Ellipticity-dependent ionization/dissociation of carbon dioxide in strong laser fields*

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(Received 23 September 2014; revised manuscript received 16 October 2014; published online 30 January 2015)

Ionization and dissociation of linear triatomic molecules, carbon dioxide, are studied in 50-fs 800-nm strong laser fields using time-of-flight mass spectrometer. The yields of double charged ions CO_2^{2+} and various fragment ions (CO^+, O^{n+}, and C^{n+} ($n = 1, 2$)) are measured as a function of ellipticity of laser polarization in the intensity range from 5.0×10^{13} W/cm^2 to 6.0×10^{14} W/cm^2. The results demonstrate that non-sequential double ionization, which is induced by laser-driven electron recollision, dominates double ionization of CO_2 in the strong IR laser field with intensity lower than 2.0×10^{14} W/cm^2. The electron recollision could also have contribution in strong-field multiple ionization and formation of fragments of CO_2 molecules. The present study indicates that the intensity and ellipticity dependence of ions yields can be used to probe the complex dynamics of strong-field ionization/dissociation of polyatomic molecules.

Keywords: laser ellipticity, strong field, ionization and dissociation, carbon dioxide

PACS: 33.80.Rv **DOI:** 10.1088/1674-1056/24/3/033302

1. Introduction

In the laser-driven electron recollision process (also called three-step process), an electron in an atom or molecular is liberated through tunneling ionization, then it is accelerated in the laser field, and finally can revisit the parent ion upon reversal of the directional vector of the laser field. This scenario, which was pioneered by Corkum in 1993,[1] has been widely viewed as the underlying mechanism of many strong-field atomic (or molecular) phenomena, such as non-sequential double ionization (NSDI),[2–4] high harmonic generation (HHG),[5,6] high-order above threshold ionization (HATI),[7–9] and population of high-lying Rydberg states.[10] With increasing ellipticity of laser polarization, recollision with the parent ions diminishes due to the greater drift momentum spread of the returning electron wavepacket. On the other hand, "long orbit" of the returning electron (or multi-return collision trajectory) would play a significant role under elliptically polarized pulses, which could be used to control the electron wavepackets.[9,11] In the past several years, it has attracted increasing attention to investigate the interaction of atoms or molecules with elliptical polarized laser fields to reveal and control the dynamics of the strong laser-driven electron recollision process.[9,11–15]

Compared to that of atoms or diatomic molecules, the mechanism of ionization/dissociation of ployatomic molecules in strong laser fields is much more complicated due to increasing degree of freedoms.[16–18] Different molecular geometries, quantum states, alignment or orientation of molecules can affect the strong-field ionization/dissociation of molecules. Studies of the interaction of polyatomic molecules with strong laser fields are quite in demand. Particularly, it is commonly accepted that when the magnitude of the laser electric fields can be comparative with that of the Coulombic field of the electron within the target molecule, tunneling ionization is dominant, which is the first step of the laser-driven electron recollision process. However, the role of the electron recollision with the parent ions in strong field ionization/dissociation of polyatomic molecules is far less understood than that in atoms.

Here we carry out an experimental study regarding strong-field ionization/dissociation of CO_2 molecules on the ellipticity of polarization of femtosecond laser fields. CO_2 has a linear structure with the carbon atom in the center. Its highest occupied molecular orbital (HOMO) is doubly degenerate π_g with two perpendicular nodal planes, which is very similar to that of diatomic molecule O_2 or polyatomic molecule such as C_6H_6. The strong-field ionization/dissociation of CO_2 is a long-standing attractive topic. Early studies focused on dissociative ionization and Coulomb explosion in linearly polarized strong laser fields.[19] Recently, several studies investigated alignment-dependent ionization of CO_2 in the tunneling region, by experimental measurements[20,21] and theoretical calculations.[22,23] It is debated that the multielectron effect,[20] the coherent core trapping effect,[23] or differ-

*Project supported by the National Basic Research Program of China (Grant No. 2013CB922200) and the National Natural Science Foundation of China (Grant Nos. 11034003 and 11274140).

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ent structures of electron orbitals[21] may be taken into account in the tunneling ionization of CO_2. In 2006, McKenna *et al.* observed evidence for recollision leading to non-sequential enhanced dissociation of CO_2^+ in a 55-fs 790-nm laser field, by employing an intensity-selective scan technique and comparing the signals from linearly and circularly polarized pulses.[24] Later, a clear non-sequential signature was observed in double ionization of CO_2 by measuring the CO_2^{2+}/CO_2^+ ratio as a function of laser intensity.[25] Very recently, the non-sequential double ionization (NSDI) of CO_2 was demonstrated to have multichannel contributions from both the ground and first excited ionic states.[26] These studies highlight the importance of laser-driven electron recollision in strong-field ionization/dissociation of CO_2 molecules.

To show further evidence of recollision in strong-field ionization/dissociation of CO_2, in this study we measure different ion yields as a function of ellipticity of laser polarization, at various intensities in the range from 5.0×10^{13} W/cm^2 to 6.0×10^{14} W/cm^2 of a 50-fs 800-nm laser field. Our results demonstrate that the NSDI dominates double ionization of CO_2 in the strong IR laser field with intensity lower than 2.0×10^{14} W/cm^2, and indicate that laser-driven electron recollision could contribute to the strong-field multiple ionization and formation of fragments of CO_2 molecules.

2. Experimental setup

The experimental setup used for femtosecond laser ionization/dissociation experiments was similar to that described in our previous studies.[27,28] Briefly, CO_2 molecules were introduced into the vacuum chamber through a leak valve to interact with the strong laser pulses. The stagnation pressure was kept at ~1 atm, and the operating pressure in the chamber was about 3×10^{-4} Pa. Linearly polarized infrared femtosecond laser pulses were generated by a Ti:Sapphire chirped-pulse amplified laser with a central wavelength of 800 nm, pulse duration of 50 fs, repetition rate of 1 kHz, and maximum pulse energy of 4 mJ. A half-wave plate and a Glan prism were inserted into the laser beam to vary the laser intensity continuously. The polarization of the laser pulse was controlled by rotating a quarter-wave plate before it was focused into the vacuum chamber to ionize the molecules. The peak intensity of the focused laser pulse was calibrated by comparing the measured saturation intensity of Xe with that calculated by the Ammosov–Delome–Krainov (ADK) model.[29] A linear time-of-flight (TOF) mass spectrometer operated under the Wiley–McLaren condition was used to detect the produced cations from strong-field ionization/dissociation. All the cations were extracted, accelerated, and finally detected by a dual microchannel plate (MCP) detector at the end of the flight about 55 cm. A 0.5 mm width slit was mounted in front of the flight tube to ensure that only those ions produced in the center portion of the focused volume were detected. Mass-resolved ion signals were recorded using a digital oscilloscope (Tektronix TDS 3054B) and sent to a PC for analysis. All experimental data were normally averaged over 10^3 laser shots.

3. Results and discussion

The TOF mass spectra of CO_2 irradiated by linearly polarized 800-nm laser fields at 1.0×10^{14} W/cm^2 show various cations, including singly and doubly charged parent ions, CO_2^+ and CO_2^{2+}, molecular fragment ions CO^+, and atomic fragment ions with different charges, O^{n+} and C^{n+} ($n=1$, 2). The ellipticity dependence of the yield of each cation was measured at various laser intensities. The results show that the yield of the parent ions CO_2^+ only slightly decreases as the ellipticity of the laser polarization is increased, which may be due to the fact that the laser peak field decreases with the ellipticity of polarization. In the following experiments, the ratio of each ion to singly charged ion was employed, in order to take into account the small decrease in field.[16] For all the present results, the ratios at different ellipticities were normalized to that at an ellipticty of 0, i.e., linear polarization.

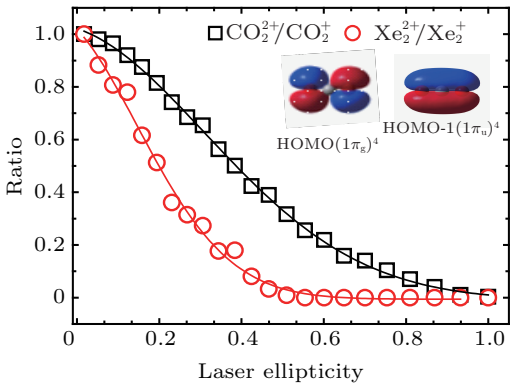


Fig. 1. (color online) Dependence of the ratio of CO^{2+} and Xe^{2+} on the ellipticity of laser polarization. The inset shows the HOMO and HOMO-1 of CO_2 molecules.

Figure 1 shows the ratio CO_2^{2+}/CO_2^+ as a function of polarization ellipticity with a laser intensity of 1.0×10^{14} W/cm^2. Since the ionization potential of the CO_2 molecule (13.77 eV) is close to that of Xe atom, we also present in the figure the ellipticity dependence of Xe^{2+}/Xe^+ at the same laser intensity for comparison. The solid curves show the best-fit Gaussian distributions to the experimental data of CO_2 and Xe. The Xe^{2+}/Xe^+ ratio is maximal for linear polarization and decreases rapidly with increasing ellipticity of the polarization. This is a general feature of the NSDI of atoms. According to the ADK model, the width of the Gaussian distribution is determined by the ionization potential and the laser intensity.

The half width at half maximum (HWHM) for Xe^{2+}/Xe^{+} distribution in this study is about 0.20, in relative agreement with previous experimental measurement.[16]

For CO_2 molecules, the ratio CO_2^{2+}/CO_2^{+} also decreases as the ellipticity of laser polarization is increased, and double ionization is completely suppressed for circular polarization, indicating an NSDI process. It is well known that for molecules, a maximum NDSI yield should be at non-zero laser ellipticity if the electron orbital of a molecule has a nodal plane along the linear laser polarization direction, leading to destructive interference between the components of the recolliding wave packets with opposite transverse momentum components in the polarization plane.[16] However, the NSDI probability of CO_2 does not display this behavior, despite the fact that its HOMO has two nodal planes perpendicular to each other (see the inset of Fig. 1). Instead, a maximal NSDI yield is at linear polarization and the distribution can be well described by a Guassian function, similar to that in atoms. This indicates that the NSDI of CO_2 is not produced via HOMO alone. Contributions from the next lower lying orbit, HOMO-1, which is a π_u symmetry, could lead to an increase in the NSDI yield at perpendicular polarization. Indeed, the multichannel contribution of strong-field ionization of CO_2 was demonstrated in two recent studies using aligned molecules.[21,26] Such a multichannel contribution does not result in an obvious destructive interference, and thus a dip of the ratio at linear polarization would not be observed.

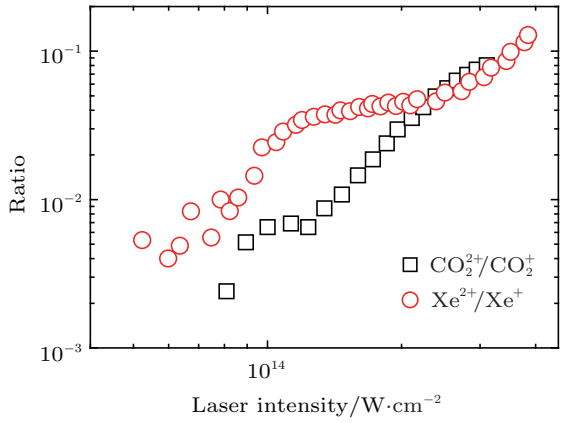


Fig. 2. (color online) The ratio of CO_2^{2+}/CO_2^{+} and Xe^{2+}/Xe^{+} as a function of laser intensity, recorded with linear polarization of the laser field.

Figure 2 shows the ratio of CO_2^{2+}/CO_2^{+} as a function of laser intensity in the range from 5.0×10^{13} W/cm^2 to 4.0×10^{14} W/cm^2 recorded at linear polarization. The result of Xe^{2+}/Xe^{+} is also plotted for comparison. A clear "knee" structure (weak dependence of the ratio on the laser intensity) in the curve of CO_2^{2+}/CO_2^{+} was observed at the intensity of $\sim 9.0 \times 10^{13}$ W/cm^2, which is a signature of the NSDI. The appearance of the "knee" structure of CO_2 occurs at roughly the same intensity as that of Xe atom, which could be attributed to their similar ionization potentials (13.78 eV for CO_2, 12.17 eV for Xe). As the intensity is further increased, sequential double ionization (SDI) dominates, and thus the yield of doubly charged ions increases again with the intensity.

To further investigate the NSDI and SDI processes for the formation of doubly charged ions, CO_2^{2+}, we measured the ellipticity dependence of the ratio of CO_2^{2+}/CO_2^{+} at five different laser intensities of 2.0×10^{14} W/cm^2, 3.0×10^{14} W/cm^2, 4.0×10^{14} W/cm^2, 5.0×10^{14} W/cm^2, and 6.0×10^{14} W/cm^2. The results are shown in Fig. 3. One can see that the CO_2^{2+}/CO_2^{+} ratio exhibits less dependence on the ellipticity of the polarization for higher laser intensity. At 6.0×10^{14} W/cm^2, the ratio at circular polarization (ellipticity is 1) only slightly decreases (less than 20%), compared to that for linear polarization (ellipticity is 0). While at low laser intensity, the double ionization is completely suppressed (see also Fig. 1). As mentioned above, the SDI process becomes the main process for strong-field double ionization of CO_2 with increasing laser intensity, which is unlike the NSDI process via laser-driven electron recollision with the parent ions that has strong ellipticity dependence. Our results can shed light on the physical mechanism of strong-field double ionization by measuring the yield of doubly charged ions with the variation of intensity and polarization of the strong laser pulses.

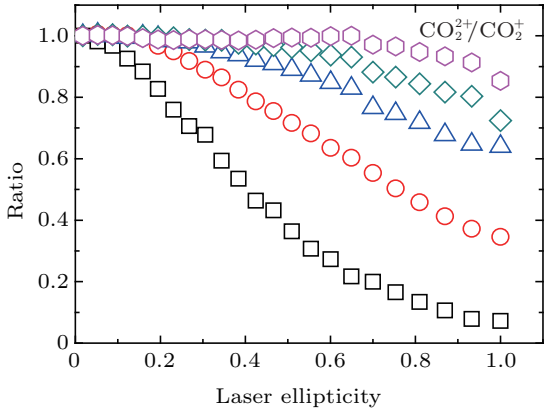


Fig. 3. (color online) Dependence of the ratio of CO_2^{2+}/CO_2^{+} on ellipticity of the laser polarization at different laser intensities: 2.0×10^{14} W/cm^2 (black square), 3.0×10^{14} W/cm^2 (red circle), 4.0×10^{14} W/cm^2 (blue triangle), 5.0×10^{14} W/cm^2 (green diamond), and 6.0×10^{14} W/cm^2 (pink hexagon).

We also investigate the ellipticity dependence of fragmental ions at different laser intensities. Figure 4 presents the results of O^{n+} ($n = 1, 2$) and CO^{+} fragments, and figure 5 presents those of O^{n+} ($n = 1, 2$) fragments. For single charged fragment ions, O^{+} and CO^{+}, the ratio decreases with increasing ellipticity of laser polarization, but the dependence is not as strong as that in NSDI. Our results indicate that O^{+} and CO^{+} could not be produced solely from dissociation of CO_2^{+}, because the CO_2^{+} yield exhibits only slight dependence on the laser ellipticity. Dissociation from double charged parent ions may also contribute to the formation of these single

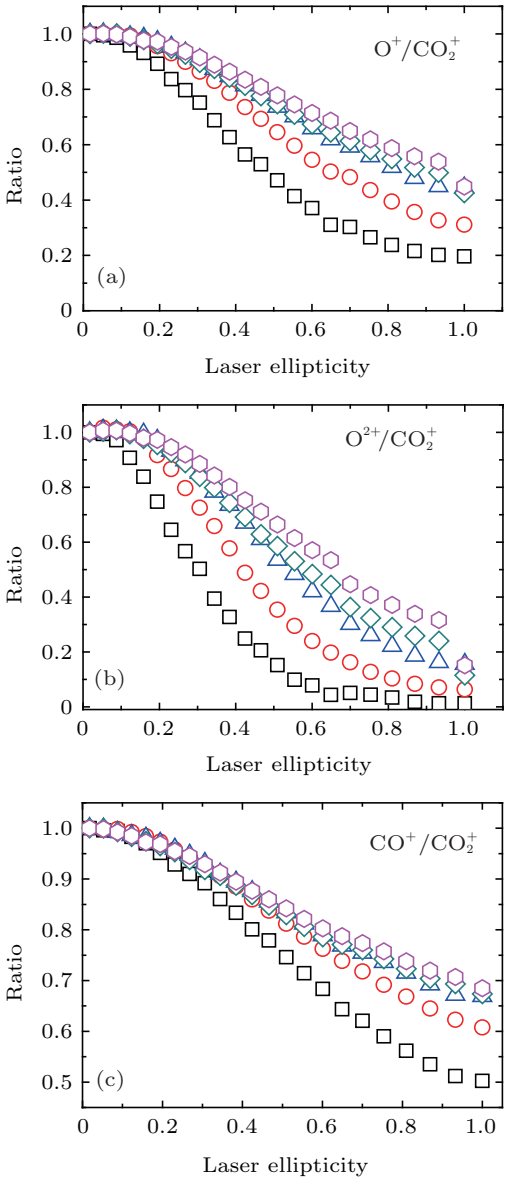


Fig. 4. (color online) Dependence of the ratio of (a) O^+/CO_2^+, (b) O^{2+}/CO_2^+, and (c) CO^+/CO_2^+ on ellipticity of the laser polarization at different laser intensities: 2.0×10^{14} W/cm^2 (black square), 3.0×10^{14} W/cm^2 (red circle), 4.0×10^{14} W/cm^2 (blue triangle), 5.0×10^{14} W/cm^2 (green diamond), and 6.0×10^{14} W/cm^2 (pink hexagon).

charged fragment ions. On the other hand, the ratio of the double charged fragment ions, O^{2+}, shows strong ellipticity dependence at each investigated laser intensity, even stronger than that of CO_2^{2+}. The O^{2+} fragments are attributed to dissociation or Coulomb explosion from highly charged CO_2 ions. If electron recollision leads to NSDI as already evidenced above, it should also play a significant role in multiple ionization of CO_2, i.e., non-sequential multiple ionization. Thus, the O^{2+} fragments from highly charged CO_2 ions would have strong ellipticity dependence as observed in Fig. 4. The results presented here clearly demonstrate the importance of the laser-driven electron recollision in strong-field multiple ionization. Similar ellipticity dependence was also observed in C fragment ions, as shown in Fig. 5. It is interesting to see that at high laser intensity of 6.0×10^{14} W/cm^2, the ratio of C^+/CO_2^+ or C^{2+}/CO_2^+ starts to increase at circular polarization, which may be due to the complicated channels of the formation of C fragment ions. Further studies with the application of more advanced coincidence measurements are in demand to reveal the underline physical mechanism.

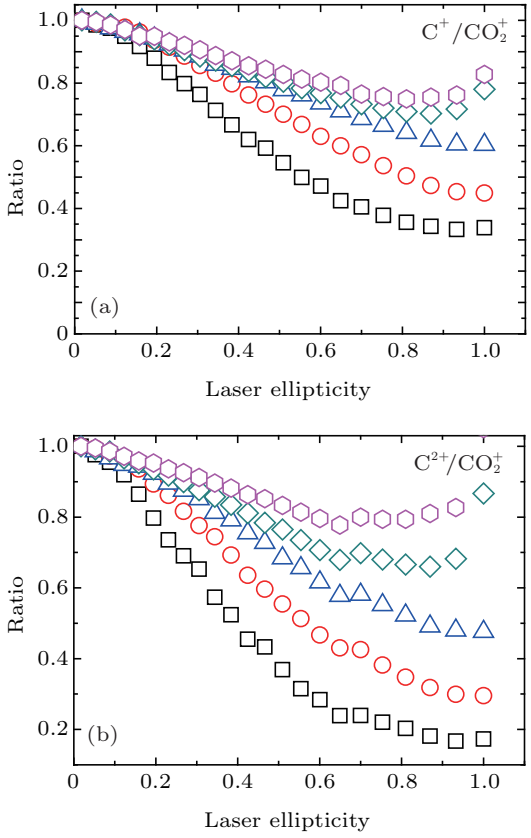


Fig. 5. (color online) Dependence of the ratio of (a) C^+/CO_2^+ and (b) C^{2+}/CO_2^+ on ellipticity of the laser polarization at different laser intensities: 2.0×10^{14} W/cm^2 (black square), 3.0×10^{14} W/cm^2 (red circle), 4.0×10^{14} W/cm^2 (blue triangle), 5.0×10^{14} W/cm^2 (green diamond), and 6.0×10^{14} W/cm^2 (pink hexagon).

4. Conclusion

In conclusion, we investigate the dependence of strong-field ionization/dissociation of CO_2 molecules on the ellipticity and intensity of the femtosecond IR laser pulses. Strong dependence of the yield of CO_2^{2+} ions on the ellipticity of polarization is observed with laser intensity below 2.0×10^{14} W/cm^2, and double ionization is completely suppressed under circular polarization. In addition, a clear "knee" structure is observed in the curve of CO_2^{2+}/CO_2^+ as a function of laser intensity. The results demonstrate the NSDI process in the strong-field double ionization of CO_2 molecules. The ellipticity dependence yields of various fragment ions were also measured at several laser intensities. The present study indicates that laser-driven electron recollision with the parent ions is of vital importance in double even multiple ionization as well as dissociation of polyatomic molecules in strong laser fields.

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