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Ionization Suppression of Heteronuclear Diatomic and Triatomic Molecules in Strong Infrared Laser Fields†

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Ionization is the fundamental process in interaction of atoms/molecules with femtosecond strong laser fields. Comparing to atoms, molecules exhibit peculiar behaviors in strong-field ionization because of their diverse geometric structures, molecular electronic orbitals as well as extra nuclear degrees of freedom. In this study, we investigate strong field single and double ionization of carbon monoxide (CO) and carbon dioxide (CO_2) in linearly polarized 50-fs, 800-nm laser fields with peak intensity in the range of 2×10^{13} W/cm^2 to 2×10^{14} W/cm^2 using time-of-flight mass spectrometer. By comparing the ionization yields with that of the companion atom krypton (Kr), which has similar ionization potential to the molecules, we investigate the effect of molecular electronic orbitals on the strong-field ionization. The results show that comparing to Kr, no significant suppression is observed in single ionization of both molecules and in non-sequential double ionization (NSDI) of CO, while the NSDI probability of CO_2 is strongly suppressed. Based on our results and previous studies on homonuclear diatomic molecules (N_2 and O_2), the mechanism of different suppression effect is discussed. It is indicated that the different structure of the highest occupied molecular orbitals of CO and CO_2 leads to distinct behaviors in two-center interference by the electronic wave-packet and angular distributions of the ionized electrons, resulting in different suppression effect in strong-field ionization.

Key words: Strong laser field, Molecule, Ionization suppression

I. INTRODUCTION

When atoms are irradiated by a strong laser field with peak intensity higher than 10^{13} W/cm^2 and pulse duration shorter than 100 fs, the electron will tunnel out through the barrier formed by the Columnb potential and the external laser field. A lot of atomic strong-field phenomena are found to be directly related to the tunneling electrons, such as high-order harmonic generation (HHG) [1], high-order above threshold ionization (HATI) [2], non-sequential double ionization (NSDI) [3], and high Rydberg state excitation (RSE) [4–6]. Comparing to atoms, for which the strong-field ionization and related phenomena have been well studied, molecules exhibit a large variety of peculiar behaviors in strong field ionization, due to their structural complexity and extra nuclear degrees of freedom. During the past decades, strong field ionization of molecules have attracted increasing experimental and theoretical interests, practically focusing on the effect of molecular structure [7], inner orbitals and nuclear motion [8, 9] on the ionization probability and related processes. Moreover, understanding the strong-field molecular ionization process is also of vital importance in application of tomographic imaging of molecular orbitals [10], molecular coherent control [11], and attosecond pulse generation [12–15].

According to strong-field theories that can well describe the tunneling ionization of atoms (*e.g.*, Ammosov-Delone-Krainov model (ADK model)), ionization probability of an atom subjected to a specific strong laser field is determined solely by its ionization potential energy (I_p). It has been long realized that homonuclear diatomic molecules (N_2 and O_2) exhibit quite distinct strong-field ionization probabilities comparing to their company atoms (*i.e.*, the atom with similar I_p to the molecule). A strong suppression is observed in the ionization probability of O_2 compared with its companion atom Xe in 800-nm laser field, while no suppression is found in N_2 molecule compared with its companion atom Ar [16]. Various theoretical methods, including Keldysh-Faisal-Reiss (KFR) [7], multi-electron screening [17], or MO-ADK (molecular version of ADK) models [18], have been proposed to address this phenomenon, and the underlying physics has remained unclear and been under debate for years. Very

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recently, Lin *et al.* performed a joint theoretical and experimental study on strong-field ionization of N_2 and O_2 as well as their companion atoms in both 800-nm and 2000-nm strong laser fields [19]. The results strongly indicate that it is the two-center interference along with different molecular orbitals of N_2 and O_2 that accounts for their different ionization suppression comparing with atoms. This mechanism also explains the absence of resonancelike enhancement (RLE) structure in HATI of O_2, which can be attributed to the constructive interference of long quantum orbits [20]. Regarding other strong physical processes which are related to the strong field ionization, previous studies have observed suppressed probability of NSDI [16] or RSE [21] of O_2 comparing to Xe, while N_2 presents similar behavior to the companion atom Ar. It is indicated that the π_g symmetry of the highest occupied molecular orbital (HOMO) of O_2 leads to a deviation of electron emission from the laser-field direction and a wider angular distribution of the tunneled electrons, resulting in a smaller probability for the electron to re-collide with ionic core to ionize another electron (for NSDI) [22, 23] or to be captured into the Rydberg state (for RSE) [21].

Little information is known about the suppressed ionization of heteronuclear diatomic molecules and polyatomic molecules in strong laser fields. Here, we investigate single and double ionization of CO and CO_2 in linearly polarized 50-fs, 800-nm laser fields with peak intensity in the range of 2×10^{13} W/cm^2 to 2×10^{14} W/cm^2 using a time-of-flight mass spectrometer. The results are compared with those of their companion atom Kr, which has similar ionization potential as the molecules, and also with the studies on N_2 and O_2 molecules reported in the literatures. The effect of molecular structure on strong field ionization is discussed based on the experimental results.

II. EXPERIMENTS

The experiment setup used for interaction of atoms and molecules with strong laser fields was similar to that described in our previous studies [24, 25]. Briefly, a gaseous sample was introduced to the reaction zone through a leak valve with an aperture of 10 μm. The base pressure of the interaction chamber was 1×10^{-7} Pa, and the operating pressure was about 3×10^{-6} Pa. A Ti:sapphire chirped-pulse amplified laser system with 1 kHz repetition rate, a maximum pulse energy of 4 mJ, a pulse duration of 50-fs and a center wavelength of 800-nm was used in the study. A half-wave plate and a Glan prism were inserted into the laser beam to adjust the laser intensity continuously. The laser polarization was controlled by rotating a quarter-wave plate before it was focused into the reaction zone by a thin lens with a focal length of 25 cm. The laser intensity was calibrated by comparing the measured saturation intensity of single ionization of Kr with that of the ADK model [26]. A linear time-of-flight (TOF) mass spectrometer is used to detect the produced cations from strong-field ionization. The distance from the laser-molecule/atom interaction zone to a dual microchannel plates (MCPs) detector is 500-mm. Mass resolved ion signals are recorded using a data acquisition card (National instruments, PXIe-5162) and are sent to a PC for analysis. All experimental data are normally averaged over 10^4 laser shots.

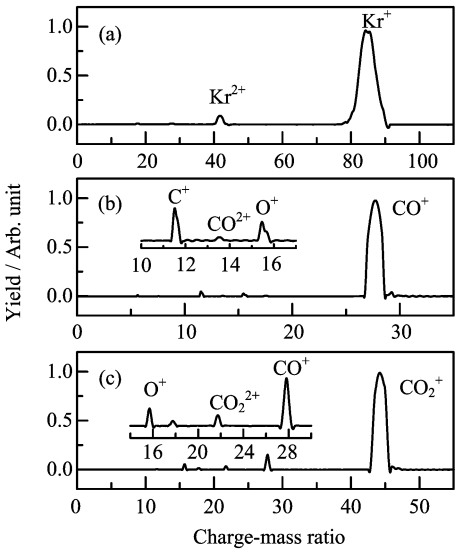

FIG. 1 Time-of-flight mass spectra of (a) Kr, (b) CO, and (c) CO_2 at 1×10^{14} W/cm^2.

III. RESULTS AND DISCUSSION

We have measured and analyzed the TOF mass spectra of Kr, CO and CO_2 irradiated by 50-fs 800-nm laser fields with intensity range of 2×10^{13} W/cm^2 to 2×10^{14} W/cm^2. In FIG. 1 we present typical TOF mass spectra of the atom and molecules recorded by linearly polarized laser fields at intensity of 1×10^{14} W/cm^2. While singly-charged parent ions are dominant in all the mass spectra, doubly-charged parent ions become visible as laser intensity larger than 5×10^{13} W/cm^2 (see the enlarged plots in FIG. 1). For the molecules, fragment ions produced by dissociative ionization can be observed, however their yields can be negligible comparing to single ionization. This is unlike the observation of CO_2 at laser intensity higher than 4×10^{14} W/cm^2, which presents significant fragmentation from dissociative ionization and Coulomb explosion of multiple-charged ions [25].

A. Strong field single ionization

We now compare the strong field ionization of molecules CO and CO_2 with that of the atom Kr.

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TABLE I The ionization potential, electronic configuration and HOMO for Kr, CO and CO_2.

	I_p/eV	Electronic configuration	HOMO
Kr	13.999	$(1s^2)(2s^2)(2p^6)(3s^2)(3p^6)(3d^{10})(4s^2)(4p^6)$	4p
CO	14.014	$(1\sigma)^2(2\sigma)^2(3\sigma)^2(4\sigma)^2(1\pi)^4(5\sigma)^2$	σ_g
CO_2	13.777	$(1\sigma_g)^2(1\sigma_g)^2(2\sigma_g)^2(3\sigma_g)^2(4\sigma_g)^2(3\sigma_u)^2(1\pi_u)^4(1\pi_g)^4$	π_g

Table I lists the I_p values, the electronic configurations and the symmetry of HOMO of the three species. Both I_ps of CO and CO_2 are close to Kr, thus a comparison investigation of the molecules to their companion atom could reveal the effect of molecular structure in strong field ionization, as previous studies on N_2 *vs.* Ar and O_2 *vs.* Xe [19]. As shown in FIG. 2(a), we note that the HOMO of CO is a bonding σ_g orbital, which is the same as that of N_2, while both HOMOs of CO_2 and O_2 have antibonding π_g symmetry. Thus we will also compare and discuss the present results with those for N_2 and O_2 in the literature to further understand the structure effect in strong-field molecular physics.

In FIG. 2(a), we show the yield of single ionization as a function of laser intensity in the range of 2×10^{13} W/cm^2 to 2×10^{14} W/cm^2, for Kr and molecules CO and CO_2. In the present study the laser field is linearly polarized along the TOF axis unless otherwise mentioned. As shown in the FIG. 2, both molecules show comparable single ionization probabilities with the companion atom Kr at all the laser intensities used in the experiment. In order to compare more clearly the behavior of the molecular strong-field ionization with that of the companion atom, we plot the ratios of yields, CO^+/Kr^+ and $CO_2{}^+/Kr^+$ in FIG. 2(b). The ratios of both CO^+/Kr^+ and $CO_2{}^+/Kr^+$ are close to 1, indicating that both molecules exhibit atom-like single ionization in strong laser fields. A close inspect shows that the ratio $CO_2{}^+/Kr^+$ is slightly smaller than CO^+/Kr^+ particularly in the laser intensities lower than 1×10^{14} W/cm^2, and a distinct dip structure appears for the curves of both ratios in the present study, comparing to the smooth curve in $N_2{}^+/Ar^+$ reported in Refs.[7, 19].

The wavelength and intensity dependence of peculiar suppressed ionization behaviors of homonuclear diatomic molecules N_2 and O_2 has been explained by two-center interference, *i.e.*, interference between ionizing wave packets emitted from the two ionic cores [19, 27]. The interference is induced by the trigonometric part, $\sin(\mathbf{k_N}\cdot\mathbf{R}/2)$ (for atomic p orbitals) or $\cos(\mathbf{k_N}\cdot\mathbf{R}/2)$ (for atomic s orbitals) inherently included in the molecular wavefunctions used in the S-matrix calculations. Here $\mathbf{k_N}$ and $\mathbf{R}$ represent the momentum of the emitted electron and internuclear distance of the diatomic molecule. Due to the fact that the vast majority of ionized electrons have small momenta, $\mathbf{k_N}\cdot\mathbf{R}/2 \ll \pi$, the trigonometric part results in constructive interference for s orbitals or destructive interference for p orbitals, leading to enhanced or suppressed ionization probability.

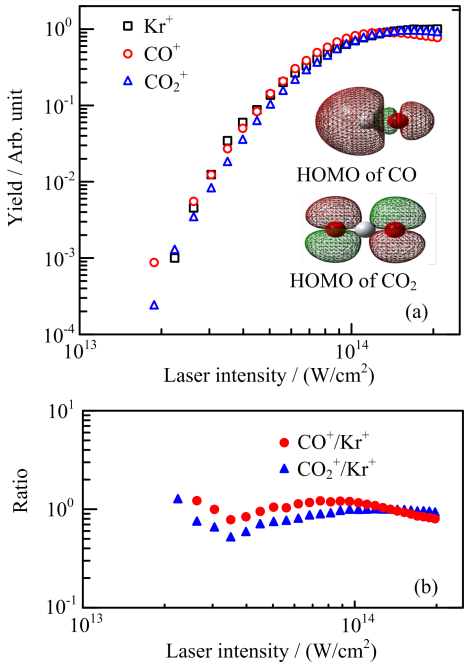

FIG. 2 (a) The single ionization yields of Kr (black hollow square), CO (red hollow circle) and CO_2 (blue hollow triangle) as a function of laser intensity. The HOMOs of the molecules are also shown. (b) The ratio between molecule and atom are shown for CO (red solid circle) and CO_2 (blue solid triangle) at different laser intensities.

According to this mechanism, it is not a surprise that CO presents nearly identical ionization probability with its companion atom Kr. Similar to N_2, the HOMO of CO is a bonding σ_g orbital, which is composed by s orbital of C atom (40%) and p orbital of O atom (60%). The admixture of both atomic s and p orbitals leads to the effect of constructive and destructive interference to be added coherently, indicating no significant suppression in the ionization as shown in FIG. 1. Additionally, the component of atomic p orbital in CO HOMO is higher than that in N_2 HOMO, thus the ratio of CO^+/Kr^+ in FIG. 2(b) is slightly smaller than $N_2{}^+/Ar^+$ which is almost constant around 1 as reported in the previous studies [7, 19].

CO_2 shows much less suppressed ionization *vs.* its companion atom Kr than O_2 *vs.* Xe, despite that both HOMOs are antibonding π_g symmetry. The ratio $O_2{}^+/Xe^+$, which increases as the laser intensity is increased [19, 21], is about one magnitude order lower than $CO_2{}^+/Kr^+$ (FIG. 2(b)) at 5×10^{13} W/cm^2. This

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phenomenon could be understood concerning the following two issues. Firstly, the electron density of CO_2 HOMO is mainly localized at two oxygen atoms, indicating the interference could occur between ionizing wavepackets emitted from the two oxygen ionic cores. The **R** value can be assumed to be the distance between oxygen atoms in CO_2 which is 2.33 Å and is about 3.6 times as large as the internuclear distance of O_2. Therefore it is expected that the destructive interference induced by the $\sin(\mathbf{k_N}\cdot\mathbf{R}/2)$ term of the atomic p orbitals in CO_2 would be less pronounced than that in O_2. Secondly, the HOMO-1 and HOMO-2 orbital of CO_2 also contributes to the strong field ionization [28], which has π_u and σ_u symmetry respectively. The contribution from lower-lying orbitals would smear out the destructive interference in strong field ionization of CO_2, resulting in a less suppression comparing to its companion atom.

Nevertheless, the behavior of strong-field single ionization of CO and CO_2 can be qualitatively addressed with the two-center interference effect, which is attributed to the different symmetries of the molecular electronic orbitals. The obvious dip structure in both CO^+/Kr^+ and $CO_2{}^+/Kr^+$ could result from the interference between different quantum orbits of the tunneled electrons, which is also indicated in the RSE process of diatomic molecules [21] and will stimulate further quantum theoretical studies to reveal the underlying physics of the phenomenon.

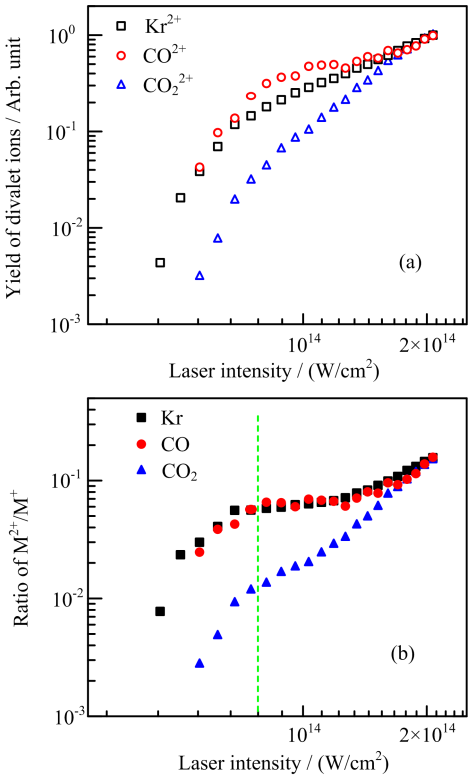

FIG. 3 (a) The double ionization yields of Kr (black hollow square), CO (red hollow circle) and CO_2 (blue hollow triangle) as a function of laser intensity. All the three curves are normalized to its maximum respectively. (b) The laser intensity dependence of M^{2+}/M^+ for Kr (black solid square), CO (red solid circle) and CO_2 (blue solid triangle). The green line indicates the "knee" structure at 8×10^{13} W/cm^2.

B. Strong field double ionization

We observed double ionization (DI) of both molecules within the laser intensities used in the experiments. FIG. 3(a) presents the DI yield as a function of laser intensity, for Kr, CO and CO_2. Each curve is normalized to its maximum respectively. A clear "knee" structure, *i.e.*, weak dependence of DI yield on the laser intensity, can be identified in each curve, which becomes more obvious if the ratio of M^{2+}/M^+ (M=Kr, CO, or CO_2) is plotted as a function of laser intensity (see FIG. 3(b)). The appearance of the "knee" structure of CO or CO_2 occurs at roughly the same intensity as that of Kr atom at $\sim8\times10^{13}$ W/cm^2 (indicated by the green line), which could attribute to their similar I_p values.

The observed "knee" structure in the curve of DI yield *vs.* laser intensity is a signature of non-sequential double ionization (NSDI) process in strong laser fields. The mechanism of atomic NSDI is well understood in the frame of so-called "three-step recollision" scenario [29]: an electron in atom first tunnels out, then is accelerated in the laser field, and finally upon reversal of the directional vector of the laser field, it can re-collide with the parent ion to release another electron in the atom. According to this mechanism, the NSDI yield would strongly depend on the laser polarization, since recollision with the parent ions diminishes due to the greater drift momentum spread of the returning electron wavepacket as laser ellipticity is increasing. In FIG. 4, we show the elliptical-dependent DI yield with laser intensity in typical NSDI region (1×10^{14} W/cm^2) and higher intensity where sequential DI dominates (2×10^{14} W/cm^2) for both atom Kr (FIG. 4(a)) and molecules CO (FIG. 4(b)) and CO_2 (FIG. 4(c)). For each plot, the experimental data at different laser ellipticities were normalized to that at an ellipticity of 0, *i.e.*, linear polarization. The solid curves show the best-fit Gaussian distributions to the experimental data. As shown in FIG. 4(a), the DI yield of Kr at 1×10^{14} W/cm^2 is maximal at linear polarization and decreases rapidly with increasing ellipticity of the polarization. This is a general feature of the NSDI of atoms as expected by the "three-step recollision" scenario. For sequential DI, however, only slightly decreases as the laser ellipticity is increased, which may be due to the fact that the laser peak field decreases with the laser ellipticity. It is interesting to see that the molecules exhibit similar dependence on the laser ellipticity as the companion atom (FIG. 4 (b) and (c)). The results demonstrate that NSDI, which is induced by laser-driven electron

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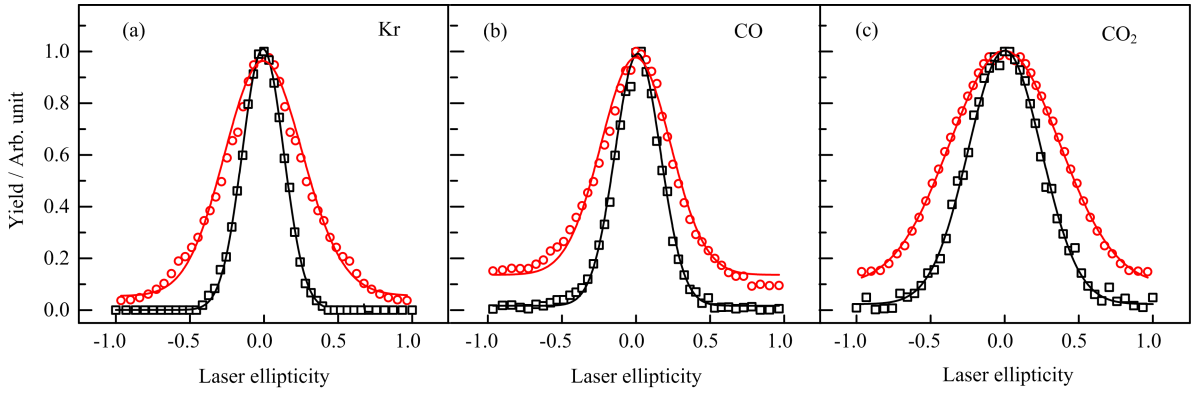

FIG. 4 The yield of double ionization as a function of laser ellipticity for (a) Kr, (b) CO, and (c) CO_2 at 1×10^{14} W/cm^2 (black square) and 2×10^{14} W/cm^2 (red circle). The solid curves show the best-fit Gaussian distributions to the experimental data.

recollision, plays a significant role in strong-field double ionization of both CO and CO_2 molecules.

It should be mentioned that it is unlikely to directly compare the DI yields of CO and CO_2 with that of the companion atom Kr because we couldn't exclude dissociation from the doubly-charged molecular parent ions. On the other hand, as discussed in the previous section, both molecules show comparable strong-field single ionization yield to the companion atom Kr. In addition, we measured the DI yields of Kr and molecules in 400-nm strong laser fields, in which the NSDI process is strongly prohibited and the fragmentation is negligible. The results further demonstrate that the sequential DI yields of both CO and CO_2 are nearly identical to that of Kr. Thus, by normalizing the curve of each measured DI yield *vs.* laser intensity to its maximum respectively, we assume little suppression in sequential DI of the molecules and we can compare the NSDI probability of each molecule to that of the companion atom Kr (see FIG. 3).

As shown in FIG. 3(b), the ratio of CO^{2+}/CO^{+} is comparable to Kr^{2+}/Kr^{+} in the whole laser intensity range used in the experiments, while the ratio of $CO_2{}^{2+}/CO_2{}^{+}$ is about one magnitude smaller than the ratio of CO^{2+}/CO^{+} or Kr^{2+}/Kr^{+} at 6×10^{13} W/cm^2 and gradually increases as the laser intensity is increased. This indicates no suppression in NSDI of CO comparing to that of the companion atom Kr, while the suppression is much more significant in NSDI of CO_2. Since NSDI is produced by re-collision with the ionic core by the tunneled electrons, its probability should be determined by the rates of both tunneling ionization and re-collision. As we have discussed above, the difference of the strong field single ionization is not so significant between the molecules and the companion atom Kr. However, the angular distribution of the tunneled electrons would be quite different because of the different HOMO structures of CO and CO_2. For CO, the bonding σ_g HOMO indicates the electron in the molecule would prefer to emit along the laser polarization direction, which is similar as the fact in N_2 which acts as a structureless atom. On the other hand, the HOMO of CO_2 is π_g symmetry, which means the electron would emit mainly at angles that deviate from the laser direction. This fact is similar to O_2 and has been demonstrated by the alignment-dependent ionization of CO_2 and O_2 [30]. Furthermore, it's worth to note that the electrons ionized from the low-lying orbits of CO_2 not only contribute to the single ionization, but also to the strong field NSDI. For HOMO-1, which has a π_u symmetry, the ionized electrons would also deviate from the laser direction and reduce the probability of re-collision. And for HOMO-2, although it has a σ_u symmetry, we believe that the contribution of HOMO-2 to the NSDI is negligible. As a result, it would be expected that more diffused wave packet as the electron returns to the ionic core for CO_2, leading to reduced probability of re-collision to induce NSDI.

The suppression of NSDI was also observed in O_2 [16], and was later explained by Jia *et al.* by the S-matrix theoretical study [22, 23] which demonstrated the effect of molecular orbital on the peculiar NSDI behaviors. For RSE process in which the tunneled electrons are even not required to return to the ionic core, it was also proved that for a wider angular distribution of the electron ionized from O_2, the electrons will be harder to be captured in high Rydberg states, leading to a relatively smaller RSE population [21]. This particular photoelectron angular distribution has also been shown to be responsible for relatively wider differential angular distribution at a cutoff energy in the HATI of O_2 compared with atoms and the N_2 molecule [31]. The present study further shows that the laser-induced electron re-collision as well as distinct angular distributions of ionized electrons from different HOMOs would play an important role in fully understanding the interaction of complexed molecules with strong laser fields.

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IV. CONCLUSION

In summary, we have performed a comparative study on strong-field ionization of molecules CO and CO_2 with their companion atom Kr. No significant suppression in strong-field single ionization has been observed for both molecules comparing to that of Kr, which could be qualitatively addressed with the two-center interference effect, *i.e.*, interference between ionizing wave packets emitted from the two ionic cores. For NSDI, different behaviors for CO and CO_2 have been identified. The NSDI of CO is comparable with that of Kr, while for CO_2, significant suppression has been observed comparing to its companion atom. This could be understood by different angular distributions of tunneled electrons which are attributed to the different symmetries of HOMOs, in analogy to the cases in NSDI, HATI and RSE processes of N_2 and O_2. Our study should shed light on the effect of molecular structure on ionization and related processes for more complex molecules in strong laser fields.

V. ACKNOWLEDGEMENTS

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