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Polarization and molecular-orbital dependence of strong-field enhanced ionization

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In this work we perform a polarization dependence study of enhanced ionization (EI) in diatomic molecules. We find that EI exists when the field polarization is parallel to the molecular axis but disappears when polarization is perpendicular. We further study EI with circular polarization and find that EI exists with circular polarization indicating that rescattering does not play a significant role for EI. Furthermore, we study molecular orbital effect on EI. We find that EI exists in σ type but not π type outmost molecular orbitals.

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I. INTRODUCTION

When molecules interact with an intense laser field, ionization can happen and usually is accompanied by dissociation. As the internuclear distance between the atomic cores is driven by the laser field to beyond the equilibrium internuclear distance, the ionization rate can increase significantly and reach a peak at a critical internuclear distance R_c. This effect is known as enhanced ionization (EI) [1,2]. Figure 1 illustrates how EI works for a diatomic molecule. A double-well potential formed by the two nuclei of the diatomic molecule is distorted by the external laser field. As the internuclear distance increases, the inner potential barrier rises and leads to two dressed states, $\sigma_+ = \sigma_g + \sigma_u$ and $\sigma_- = \sigma_g - \sigma_u$ [1], as shown in Fig. 1(a). At R_c there is a significant probability that the electron is trapped in the upper potential well, and this phenomenon is called electron localization [2]. When this happens, the tunneling ionization rate of the electron from the upper potential well (the σ_+ state) increases significantly, leading to EI, because the electron only needs to tunnel through the inner potential barrier that is much smaller than the outer one, as illustrated by paths A and B in Fig. 1(b), respectively [1,2]. When the internuclear distance extends beyond R_c [Fig. 1(c)], the ionization rate drops to be atomiclike.

In the past two decades, EI has been actively studied and significant advancements have been made [1–18]. EI has been observed in a number of molecules as well as clusters [7,12,14–18]. Recently we directly observed EI in two commonly studied molecules, N_2 and CO [18]. Our results show that, for double ionization induced dissociation, EI mostly exists in a slow dissociation channel with a lower kinetic energy release (KER) but not in the commonly seen dissociation channel with a higher KER for both N_2 and CO [18]. The successful identification of the EI channels in these molecules further improves our understanding of EI [6,18–23].

However, systematic experimental studies of a number of fundamental properties of EI have been lacking. Although theoretical exploration of EI has been greatly extended and recent advancements have shown interesting effects of EI such as molecular orbital type dependence of EI [9], experimental investigation on these studies is needed. Furthermore, so far no experimental studies have been performed on the polarization dependence of EI.

In this work we perform a polarization dependence study of EI in diatomic molecules. We find that EI exists when the field polarization is parallel to the molecular axis but disappears when polarization is perpendicular. We further study EI with circular polarization and find that EI exists in circular polarization indicating that rescattering does not play a significant role for EI. In addition, we study molecular orbital effect on EI. We find that EI exists in σ type but not π type outmost molecular orbitals.

II. EXPERIMENTAL SETUP

Experimentally, ion detection and collection is realized with a recently modified TOF spectrometer, which consists of a 3.1-cm-long extraction, a 1.0-cm-long acceleration, and a 48.1-cm-long drift region [18,24]. Ions are detected with a microchannel plate as a function of the flight time. The signal is further amplified, discriminated, and sent to a boxcar or a multihit time digitizer to produce ion yields and TOF mass spectra. The chamber base pressure is less than 5.0×10^{-10} Torr. The laser used is a Ti:sapphire system that delivers pulses of 1.0–1.2 mJ/pulse at a 1 kHz repetition rate with the central wavelength at 800 nm. By adjusting the bandwidth of the seed pulses and dispersion in the amplifier cavity, we manage to generate two pulse durations at 45 and 68 fs. Both pulse durations are virtually dispersion free with both their second- and third-order dispersions compensated. For the time-resolved pump-probe experiments in this study, a Mach-Zehnder arrangement is used with two beams propagating collinearly into the chamber. The delay stage has a 0.1-μm-resolution that gives a 0.67-fs temporal step.

III. RESULTS AND DISCUSSION

First, we study polarization dependence of EI. Figure 2(a) shows the TOF spectra of N^+ ions from N_2 obtained with linearly polarized 68-fs pulses with field polarization parallel to the TOF axis. The voltage plates for extracting and accelerating ions each has a 2.5-mm pinhole opening to allow only ions having their velocity aligned with the TOF axis to pass through, which guarantees that the field polarization is parallel to the molecular axis when the field is parallel to the TOF axis. In Fig. 2(a) we clearly see two pairs of $N^+ + N^+$ channels, a fast channel with KER of 7.0 eV, and a slow channel with KER of 3.8 eV, labeled as $N(1,1)_{\text{fast}}$ and

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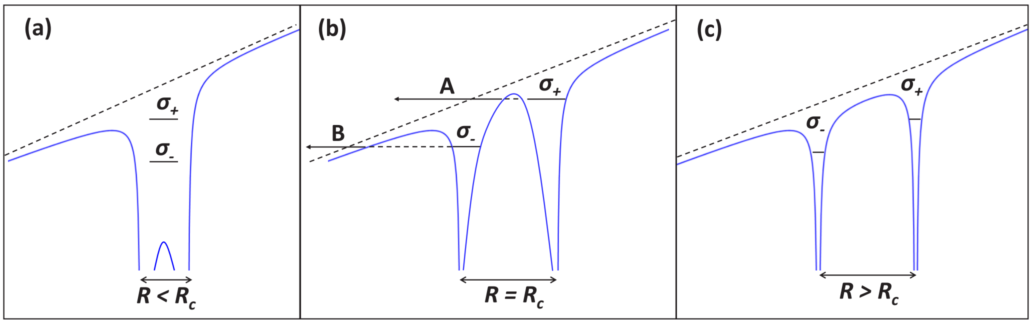

FIG. 1. The double-well potential of a diatomic molecule distorted by the external laser field at three different internuclear distances: (a) $R < R_c$, (b) $R = R_c$, and (c) $R > R_c$. In (a), the electron is in a superposition of two dressed states, $\sigma_+ = \sigma_g + \sigma_u$ and $\sigma_- = \sigma_g - \sigma_u$ [1]. In (b), paths A and B indicate tunneling ionization through the inner and outer potential barrier, respectively.

$N(1,1)_{slow}$. As discussed in Ref. [18], $N(1,1)_{slow}$ is formed through EI via an intermediate state $N^+ + N$, while $N(1,1)_{fast}$ is formed through nonsequential double ionization (NSDI). Figure 3 illustrates the pathways of the two channels. Due to a large number of electronic states that could be involved in forming the $N^+ + N$ and $N^+ + N^+$ dissociation channels and the exact information is currently unavailable, here we only plot selected states that lead to dissociation limits with similar KER as our $(N^+ + N^+)_{fast}$ (7.0 eV) ($X\,^1\Sigma^+{}_g$, $A\,^3\Sigma^-{}_g$, and $a\,^3\Pi_u$) and $N^+ + N$ (0.6 eV) ($C\,^2\Sigma^+{}_u$ and $D\,^2\Pi_g$) [24–26]. The potential energy curves in Fig. 3 are taken from Refs. [25,26]. We also plot two schematic curves (marked by "schematic") to represent the dissociation states that lead to $N^+ + N$ and $N^+ + N^+$ for the purpose of illustration. As marked by respective arrows, $(N^+ + N^+)_{fast}$ is formed through NSDI from neutral molecule N_2 and $(N^+ + N^+)_{slow}$ is from EI via an intermediate state $N^+ + N$ [18]. Note, similar labels will be used throughout this paper, e.g., N(1,0) represents N^+ from the $N^+ + N$ channel and $(N^+ + N^+)_{slow}$ represents the entire slow $N^+ + N^+$ channel. We also plot the TOF spectra of N^+ ions obtained with the same polarization condition but with shorter 45-fs pulses in Fig. 2(b). Compare Figs. 2(a) and 2(b), $N(1,1)_{slow}$ is clearly visible in the 68-fs spectrum but unclear in the 45-fs spectrum, showing that the EI channel will be significantly reduced if the pulse duration is not sufficient to allow the molecular ion N_2^+ to reach R_c that takes about 43 fs

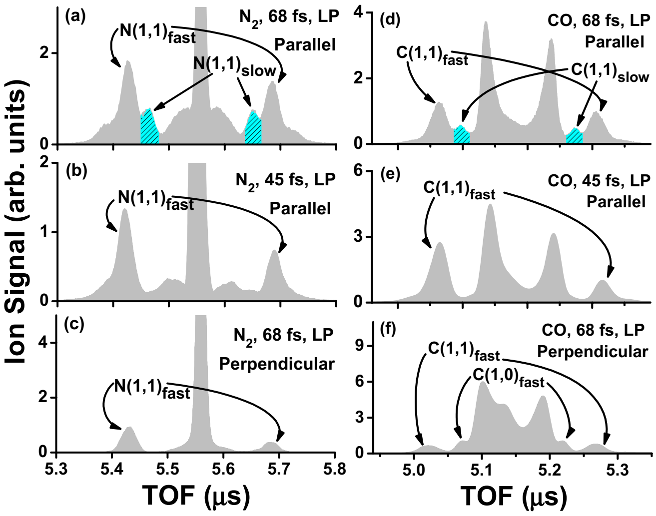

FIG. 2. (a) and (b) TOF spectra of N^+ from N_2 obtained with linearly polarized (a) 68-fs and (b) 45-fs pulses with field polarization parallel to the molecular axis. (c) TOF spectra of N^+ from N_2 obtained with linearly polarized 68-fs pulses with field polarization perpendicular to the molecular axis. (d) and (e) TOF spectra of C^+ from CO obtained with linearly polarized (d) 68-fs and (e) 45-fs pulses with field polarization parallel to the molecular axis. (f) TOF spectra of C^+ from CO obtained with linearly polarized 68-fs pulses with field polarization perpendicular to the molecular axis.

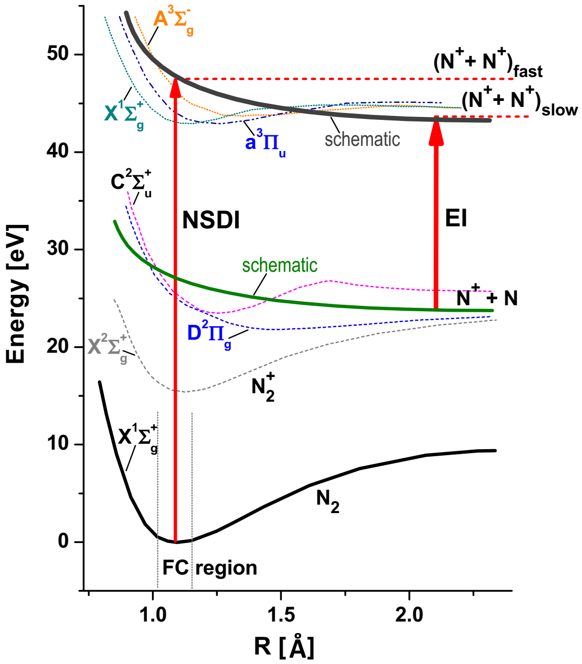

FIG. 3. Illustration of the pathways for $(N^+ + N^+)_{fast}$ and $(N^+ + N^+)_{slow}$. The potential energy curves are taken from Refs. [25,26]. Due to a large number of dissociation states leading to $N^+ + N^+$ and $N^+ + N$, only selected states that lead to similar KER as our $(N^+ + N^+)_{fast}$ (7.0 eV) and $N^+ + N$ (0.6 eV) are plotted here. We also plot two schematic curves (marked by "schematic") to represent the dissociation states that lead to $N^+ + N$ and $N^+ + N^+$ for the purpose of illustration. As marked by respective arrows, $(N^+ + N^+)_{fast}$ is formed through NSDI from neutral molecule N_2 and $(N^+ + N^+)_{slow}$ is from EI via an intermedia state $N^+ + N$.

[1,6,8,18]. This is again consistent with previously reported in Ref. [18].

To contrast the EI efficiency in different pulse durations and polarizations, we reproduce from Ref. [18] the TOF spectra of C^+ ions of CO in linearly polarized 68- and 45-fs pulses with field polarization parallel to the molecular axis, as shown in Figs. 2(d) and 2(e), respectively. Two $C^+ + O^+$ channels are clearly seen in Fig. 2(d), $C(1,1)_{\text{fast}}$ and $C(1,1)_{\text{slow}}$. As discussed in Ref. [18], $C(1,1)_{\text{slow}}$ is formed through EI but $C(1,1)_{\text{fast}}$ is not. Compare Figs. 2(d) and 2(e), $C(1,1)_{\text{slow}}$ is clearly visible in 68-fs pulses but again not clear in 45-fs pulses, since it takes about 46 fs for the molecular ion CO^+ to reach R_c [18]. Therefore, the results of N_2 and CO in Fig. 2 show that EI exists when the field polarization is parallel to the molecular axis and the pulse duration is sufficiently long.

When we change the field polarization to be perpendicular to the molecular axis by rotating the beam polarization by 90 deg, the EI-induced $(N^+ + N^+)_{\text{slow}}$ channel disappears. The results are shown in Fig. 2(c) with linearly polarized 68-fs pulses with field polarization perpendicular to the molecular axis. No clear signal of $(N^+ + N^+)_{\text{slow}}$ is seen on the TOF spectrum in Fig. 2(c). We have tested for a series of different intensities but did not observe any signal from $(N^+ + N^+)_{\text{slow}}$. Not surprisingly, $(N^+ + N^+)_{\text{slow}}$ is also missing in linearly polarized 45-fs pulses in the perpendicular geometry.

According to the EI theory, one key factor to allow EI to occur is the distorted double-well potential of the molecule from which the electrons in the upper potential well tunnel out at R_c leading to EI [1,2]. When the field polarization is perpendicular to the molecular axis, however, only atomlike single well potential is seen by the field, and, therefore, no EI will take place. This observation is verified again by repeated experiments with CO using linear 68-fs pulses in the perpendicular geometry. The results are shown in Fig. 2(f), where no clear signal of $(C^+ + O^+)_{\text{slow}}$ is seen. Note, the two small peaks adjacent to the two prominent C(1,0) peaks are the $C(1,0)_{\text{fast}}$ channel as identified in Refs. [27,28]. Therefore, our experiments show that EI most likely is turned off when the field polarization is perpendicular to the molecular axis.

Next, we switch to circular polarization. Figures 4(a) and 4(b) show the TOF spectra of C^+ ion fragments of CO obtained with circularly polarized 68- and 45-fs pulses, respectively. We can see that the $(C^+ + O^+)_{\text{slow}}$ channel is clearly seen in Fig. 4(a) with circularly polarized 68-fs pulses, consistent with Fig. 2(d). In contrast, $(C^+ + O^+)_{\text{slow}}$ is not clear in circularly polarized 45-fs pulses, similar to the results in Fig. 2(e) where linear 45-fs pulses are used and this is because the 45-fs pulse does not provide enough time for the molecular ion to reach R_c for EI to happen [1,6,8]. Therefore, our results show that EI effectively exists in circularly polarized light when the pulse duration is sufficiently long.

We also perform a time-resolved pump-probe study on CO using circularly polarized 45-fs pulses. The results are shown in Fig. 5 with pump-probe TOF spectra of C^+ and O^+ ion fragments at different time delays, for example, 36 and 53 fs are shown here. We can see clear signal enhancements in both the C^+ and O^+ peaks that correspond to $(C^+ + O^+)_{\text{slow}}$ in the 53-fs spectrum compared to the spectrum at a shorter time delay of 36 fs. These time-dependent signal enhancements in $(C^+ + O^+)_{\text{slow}}$ is consistent with our previously reported pump-probe experiments using linearly polarized pulses [18]. Note, a clear depletion is seen in the $O(1,0)_{\text{slow}}$ channel in Fig. 5 along with the enhancements in $(C^+ + O^+)_{\text{slow}}$ and this indicates $O(1,0)_{\text{slow}}$ as the precursory state that forms $(C^+ + O^+)_{\text{slow}}$, as discussed in details previously [18].

Similarly, we repeat the circular polarization experiments for N_2 and observe that the EI-induced $(N^+ + N^+)_{\text{slow}}$ is clearly seen in circularly polarized 68-fs pulses but missing in the circularly polarized 45-fs pulse, as shown in Figs. 4(c) and 4(d). Therefore, both results of CO and N_2 in our circular polarization experiments show that EI can efficiently occur in circular polarization. The fact that EI consistently occurs in circularly polarized light also indicates that rescattering does not play a significant role in EI [29,30].

Lastly, we study molecular orbital effect on EI. Previous theoretical studies of EI showed that EI-induced ionization rate maximizing at R_c, i.e., a R_c-dependent EI peak, only occurs in molecules with σ-type highest occupied molecular orbitals (HOMO) [9]. This is because σ-type electrons are concentrated along the internuclear axis and experiencing, taking diatomic molecules for example, a double-well potential, which allows electron localization and the strong coupling between the charge resonance states and the laser field to happen at R_c [9]. For molecules with other types

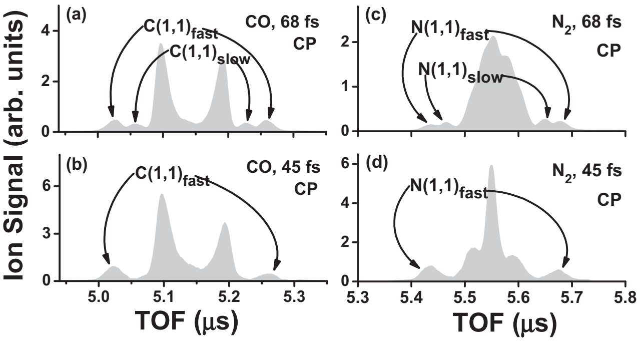

FIG. 4. (a) and (b) TOF spectra of C^+ from CO obtained with circularly polarized (a) 68-fs and (b) 45-fs pulses. (c) and (d) TOF spectra of N^+ from N_2 obtained with circularly polarized (c) 68-fs and (d) 45-fs pulses.

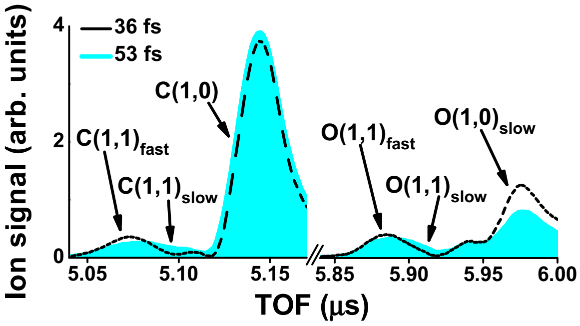

FIG. 5. TOF spectra of C^+ and O^+ ion fragments in the pump-probe experiment with circularly polarized 45-fs pulses at different probe delays of 36 and 53 fs. The $(C^+ + O^+)_{\text{slow}}$ channel is enhanced at 53 fs compared to that at 36 fs.

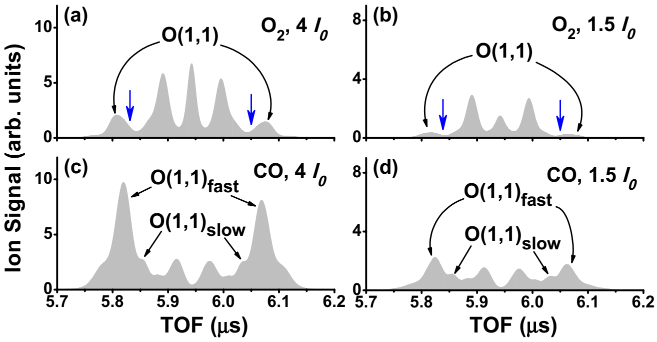

FIG. 6. (a) and (b) TOF spectra of O^+ from O_2 obtained with linearly polarized 68-fs pulses at (a) 4 I_0 and (b) 1.5 I_0 with $I_0 = 10^{14}\ \mathrm{W/cm^2}$. If there is an EI-induced $O^+ + O^+$ channel from O_2, the signal peaks will appear at the locations marked by the arrows in (a) and (b). See text for more details. (c) and (d) TOF spectra of O^+ from CO obtained with linearly polarized 68-fs pulses at (c) 4 I_0 and (d) 1.5 I_0.

of HOMO where electrons are not concentrated along the internuclear axis, the ionization rate does not have a R_c-dependent maximum, instead, only a monotonic increase with increasing internuclear distance under certain laser conditions [9]. We have explicitly shown the R_c-dependent EI channels in two molecules with σ-type HOMO, CO and N_2, as discussed above and in Ref. [18]. To experimentally investigate the molecular orbital type dependence of EI in molecules with non-σ-type HOMO, we perform a comparison study in another diatomic molecule O_2, since O_2 has a π-type HOMO.

In Figs. 6(a) and 6(b) we plot the TOF spectra of O^+ ions from O_2 using linearly polarized 68-fs pulses at intensity of (a) 4 I_0 and (b) 1.5 I_0 with $I_0 = 10^{14}\ \mathrm{W/cm^2}$. We also plot the TOF spectra of O^+ from CO under similar laser conditions in Figs. 6(c) and 6(d) as comparison, since the O^+ ions from both molecules have the same mass-over-charge ratio and therefore have similar flight time on the TOF spectra. In Figs. 6(c) and 6(d) we clearly see two O(1,1) peaks of CO, the EI-induced $O(1,1)_{slow}$ channel, and the commonly seen $O(1,1)_{fast}$ channel. However, in Figs. 6(a) and 6(b), there is only one $O^+ + O^+$ channel from O_2, which is the commonly seen $O^+ + O^+$ channel with a KER of 6.5 eV.

If we apply the generalized analytic EI model to O_2 [1,6,8], the R_c for O_2^+ is estimated to be 4.3 Å. The expansion time for O_2^+ from equilibrium internuclear distance R_e to R_c is estimated to be 51 fs [6], which is shorter than our 68-fs pulse and therefore, the R_c is likely to be reached in our 68-fs pulse. The Coulomb repulsion energy at R_c for a double ionization induced dissociation $O^+ + O^+$ channel is estimated as $(14.4 \times p \times q)/R_c = 3.35\,\mathrm{eV}$. Take the kinetic energy of the precursory state $O^+ + O$ (about 1.0 eV) into account, an EI-induced $O^+ + O^+$ channel from O_2, if occurs, would have a KER of 3–4.5 eV. This channel should be distinguishable on the TOF spectra from the commonly seen $O^+ + O^+$ channel with a KER of 6.5 eV, according to our previous study of $(C^+ + O^+)_{slow}$ and $(C^+ + O^+)_{fast}$ from CO as discussed above. Take 4.35 eV as an example for the KER of the imaginary EI-induced $O^+ + O^+$ channel, the corresponding signal peaks will appear at 5.83 μs (forwarding ions) and 6.05 μs (backwarding ions) on the TOF spectra, as marked by arrows in Figs. 6(a) and 6(b). However, we do not see any clear signal at these positions at either a higher [Fig. 6(a)] or a lower intensity [Fig. 6(b)]. Therefore, we conclude that the EI efficiency in O_2 is significantly low, if not zero. We believe that this conclusion holds for other molecules with π-type HOMO.

IV. CONCLUSIONS

In summary, we perform a polarization dependence study of enhanced ionization in diatomic molecules. We find EI exists when the field polarization is parallel to the molecular axis but disappears when the polarization is perpendicular. We further study EI with circular polarization and find that EI consistently exists in circular polarization indicating that rescattering does not play a significant role for EI. Furthermore, we study molecular orbital effect on EI and find that EI exists in σ type but not π type outmost molecular orbitals.

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