

ARTICLE

Laser-induced Alignment and Coulomb Explosion of CO₂[†]

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Dynamic processes of CO₂ are experimentally studied in intense femtosecond laser fields with laser intensity varying from 1×10^{13} W/cm² to 6×10^{14} W/cm². When the laser intensity is below the ionization threshold, a coherent rotational wave-packet is formed for CO₂ at room temperature through nonadiabatic rotational excitation. The evolution of the wave-packet leads to transient alignment. The field-free alignment revives periodically after the laser pulse is over. The revival structure can be modified by a second laser pulse for the rotational wave-packet through precisely adjusting the time delays between the two laser pulses. When the laser intensity exceeds the ionization threshold, ionization and Coulomb explosion occur. The atomic ions C^{m+} ($m=1-3$) and Oⁿ⁺ ($n=1-3$) observed in the experiment exhibit highly anisotropic angular distributions relative to the laser polarization. Using two linearly polarized laser pulses with crossed polarization, we conclude that the anisotropic angular distribution results from dynamic alignment, in which the rising edge of the laser pulse aligns the neutral CO₂ along the laser polarization direction prior to ionization.

Key words: Rotational wave-packet, Field-free alignment, Coulomb explosion

I. INTRODUCTION

With the development of ultrashort and ultraintense pulsed laser technologies, femtosecond lasers have become powerful tools to probe and control molecular dynamics. The property of the ultrashort temporal width enables the molecular dynamic processes to be monitored in real time on femtosecond time scale. For example, the transition states of chemical reactions can be measured using femtosecond pump-probe technique [1]. The transient molecular structure can be imaged with the diffraction of the ultrafast electron pulses generated by femtosecond laser pulses and a back-illuminated photocathode [2,3]. The property of ultraintense intensity enables the molecular behaviors to be changed after intense laser irradiation. In other words, the molecular dynamics can be controlled by intense femtosecond laser pulses. For example, by using phase- and amplitude-shaped intense femtosecond laser pulses, impulsive orientation and alignment of NO molecules are achieved [4].

Understanding the behavior of molecules in strong laser fields is crucial to manipulate the internal and the external degrees of freedom of molecules using femtosecond lasers [5]. 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 [6,7]. Because the laser pulse duration is much shorter than the molecular rotational period, such transient alignment can be revived periodically after the exciting laser is over. As the components of the wave-packet evolve with different frequencies, the coherent rotational wave-packet created by one strong femtosecond laser pulse can be further modified by applying another strong femtosecond laser pulse after a certain time delay [8–12].

When the laser intensity exceeds the ionization threshold, the potential energy surface (PES) of a molecule could be deformed in terms of the formation of light dressed PESs. By controlling the direction of nuclear motion on the dressed PESs, the geometrical structure of molecules might be modified [5,13,14]. During such structural deformation, the external field asymmetrically distorts the potential and forms a potential barrier on one side of the molecules [15,16]. Tunneling ionization occurs and singly, doubly, and multiply charged parent ions are formed. Due to Coulomb repulsive force, the multiply charged parent ions will explode into multiply charged atomic ions with large kinetic energies [17,18]. The angular distributions of the exploding fragments have been observed to be highly anisotropic relative to the laser polarization, which is attributed to alignment prior to ionization or angle-dependent ionization. The former is also called dynamic alignment and the latter geometric alignment [17,18].

CO₂, an important linear triatomic molecule with two centers, its dynamic processes have been exten-

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sively studied in strong femtosecond laser fields. When the laser intensity is below the ionization threshold, a coherent rotational wave-packet is formed for CO₂ irradiated by one strong femtosecond laser pulse. The evolution of the wave-packet periodically displays transient alignment and anti-alignment, which have been observed by many research groups [19–24]. Structural deformation of CO₂ molecules in an intense laser field is measured by Coulomb explosion imaging [25–27]. The angular dependence of the ionization probability is directly obtained by measuring the ion yield of the aligned CO₂ molecules. It is seen to have a sharp preference for ionization when aligned at 45° to the laser field [28]. The high-order harmonic generations from aligned CO₂ molecules have been measured and the intermolecular quantum interference has been identified [29–32]. Coulomb explosion of CO₂ are also studied and the explosion pathways are identified. Anisotropic angular distributions are observed for the exploding fragments relative to the laser polarization [33–37].

In this work, we experimentally investigate the alignment, ionization and Coulomb explosion of CO₂ in intense femtosecond laser pulses with laser intensity varying from 1×10^{13} W/cm² to 6×10^{14} W/cm². The revival structures are measured for the rotational wave-packet created by one strong femtosecond laser pulse with the intensity below ionization threshold of molecules. In addition, we demonstrate that the revival structure can be further shaped by another laser pulse for the rotational wave-packet through controlling the time delays between the two laser pulses. Coulomb explosion is also studied for CO₂ in intense femtosecond laser pulses with laser intensity exceeding the ionization threshold of molecules. The atomic ions C^{*m*+} (*m*=1–3) and O^{*n*+} (*n*=1–3) observed in the experiment exhibit highly anisotropic angular distributions relative to the laser polarization. Using two linearly polarized laser pulses with crossed polarization, we conclude that the anisotropic angular distribution results from dynamic alignment.

II. EXPERIMENTS

The experiment setup is described in our previous work [38]. The Ti:sapphire chirp-pulsed amplifier (TSA-10, Spectra-Physics Inc., USA) used in the experiment delivers laser pulses with a central wavelength of 800 nm and pulse duration of 110 fs at a repetition rate of 10 Hz. The maximum pulse energy is 6 mJ and the laser polarization is horizontal. For the field induced alignment measurement, the above laser beam is divided into two parts to provide a strong energy pump beam and a weak energy probe beam. Both the pump beam and the probe beam are focused with a 30 cm focal length lens into a 20 cm long gas cell at a small angle. The gas cell is filled with CO₂ at room temperature under one atmospheric pressure. The field-free aligned

molecules induced by the strong pump laser cause birefringence and depolarize the probe laser. The depolarization of the probe laser represents the alignment degree. Different from typical weak field polarization technique [39], the weak probe laser is a slightly elliptically polarized and a heterodyne alignment signal is obtained in the current measurement. The pure heterodyne alignment signal is obtained by subtracting the two heterodyne signals measured respectively by a left-handed and right-handed elliptically polarized probe laser. The signal is proportional to $(\langle \cos^2 \theta \rangle - 1/3)$, where θ is the polar angle between the molecular axis and the laser polarization direction. Thus, the experimental data directly reproduces the revival structure of the rotational wave-packet.

While for ionization and Coulomb explosion measurement, a beam splitter is used to divide the laser beams into two parts with equal intensity and a half-wave plate is inserted into one of the laser beams to rotate its linear polarization. The relative optical paths of the two laser beams are precisely adjusted using an optical translational stage controlled by a stepping motor. Then the two laser beams are recombined by a beam splitter. The spatial and temporal overlap of the two laser beams could be detected by observing the interference patterns after recombination. The recombined laser beams are focused into the chamber of a time-of-flight spectrometer by a lens with a focal length of 150 mm. A gaseous sample is introduced into the chamber via a pulsed valve (Park Inc., USA) with a 0.2 mm orifice. The ions produced in the laser beam are accelerated by a two-stage electric field and detected by a micro-channel plate (MCP). The signals are typically averaged over 256 laser pulses and recorded using a 1 GHz data acquisition card (DP110, Acqiris Digitizers, Switzerland).

III. RESULTS AND DISCUSSION

A. Rotational excitation of CO₂ with laser intensity below the ionization threshold

When linear molecules are irradiated by strong femtosecond laser pulses with laser intensity below the ionization threshold, nonadiabatic rotational excitation of molecules generates a coherent rotational wave-packet. Evolution of the wave-packet exhibits transient alignment or anti-alignment after the laser pulse is over. In the present measurement, we monitor the evolution of the rotational wave-packet using the pure heterodyne weak field polarization technique [38]. The pure heterodyne alignment signal is obtained by subtracting the two heterodyne signals measured respectively by a left-handed and right-handed elliptically polarized laser pulse. The experimental data directly reproduce the revival structure of the wave-packet.

Figure 1(a) shows the alignment signal for CO₂ at

room temperature irradiated by one 800 nm, 110 fs laser pulse at an intensity of 10^{13} W/cm². The laser intensity is estimated by measuring the focus size using a standard knife edge method. The classical rotational period T_r of molecules is determined by the equation $T_r=1/2B_0c$, where B_0 is the rotational constant in the ground vibronic state and c is the speed of the light. B_0 is 0.3902 cm⁻¹ for CO₂ and T_r is therefore determined to be 42.7 ps. The alignment signal repeats every molecular rotational period. Because the two O atoms are indistinguishable for CO₂ molecules, only even J states are populated due to the requirement of the symmetrization of the wave function, which lead to the observation of the alignment and antialignment around the time of quarter revival times. This work indicates that molecules can be aligned along the laser polarization by femtosecond laser pulses and macroscopic ensembles of highly aligned molecules can be obtained under field-free conditions.

After the laser is over, different components evolve with different frequencies for the rotational wave-packet. However, at the half or full revival time, all the components of the wave-packet evolve in phase and the wave-packet exhibits alignment and antialignment. If another laser pulse is applied around this time, it causes the same effects on all components of the wave-packet. Depending on the precisely time delay between two laser pulses, the wave-packet can be enhanced or annihilated when the second laser pulse is applied at half revival time. The enhancement of the alignment degree has been carefully studied by two laser pulses or a trains of laser pulses [8–10]. Here, we demonstrate that the wave-packet can be annihilated by the second

laser pulse.

Figure 1(b) shows the pure heterodyne alignment signal of CO₂ at room temperature irradiated by second laser pulses alone. The laser intensity is 8×10^{12} W/cm². The revival structure is very similar to that shown in Fig.1(a), which is formed by the first laser pulse alone. Figure 1(c) shows the pure heterodyne alignment signal of CO₂ at room temperature irradiated by two laser pulses separated by 21.36 ps. It can be seen that the wave-packet created by the first laser pulse is approximately annihilated by the second laser pulse. It should be mentioned that the energy of the second laser pulse is a little lower than that of the first one for achieving the best annihilation in the experimental measurement.

For a general case, the second laser pulse is applied at arbitrary times. The second laser pulse will cause different effects on the different components of the wave-packet created by the first laser pulse. Figure 2 (a) and (b) show the alignment signal of the wave-packet created by one 800 nm, 110 fs laser pulse at an intensity of 10^{13} W/cm². Figure 2(c) show the pure heterodyne alignment signal of the total wave-packet created by two identical laser pulses with a time delay of 12.98 ps. It can be seen that three serials of revival structures are observed for the wave-packet created by two laser pulses with a time delay of 12.98 ps. The revival structures located at $t+\Delta t$ ($t=nT_r/4$) is generated by the second laser pulse, where n is an integer number. The rotational wave-packet created by the first laser pulse is split into two wave-packets, their revival structures respectively located at $nT_r/4$ and $nT_r/4+2\Delta t$. The split originates from that different components of the wave-packet created by the first laser pulse have differ-

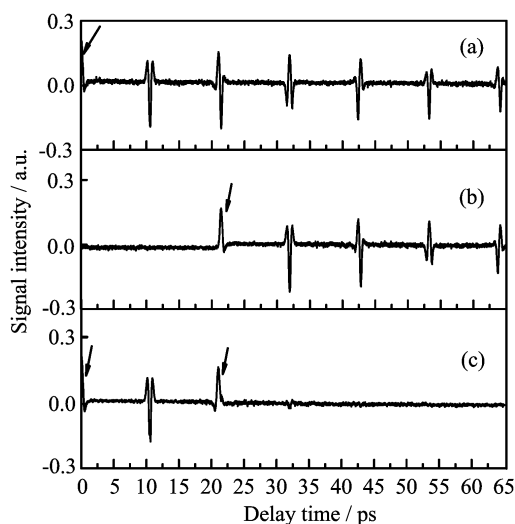


FIG. 1 Alignment signal for CO₂ at room temperature irradiated by (a) the first laser pulse alone, (b) the second laser pulse alone, and (c) both of the two laser pulses with a separated time of 21.36 ps. The arrows indicate the timing that the pulses are applied.

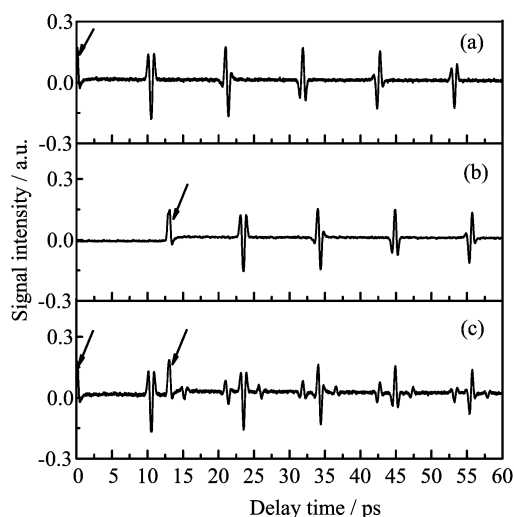


FIG. 2 Alignment signal for CO₂ at room temperature irradiated by (a) the first laser pulse alone, (b) the second laser pulse alone, and (c) both of the two laser pulses with a separated time of 12.98 ps. The arrows indicate the timing that the pulses are applied.

ent phases when the second laser pulse is applied.

B. Coulomb explosion of CO₂ with laser intensity exceeding the ionization threshold

When the laser intensity exceeds the ionization threshold of molecules, tunneling ionization leads to the formation of singly, doubly, and multiply charged parent ions. Due to Coulomb repulsive force, the multiply charged parent ions explode into atomic ions with large kinetic energies. Figure 3 shows typical mass spectra of CO₂ irradiated by (a) perpendicular polarized laser pulses at an intensity of $6. \times 10^{14}$ W/cm² and (b) parallel polarized laser pulses at an intensity of 4.5×10^{14} W/cm². The pulse duration is 110 fs and the central wavelength is 810 nm. It can be seen that strong singly and doubly charged parent ions exhibit similar shapes and intensities for both parallel and perpendicular laser polarizations. However, the atomic ions C^{m+} ($m=1-3$) and O^{n+} ($n=1-3$) exhibit different shapes and intensities depending on the laser polarizations. When the laser polarization is perpendicular to the TOF axis, C^{m+} ($m=1-3$) exhibit double peaks. However, the peaks of O^{n+} ($n=1-3$) are very weak. While the laser polarization is parallel to the TOF axis, O^{n+} ($n=1-3$) exhibit obvious double peaks. The splitting double peaks indicate that the atomic ions have large kinetic energies and are produced through Coulomb explosion.

When these ions are ejected in the direction perpendicular to the TOF axis, they will miss the micro-channel plate and cannot be collected by the detector. While these ions are ejected in the direction parallel to the time-of-flight axis, they will produce two peaks in the mass spectra. 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 backward the detector and is reversed by the extraction field. The differences of mass spectra for parallel and perpendicular polarizations demonstrate that the angular distributions of the atomic ions are highly anisotropic relative to the laser polarization. The atomic ions O^{n+} ($n=1-3$) ions are ejected along the laser polarization direction and the C^{m+} ($m=1-3$) ions perpendicular to the laser polarization direction during Coulomb explosion.

The anisotropic angular distributions of the atomic ions have been studied by many research groups [17,18]. Two mechanisms are proposed to interpret the anisotropic angular distributions, one is dynamic alignment and the other is geometric alignment. Alnaser and Voss *et al.* observed that the atomic ions peak at $\theta=0^\circ$ for N₂ and $\theta=40^\circ$ for O₂ when they studied Coulomb explosion of these molecules irradiated by 8 fs laser pulses at an intensity of 2×10^{14} W/cm² [39,40]. With the theoretical simulation, they concluded that the anisotropic angular distributions of the atomic ions

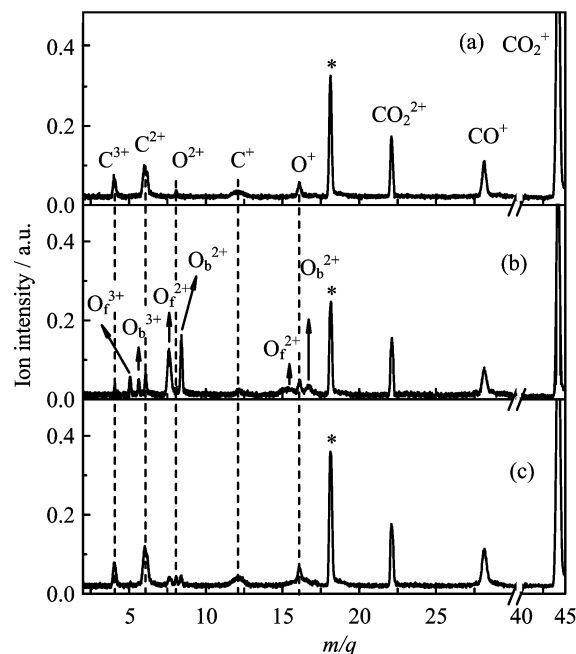


FIG. 3 Typical TOF mass spectra of CO₂ irradiated by (a) the first perpendicular polarized laser pulse alone, (b) the second parallel polarized laser pulse alone, and (c) both of the two laser pulses with a separated time of 1 ps. The peak denoted with an asterisk originates from the residual water in the vacuum chamber.

result from the geometric alignment. The angular distributions of the atomic ions reflect the angle-dependent ionization of N₂ and O₂ in an 8 fs laser field. However, Huang *et al.* observed that the atomic ions peak along the laser polarization for both N₂ and O₂ when these molecules are irradiated by 110 fs laser pulses at an intensity of 6×10^{14} W/cm² [41]. Using two crossed linearly polarized laser pulses, they concluded that the anisotropic angular distributions result from dynamic alignment, in which the rising edge of the laser pulses aligns the neutral molecules along the laser polarization direction. The angular distributions of the atomic ions reflect the degree of the alignment of molecules prior to ionization.

For CO₂ molecules, when the intensity is below the molecular ionization threshold, we have demonstrated that the molecule can be aligned along the laser polarization direction by 110 fs laser pulses using the pure heterodyne weak field polarization technique. When the intensity is much higher than the molecular ionization threshold, Coulomb explosion occurs for CO₂ molecules irradiated by 110 fs laser pulses. Meantime, we have observed that the O^{n+} ($n=1-3$) ions are ejected along the laser polarization and the C^{m+} ($m=1-3$) ions perpendicular to the laser polarization during Coulomb explosion. Therefore, we speculate that our observed anisotropic angular distributions of the atomic ions result from dynamic alignment. In order to confirm our speculation, we use double pulse method to evaluate the

contribution of dynamic alignment.

Figure 3(c) shows the characteristic mass spectra of CO₂ irradiated by two laser pulses with a separate time of 1 ps. The first laser pulse is perpendicular polarized and has an intensity of 6×10^{14} W/cm². The second laser pulse is parallel polarized and has an intensity of 4.5×10^{14} W/cm². It can be seen that the mass spectra in Fig.3(c) is similar to that in Fig.3(a). The signal intensities of Oⁿ⁺ ($n=1-3$) ions produced by the second laser pulse are greatly decreased by the first laser pulse during the double pulse experiment. The observation verifies that dynamic alignment dominates under our experimental condition. All molecules within the laser focus are aligned by the rising edge of the first laser pulse. At the peak of the laser intensity, these aligned molecules are ionized and multiply charged parent ions are produced. Coulomb explosion occurs immediately for the multiply charged parent ions. The atomic ions Oⁿ⁺ ($n=1-3$) ions fly along the laser polarization and the C^{m+} ($m=1-3$) ions perpendicular to the laser polarization. Because most of the neutral molecules have been depleted by the first laser pulse, the signal generated by the second laser pulse is greatly decreased in the presence of the first laser pulse.

IV. CONCLUSION

In summary, we experimentally investigate the alignment, ionization and Coulomb explosion of CO₂ in intense femtosecond laser pulses with laser intensity varying from 1×10^{13} W/cm² to 6×10^{14} W/cm². When the laser intensity is below the ionization threshold, nonadiabatic rotational excitation of molecules generates a coherent rotational wave-packet. The evolution of the wave-packet periodically displays alignment and anti-alignment after the laser pulse is over. The revival structure can be modified by a second laser pulse. Depending on the time delay between the two laser pulses, the wave-packet created by the first laser pulse can be enhanced, annihilated or split by the second laser pulse. When the laser intensity exceeds the ionization threshold, ionization and Coulomb explosion occur. The atomic ions C^{m+} ($m=1-3$) and Oⁿ⁺ ($n=1-3$) observed in the experiment exhibit highly anisotropic angular distributions relative to the laser polarization. Using two linearly polarized laser pulses with crossed polarization, we conclude that the anisotropic angular distribution results from dynamic alignment, in which the neutral CO₂ is aligned along the laser polarization direction prior to ionization by the rising edge of the laser pulse. At the peak of the laser intensity, these aligned molecules are ionized and multiply charged parent ions are produced. The atomic ions Oⁿ⁺ ($n=1-3$) ions produced in Coulomb explosion fly along the laser polarization direction and the C^{m+} ($m=1-3$) ions perpendicular to the laser polarization direction.

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