Electronic and Nuclear Wave Packet Dynamics of Molecular Systems in Intense Laser Fields

Hirohiko Kono, Yukio Sato, and Yuichi Fujimura

Department of Chemistry, Graduate School of Science, Tohoku University, Sendai 980-8578, Japan  E-mail: kono@mcl.chem.tohoku.ac.jp

Abstract. Intramolecular electronic dynamics and tunnel ionization of H$_2$ in an intense laser field (I = 10$^{14}$ W cm$^{-2}$ and $\lambda$ = 760 nm) were examined with accurate evaluation of three-dimensional two-electron wave packet dynamics. Based on the results of population analysis of "field-following" adiabatic states, an electrostatic model in which each atom in a molecule is charged by laser-induced electron transfer and ionization proceeds via the most unstable atomic site is proposed. This simple electrostatic approach is applied to investigate the structural deformations of CO$_2$ cations in an intense field.

INTRODUCTION

Interaction of atomic or molecular systems with high-power laser fields results in nonperturbative electronic dynamics such as above-threshold ionization and tunnel ionization [1]. In a high-intensity and electronically nonresonant long-wavelength regime (intensity I > 10$^{13}$ W/cm$^2$ and wavelength $\lambda$ > 700 nm), a laser electric field significantly distorts the Coulombic potential that the electrons are placed in. The distorted potential forms a "quasistatic" barrier through which an electron or electrons can tunnel. In the case of molecules, a large part of the electron density is transferred among nuclei within a half optical cycle [2]. Consequently, tunnel ionization proceeds through the most unstable atomic site [3]. Laser-induced intramolecular electron transfer, which is governed by electron-electron repulsion and molecular structure (e.g., bond length), also induces structure deformation of molecules [2]. Resultant structure deformations in turn change the electronic response to the field, e.g., intramolecular electron transfer followed by tunnel ionization [4]. It is known that tunnel ionization is greatly enhanced at a specific internuclear distance that is much longer than the equilibrium internuclear distance $R_e$ (known as enhanced ionization) [5].

To quantitatively understand the ultrafast electronic dynamics in intense fields, it is necessary to solve the time-dependent Schrödinger equation for the electronic degrees of freedom of a molecule. We have developed an efficient grid point method, the dual transformation method [6], for accurate estimation of propagation of an electronic wave packet. In this method, both the wave function and the Hamiltonian are transformed consistently to overcome the numerical difficulties arising from the divergence of the Coulomb potentials. We have applied this method to small molecular systems such as H$_2$ and H$_2$ [3,4]. To the best of our knowledge, our treatment of H$_2$ is the first
accurate evaluation of two-electron dynamics of a molecule. The vibrational degree of freedom is also incorporated in the calculation of H$_2^+$ without resorting to the Born-Oppenheimer approximation [3].

In this paper, we present the results of a theoretical investigation of intense-field-induced phenomena of H$_2$. In a low frequency case ($\lambda > 700$ nm), the localized ionic (bond) states H$^+$H$^-$ and H$^-$H$^+$ appear alternately according to the laser cycle, through which tunnel ionization proceeds. The electron pair is always created around the proton at which the dipole interaction energy with the field becomes lower (descending well). It is expected that the clarified mechanism of enhanced ionization for H$_2$ would serve as a prototype of tunnel ionization of multi-electron molecules. We have also investigated the electronic dynamics of H$_2$ in a high-frequency case in which the laser frequency $\omega$ is larger than the energy difference $\omega_{X-B}$ between the ground state $X^1\Sigma^+_g$ and first excited state $B^1\Sigma^+_u$ ($\lambda_{X-B} \sim 100$ nm). For $\omega > \omega_{X-B}$, the electron pair is formed in the ascending well. This indicates that it is possible to control the ionization probability in various ways.

Levis et al. recently reported selective bond dissociation and rearrangement of polyatomic molecules with optically tailored, intense-field laser pulses [7]. Intense laser chemistry has now opened up. The characteristic features of electronic dynamics of H$_2$ in an intense laser field leads to a simple electrostatic concept that the dynamics of bound electrons and the subsequent ionization process of a multi-electron polyatomic molecule can be clarified in terms of "field-following" adiabatic states. The properties of adiabatic states of multi-electron molecules can be determined by \textit{ab initio} molecular orbital (MO) methods. The calculated time-dependent potentials are used to evaluate the nuclear dynamics (such as bond stretching) until the next ionization process. We apply this approach to a CO$_2$ molecule in an intense field and show that the two C-O bonds can be symmetrically stretched in the CO$_2^{+}$ stage.

**ELECTRONIC DYNAMICS OF H$_2$**

In this section, we present the results of an investigation of the two-electron wave packet dynamics of H$_2$ in intense fields. The position of the $j$th electron is designated by cylindrical coordinates ($\rho_j$, $z_j$, and $\varphi_j$). The $z_1$ and $z_2$ axes are parallel to the molecular axis. The internuclear distance $R$ is fixed. We assume that the molecule is aligned parallel to the field polarization direction [8]. Electron transfer in H$_2$ is then characterized by motion along the molecular axis. To represent the wave packet $\psi(t)$, we therefore employ the reduced density $\tilde{P}(z_1,z_2)$ obtained by integrating $|\psi(t)|^2$ over the degrees of freedom other than $z_1$ and $z_2$. As an example, $\tilde{P}(z_1,z_2)$ of the state $X$ at $R=4$ a.u. is drawn in Fig. 1(a). The reduced density clearly demonstrates that the covalent components around $z_1 = z_2 = \pm R/2$ are dominant at $R = 4$ a.u. The localized ionic components $|H^+H^-\rangle$ and $|H^-H^+\rangle$ contained in $\psi(t = 0)$ are both 19%. Here, $|H^+H^-\rangle$ and $|H^-H^+\rangle$ are defined as H$^-$ ions located at $z_1 = z_2 = \pm R/2$.

We first discuss a case in which the laser frequency $\omega$ is much smaller than $\omega_{X-B}$. The laser electric field $\mathcal{E}(t)$ that the H$_2$ interacts with is assumed to be $f(t)\sin(\omega t)$,
where $f(t)$ is the pulse envelope at time $t$. The parameters are as follows: $\omega = 0.06$ a.u. ($\lambda = 760$ nm); $f(t)$ is linearly ramped so that $f(t)$ attains its maximum $f_0 = 0.12$ a.u. after one cycle. The corresponding intensity $I$ is given by $3.5 \times 10^{16} f^2(t)$ W cm$^{-2}$. The ionic component around the descending potential well, where $z E(t) < 0$, increases as the field approaches the first local maximum at $t = \pi / 2\omega = 26.2$ a.u. = 0.634 fs. At $t = \pi / 2\omega$, $E(t) = 0.03$ a.u. and $|\langle \psi | H^+ H^- \rangle|^2 = 0.31$. See Fig. 1(b). The laser field forces the two electrons to stay near a nucleus for almost a half cycle. The packet at $t = \pi / \omega$ is nearly identical to the initial one shown in Fig. 1(a), indicating that the electronic response to the field is basically adiabatic. Until then, no ionization current is observed. A quarter cycle later, as shown in Fig. 1(c), the density around $z_1 = z_2 = R / 2$ becomes as large as $|\langle \psi | H^+ H^- \rangle|^2 = 0.54$ because of the stronger field $E(t = 3\pi / 2\omega) = -0.09$ a.u. The ionic character increases as $|E(t)|$ increases.

As indicated by the broken line in Fig. 1(c), an electron is ejected from the localized ionic structure $H^+ H^-$. If the two nuclei are far from each other, the ionization potential of the localized ionic structure is considered to be as low as $I_p(H^-) = 0.75$ eV. The

![Figure 1](image-url). Snapshots of the electronic wave packet dynamics of $H_2$ in an intense field at (a) $t = 0$, (b) $\pi / 2\omega$, and (c) and (c') $3\pi / 2\omega$ ($\omega = 0.06$ a.u.). The coordinates $z_1$ and $z_2$ for the two electrons are parallel to the molecular axis. The reduced density $P(z_1, z_2)$ is represented by a contour map. The instantaneous field strengths are 0.03 a.u. at $t = \pi / 2\omega$ and -0.09 a.u. at $t = 3\pi / 2\omega$. The snapshots (a)-(c) are those obtained for $R = 4$ a.u. As shown in (a), the covalent components around $z_1 = -z_2 = \pm R / 2$ are dominant in the initial ground state at $R = 4$ a.u. As shown in (b) and (c), an ionic component $H^+ H^-$ or $H^{-} H^+$ is created near the descending well. As indicated by the broken line in (c), an electron is ejected from the structure $H^+ H^-$. The reduced density at $t = 3\pi / 2\omega$ for $R = 1.6$ a.u. is drawn in (c').
localized ionic structure is hence unstable and is regarded as a doorway state to ionization. The ionization current from the covalent structure, denoted by a dotted line in Fig. 1(c), is relatively small. At \( R = 4 \) a.u., the rate of ionization from a pure localized ionic state is at least five-times greater than that from a pure covalent state. We have calculated the wave packet dynamics at different values of \( R \) for the same pulse. The reduced density at \( t = 3\pi/2\omega \) for \( R = 1.6 \) a.u. (\( = R_c \)) is shown in Fig. 1(c'). Despite the intense field strength at \( t = 3\pi/2\omega \), the ionization current is very small. The change in \( R \) from 4 a.u. to 1.6 a.u. reduces the ionization rate by about two orders of magnitude. As \( R \) decreases, the rate of ionization from \( \text{H}^-\text{H}^+ \) decreases owing to the more attractive force of the distant nucleus, while the rate of a covalent state is almost independent of \( R \). On the other hand, the population of the \( \text{H}^-\text{H}^+ \) created increases as \( R \) decreases. For instance, for \( R = 1.6 \) a.u., \( |\langle \psi | \text{H}^-\text{H}^+ \rangle|^2 \) is as large as 0.74 at \( t = 3\pi/2\omega \) (at \( t = 0 \), \( |\langle \psi | \text{H}^-\text{H}^+ \rangle|^2 = 0.58 \)). As a result, ionization is enhanced at the critical distance \( R_c = 4-6 \) a.u. The explanation that the created \( \text{H}^+\text{H}^- \) is the main doorway state to ionization is reinforced by the fact that ionization is suppressed for triplet states (\( \text{H}^+\text{H}^- \) is not created owing to the Pauli exclusion principle).

We have examined the intramolecular electronic dynamics that governs the ionization process by analyzing the populations of field-following adiabatic states defined as eigenfunctions of the instantaneous electronic Hamiltonian \( H_{elec}(t) \) including the interaction with \( E(t) \) \[9\]. The effective \( H_{elec}(t) \) for \( \text{H}_2 \) is constructed from the three lowest electronic states, \( X, B, \) and \( EF \). One of the three resultant adiabatic states is the lowering ionic adiabatic state characterized by \( \text{H}^-\text{H}^+ \) or \( \text{H}^+\text{H}^- \), which decreases in energy by \( -|\mathcal{E}(t)|R \); on the other hand, the energy of the covalent adiabatic state \( \text{H} \cdot \text{H} \) is relatively insensitive to \( |\mathcal{E}(t)| \). Therefore, the lowering ionic state and the covalent character-dominated initial state can cross each other in energy. By solving the time-dependent Schrödinger equation for the effective \( H_{elec}(t) \), as shown below, we have found that the difference in ionization dynamics between small \( R \) and the \( R \) cases originates in the character of the level crossing of the lowest two adiabatic states.

The initial ground state is adiabatically connected with the lowering ionic state after the crossing. As \( |\mathcal{E}(t)| \) increases, the lowest adiabatic state starting from the \( X \) state becomes more ionic. In the case of \( R \leq 4 \) a.u., the energy gap at the avoided crossing between the two lowest adiabatic states is as large as that at zero field strengths because the transition dipole moment of the \( B-X \) transition, which enlarges the gap, increases as \( \sim R/\sqrt{2} \) up to \( R = 3 \) a.u. As a result, nonadiabatic transitions to upper adiabatic states hardly occur. Ionization occurs from the lowest adiabatic state characterized by \( \text{H}^+\text{H}^- \) or \( \text{H}^+\text{H}^- \). Around \( R = 6 \) a.u., the ionization from the covalent state competes with that from the localized ionic state. As \( R \) increases further, the electron density transferred between the nuclei is suppressed: the main character becomes covalent. The ionization at large \( R \) (\( \geq 8 \) a.u.) mainly proceeds from the remaining covalent component, which outmeasures the created ionic component.

We have also investigated the electronic dynamics of \( \text{H}_2 \) in a high-frequency case in which \( \omega \) is close to \( \omega_{X-B} \). Even after the incident pulse has fully decayed, \( \text{H}^+\text{H}^- \) and \( \text{H}^-\text{H}^+ \) appear alternately with a period of \( 2\pi/\omega_{X-B} \) (\( \sim 500 \) attoseconds). Contrary
to the long wavelength case, for \( \omega > \omega_{X-B} \), the electron pair is formed in the ascending well. We have found that ionization is enhanced or suppressed according to whether \( \omega \) is just below \( \omega_{X-B} \) (case I) or just above \( \omega_{X-B} \) (case II). In case I, the first step is the formation of an electron pair in the descending well. The subsequent ionization process is regarded as a hybrid of tunnel ionization and multiphoton ionization. In case II, the electron pair is formed in the ascending well from which ionization is greatly suppressed. This is in sharp contrast to the low-frequency case in which the ascending well is most unstable to ionization. We have numerically demonstrated that two-electron dynamics and ionization can be controlled by adjusting the time lag between two phase-locked ultrashort pulses.

**INTENSE FIELD CHEMISTRY OF \( \text{CO}_2 \)**

The characteristic features of electronic dynamics of \( \text{H}_2 \) in an intense laser field leads to a simple electrostatic view that each atom in a molecule is charged by field-induced electron transfer and that ionization proceeds via the most unstable atomic site. In the low-frequency case (\( \lambda > 700 \) nm), the dynamics of bound electrons can be described in terms of field-following adiabatic states. The properties of adiabatic states can be determined by \textit{ab initio} MO methods. While the time-dependent adiabatic potentials calculated by an MO method are used to evaluate the nuclear dynamics until the next ionization process, the charge distributions on individual atomic sites are used to estimate the ionization probability. For \( \omega_{\text{elec}} > \omega \), where \( \omega_{\text{elec}} \) represents the characteristic frequency for the electronic transitions, the vibrational motion is mainly determined by the time-dependent potential of the lowest adiabatic state. Here, we ignore field-induced potential crossing at large internuclear distances.

Ultrafast field-induced deformation of molecular structure has been observed for various molecules such as \( \text{CO}_2 \). Hishikawa \textit{et al.} [10] have experimentally determined the geometry of \( \text{CO}_2^+ \) in a 1.1 PW cm\(^{-2}\), 100 fs pulse (\( \lambda = 795 \) nm) just before undergoing Coulomb explosions, namely, that the C-O bond length \( R_{\text{C-O}} \) is stretched to about 3.0 a.u. from the equilibrium value of \( \text{CO}_2 \) (\( R_e = 2.3 \) a.u.), and the amplitude of bending is much larger in comparison with \( \text{CO}_2 \) (the observed mean amplitude of the deviation angle \( \theta \) from the linear structure being \( \sim 40^\circ \)). We apply the above approach to investigate the deformation stage of \( \text{CO}_2 \). The adiabatic potential surfaces and charge distributions of \( \text{CO}_2 \) and its cations are calculated by using the full-optimized reaction space multiconfiguration self-consistent-field method with the 6-311+G(d) basis set [2]. The total charge is assigned to each atom by Mulliken population analysis.

A linearly polarized laser field can align molecules [8]. To achieve high alignment in the polarization direction by adiabatic following of the molecular rotation to the pulse envelope, the pulse duration must be longer than \( 2\pi/2B \approx 40 \) ps, where \( B(=0.39 \text{ cm}^{-1}) \) is the rotational constant of \( \text{CO}_2 \). Since the pulse duration used in the experiment \((\approx 100 \) fs\) is shorter than \( 2\pi/2B \), the alignment is incomplete. The ionization rate has, however, a maximum at the geometry where the molecular O-O axis is parallel to the polarization direction. This parallel case should be studied as the main spatial
configuration. When a molecule is perpendicular to the polarization direction, the electric field does not stretch the bonds, while it may cause bending.

We now apply the above-described electrostatic model to the CO\textsubscript{2} case. Consider a linear molecule \( O^p+ C^O^+ O^z+ \) placed parallel to \( E(t) \). Numerical calculations indicate that the charge on C does not change in an intense field. Thus, the charge transfer ionic state \( O^{(p-1)+} C^O^+ O^{(z+1)+} \) is expected to be a doorway state to tunnel ionization. The field strength \( E_c \) required for its creation is then given by \[ E_c = \left| I_p(O^{2+}) - I_p(O^{(p-1)+}) - (Z + 1 - P)/R_{O-O} \right|/R_{O-O}, \] (1)

where \( R_{O-O} \) is the O – O bond length. An appreciable amount of charge is transferred between the O atoms when \( |E(t)| \) exceeds the value \( E_c \). The estimated value of \( E_c \) serves to predict the intensity required for tunnel ionization.

(i) Neutral CO\textsubscript{2} The charges on the three atoms in the lowest adiabatic state of linear CO\textsubscript{2} are calculated as a function of the field strength. At \( R_{C-O} = R_c \), the charges of O and C at zero fields are -0.22 and +0.45, respectively. The charge distribution of the main electronic configuration is expressed as \( O^0+ C^O^+ O^0+ \). When a field is applied, an appreciable amount of negative charge is transferred from the O atom in the ascending well to the O atom in the descending well. From Eq. (1), \( E_c = 0.05 \) a.u. at \( R_{C-O} = R_c \). The calculated charge of the O atom in the descending well is -0.83 at \( E(t) = 0.1 \) a.u. > \( E_c \). The ionic component \( O^-C^+ \) becomes dominant beyond \( E_c \). We thus expect for CO\textsubscript{2} that tunnel ionization via the ionic structure occurs somewhere around \( E_c \).

Experimentally, CO\textsubscript{2}\textsuperscript{+} appears at an intensity around \( 3.5 \times 10^{13} \) W cm\(^{-2} \) (\( E \approx 0.03 \) a.u.).

We have calculated the potential surface of the lowest adiabatic state of CO\textsubscript{2} as a function of the two C-O bond distances \( R_1 \) and \( R_2 \) and the bond angle \( \theta \). There are two types of bond stretching: symmetric stretching in which \( R_1 = R_2 \) and asymmetric stretching in which one C-O bond is longer than the other (e.g., \( R_1 > R_2 = R_c \)). The adiabatic potential of CO\textsubscript{2} is greatly distorted in an intense field; the dissociation energy for a C-O bond is 7 eV at zero fields and is reduced to 4 eV at \( E(t) = 0.1 \) a.u. (> \( E_c \)). However, bond stretching hardly occurs for the following reason.

The potential of the lowest adiabatic state, \( V_0(R_1, R_2, t) \), can be approximated as \( V_0(R_1, R_2, t) = V_0(R_1, R_2) - \mu(R_1 - R_2)E(t) - \alpha(R_1 + R_2)E^2(t)/2, \) (2)

where \( V_0(R_1, R_2) \) is the adiabatic potential at zero fields. Because of the second term, at a moment, the barrier for asymmetric dissociation is greatly reduced as mentioned above. It should, however, be noted that the following inequality in temporal or energy scale holds:

\( \omega_{\text{elec}} > \omega > \omega_{\text{vib}} \), (3)

where \( \omega_{\text{vib}} \) is the vibrational frequency. The experimental condition \( \omega > \omega_{\text{vib}} \) means that the change in \( E(t) \) is too fast for the vibrational motion to follow \( E(t) \) adiabatically. Since \( \omega > \omega_{\text{vib}} \) and \( f(t) \) does not change in one optical cycle, i.e., \( \omega > f(t)/f(t) \), \( V_0(R_1, R_2, t) \) can be replaced with the cycle-averaged effective potential \( \overline{V_0}(R_1, R_2, t) \):

\[ \overline{V_0}(R_1, R_2, t) = V_0(R_1, R_2) - \alpha(R_1 + R_2)f^2(t)/4. \] (4)

Thus, the second term in Eq. (2) disappears by cycle averaging. Since \( \omega_{\text{vib}} > \)
\( \dot{f}(t) / f(t) \), the \( v \)th vibrational state at zero fields is adiabatically transferred to the \( v \)th state of \( V_0(R_1, R_2, t) \). Up to \( \dot{f}(t) \approx 0.15 \) a.u. (\( \gg E_c \)), the equilibrium geometry of \( V_0(R_1, R_2, t) \) is almost equal to that of \( V_0(R_1, R_2) \). We thus conclude that the neutral CO\(_2\) in an intense field takes a stable linear structure around the zero-field equilibrium geometry.

In conclusion, in the CO\(_2\) and CO\(_2^+\) stages, ionization occurs before the field intensity becomes high enough to deform the molecule. 

(ii) CO\(_2^+\) The geometry of CO\(_2^+\) just after ionization of CO\(_2\) is expected to be nearly equal to that of CO\(_2\). Two positive charges in CO\(_2^+\) are nearly equally distributed among the three atoms as O\(^+\)C\(^+\)O, O\(^+\)C\(^+\)O, and O\(^+\)C\(^+\)O. An ionic structure favorable for tunnel ionization is O\(^+\)C\(^+\)O\(^2+\) created from O\(^+\)C\(^+\)O. From Eq. (1), \( E_c = 0.18 \) a.u. at \( R_1 = R_2 = R_e \). Before the field intensity reaches the large threshold value of 0.18 a.u., structural deformation of CO\(_2^+\) can occur before ionization.

We examined the nuclear wave packet dynamics on \( V_0(R_1, R_2, t) \) of CO\(_2^+\). A realistic pulse shape is employed: \( f(t) = f_0 \sin^2(\pi t/T_{\text{pulse}}) \) for \( 0 \leq t \leq T_{\text{pulse}} \), where \( f_0 = 0.19 \) a.u. and \( T_{\text{pulse}} = 194 \) fs. The frequency \( \omega \) was set at 0.06 a.u. We simulated a case in which the initial wave packet is created on the lowest adiabatic state of CO\(_2^+\) when the field envelope \( f(t) \) reaches \(-0.03\) a.u. (\( t = 24 \) fs); vertical transition from the ground vibrational state of CO\(_2\) was assumed. In Fig. 2, the position of the initial wave packet prepared at \( t = 24 \) fs is marked with \( \times \), and the instantaneous potential surface at \( E(t = 84.7 \) fs) = 0.18 a.u. is shown. The dissociation energy at zero fields is \(-10.7 \) eV in the case of symmetric stretching and ca. 1.0 eV in the case of asymmetric stretching.

![Figure 2](image-url)
From the viewpoint of dissociation energy, it is expected that asymmetric stretching is predominant. However, the wave packet at $t = 84.7 \text{ fs}$ denoted by bold solid lines shows that symmetric stretching occurs as well as asymmetric stretching. The appearance of simultaneous two-bond stretching can be explained by using the cycle-averaged potential $V_0(R_1, R_2, t)$ of $\text{CO}_2^{2+}$. In $\text{CO}_2^{2+}$, the stabilization energy $-\alpha(R_1 + R_2)f^2(t)/4$ due to the induced dipole moment is large in comparison with $V_0(R_1, R_2)$. Consequently, a deep valley along the symmetric stretching coordinate is formed in $V_0(R_1, R_2, t)$: the symmetric dissociation becomes barrierless. After bond stretching, ionization to $\text{CO}_2^+$ occurs owing to enhanced ionization at larger values of $R_{\text{C-O}}$.

We also propose that the experimentally observed two-body breakup to $\text{CO}^+ + \text{O}^+$ is due to asymmetric stretching on the adiabatic potential of $\text{CO}_2^{2+}$. The asymmetric stretching originates from the difference in the equilibrium internuclear distance between $\text{CO}_2^{2+}$ and $\text{CO}_2$ (or $\text{CO}_2$). A part of the wave packet propagates toward the two-body breakup from the Franck-Condon region, as shown by the wave packet at $t = 48 \text{ fs}$ denoted by dotted contour lines in Fig. 2. Although the instantaneous potential is dissociative for the asymmetric channels, asymmetric stretching is not enhanced by a field because of the disappearance of $-\mu(R_1 - R_2)E(t)$ in $V_0(R_1, R_2, t)$. As the field intensity is increased, the field-induced symmetric stretching becomes dominant.

The induced dipole of the lowest adiabatic state that shifts the energy downward becomes smaller as the molecule becomes more bent; the curvature of the potential with $\theta$ is larger at nonzero fields than at zero fields. Therefore, a large-amplitude bending motion is expected to be hardly induced. However, the curvature of the potential with $\theta$ is smaller in $\text{CO}_2^{2+}$ than in $\text{CO}_2$. Hence, the wave packet prepared on the lowest adiabatic state of $\text{CO}_2^{2+}$ from $\text{CO}_2$ spreads out along $\theta$. The experimentally observed structure of $\text{CO}_2^{2+}$ just before Coulomb explosions originates from the field-induced bond stretching of $\text{CO}_2^{2+}$ accompanied by a large-amplitude bending motion. The proposed approach is simple but has wide applicability in predicting the electronic and nuclear dynamics of polyatomic molecules in intense laser fields.

REFERENCES


