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Molecular dynamics of CO in few-cycle laser fields

Qingqing Liang, Chengyin Wu*, Zhifeng Wu, Min Liu, Yongkai Deng, Qihuang Gong*

State Key Laboratory for Mesoscopic Physics and Department of Physics, Peking University, Beijing 100871, People's Republic of China

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ABSTRACT

We experimentally study the ionization, fragmentation and Coulomb explosion of CO using 6 fs laser pulses. Different from previous observations in tens or hundreds of femtoseconds laser pulses, strong charge asymmetric dissociation and CO^{2+} are observed in the current intense few-cycle laser field.

1. Introduction

Recent advances in femtosecond laser technology have made it possible for laboratories to have few-cycle femtosecond lasers with intensities around or higher than 10^{14} W/cm^2. The behaviors of molecules in such an ultrashort laser field have attracted much attention [1–8]. These studies have greatly improved the understanding of laser–molecule interaction mechanism. Now, few-cycle femtosecond laser pulses have become powerful tools to probe and control the behaviors of molecules. Alnaser et al. [9] measured the anisotropic angular distributions of ionic fragments of N_2 and O_2 by using 8 fs laser pulses at intensities in the rescattering double ionization regime. The angular-dependent ionization rate is obtained by the angular distribution of the ionic fragments. Baldit et al. [10] studied molecular multiple ionization of N_2 using 10 fs laser pulses at an intensity of 2×10^{15} W/cm^2. They found that Coulomb explosions occur at short internuclear distances close to the neutral molecular equilibrium distance in the few-cycle laser pulse. Their work opens the route to Coulomb explosion imaging of neutral molecules. Recently, Niikura et al. [11] demonstrated the control of vibrational wave-packet with intense, few-cycle laser pulses. They successfully manipulated the branching ratio of the wave-packet propagating in the bound state to the dissociation continuum by adjusting the time delay of the control laser pulse. Control of electron localization was also demonstrated in molecular dissociation by a phase-stabilized few-cycle laser pulse [12]. Electron localization controlled by the subcycle laser electric field results in the asymmetric ejection of the ionic fragment.

Multielectron dissociative ionization (MEDI) of molecules has been extensively studied experimentally and theoretically [13–19]. One of the most important findings is that the kinetic energies releases (KERs) of the exploding fragments measured in the experiment are much lower than those predicted by Coulomb explosion occurring at the neutral molecular equilibrium distance [13,14]. This finding is explained by the well-known charge resonance enhanced ionization, where the molecular ionization rate at the critical internuclear distance is many orders of magnitude greater than that at other internuclear distances [15,16]. In the process of MEDI, additional electrons are stripped away and highly charged parent ions are produced. Due to Coulomb repulsive force, the highly charged parent ions explode. Explosion pathways are divided into charge asymmetric dissociation (CAD) and charge symmetric dissociation (CSD). CSD dominates in the process of MEDI for the laser pulse with the pulse duration long enough for the molecule to reach the critical internuclear distance [17,18]. Guo et al. observed CAD of homonuclear diatomic molecules with near-infrared 30 fs laser pulse for the first time even though the signals of these channels are very weak [19]. They concluded that CAD is a natural result of strong field excitation and ionization. It was predicated that CAD will be observed easily in a few-cycle laser field. In this article, we experimentally studied the dynamic behaviors of CO by 6 fs laser pulses. The laser intensity varies from 1×10^{14} to 2×10^{15} W/cm^2. The observations support previous theoretical predictions for few-cycle laser fields that strong CAD is easily observed and Coulomb explosions occur near the equilibrium distance of the neutral molecule.

2. Experimental setup

The experimental setup is described in detail in our previous paper [20]. The commercial femtosecond laser amplifier (Femto-

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* Corresponding authors.
E-mail addresses: cywu@pku.edu.cn (C. Wu), qhgong@pku.edu.cn (Q. Gong).

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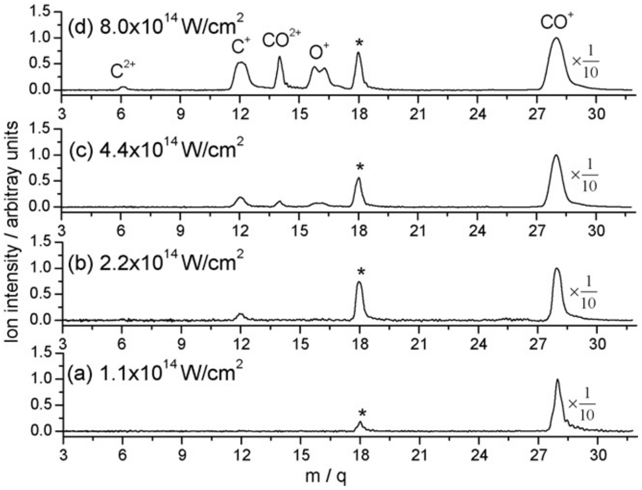

Fig. 1. Time-of-flight mass spectra of CO irradiated by 6 fs laser pulses with different intensities. The spectra are normalized to the intensity of CO^+. The peak denoted with an asterisk originates from the residual water in the vacuum chamber.

lasers GmbH) delivers laser pulses with a central wavelength of 780 nm and a pulse duration of 27 fs at a repetition rate of 3 kHz. Then the laser spectrum is broadened in a 1 m-long hollow fiber filled with 2 atm neon. After collimation by an $f = 1$ m concave silver mirror, the laser beam is recompressed using another five broadband chirped mirrors. The laser pulse duration is monitored by a commercial interferometric autocorrelator and the shortest laser duration is about 6 fs. Finally, the 6 fs laser beam is focused by a concave mirror with a focal length of 75 mm, which is put into the chamber of a time-of-flight spectrometer. A gaseous sample is introduced into the chamber via a pulsed valve (Park Inc., USA) with a 0.2 mm orifice. The vacuum pressure is maintained to be lower than 5×10^{-7} mbar when gaseous molecules are introduced. There are several tens of molecules within the laser–molecule interaction region under our experimental conditions. 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 mass spectra and recorded using a 1 GHz data acquisition card (DP110, Acqiris Digitizers, Switzerland).

3. Results and discussion

Fig. 1 illustrates the typical time-of-flight mass spectra of CO irradiated by 6 fs linearly polarized laser pulses. The laser intensities vary from 1.1×10^{14} to 8.0×10^{14} W/cm^2. The laser polarization direction is parallel to the flight tube of the time-of-flight mass spectrometer. The H_2O^+ and H^+ ions originate from the residual water in the vacuum chamber. The mass spectra are normalized to the intensity of the singly charged parent ion CO^+. When the laser intensity is lower than 2.0×10^{14} W/cm^2, only the parent ion CO^+ is observed. When the laser intensity is further increased, C^+, O^+, CO^{2+} and C^{2+} show up in turn. The appearance sequences for these ions are similar to those reported in the single photon ionization mass spectra [21]. These similarities make us conclude that these ionic fragments have the same formation mechanisms as those in the single photon ionization dissociation processes. They are formed through the direct dissociation of the parent ions in different excited electronic states in the current few-cycle laser field.

The neutral CO molecule has the electronic configuration $1\sigma^2 2\sigma^2 3\sigma^2 4\sigma^2 1\pi^4 5\sigma^2$. Laser-induced ionization of an inner-valence electron or multielectron excitation will result in the parent ion in an excited electronic state. The parent ion thus formed is unstable and dissociates into ionic fragments. The assignments of these excited electronic states have been extensively studied [21–24]. The inner-valence electron has large binding energy and is difficult to be stripped away by the laser field. Chu and Chu [25] utilized an all-electron *ab initio* study of the ionization of diatomic molecules and showed that inner-valence electrons contribute to the molecular ionization in intense laser fields. They concluded that it is more difficult to ionize the inner-valence electron at lower laser intensity because of its high binding energy. However, the ionization probability for the inner-valence electron increases with increasing laser intensity. For the laser pulse with the pulse duration of tens or hundreds of femtoseconds, ionization of the highest occupied molecular orbit electron is dominant due to the long rising time. However, the laser electric field reaches its peak intensity very rapidly for the few-cycle laser pulse, ionization of an inner-valence electron or multielectron excitation becomes possible in such an ultrashort laser field. The laser–molecule interaction invokes vertical transitions of the neutral molecule to different excited electronic states of the parent ion, which dissociates into ionic fragments.

It should be pointed out that the KER of O^+ is higher than that of C^+ when the laser intensity is 8.0×10^{14} W/cm^2 according to the widths of these ionic peaks shown in Fig. 1(d). These results indicate that these two fragments come from the dissociation of CO^+ in different electronic states. This conclusion is consistent with the fact that the appearance intensity of O^+ is higher than that of C^+. Because O^+ and CO^{2+} emerge at nearly the same laser intensity as shown in Fig. 1(c), we conclude that the energy levels of the excited electronic states of CO^+, which will dissociate into O^+ and a neutral C atom, are as high as that of the ground electronic state of CO^{2+}.

When the laser intensity is further increased, ionization of many electrons leads to the production of highly charged parent ions. Due to Coulomb repulsive force, these highly charged parent ions explode into atomic ions with large kinetic energies. Fig. 2 shows the time-of-flight mass spectra of CO irradiated by linearly polarized 6 fs laser pulses at an intensity of 2×10^{15} W/cm^2. The laser polarization direction is (a) parallel and (b) perpendicular to the flight tube of the time-of-flight mass spectrometer. Highly charged atomic ions C^{p+} and O^{q+} are observed and (p, q) refers to the dissociation channel $CO^{(p+q)+} \rightarrow C^{p+}+O^{q+}$. These atomic ions exhibit different intensity distributions for laser pulses with parallel and perpendicular polarizations. The differences are attributed to the angular distributions of the atomic ions produced in the Coulomb explosion. When these ions are ejected in the direction parallel to the flight

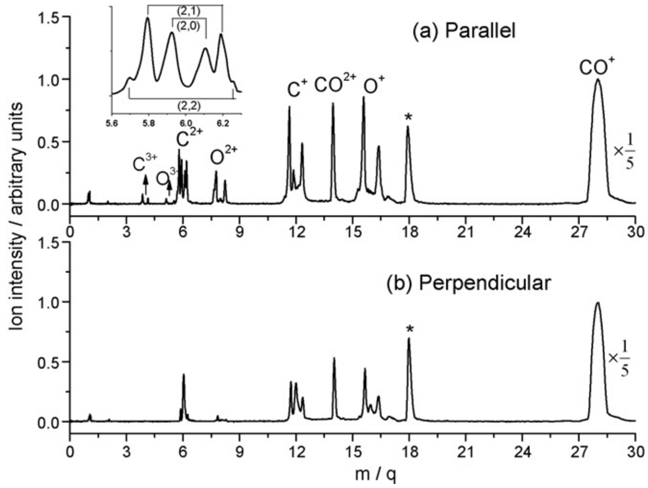

Fig. 2. Time-of-flight mass spectra of CO irradiated by 6 fs laser pulses at an intensity of 2×10^{15} W/cm^2. The laser polarization is (a) parallel and (b) perpendicular to the time-of-flight mass spectrometer axis. The mass spectra are normalized to the intensity of CO^+. The peak denoted with an asterisk originates from the residual water in the vacuum chamber.

tube, they produce two peaks in the mass spectrum corresponding to the ions with initial velocities toward and away from the detector. When these ions are ejected in the direction perpendicular to the flight tube, only the ions with low kinetic energies are collected by the detector. Comparing with the mass spectra recorded in the 110 fs laser field [26,27], each highly charged atomic ion has several peaks in the 6 fs laser field. In addition, strong peak is observed for the doubly charged parent ion CO^{2+}.

The atomic ion C^{2+} exhibits three pairs of peaks as shown in the inset of Fig. 2(a). By measuring the time differences of the forward and the backward peaks of the ion in the mass spectra, KERs of C^{2+} are determined to be 3.2, 15.5 and 30.0 eV for the inner, middle and outer pair double peaks, respectively. The C^{2+} ions with different KERs are produced through different channels. Lavancier et al. [13] reported the observation of C^{2+} ions with KER of 3.75 ± 0.40 eV. These ions are proposed to be generated by the charge asymmetric dissociation $CO^{2+} \rightarrow C^{2+} + O$ because of no observation of correlated atomic oxygen ions. Considering the experimental error, we suppose that the C^{2+} ion with KER of 3.2 eV is produced from the same CAD channel in the current experiment. The C^{2+} ion with KER of 15.5 and 30.0 eV are from CSD channels $CO^{3+} \rightarrow C^{2+} + O^{+}$ and $CO^{4+} \rightarrow C^{2+} + O^{2+}$, respectively. Comparing with previous reports in tens or hundreds of femtoseconds laser fields [26,27], the CAD channel (2, 0) is greatly enhanced and KERs of the atomic ions are greatly increased for the (2, 1) and (2, 2) channels under few-cycle laser fields.

CAD is a natural result of strong field excitation and ionization in the tunneling regime [19]. However, CAD channel is hardly observed in the near-infrared radiation with long pulse duration [26,27]. The strong CAD observed in the current experiment might result from the following reasons. First, enhanced ionization is greatly suppressed in the few-cycle laser field. It is well known that Coulomb explosion proceeds through three different processes: enhanced ionization, sequential ionization and rescattering double ionization [28]. Enhanced ionization is dominant for long laser pulses and CSD dominates the process of enhanced ionization [29]. Therefore, the CAD is hidden by the dominant CSD channel in the Coulomb explosion of molecules in long laser pulses. However, the internuclear distance is frozen near the neutral molecular equilibrium distance within the laser pulse duration for few-cycle laser pulses and enhanced ionization can be neglected. Secondly, post-dissociative ionization can not occur in few-cycle laser pulses. The final stage of ionization in post-dissociative ionization occurs at a separation well beyond the critical nuclear distance. Therefore, post-dissociative ionization modifies the nascent charge-asymmetry into the charge-symmetry in long laser pulses and results in the absence of CAD in long laser pulses experiments [30]. However, the laser pulse is over before the parent ion dissociates into atomic ions for few-cycle laser pulses. Post-dissociative ionization is cut off for few-cycle laser pulses. Thirdly, the internuclear distance is frozen near the equilibrium distance of the neutral molecule within the pulse duration for few-cycle laser pulses. Therefore, vertical transition dominates in the few-cycle laser field [31]. It is known that the molecular ionization rate is determined by the laser intensity and internuclear separation [32]. The rising time is very long for the laser pulse with the pulse duration of tens or hundreds of femtoseconds. The neutral molecule is ionized through the tunneling by the rising edge of the laser pulse. The internuclear distance of the parent ion becomes larger when the transient laser field reaches the intensity required by CAD channel. Under this condition, nonvertical ionization is dominant. The nonvertical ionization favors CSD [13,14,17,18] and results in the absence of CAD for the long laser pulse. However, for few-cycle femtosecond laser pulses, the laser field reaches its peak intensity very quickly and the internuclear distance is frozen within the laser pulse duration. Vertical ionization dominates in the few-cycle laser field, which leads to the strong CAD observed in the current experiment. This is consistent with previous reports that vertical ionization favors CAD drawn in short wavelength radiation [33].

From above discussions, we know that CAD is not easily observed in long laser pulse because of its long rising time. Many dynamic processes, such as enhanced ionization, post-dissociative ionization and stretch of the chemical bond, occur before the laser reaches its peak intensity. These dynamic processes favor CSD. However, the internuclear distance is frozen near the neutral molecular equilibrium distance within the pulse duration for few-cycle laser pulses. The above dynamic processes are greatly suppressed. Vertical ionization becomes dominant, which leads to the strong CAD observed in the 6 fs laser field.

Comparing with the mass spectra of CO in the 110 fs laser field, another striking difference is that strong CO^{2+} peak is observed in the current few-cycle laser field. The CO^{2+} has been studied using various experimental methods, such as photoionization with synchrotron radiation, electron-impact ionization, fast ion collision, etc. [34–38]. With the help of theoretical calculations, it is commonly accepted that CO^{2+} can exist stably in some metastable states [39–42]. These stable dications are produced via vertical transitions within the Franck–Condon region. They predissociate to the $C^{+} + O^{+}$ via the repulsive ${}^{3}\sum^{-}$ state at large internuclear distance. However, the study of CO^{2+} is scarce with near-infrared femtosecond laser pulse. The yield of CO^{2+} strongly depends on pulse durations and laser intensities. CO^{2+} is scantly seen in previous 110-fs laser pulses [27]. An abundance of CO^{2+} is observed by 55-fs laser pulse with the intensity in the non-sequential double ionization regime [43]. However, strong peak of CO^{2+} is observed in the current few-cycle laser field with the laser intensity beyond the non-sequential double ionization regime. According to field-assisted bond-stretching model [31], the internuclear distance of the singly charged parent ion depends on the laser pulse duration when the double ionization occurs. The rising time is very long for the 110-fs laser pulse, the internuclear distance of CO^{+} is stretched with the assistance of the laser field. The ionization rate of CO^{+} becomes larger with increasing the internuclear distance. Nonvertical ionization dominates under this condition. The ion CO^{2+} produced at large internuclear distance is unstable. The fast dissociation results in the low yield of the ion CO^{2+} in the 110-fs laser pulse. While for the 55-fs laser pulse, the change of the internuclear distance is very small for non-sequential double ionization process. Therefore, an abundance of CO^{2+} is observed by 55-fs laser pulse with the intensity in the non-sequential double ionization regime. While for the current few-cycle laser field, the internuclear distance is frozen within the laser pulse duration. Vertical ionization dominates and strong peak of CO^{2+} is observed for a large range of laser intensities.

The internuclear distance upon explosion can be obtained from the measurement of KERs of the exploding fragments. The middle pair double peaks of C^{2+} with KER of 15.5 eV are from $CO^{3+} \rightarrow C^{2+} + O^{+}$. The outer pair double peaks of C^{2+} with KER of 30.0 eV are from $CO^{4+} \rightarrow C^{2+} + O^{2+}$. Using a pure Coulomb explosion model, the KER of C^{p+} is $E(C^{p+}) = \frac{m_O}{m_C + m_O} \frac{pq}{R}$ for explosion pathway (p, q) occurring at R. The KERs of C^{2+} are 14.6 and 29.2 eV respectively for (2, 1) and (2, 2) channel when the explosion occurs at the neutral molecular equilibrium distance (1.128 Å). The experimental measured values are around those of the theoretical values. These results show that the explosion occurs around the neutral molecular equilibrium distance and supports the conclusion that vertical ionization dominates in the current few-cycle laser field. The small discrepancy of KERs between the experimental measured values and the theoretical predicted values might origin from the deviation between the real potential and the pure Coulomb potential. This deviation of the potential energy curve from a pure Coulomb potential has been reported for highly charged parent ions at short internuclear distances [44].

4. Conclusion

We experimentally study dynamic processes of CO in intense few-cycle laser fields. The laser electric field reaches its peak intensity very rapidly for such ultrashort laser pulses and the internuclear distance is frozen near the neutral molecular equilibrium distance within the pulse duration. The vertical ionization leads to the observations of the strong CAD (2, 0) channel and CO^{2+}. The experimental measured kinetic energies for the exploding fragments support the conclusion that vertical excitation and ionization dominate in the few-cycle laser field.

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