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Research paper

Three-body fragmentation dynamics of carbon-dioxide dimers induced by intense femtosecond laser pulses

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ABSTRACT

We experimentally studied three-body fragmentation dynamics of $(CO_2)_2^{3+}$ generated by intense femtosecond laser fields. Three-dimensional momentum vectors as well as kinetic energies were measured for correlated fragmental ions using the technology of coincidence measurement. The results demonstrate that sequential fragmentation channel dominates for three-body fragmentation of $(CO_2)_2^{3+}$, in which the weak van der Waals bond breaks first and then one strong covalent bond.

1. Introduction

Many-body fragmentation dynamics is very complicated for polyatomic molecules [1–3]. One of the big challenges is that many-body fragmentation can occur through a concerted or a sequential fashion. In the case of concerted fragmentation process, all reaction products are generated in a single kinetic event. Instead, in the case of sequential fragmentation process, the reaction products are generated one after the other with a time interval longer than the rotational period of the intermediate. Maul and Gericke [4] summarized the most common experimental procedures and theoretical models that have been used to identify the concerted fragmentation channel and the sequential fragmentation channel for the photo-induced three-body fragmentation dynamics. Very recently, three-body fragmentation dynamics of molecular ions has attracted much attention because of the development of some experimental technologies. Firstly, multiple charged molecular ion can be easily generated in the laboratory by synchrotron radiation [5], collision of charged ions [6,7], electron impact [8,9], or intense laser pulses irradiation [10–15]. Due to Coulomb repulsive force, multiple charged molecular ions are usually unstable and will dissociates into fragmental ions with high kinetic energies. Thus these multiple charged molecular ions are suitable candidates for studying many-body fragmentation dynamics. Secondly, the improvement of many-body coincidence measurement technologies makes possible the direct determination of internal quantum states and three-dimensional momentum distributions of correlated fragment ions [16–18]. Thus the concerted fragmentation channel and the sequential fragmentation channel can be precisely identified and separated.

As a prototype system of three-body fragmentation, the process of $CO_2^{3+} \rightarrow O^+ + C^+ + O^+$ has been extensively studied. In these studies CO_2^{3+} is generated through synchrotron radiation [19], slow highly charged ions collision [20], fast heavy-ion beams collision [6], intense femtosecond laser pulses irradiation [21] as well as electron impact [22]. Among which it has been confirmed that concerted fragmentation channel and sequential fragmentation channel coexist when CO_2^{3+} is generated through slow highly charged ions collision [20], intense femtosecond laser pulses irradiation [21] as well as electron impact [22]. Because the laser parameters can be easily adjusted, it becomes a hot topic for studying many-body fragmentation dynamics induced by intense femtosecond laser pulses. However, the laser-molecule interaction is very complicated in intense laser fields. The molecular reaction pathway is determined by the correlation dynamics of electron–electron, electron–nuclear, and nuclear–nuclear in intense laser fields. Even though both concerted fragmentation channel and sequential fragmentation channel exist when CO_2^{3+} is generated by intense femtosecond laser pulse irradiation [21], recent experimental results show that only concerted fragmentation channel exists for CO_2^{n+} ($n = 4$–6) [23]. In addition, it is concluded that all the atomic ions generated in the three-body fragmentation channel of CO_2^{n+} ($n = 3$–6) are in the ground electronic state. In contrast, it is confirmed that the concerted fragmentation channel dominated for both Ar_3^{3+} and Ar_3^{n+} ($n = 4$–6) generated by intense laser pulses,

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in which the atomic ions can be in the ground electronic state as well as highly Rydberg state [24].

In this Letter, we experimentally studied three-body fragmentation dynamics of $(CO_2)_2^{3+}$ generated by intense femtosecond laser fields. Carbon-dioxide dimers are aggregates of two CO_2 molecules bound by a van der Waals bond [25]. They contain two kinds of bonds, a very weak van der Waals bond and strong covalent bonds. The results clearly demonstrate that sequential fragmentation channel dominate for three-body fragmentation of $(CO_2)_2^{3+}$ generated by intense laser fields, in which the weak van der Waals bond breaks first and then one strong covalent bond.

2. Experimental setup

The experiment was carried out using the combination of a home-built reaction microscope [26] and an intense femtosecond laser amplifier. The CO_2 dimers were generated by the supersonic expansion of CO_2 gas through a 30 μm nozzle with a driving pressure of 8 bars. The laser pulse with 25 fs duration centered at 780 nm was produced from a Ti:Sapphire laser system (Femtolasers, GmbH). The ions produced in the laser-molecule interaction were collected by a temporal and position-sensitive detector (RoentDek, Germany). By measuring the time of flight and the position in the detector, the ion can be identified and the initial three-dimensional momentum can be determined. To ensure all fragmental ions originate from the same target molecule, we control the reaction chamber pressure low enough so that there is less than one ionization event for each laser pulse. One of the advantages of reaction microscope is that the data of all reaction channels can be recorded in one experiment. In the off-line analysis, the data from different reaction channels can be disentangled by designing some constraints to filter the experimental data.

3. Results and discussion

Fig. 1(a) shows the time-of-flight mass spectra of supersonic carbon dioxide beam irradiated by 780 nm, 25 fs laser pulses at an intensity of 4×10^{14} W/cm^2. The peaks can be assigned to H^+, H_2^+, C^{2+}, C^+, O^+, H_2O^+, CO_2^{2+}, CO^+, CO_2^+ and $(CO_2)_2^+$ according to their mass to charge ratios. Among which the ions, H_2O^+ and H_2^+ and H^+, originate from the residual gas H_2O and H_2 in the vacuum chamber. The mass spectra are consistent with our previous report [27]. Most of peaks are broad, such as C^{2+}, C^+, O^+, and CO^+. The broad peak indicates that the corresponding ion has a large kinetic energy (KE), which should be generated through dissociative ionization of CO_2 monomer or CO_2 cluster. These channels can be disentangled by designing some constraints to filter the experimental data. Here, the following fragmentation channels are selected out to analyze the three-body fragmentation dynamics of $(CO_2)_2^{3+}$.

$$(CO_2)_2^{3+} \rightarrow CO_2^+ + CO^+ + O^+ \quad (1)$$

$$(CO_2)_2^{3+} \rightarrow CO_2^+ + CO_2^{2+} \quad (2)$$

$$CO_2^{2+} \rightarrow CO^+ + O^+ \quad (3)$$

Fig. 1(b)–(d) shows the time-of-flight mass spectra from the channels (1)-(3), respectively. In order to select out the events from these channels, we designed some constrains to filter the experimental data. For example, three constrains are applied to select out the events from channel (1). (a) Only one CO_2^+ ion, one CO^+ ion and one O^+ ion were detected in one laser pulse. (b) The sum-momentum of the three ions was less than 10 atomic units to meet the requirement of the momentum conservation of three-body fragmentation. (c) The momentum of the CO_2^+ ion was larger than 0.5 eV to eliminate false coincident events, in which two CO_2 monomers are ionized by the same laser pulse and the CO_2^+ ion is stable and the CO_2^{2+} ion further dissociates into CO^+ and O^+. It should be mentioned that such process cannot completely rule out the contamination of false coincidence events, such as from the dissociative channel of $(CO_2)_3^{3+}$ with one fragment is neutral CO_2 molecule. According to our data analysis, the contamination is evaluated to be smaller than 5%.

Fig. 2 show the KE of each fragmental ion as well as the total KE measured in the center of mass coordinate for channels (1)–(3). It can be seen that the CO_2^+ KE distribution generated from channel (1) is similar to that generated from channel (2). The little discrepancy at low-energy side might come from the contamination of

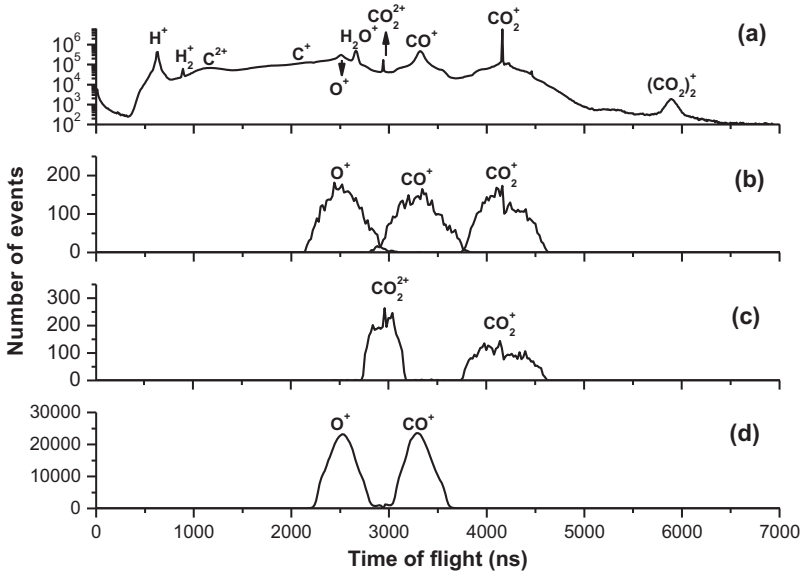

Fig. 1. (a) Time-of-flight mass spectra of supersonic carbon dioxide beam irradiated by 780 nm, 25 fs laser pulses at an intensity of 4×10^{14} W/cm^2. The ions are generated through various reaction channels, among which the channels of (b) $(CO_2)_2^{3+} \rightarrow CO_2^+ + CO^+ + O^+$; (c) $(CO_2)_2^{3+} \rightarrow CO_2^+ + CO_2^{2+}$; (d) $CO_2^{2+} \rightarrow CO^+ + O^+$ are selected out to analyze further.