass spectrum with the parallel laser preceding the perpendicular laser and that with the parallel laser following the perpendicular laser.

Figure 2 shows typical results of the double-pulse experiment. Figure 2a,b is the single-pulse mass spectra of O_2 irradiated by a single 110-fs, 800-nm laser pulse at an intensity of 6×10^{14} W/cm^2. The laser polarization was (a) perpendicular and (b) parallel to the time-of-flight axis. Strong singly and doubly charged parent ions had similar shapes and intensity for both parallel and perpendicular laser polarizations. However, the atomic ions O^+ and O^{2+} 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, these atomic ions 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 can be attributed to the large
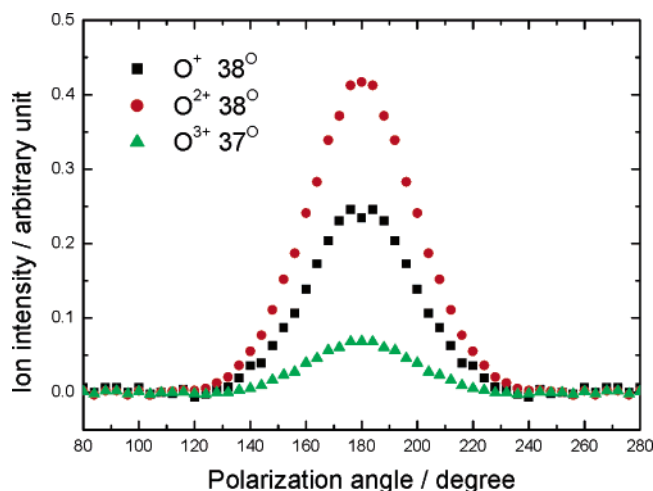
Figure 2. TOF mass spectra of O_2 irradiated by 800-nm, 110-fs laser pulses (a) perpendicular polarized laser alone; (b) parallel polarized laser alone; (c) double pulse with perpendicular polarized laser preceding parallel polarized laser by 500 fs (black line) and double pulse with parallel polarized laser preceding perpendicular polarized laser by 500 fs (red line). The laser intensities were 6×10^{14} W/cm^2 for both perpendicular and parallel laser pulses.

Figure 3. TOF mass spectra of N_2 irradiated by 800-nm, 110-fs laser pulses (a) perpendicular polarized laser alone; (b) parallel polarized laser alone; (c) double pulse with perpendicular polarized laser preceding parallel polarized laser by 500 fs (black line) and double pulse with parallel polarized laser preceding perpendicular polarized laser by 500 fs (red line). The laser intensities were 6×10^{14} W/cm^2 for both perpendicular and parallel laser pulses.

kinetic energy of the fragmental ions produced in the Coulomb explosion. When these ions are ejected in the direction perpendicular to the TOF axis, they will miss the microchannel plate and cannot be collected by the detector. However, when these ions are ejected in the direction parallel to the TOF axis, they will produce two peaks in the mass spectrum. The early-arriving peak corresponds to the forward ions with initial velocities toward the detector. The late one corresponds to the backward ions with initial velocities moving away from the detector and is reversed by the extraction field. The difference between Figure 2a and 2b implied that the distributions of atomic ions are highly anisotropic, having a maximum along the laser polarization and a minimum perpendicular to it. The origin for such anisotropic angular distributions was evaluated by the double-pulse method.

Figure 2c shows the characteristic double-pulse mass spectra for irradiation by both the parallel and the perpendicular polarized laser pulses. The laser intensity was 6×10^{14} W/cm^2 for both the parallel and perpendicular polarized laser. The black line represents the double-pulse mass spectrum with the perpendicular laser preceding the parallel polarized laser by 500 fs, and the red one represents the double-pulse mass spectrum with the parallel laser preceding the perpendicular laser by 500 fs. It can be seen that the double-pulse mass spectrum depends on the timing between the parallel laser and the perpendicular laser. When the parallel laser preceded the perpendicular laser, the double-pulse mass spectrum was similar to the single-pulse mass spectrum for irradiation by the parallel laser alone. When the perpendicular laser preceded the parallel laser, the double-pulse mass spectrum was similar to the single-pulse mass spectrum for irradiation by the perpendicular laser alone. The results demonstrated that the first laser pulse had ionized and Coulomb-exploded almost all molecules in the laser focus and indicated that dynamic alignment was dominant. Our conclusions are also consistent with reports of Voss et al.[27] They observed that the atomic ions peaked at $\theta = 40°$ when geometric alignment dominated for O_2 by an 8-fs laser at an intensity of 2×10^{14} W/cm^2.

Similar results were also observed for N_2, which are shown in Figure 3. These results demonstrated that dynamic alignment dominated for these molecules under our experimental condi-

tions, in which the rising edge of the laser pulse aligned the molecules along the laser polarization direction prior to ionization. Therefore, the angular distribution represented the alignment degree of molecules prior to ionization.

Discussion

Voss et al.[27] observed that the atomic ions peaked at $\theta = 0°$ for N_2 and $\theta = 40°$ for O_2 by an 8-fs laser at an intensity of 2×10^{14} W/cm^2. They concluded that geometric alignment dominated and the angular distribution of the exploding fragments therefore represented the angle-dependent ionization of molecules. The results agreed with theoretical prediction by the molecular ADK theory, the maximum ionization occurred at $\theta = 0°$ for N_2 and $\theta = 40°$ for O_2.[25] We observed that the atomic ions peaked at $\theta = 0°$ for both N_2 and O_2; the results implied that the geometric alignment was not significant under our experimental conditions. Using a double-pulse method with crossed polarization, we confirmed that dynamic alignment dominated, in which the rising edge of the laser aligned the molecules prior to ionization. Coexistence of alignment and ionization require that the degree of alignment should be taken into account when the ion yield of these molecules is qualitatively compared with their companion atoms. Because maximum ionization occurs at $\theta = 0°$ for N_2 and $\theta = 40°$ for O_2, the ion yield will be greater for N_2 and smaller for O_2 than the counterparts at random orientation when the molecules are aligned along the laser polarization prior to ionization. The current double-pulse experiment demonstrated that, for our commonly used laser with tens of femtoseconds duration, the rising edge of the laser pulse aligns the molecules along the laser polarization direction. Then the aligned molecules are ionized at the peak of the laser intensity. While the ionization probability is minimum for O_2 molecules with the axes along the laser polarization, the ion yields are therefore smaller than those at random orientation. Such alignment prior to ionization can, at least, partially explain the suppression of O_2 relative to the companion Xe atom with nearly the same ionization potential. Further theoretical calculation indicates that the longer the laser pulse width, the higher the alignment degree prior to ionization.[37] It is therefore expected that the suppression becomes higher for longer laser pulses. Such expectation was

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Figure 4. fwhms of the angular distributions for O^+, O^{2+}, and O^{3+} produced by O_2 with an 800-nm, 110-fs laser at an intensity of 3×10^{15} W/cm^2.
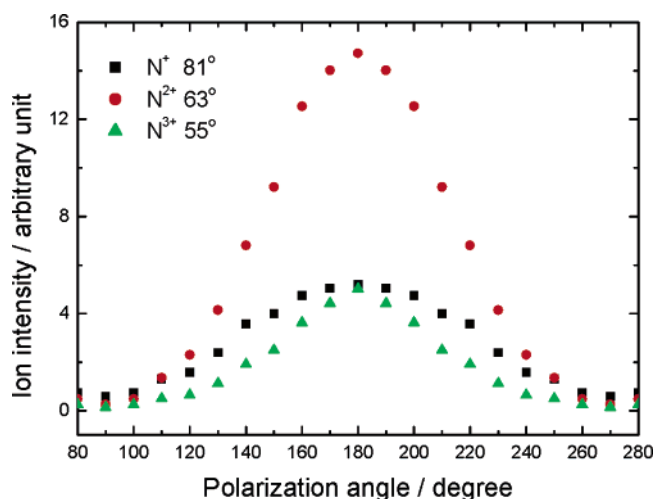

Figure 5. fwhms of the angular distributions for N^+, N^{2+}, and N^{3+} produced by N_2 with an 800-nm, 110-fs laser at an intensity of 3×10^{15} W/cm^2.

also verified by the experimental results that the ion yield ratio for pairs O_2/Xe became smaller when the laser pulse width became longer.[10]

Under our experimental conditions, the anisotropic angular distribution of the exploding fragments resulted from the dynamic alignment. Thus, the angular distribution of the exploding fragments represented the alignment degree of molecules prior to ionization by the rising edge of the laser pulse. We also measured the angular distribution of the exploding fragments. By rotating the half-wave plate, the polarization direction of the linear laser beam was changed relative to the TOF axis. Subsequently, the angular distribution of ions was obtained through measuring the ion intensity at different laser polarization angles. The full widths at half-maximum (fwhm) of ions represent the alignment degree of molecules prior to ionization. The smaller the fwhm is, the higher the degree of the alignment. The results of O^{n+} ($n = 1$, 2, 3) and N^{n+} ($n = 1$, 2, 3) are shown in Figure 4 and Figure 5. The atomic ions had a maximum when the laser was parallel polarized and a minimum with a perpendicular polarized laser. The fwhms were 38°, 38°, and 37° for O^+, O^{2+}, and O^{3+}, and 81°, 63°, and 55° for N^+, N^{2+}, and N^{3+}, respectively. Because the polarizability anisotropy (representing the difference between the polarizabilities in the directions parallel and perpendicular

to the molecular axis) of O_2 is larger than that of N_2, it is expected that the alignment degree is higher for O_2 than that for N_2 under the same laser conditions, which agreed with our measurement that fwhms are smaller for O_2 relative to their counterparts for N_2.

It should be noted that the angular distributions were almost equal for O^+, O^{2+}, and O^{3+} and became narrower for N^+, N^{2+}, and N^{3+}. Our above-mentioned double-pulse experiment clearly demonstrated that dynamic alignment dominated for both N_2 and O_2, so why did the angular widths display different behaviors for exploding fragments of N_2 and O_2? Tong et al.[37] recently pointed out that there is an important post ionization alignment effect of molecular ions in addition to the dynamic alignment of neutral molecules before ionization. Such post ionization alignment results from the breakup of a rotating linear rotor. Alignment is generally higher for higher charged molecular ions. The angular width therefore becomes narrower for higher-charged atomic ions if post ionization alignment plays an important role at the falling edge of the pulse. In the present study, the same width of the angular distributions for all O^{n+} ($n = 1, 2, 3$) implied that alignment prior to ionization dominates and post ionization alignment can be neglected for O_2. However, for N_2, the narrower angular widths for N^+, N^{2+}, and N^{3+} indicated that post ionization alignment has an important effect at the falling edge of the pulse. In other words, comparing with alignment before ionization, post ionization alignment plays an important role for N_2 even though it can be neglected for O_2.

Conclusions

In summary, we have experimentally studied field-induced alignment of O_2 and N_2 molecules in a femtosecond laser field with intensity varying from 10^{13} to 10^{15} W/cm^2. When the laser intensity is below the ionization threshold, the interaction between the induced dipole moment and the laser field aligns the molecular axis along the laser polarization. We demonstrated that such alignment could be periodically revived under field-free conditions using weak field polarization spectroscopy. When the laser intensity exceeded the ionization threshold, multielectron ionization and Coulomb explosion occurred. Using two linearly polarized laser pulses with crossed polarization, we demonstrated that the rising edge of the laser pulse aligned the molecules along the laser polarization direction prior to ionization and resulted in the anisotropic angular distribution of the exploding fragments. Coexistence of alignment and ionization indicated that the degree of alignment should be taken into account when the ion yield of these molecules is qualitatively compared with their companion atoms in tens of femtoseconds laser pulse fields. The fwhms, which represented the alignment degree of molecules prior to ionization, were also measured for the exploding fragments. They are similar for O^{n+} ($n = 1, 2, 3$) and become narrower for N^{n+} ($n = 1, 2, 3$). Such differences were interpreted by the relative importance between alignment prior to ionization and post ionization alignment.

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