

Chapter 2

Enhanced Ionization of Molecules in Intense Laser Fields

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Abstract Molecules exposed to intense ultrashort laser pulses undergo rapid ionization at critical large internuclear distances requiring nonperturbative models to explain the highly nonlinear nonperturbative response. Simple quasistatic models allow for the prediction of these critical internuclear distances. It is shown that the mechanism for enhanced ionization is due to overbarrier ionization of the Stark-shifted LUMO (lowest unoccupied molecular orbital) in both symmetric diatomic and triatomic molecules. In nonsymmetric molecules, permanent dipole moments contribute to the strong Stark-shifts of electron orbitals and enhanced ionization occurs through resonances of the HOMO (highest occupied molecular orbital) and LUMO, corresponding to charge transfer. Double ionization is also enhanced at certain critical distances but involves considerable excitations of intermediate electronic states.

2.1 Introduction

Advances in current laser technology are providing new tools for experimentalists and challenges for theorists to explore a new regime of light–matter interaction, the nonlinear nonperturbative interaction of atoms and molecules with intense ($I \geq 10^{+14}$ W/cm²) few-cycle laser pulses. Much of the earlier experimental and theoretical work in this new nonperturbative regime of radiation–atom interaction in atomic systems has been summarized by Brabec and Krausz [1], Corkum and

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Krausz [2]. Thus in the atomic case new nonperturbative optical phenomena have been found such as above-threshold ionization (ATI), [1], tunnelling ionization predicted as early as 1980 [3–5] and is the basis of recollision physics [6] which has been confirmed experimentally recently [7]. Another important nonlinear process, high-order harmonic generation (HHG), is the current most convenient source of attosecond ($1 \text{ asec} = 10^{-18} \text{ s}$) pulses [8]. This rapid development of ultrashort intense laser pulses allows nowadays for shaping and focussing such pulses to higher intensities creating electric fields E greater than the atomic unit, a.u., $E_0 = 5 \times 10^9 \text{ V cm}^{-1}$ at the 1s orbit radius $a_0 = 0.0529 \text{ nm}$ of the H atom. This corresponds to an a.u. of intensity $I_0 = cE_0^2/8\pi = 3.54 \times 10^{16} \text{ W/cm}^2$. Such pulses can be compressed to times of several femtoseconds ($1 \text{ fs} = 10^{-15} \text{ s}$), thus creating radiative coherences that are shorter than molecular nonradiative (radiationless) relaxation times [9, 10]. The Schwinger limit, 10^{29} W/cm^2 , is the limit of instability of matter by tunneling from the vacuum itself thus creating electron–positron pairs [11], and electron–positron pairs can now be created from vacuum at smaller intensities, $I \sim 10^{26} \text{ W/cm}^2$ [12].

Whereas at the end of the twentieth century the focus was on femtosecond (fs) photochemistry and photophysics culminating with a Nobel prize to A. H. Zewail (Caltech) for “femto chemistry” [13], a major effort is now underway to develop single attosecond (asec) optical pulses based on the nonperturbative physics of HHG in atoms [8] and molecular high order harmonic generation (MHOHG) in molecules [14]. These new asec pulses presage the control of electron motion in molecules, the “Holy Grail” of many atomic and molecular sciences from chemistry to biology to material sciences [15–17].

The intensities discussed in this chapter, $10^{14} \leq I \leq 10^{15} \text{ W/cm}^2$ correspond to fields approaching the internal Coulomb potentials V_0 or corresponding electric fields E_0 (Table 2.1) in atoms and molecules, thus introducing considerable distortions of intermolecular potentials. Using a dressed photon state representation, such strong radiatively induced distortions create, laser induced molecular potentials (LIMP’S), leading to “bond-softening” via laser-induced avoided crossings of molecular potentials [18, 19]. At these high intensities, one needs to consider further ionization and above-threshold dissociation (ATD), the equivalence of ATI in atoms. Furthermore, the quasistatic picture of atomic tunneling ionization [3, 5, 6] needs to be modified in view of the multi-centre nature of electron potentials in molecules and the presence of large radiative transition moments, originally described by Mulliken [20] as charge resonance (CR) transitions [18, 19]. One of the fundamental differences between intense field ionization of atoms and molecules is “enhanced ionization” which occurs at critical internuclear distances R_c greater than equilibrium distances R_e for molecules exposed to intense laser pulses [21–24]. Quasistatic models of enhanced ionization have been successful in predicting values of R_c . The main tenet of this model, which differs fundamentally from quasistatic atomic ionization, is that laser-induced charge resonance (CR) effects localize temporarily the electron by charge transfer [22–24] due to the presence of large electronic transition moments in molecules resulting in adiabatic electronic charge transfer across the whole length of a molecule.

Table 2.1 Atomic units ($e = \hbar = m_e = 1$; $c = 137.036$)

Potential energy	$V_0 = e^2/a_0 = 1$ Hartree = 27.2 eV
Electric field	$E_0 = e/a_0^2 = 5.14 \times 10^9$ V cm $^{-1}$
Intensity	$I_0 = cE_0^2/8\pi = 3.54 \times 10^{16}$ W cm $^{-2}$
Distance	$a_0 = 0.0529$ nm
Velocity	$v_0 = 2.19 \times 10^8$ cm s $^{-1}$
Time	$t_0 = a_0/v_0 = 24.2 \times 10^{-18}$ s = 24.2 attoseconds
	$t_c = \hbar/m_e c^2 = 1.29 \times 10^{-21}$ s = 1.29 yoctoseconds

2.2 Quasistatic Models: Diatomic One-Electron Systems in Strong Fields

The first simple analytic formulae for atomic multiphoton ionization beyond usual perturbation (Fermi Golden Rule) theory were obtained by Keldysh [3] who showed that a parameter γ , today called the Keldysh parameter [25, 26], allows to separate the perturbative multiphoton regime from the nonperturbative quasistatic tunnelling regime. This parameter is obtained from the low frequency limit of the transition probability from an initial bound state to a Volkov state, the state of a free electron “dressed” by a laser field [25]. This is essentially a time-dependent distorted wave born approximation (DWBA), where the Coulomb potential is retained in the initial bound state and the laser field only in the final continuum state. The Keldysh parameter is then defined by

$$\gamma = (I_p/2U_p)^{1/2}, \quad (2.1)$$

where I_p is the ionization potential and U_p is the ponderomotive energy, $U_p = I_0/4\omega^2$, the average kinetic energy of a free electron in a linearly polarized field $E(t) = E_0 \cos(\omega t)$ and peak intensity $I_0 = cE_0^2/8\pi$ at frequency ω . The ponderomotive energy U_p and ponderomotive radius α_p are in fact two additional important parameters in laser-induced electronic processes. Solving the classical equations of motion in a monochromatic time-dependent field $E(t) = E_0 \cos(\omega t + \phi)$ gives in a.u. (Table 2.1)

$$\ddot{z} = -E_0 \cos(\omega t + \phi), \dot{z}(t) = -\frac{E_0}{\omega} [\sin(\omega t + \phi) - \sin \phi] \quad (2.2)$$

$$z(t) = -\frac{E_0}{\omega^2} [\cos \phi - \cos(\omega t + \phi) - \omega t \sin \phi], \quad (2.3)$$

where ϕ is the initial laser phase called the CEP (carrier envelope phase) at which moment an electron is ionized at time $t = 0$ assuming an initial zero velocity $\dot{z}(0) = 0$ as in the tunnelling model [4], from which one can estimate initial ionization rates using a static E field rate [3, 4, 25]. Tunneling ionization has now been detected in atoms from the modulation of the ionized electron density by the

intense laser field [7]. These classical equations define the ponderomotive radius α_p and ponderomotive energy U_p (in a.u.)

$$\alpha_p = E_0/\omega^2, \quad U_p = \frac{1}{4}\alpha_p^2\omega^2 = E_0^2/4\omega^2 \quad (2.4)$$

The Keldysh parameter γ , (2.1) is a ratio of two energies: I_p the minimum energy to ionize the electron and $2U_p$, the maximum kinetic energy, $\frac{1}{2}\dot{z}^2$, acquired by a free electron in the laser field. This simple classical quasistatic model described by (2.2)–(2.4) where one assumes an electron is ionized with initial zero velocity $\dot{z}(t_0) = 0$, the basis of tunnelling ionization models [1, 4], allows us to deduce the laser-induced dynamics of the electron after ionization. Thus for $\lambda = 1,064 \text{ nm}$ ($\omega = 0.043 \text{ a.u.}$) and intensity $I = 1 \times 10^{14} \text{ W/cm}^2 = 3 \times 10^{-3} \text{ a.u.}$, one obtains $\alpha_p = 29 \text{ a.u.} = 1.53 \text{ nm}$. This maximum displacement α_p determines the minimum size of numerical grids [26] and U_p determines the corresponding spatio-temporal steps Δx , Δt (from uncertainty principle relations) in computations at high intensities. Equation (2.2) also allows predicting the maximum average kinetic energy to be $\frac{1}{2}\langle\dot{z}(t)\rangle^2 = 3U_p$ at CEP phase $\varphi = \pi/2$ ($\langle\cos^2\omega t\rangle = 1/2$, $\langle\cos\omega t\rangle = 0$). Such high energies acquired during ionization was observed and explained in microwave ATI using the simple model of (2.2)–(2.3) [27]. In practice, ionization occurs at different phases $\phi = \omega t_0$, i.e., at time t_0 , the instant the electron is created in the field. As we show later, t_0 , determines the electron trajectories for collision with neighbouring ions and recollision with the parent ion in both linear and circularly polarized laser fields. Initial *zero* velocity, $\dot{z}(t_0) = 0$ will occur at the instant of tunnel ionization [4–6]. The model can also be extended to *nonzero* initial velocity, $\dot{z}(t_0) > 0$ as in preionization with X-ray-VUV pulses [28] and to circularly polarized light [29].

The quasistatic model allows further for establishing the critical or minimum field E_m where above-barrier ionization occurs instead of under-barrier tunnelling ionization [21–24]. This is illustrated in Fig. 2.1a for a one-electron atom and Fig. 2.1b for a single valence electron in a diatomic molecule such as H_2^+ , in the presence of a static electric field E . In the atomic case, where a single “active” electron is bound by an effective nuclear charge $q+$, the total potential along the field polarization, i.e., z -axis, can be written as $V(z) = -q/|z| - Ez$. As seen from Fig. 2.1a, the electric field distorts the atomic Coulomb potential thus producing a barrier with a maximum at $z_m = (q/E_m)^{1/2}$ obtained from $\partial V/\partial z = 0$. Setting $V(z_m) = -I_p$, where I_p is the ionization potential, one obtains the minimum field $E_m = I_p^2/4q$ for above-barrier ionization. For hydrogenic levels, $I_p = -\varepsilon_n = q^2/2n^2$, for a level of principal quantum number n . For such a level, the minimum electric field E_m for atomic above-barrier ionization is $E_m = q^3/(2n)^4 \text{ a.u.}$ or intensity $I_m = cE_m^2/8\pi = cq^6/(8\pi(2n)^8) \text{ a.u.}$ Thus for the $n = 1$ (1s) level of H, $I_p = 1/2 \text{ a.u.}$, $I_m = 1.4 \times 10^{14} \text{ W/cm}^2$. For the Th^{+89} ion in its $n = 2$ level, $I_m = 90^6/2^{16} \text{ a.u.} = 2.84 \times 10^{23} \text{ W/cm}^2$. Such superintense fields are being currently considered in the European ELI (extreme light infrastructure) project [30]. The theoretical description of super intense field electron ionization

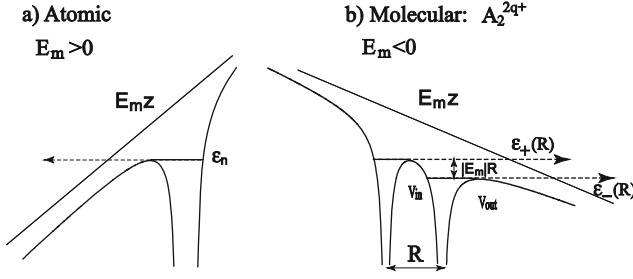


Fig. 2.1 Above barrier ionization in (a) atoms and (b) molecules via Stark-shifted LUMO, $\epsilon_+(R)$ and HOMO, $\epsilon_-(R)$

requires applying the relativistic Dirac equation to such processes and is becoming a new active field of research [31–33].

Molecular above-barrier ionization leads to a new concept, CREI (charge resonance enhanced ionization), for which recent experiments are summarized in [21]. As illustrated in Fig. 2.1b, a molecular electronic potential is a multicenter potential. Atomic energy levels are transformed into delocalized molecular orbitals, MO's, which at large internuclear distance R become linear combinations of atomic orbitals. In the case of H_2^+ , the two most important MO's in low frequency multiphoton processes are the HOMO, highest occupied MO and LUMO, lowest unoccupied MO, as these couple radiatively through an electronic transition moment $\langle \text{HOMO} | z | \text{LUMO} \rangle = R/2$ [19, 20]. In the presence of a static field, the LUMO is Stark-shifted in energy by $+ER/2$ whereas the HOMO's energy is lowered by $-ER/2$. Thus, at some critical internuclear distance R_c and critical field E_m , the LUMO, which can be populated nonadiabatically by the field [34, 35], ionizes directly over the barrier between the two nuclei. This simple quasistatic model was first proposed by Codling et al. [36–38] neglecting the Stark energies of both HOMO and LUMO, but rather emphasizing the localization (suppression of the tunnelling) between the field free wells, (Fig. 2.1b). Inclusion of the Stark energy shift of the LUMO allows for analytical estimates of R_c and E_m for above-barrier ionization in symmetric diatomic molecules [23, 39]. Inclusion of static dipole moments such as in HeH^{2+} further generalizes the concept of CREI in nonsymmetric molecules [40].

We follow the original 1-D model of one-electron molecular ionization in A_2^{2q+} systems, (Fig. 2.2b), where $q+$ is the charge on each nucleus [23, 39]. Exact Born–Oppenheimer (static nuclei) time-dependent Schrödinger, TDSE, simulations of highly charged diatomic molecular ions exhibit ionization maxima for internuclear distances $6 \leq R \leq 10$ a.u. at “critical” distances R_c for laser polarization parallel to the internuclear axis. Ionization from MO's with densities perpendicular to the intermolecular axis also induces abrupt increase of ionization into atomic plateaus at $R_c \sim 8$ a.u. [40, 41]. The necessary conditions for CREI to occur at R_c is that the upper Stark shifted electronic eigenstate energy $\epsilon_+(R)$ exceeds the two potential barriers in Fig. 2.1b, i.e.

$$V_{\text{out}}(R_c) \leq V_{\text{in}}(R_c) \leq \varepsilon_+(R_c), \quad (2.5)$$

where V_{in} and V_{out} are the inner and outer barriers of field modified the two-center Coulomb potential at the critical field strength E_m ,

$$V(z, R) = \frac{-q}{|\frac{R}{2} + z|} - \frac{q}{|\frac{R}{2} - z|} + E_m z, \quad (2.6)$$

and

$$\varepsilon_+(R) = -qI_p - q/R + E_m R/2. \quad (2.7)$$

$E_m R/2$ is the energy Stark shift of the LUMO (Fig. 2.1b), I_p is the ionization potential of the neutral atom and $\frac{-q}{R}$ is the electron Coulomb attraction with a neighbouring ion. Ionization potentials of electrons from the same shell are assumed to scale as $I(\text{ion}) = qI_p$ for an ion of charge $q-1$. The potential (2.6) has maxima at z_{in} and z_{out} for $E_m < 0$ as in Fig. 2.1b which are given by [23],

$$z_{\text{out}} = -\left(R/2 + E_q^{-1/2}\right), \quad z_{\text{in}} = E_q R^3/32 \quad (2.8)$$

where $E_q = |E_m|/q$.

Solution of the CREI conditions (2.5) by setting $\varepsilon_+(R_c) = V_{\text{out}}(R_c) = V_{\text{in}}(R_c)$ gives [23, 39],

$$R_c = 4.07/I_p, \quad |E_m| = 0.139I_p^2 \quad (2.9)$$

Neglecting the Stark-shift of the LUMO gives the value of $R_c = 3/I_p$ and defines the onset of electron localization due to negligible electron tunnelling between the potential double wells in the unperturbed field-free molecule [23, 24]. Equation (2.8) shows the importance of the static field E_m in Stark-shifting the LUMO energy to $\varepsilon_+(R)$ above the field modified barriers illustrated in (Fig. 2.1b). Nevertheless, the end result is the surprising independence of R_c from field strengths E_m and nuclear charge $q+$, thus establishing CREI as a *universal* intense field nonperturbative phenomenon [21, 34–60]. A simple physical explanation of CREI also emerges from the quasistatic image, Fig. 2.1b. The ionizing electron in the Stark-shifted LUMO of energy $\varepsilon_+(R)$ is initially accelerated by the neighboring ion whereas the electron in the HOMO, $\varepsilon_-(R)$, is initially decelerated by its parent ion as it leaves it, thus making ionization from the LUMO the dominant mechanism for CREI.

Nonsymmetric molecules such as the one-electron HeH^{2+} [47, 48] present a different nonlinear nonperturbative response due to the presence of a permanent dipole moment. This is illustrated in Fig. 2.2a for $E_m > 0$ and Fig. 2.2b for $E_m < 0$. Thus, the HOMO is displaced upward ($\uparrow$) to $\varepsilon_+(R)$ by $|E_m|R/2$ for $E_m < 0$ (Fig. 2.2b) and becomes resonant (degenerate) with the LUMO, $\varepsilon_-(R)$ which is shifted downward ($\downarrow$). Thus, enhanced ionization, EI, can occur by resonance between these two levels. In the case of the opposite positive field, $E_m > 0$ (Fig. 2.2a), both HOMO and LUMO can never cross, thus suppressing the resonance and enhanced ionization. This qualitative picture of CREI in symmetric molecules

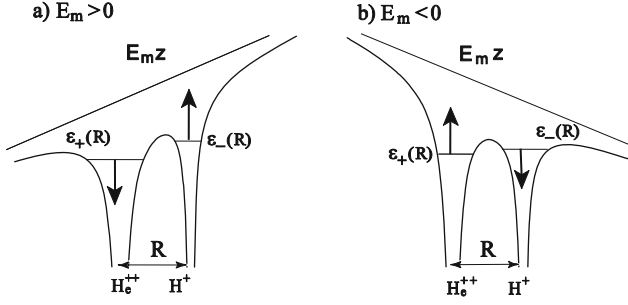


Fig. 2.2 Enhanced ionization in HeH^{2+} by Stark-shifted ϵ_+ (LUMO), ϵ_- (HOMO)

illustrated in Fig. 2.2 emphasizes the importance of resonances in these molecules with nonvanishing dipole moment. In fact, an avoided crossing between the HOMO and LUMO occurs for $E_m > 0$ (see Fig. 10 in [47]), inducing a new harmonic generation spectrum at high intensities [48]. Using the similar static Stark-displaced potential model as in the symmetric diatomic case above, one readily obtains the critical distance R_c for enhanced ionization in nonsymmetric systems [47] to occur for $E_m < 0$ at

$$R_c = \frac{(I_1 - I_2) + [(I_1 - I_2) - 4|E_m|(Z_1 - Z_2)]^{1/2}}{2|E_m|}. \quad (2.10)$$

where I_1 and I_2 are the ionization potentials of the different heteroatoms with $I_1 > I_2$ and effective nuclear charges Z_1 , Z_2 . Numerical TDSE simulations for HeH^{2+} where $Z_1 = 2$ and $Z_2 = 1$ show for this nonsymmetric system dependence of R_c on the CEP phase ϕ of the field strength E_m and charge separation $Z_1 - Z_2$ as predicted by (2.10). Clearly in this nonsymmetric case, the critical distance R_c for enhanced ionization depends on the CEP, ϕ , the field amplitude $|E_m|$, and the I_p difference between the heteroatoms, thus offering the possibility of controlling this highly nonlinear nonperturbative electron response in nonsymmetric molecules. The simple one-barrier models of enhanced ionization illustrated in Figs. 2.1–2.2 corresponding to quasistatic (electrostatic) models are confirmed by exact 3-D calculation of ionization rates using time-dependent Schrödinger equations, TDSE's for one-electron molecules H_2^+ [41–43] and HeH^{2+} [47]. In Fig. 2.3 we illustrate the ionization rate for H_2^+ as a function of internuclear distance R (a.u.) at intensity $I = 2 \times 10^{14} \text{ W/cm}^2$ for a linearly polarized pulse, wavelength $\lambda = 800 \text{ nm}$ ($\omega_L = 0.0567 \text{ a.u.}$, $\tau_L = 2.67 \text{ fs}$) pulse duration five cycles. The circularly polarized pulse is in the (x, y) plane of the molecule perpendicular to the internuclear (z) R axis. The linearly polarized pulse has its electric field parallel or perpendicular to the (z) R axis. The parallel linearly polarized pulse results (+) show two maxima at $\lambda = 800 \text{ nm}$ with ionization rates $\sim 1 \times 10^{13} \text{ s}^{-1}$. Since $I_p(\text{H}(1s)) = 0.5 \text{ a.u.}$, (2.9) predicts $R_c \sim 8 \text{ a.u.}$ The first peak at $R \simeq 6 \text{ a.u.}$ in Fig. 2.3 a.u. corresponds to a photon resonance at energy $\sim 10 \text{ eV}$ for the ground $\sigma_g(1s)$ to excited $\sigma_u(1s)$

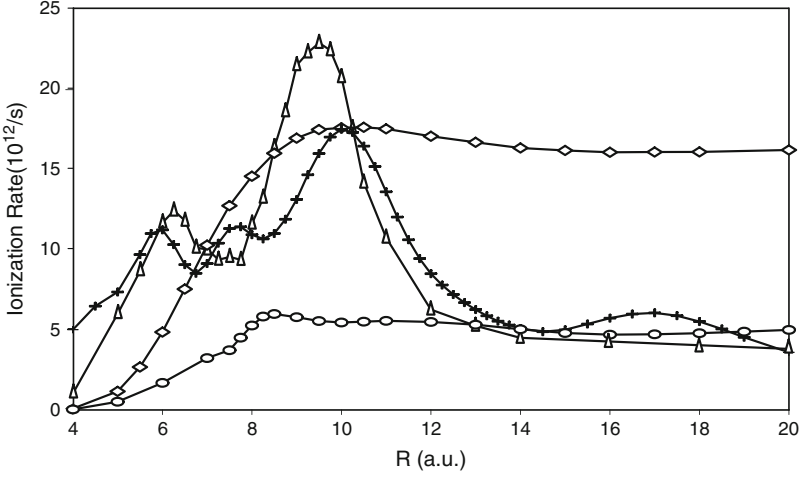


Fig. 2.3 Ionization rates for H_2^+ vs R for (i) $I = 10^{14} \text{ W/cm}^2$, linear polarization parallel to R (z-axis): (a) (plus): $\lambda = 800 \text{ nm}$; (b) (open triangle): $\lambda = 1,064 \text{ nm}$, and (c) (open circle): $\lambda = 800 \text{ nm}$, perpendicular to R and (ii) $I = 2 \times 10^{14} \text{ W/cm}^2$, circular polarization in x - y plane, $\lambda = 800 \text{ nm}$: (open diamond)

CR transition in H_2^+ [39–42]. Clearly, the region $R \geq 8 \text{ a.u.}$ is dominated by atomic transitions for perpendicular linear and circularly ($\diamond$) polarized pulses which are constant for large R . For parallel linearly polarized pulses the double CREI maximum at $8 \leq R \leq 10 \text{ a.u.}$ at $\lambda = 800 \text{ nm}$ (+) becomes a single maximum at longer wavelength $\lambda = 1,064 \text{ nm}$ (Δ) thus eliminating multiphoton resonances and rendering the static model of (2.8)–(2.9) more rigorous. Thus in Fig. 2.3 clear maxima at $R = 6 \text{ a.u.}$ with ionization rate $\sim 10^{13} \text{ s}^{-1}$ and $R = 9 - 10 \text{ a.u.}$ with rate $2 \times 10^{13} \text{ s}^{-1}$ correspond to lifetimes of $\leq 100 \text{ fs}$ with $I'_p \cdot s = 0.5 + \frac{1}{R} \sim 0.7 - 0.6 \text{ a.u.}$ ($\sim 20 - 16 \text{ eV's}$)

The nonsymmetric HeH^{2+} one-electron molecular system contrary to the symmetric H_2^+ system is influenced strongly by a permanent dipole moment in the ground state configuration $\text{He}^+ (1s) \text{ H}^+$ which changes polarity in the first excited state $\text{He}^{++} \text{ H} (1s)$ following charge transfer [47, 48]. The resulting ionization probabilities are illustrated in Fig. 2.4 at intensities $I = 10^{15}$, 3×10^{15} , and $5 \times 10^{15} \text{ W/cm}^2$ for CEP's $\phi = 0$ and π , i.e., for $E_m > 0$ and < 0 , respectively. As predicted by (2.10) and illustrated in Fig. 2.2, only for negative field $E_m < 0$, $\phi = \pi$, the HOMO ($\text{He}^+(1s)$) and LUMO ($\text{H}(1s)$) cross, resulting in enhanced ionization at $4 \leq R_c \leq 6 \text{ a.u.}$ for intensities $5 \times 10^{15} \geq I \geq 10^{15} \text{ W/cm}^2$. Such field-amplitude-dependent ionization rates demonstrate the importance of laser control of charge transfer, CR, at high intensities in nonsymmetric molecules. Enhanced ionization predicted theoretically [22–24] has been confirmed experimentally [21], in diatomics and complex molecules [56]. Furthermore, such a quasistatic model of Coulomb explosion of clusters [57] suggests that enhanced ionization may be

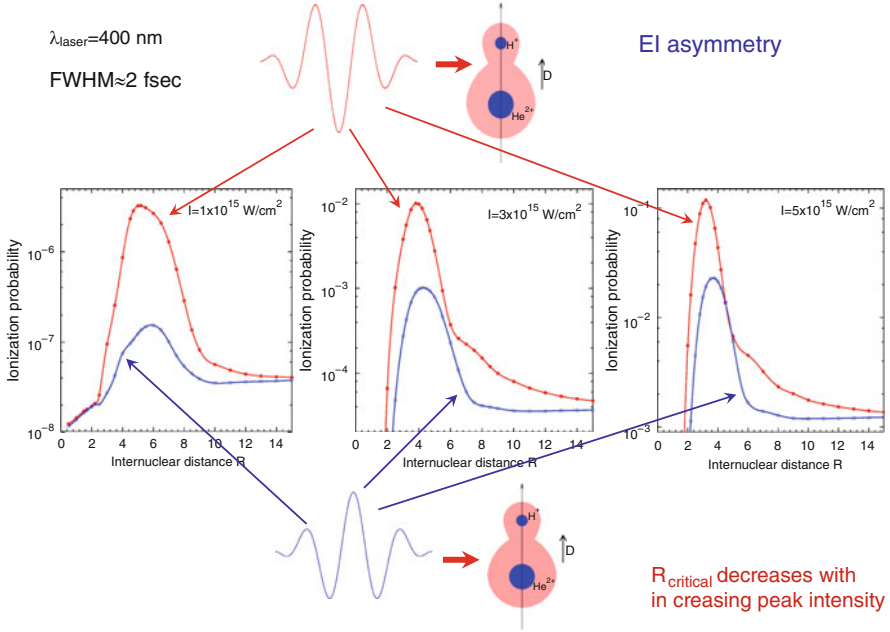


Fig. 2.4 HeH_2^+ ionization probabilities for: fields $E(\tau, \phi)$ and CEP $\phi = 0$ (blue), π (red) at peak intensities $I = 10^{15}$, 3×10^{15} , and $5 \times 10^{15} \text{ W/cm}^2$, as a function of internuclear distance R with maxima at R_c (R_{critical})

a mechanism for laser fusion [58, 59]. In nonsymmetric molecules, the strong orientation and CEP dependence of enhanced ionization as shown in simulations of one-electron nonsymmetric molecular systems suggest applications in the control of ionization of complex molecules [60], in the laser-assisted population inversion in heavy ion collisions for X-ray laser production [61] and finally in the imprinting of nanostructures in heteroatomic lattices with ultrashort intense few-cycle pulses [62]. With the extension of the CREI model to one-electron nonlinear molecules such as H_3^{2+} [63], multi-center systems [64] have predicted this nonlinear nonperturbative phenomenon to be unique to molecules, i.e., over-barrier ionization results in complete ionization at critical distances R_c . Nuclear motion of H_2^+ for which the vibrational period $\tau_v \sim 10 \text{ fs}$ has the tendency to “wash out” the double peak structure of the ionization shown in Fig. 2.3 [65, 66]. Exact non-Born-Oppenheimer simulations of one-dimensional one-electron model systems H_2^+ and H_3^{2+} with quantum moving nuclei show that coherent excitations of the HOMO and LUMO occur during the nuclear motion at the critical distances R_c [67]. Such coherent efforts during CREI resulting in two-surface electron population dynamics have been observed recently by *asec* strobing [68] and modeled successfully with non-Born-Oppenheimer electron—nuclear TDSE’s [69]. Such non-adiabatic simulations have shown highly correlated electron-nuclear motion in dissociating H_2^+ [70] resulting

in multiple ionization bursts on *asec* time scale due to strong field-modulated diffraction effects [71] due to laser-induced electron diffraction, LIED [72].

2.3 Double Ionization in Strong Fields

In Sect. 2.2, we have described quasistatic above-barrier simple electron ionization in one-electron molecules for which analytic models were obtained to derive expressions for the critical distance R_c for CREI in symmetric molecules and enhanced ionization in nonsymmetric molecules. Double ionization introduces a new phenomenon—nonsequential double ionization, NSDI, observed as early as 1992 with optical tunnelling of He [73] and explained based on the 3-step model of tunneling ionization, where electron propagation in an intense laser pulse is followed by recollision with maximum energy $3.17 U_p$ [4]. Tunneling ionization imposes the initial velocity condition $\dot{z}(t_0) = 0$ at time t_0 . This model has been generalized to nonzero initial velocity $\dot{z}(t_0) \neq 0$ where it was shown that the return collision energy E_c never exceeds $3.17 U_p$ i.e., $E_c \leq 3.17 U_p$ [28]. Collision with neighboring ions can, however, lead to energies exceeding $E_c > 3.17 U_p$ and is R dependent [57, 74–76]. In such processes, charge transfer or ionic states, such as $H^+ H^-$ in H_2 , $O^+ O^-$ in O_2 , produce very large transition moments emphasized early by Mulliken [20]. Ab initio calculations of H_2 in strong fields have proven the existence of $H^+ H^-$ as a precursor to enhanced ionization [77–79] as well as in the triatomic H_3^+ in its linear [80] and nonlinear [81] geometry. 1-D non-Born-Oppenheimer simulations of H_2 have also identified the CR state $H^+ H^-$ as an important doorway state in the Coulomb explosion of this system [82]. The presence of a pair of electrons implies considerable electron correlation in the nonlinear nonperturbative response of molecules to intense ultrashort pulses [83–85]. However in molecules, charge transfer in ionic states such as $H^+ H^-$ and $O^+ O^-$ as recently suggested in O_2 time-resolved dissociative ionization [86] implies dipole moments which will introduce large Stark-shifts in the molecular potentials at high laser intensities. It was found earlier that the dipole moment of asymmetric charge distributions interacts with intense external fields leading to charge asymmetric dissociation of highly charged ions [87]. Ionization from these asymmetric charge distributions can lead to shake up excitation of the remaining bound electrons [88] by interaction with the ionized electron. CEP sensitivity of these charge transfer, CR, processes has been now confirmed in dissociative ionization of CO [89], and correlated two-electron ab initio calculations in H_2 show evidence of enhanced excitation in single and double ionization at the CREI critical distance R_c [84]. Ionic states act therefore as doorway states for intense laser-field-enhanced ionization of two-electron molecules. For H_2 , one must consider at least the two electronic states, $1\sigma_g^2(X^1\Sigma_g^+)$, $1\sigma_g 1\sigma_u(B^1\Sigma_u^+)$ and $1\sigma_u^2(E, F^1\Sigma_q^+)$ electronic configurations [77–79, 84]. The asymptotic dissociation products ϕ_1 , ϕ_2 , and ϕ_3 for these states are expressed in terms of 1s atomic orbitals a and b on each H atom [90],

$$\phi_1(1, 2) = [a(1) a(2) + b(1) b(2) + a(1) b(2) + b(1) a(2)]/2, \quad (2.11)$$

$$\phi_2(1, 2) = [a(1) a(2) - b(1) b(2)]/\sqrt{2} \quad (2.12)$$

$$\phi_3(1, 2) = [a(1) a(2) + b(1) b(2) - a(1) b(2) - b(1) a(2)]/2 \quad (2.13)$$

Both ground ϕ_1 and excited ϕ_3 states are linear combinations of *ionic* (aa, bb) and *covalent* (ab) configurations whereas ϕ_2 is purely ionic. The electronic transition moments between these states are thus coupled by the same radiative matrix element $ER/\sqrt{2}$ for a field strength E (intensity $I = cE^2/8\pi$). Diagonalization of the 3×3 energy matrix yields readily the *adiabatic* energies and states [78]

$$\varepsilon_0 = 0, \phi_0 = [a(1) b(2) + b(1) a(2)]/\sqrt{2} \quad (2.14)$$

$$\varepsilon_+ = ER, \phi_+ = b(1) b(2), \quad (2.15)$$

$$\varepsilon_- = -ER, \phi_- = a(1) a(2), \quad (2.16)$$

The ground state is ϕ_- corresponding to the ionic state $H_a^- H_b^+$ whose energy agrees with the electrostatic energy of a charge displaced (transferred) through the distance R by a field of amplitude E . The ionic states ϕ_-, ϕ_+ acquire additional Coulomb energy. Thus, if we consider the general charge transfer $A^{q+} A^{q+} A^{(q-1)+} A^{(q+1)+}$, the initial total energy is

$$\varepsilon_{q,q}(R) = -2I_p(q) + q^2/R, \quad (2.17)$$

where $I_p(q)$ is the ionization potential of the ion A^{q+} and q^2/R is the ion-ion repulsion. The final energy of the charge transfer state in the field E is

$$\varepsilon_{q-1,q+1}(R) = -I_p(q-1) - I_p(q) + (q+1)(q-1)/R - ER, \quad (2.18)$$

and the energy difference is

$$\Delta\varepsilon(R) = \Delta I_p - 1/R - ER, \quad (2.19)$$

where $\Delta I_p = I_p(q) - I_p(q-1)$. The critical field strength E_c required for the crossing of the covalent $A^{q+} A^{q+}$ and ionic $A^{(q-1)+} A^{(q+1)+}$ state at critical distance R_c is determined from $\Delta\varepsilon(R_c) = 0$

$$E_c = (\Delta I_p - 1/R_c)/R_c. \quad (2.20)$$

Solutions of the TDSE for H_2 in 1-D [78], and 3-D [84] show in fact that the ionic state $H_a^- H_b^+$ due to its large dipole moment indeed becomes the ground state for fields $E > E_c$ and that the ionic state created determines the ionization process. In H_2 since $\Delta I_p = I_p(H) - I_p(H^-) = 0.47$ a.u., one obtains $R_c = 5$ a.u. at intensity $I = 10^{14}$ W/cm² ($E = 0.053$ a.u.) in agreement with 3-D TDSE's [84]. We note that a similar model of charge transfer at critical distance R_c for critical fields E_m

applies to nonsymmetric molecules and is derived in [46] giving (2.10). The charge transfer model for H_2 , (2.11)–(2.20), has been applied to linear H_3^{2+} , H_4^{3+} , H_4^{2+} [61], H_3^+ with static [80] and H_3^{2+} [67] and H_2 [91] with moving (non-Born-Oppenheimer) nuclei. In the latter non-Born-Oppenheimer simulations, coherent excitations of the HOMO and LUMO in the CREI region occur and influence both the ATI spectra as well as the Coulomb explosion proton kinetic energies. The presence of such charge asymmetric states has been shown to occur especially for highly charged states at high intensities, $I \geq 5 \times 10^{14} \text{ W/cm}^2$ in I_2 leading to both charge symmetric and asymmetric dissociation products [87].

Non-symmetric molecules as discussed for the one-electron HeH^{2+} have permanent dipole moments [47, 48] with the effect that enhanced ionization is CEP sensitive as illustrated in Figs. 2.2 and 2.4. This effect persists in the two-electron case [92], HeH^+ with the two configurations $He-H^+$ and He^+-H with dipole moments $4R/5$ and $-R/5$, respectively. In Fig. 2.5a we report the total single, P_{SI} and double, P_{DI} , ionization probabilities at intensity $I = 10^{15} \text{ W/cm}^2$ and wavelength $\lambda = 800 \text{ nm}$ ($\omega = 0.057 \text{ a.u.}$), obtained by projection on all bound electronic states. Figure 2.5b reports the accompanying single, P_{SE} and double P_{DE} , electron excitations obtained by projecting the exact 6-D two-electron wavefunction on appropriate field-free electronic states of the HeH^+ molecule [92]. Figure 2.5 shows clearly enhancement of one- and two-electron ionizations and excitations at a critical distance $3 \leq R_c \leq 4 \text{ a.u.}$ We now use as in previous sections a simple quasistatic one-electron approximation model to derive an analytical expression for R_c of HeH^+ ionization in intense fields. At large R , the field-free one-electron ground state energy level of HeH^+ becomes $\epsilon_1 = -I_p(H) - Z_1/R$ corresponding to $He-H^+$ configurations [93]. The first excited state He^+-H has energy $\epsilon_2 = -I_p(H) - Z_1/R$. In the presence of a negative electric field, $|E|$, ϵ_1 and ϵ_2 are Stark-shifted via the dipole moments of HeH^+ and He^+H respectively:

$$\epsilon_1 = -I_p(He) - Z_2/R + (4R/5)|E_m| \quad (2.21)$$

$$\epsilon_2 = -I_p(H) - Z_1/R - (R/5)|E_m|. \quad (2.22)$$

Setting $Z_1 = Z_2 = 1$ corresponds to the nuclear charges of He^+ and H^+ . Crossing of the two electronic states with energies ϵ_1 and ϵ_2 occurs for negative E_m since $I_p(He) > I_p(H)$, resulting in efficient over-barrier ionization as in HeH^{2+} [47]. This crossing defines the critical distance for enhanced ionization

$$R_c = (I_p(He) - I_p(H)) / |E_m|, \quad (2.23)$$

as a function of the field E_m . At intensity $I = 10^{15} \text{ W/cm}^2$, $E_m = 0.17 \text{ a.u.}$ and $R_c = 3 \text{ a.u.}$ in good agreement with Fig 2.5a [92]. Adding polarizabilities of He and H as suggested recently [94] can be shown to have negligible effect. Equations (2.21)–(2.22) show that it is the permanent dipole moment and the electric field which dominate the nonlinear nonperturbative ionization of asymmetric molecules.

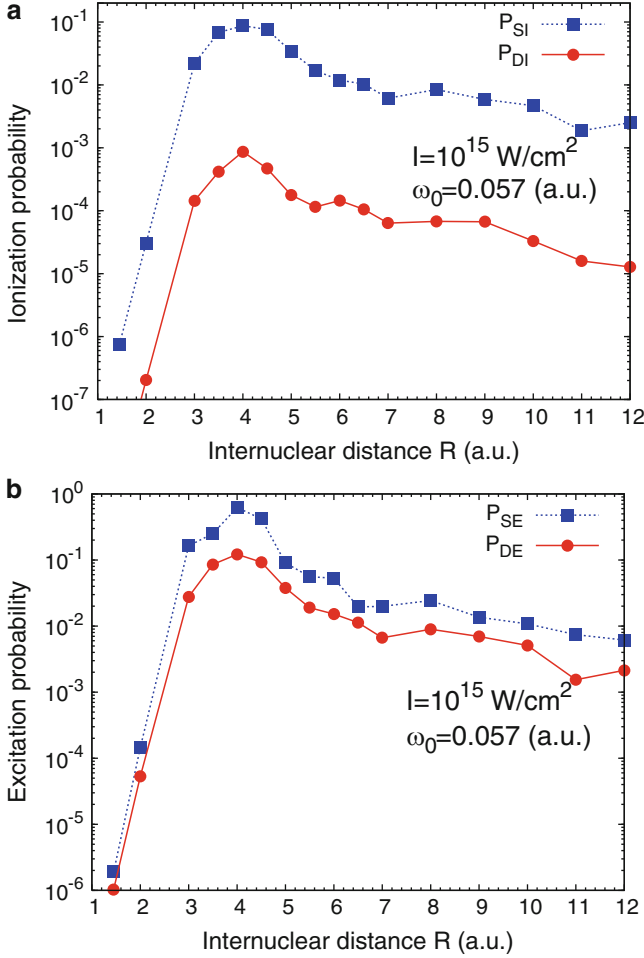


Fig. 2.5 Ionization and excitation probabilities for HeH^+ as a function of internuclear distance R at intensity $I = 10^{15} \text{ W/cm}^2$, $\omega_0 = 0.057 \text{ a.u.}$ ($\lambda = 800 \text{ nm}$). (a) total single P_{SI} and double P_{DI} ionization; (b) total single P_{SE} and double P_{DE} excitation

2.4 Coulomb Explosion Imaging of CREI in Triatomics

Time-resolved Coulomb explosion imaging (CEI) is now an established technique to make molecular movies with sub-Angstrom spatial resolution and sub-5fs temporal resolution [95,96]. CEI of diatomics has shown that molecular bonds stretch towards a critical internuclear distance R_c discussed in the previous sections, where ionization rates are greatly enhanced [21]. This also explains the explosion of large clusters creating highly charged states [57] which can catalyze nuclear fusion [58, 59]. In symmetric diatomics, one-electron systems such as $\text{H}_2^+/\text{D}_2^+$, the enhancement of

ionization is explained in Sect. 2.2 as CREI in terms of charge resonance, CR, states which are strongly coupled by the laser field at large internuclear distances called critical distances R_c [21–24]. At such internuclear distances, the energy difference between two CR states approaches the incident high intensity photon energy $h\nu$ giving rise to electron *localization* by creating a coherent superposition of opposite parity states [67–71]. For triatomic molecules such as CO_2 , the experimental confirmation of a critical geometry for CREI has been recently reported by CEI [97]. Previous experimental results for CO_2 [98, 99] have been compared to over-barrier ionization models discussed for diatomics in previous sections. Numerical simulations of nuclear motion on field dressed molecular potentials such as CO_2^{2+} but without ionization have revealed bending and symmetric stretching on a time scale of ~ 100 fs [60]. Using 3-D ion momentum coincidence measurements of the three-body dissociative ionization of CO_2 to CO_2^{n+} ($n = 3 - 6$) with few-cycle linearly polarized 800-nm laser pulses, a systematic study of dissociative multiple ionization dynamics has been achieved as a function of pulse duration [97]. This has been supplemented by nonlinear nonperturbative time-dependent density functional theory (TDDFT) calculations [100] allowing for the identification of the relevant CR states and thus the underlying mechanism for CREI in CO_2 . It has been previously demonstrated for diatomics that by using few-cycle pulses, CREI can be suppressed since the molecular ion does not have time to reach R_c within the duration of the pulse [101].

Figure 2.6 illustrates the kinetic energy release, KER of the charge state of CO_2 as a function of pulse duration for the $(1, 1, 1 \div \text{O}^+ + \text{C}^+ + \text{O}^+)$, $(1, 1, 2 \div \text{O}^+ - \text{C}^+ - \text{O}^{2+})$, $(1, 2, 2 \div \text{O}^+ - \text{C}^{2+} - \text{O}^{2+})$, and finally, $(2, 2, 2 \div \text{O}^{2+} - \text{C}^{2+} - \text{O}^{2+})$ channels. It is observed that the KER becomes significantly dependent on the pulse duration only for the $4+$ and higher charge states. With 7 fs duration and for final charge states higher than $3+$, the observed KER is close to the value expected for Coulomb explosion from the CO_2 equilibrium geometry. For longer pulse duration, the total KER impact decreases and becomes almost independent for pulse duration $\tau > 100$ fs [97]. Similar results occur for the KER pulse duration of D_2 with pulse duration > 40 fs [101]. In the D_2 case, the KER from Coulomb explosion becomes independent of pulse duration once the molecular ion stretches to R_c where CREI is known to be operative. Simulation of the Coulomb explosion of the $(2, 2, 2)$ channel using the measured moments and a numerical data inversion algorithm [102] allows to retrieve the molecular structure at 7 fs, ($\langle R_{\text{CO}} \rangle = 1.3 \text{ \AA}$ and $\langle \theta_{\text{OCO}} \rangle = 168^\circ$) very close to the equilibrium geometry ($\langle R_{\text{CO}} \rangle = 1.16 \text{ \AA}$ and $\langle \theta_{\text{OCO}} \rangle = 172^\circ$) [102]. For a pulse duration longer than 100 fs, the experimental observations lead to a molecular structure of ($\langle R_{\text{CO}} \rangle = 2.1 \text{ \AA}$ and $\langle \theta_{\text{OCO}} \rangle = 163^\circ$) at which CREI occurs.

Since the mechanism for CREI relies on strong radiative interactions between “frontier” orbitals (Sects. 2.2–2.3), HOMO’s and LUMO’s, nonlinear nonperturbative TDDFT calculations for CO_2 [100] have been used to calculate the ionization rates and relevant electronic transition moments as a function of R_{CO} for intensities $I = 8 \times 10^{14} \text{ W/cm}^2$, $\lambda = 800 \text{ nm}$ and $\tau = 6$ optical cycles. The numerical results are summarized in Fig. 2.7. In Fig. 2.7a, perpendicular and parallel to

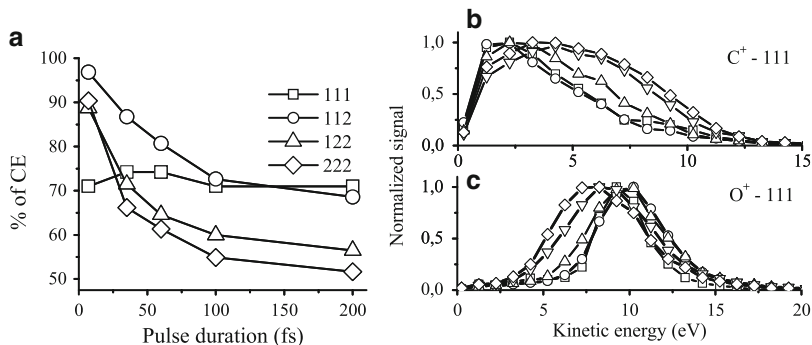


Fig. 2.6 Experimental kinetic energy release, KER, spectra as a function of laser pulse duration: (a) ratio of KER vs energy at equilibrium ($R_{CO} = 1.16 \text{ \AA}$), (b) C^+ , and (c) O^+ : KER for (1,1,1 charge state). Pulse duration and intensity: open square—7 fs, 5×10^{14} ; open circle—35 fs, 5×10^{14} ; open triangle—60 fs, 3×10^{14} ; inverted triangle—100 fs, 3×10^{14} ; open diamond—200 fs, $3 \times 10^{14} \text{ W/cm}^2$

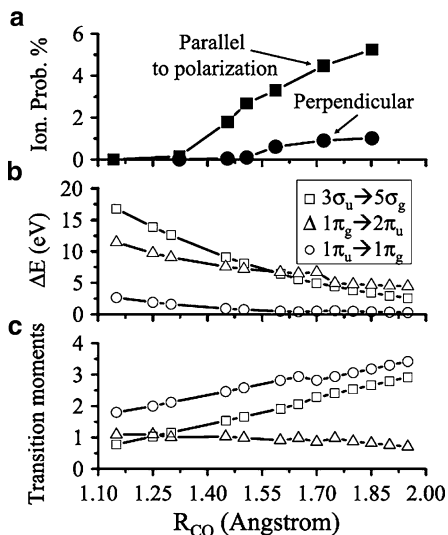


Fig. 2.7 TDDFT simulations (a) total ionisation probability for parallel and perpendicular orientations, (b) Energy gaps between KS orbitals, (c) transition moments (a.u.) for parallel transitions as a function of R_{CO}

laser polarization ionization rates confirm the dominance of parallel ionization. Figure 2.7b shows the energy separation of relevant orbitals as a function of R_{CO} . and Fig. 2.7c reports the corresponding transition moments. Of note is that the $1\pi_u$ (HOMO-2) $\rightarrow$ $1\pi_g$ (HOMO) transition behaves as a CR transition with a transition moment which scales as $R_{CO}/2$ between the antibonding $1\pi_g$ and the inner shell bonding $1\pi_u$. These two orbitals become nearly degenerate at $R_{CO} > 2 \text{ \AA}$

with an $R/2$ transition moment, a signature of CREI. Another similar dominant CR transition is the $3\sigma_u$ (HOMO-1) to $5\sigma_g$ (LUMO). The numerical simulations, limited to $R_{CO} \leq 2$ a.u. because of energy degeneracy of orbitals for large R_{CO} , nevertheless show that it is the $3\sigma_u$ orbital which contributes mostly to the ionization as its density is mainly localized along the internuclear axis. Even though the ionization potential I_p of the $3\sigma_u$ orbital is higher than that of the $1\pi_g$, its ionization rate is larger due to its geometry, a general phenomenon at high intensities [100].

To summarize, as the CO_2^{3+} breaks apart through dissociative ionization, the numerical simulations confirm that the ionization rate increases rapidly when the molecule is aligned along the laser polarization axis because of strong, nonperturbative radiative coupling of the main CR states, $3\sigma_u \rightarrow 5\sigma_g$. The increasing ionization rate, linearly varying transition moments and the increasing degeneracy of orbitals as a function of R_{CO} in Fig. 2.7 allow us to characterize the dynamics of the dissociative ionization as due to CREI. This requires CO_2^{3+} to stretch with a time equal to or longer than 100 fs to the critical distance $R_{CO} > 2$   . For shorter pulses, CREI is suppressed due to less ionization at shorter distances.

In conclusion, in spite of highly nonlinear, nonperturbative response of molecules to extreme intense electromagnetic pulses, simple classical models have been the guiding models for understanding laser induced electron recollision, LIERC [4, 28, 103] laser imprinting of nanostructures [62], molecular ATI [67], laser induced electron diffraction LIED [72, 104], and charge-resonance-enhanced ionization CREI, reviewed in this chapter. Other examples are HHG [76, 105], time-resolved photoelectron holography [106] and in many cases, controlling the physical processes that occur in this new nonperturbative highly nonlinear regime of laser molecule interaction [107].

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References

1. T. Brabec, F. Krausz, *Rev. Mod. Phys.* **72**, 545 (2000)
2. P. Corkum, F. Krausz, *Nature Phys.* **3**, 381 (2007)
3. L. Keldysh, *Sov. Phys. JETP* **20**, 1307 (1965)
4. P.B. Corkum, *Phys. Rev. Lett.* **71**, 1994 (1993)
5. P.B. Corkum, N.H. Burnett, F. Brunel, *Phys. Rev. Lett.* **62**, 1259 (1989)
6. P.B. Corkum, *Phys. Today* March 36 (2011)
7. A.J. Verhaef et al., *Phys. Rev. Lett.* **104**, 163904 (2010)
8. F. Krausz, M.Y. Ivanov, *Rev. Mod. Phys.* **81**, 163 (2009)
9. J.I. Steinfeld, *Molecules and Radiation* (MIT Press, Cambridge, USA, 1985)
10. H. Lefebvre-Brion, R.W. Field, *Spectra and Dynamics of Diatomic Molecules* (Elsevier, Holland, 2004)
11. W. Greiner, B. Muller, J. Rafelski, *Quantum Electrodynamics of Strong Fields: Introduction to Modern Relativistic Quantum Mechanics* (Springer, Berlin, 1985)

12. S.S. Bulanov et al., Phys. Rev. Lett. **104**, 220404 (2010)
13. A.H. Zewail, J. Phys. Chem. A. **104**, 5660 (2000)
14. A.D. Bandrauk, S. Barmaki, S. Chelkowski, G.L. Kamta, in “*Progress in Ultrafast Intense Laser Science*”, vol III, Chap. 9, ed. By K. Yamanouchi et al. (Springer, N. Y., 2007)
15. S. Chelkowski, G. Yudin, A.D. Bandrauk, J. Phys. B. **39**, S409 (2006)
16. A.D. Bandrauk, J. Manz, M. Vrakking, Chem. Phys. **366**, 1 (2009)
17. Z. Chang, “*Fundamentals of Attosecond Optics*” (CRC Press, Taylor-Francis, USA, 2011)
18. A. Zavriyev, P.H. Bucksbaum, in “*Molecules in Laser Fields*”, Chap. 2, ed. by A.D. Bandrauk (Marcel Dekker Inc., NY, 1994)
19. A.D. Bandrauk et al, in “*Molecules in Laser Fields*”, Chap. 4, ed. by A.D. Bandrauk (Marcel Dekker Inc., NY 1994)
20. R.S. Mulliken, J. Chem. Phys. **7**, 20 (1939)
21. C. Cornaggia in “*Progress in Ultrafast Intense Laser Science*”, vol VI, Chap. 1, ed. by K. Yamanouchi et al (Springer, Tokyo 2011)
22. T. Zuo, A.D. Bandrauk, Phys. Rev. A **54**, 3254 (1996)
23. S. Chelkowski, A.D. Bandrauk, J. Phys. B **28**, L723 (1995)
24. T. Seideman, M.Y. Ivanov, P.B. Corkum, Phys. Rev. Lett. **75**, 2819 (1995)
25. N.B. Delone, V.P. Krainov, “*Multiphoton Processes in Atoms*” (Springer, Berlin, 1994)
26. M.J. DeWitt, R.J. Levis, J. Chem. Phys. **108**, 1, (1998)
27. T.F. Gallagher, Phys. Rev. Lett. **61**, 2304 (1988)
28. S. Chelkowski, J. Goudreau, A.D. Bandrauk, J. Mod. Opt. **52**, 411 (2005)
29. A.D. Bandrauk, S. Chelkowski, K.J. Yuan, Intl. Rev. Atom Molec. Phys. (2011)
30. G. Mourou, T. Tajima, Science **331**, 41 (2011)
31. U.W. Rathe, C.H. Keittel, M. Protopapas, P.L. Knight, J. Phys. B **30**, L531 (1997)
32. S. Selsto, E. Lindroth, J. Bengtsson, Phys. Rev. A **79**, 043418 (2009)
33. A. Maquet, R. Grobe, J. Mod. Opt. **49**, 2001 (2002)
34. A.D. Bandrauk, H. Kono, in “*Advances in Multiphoton Processes*”, vol 15, ed. by S.H. Lin (World Scientific, Singapore, 2000)
35. P. Dietrich, M.Y. Ivanov, F.A. Ilkov, P.B. Corkum, Phys. Rev. Lett. **77**, 4150 (1996)
36. K. Codling, L.J. Frasinski, P.A. Hatherly, J. Phys. B **22**, L321 (1989)
37. J.H. Posthumus et al., J. Phys. B. **28**, L 349 (1995)
38. J.H. Posthumus, Rep. Prog. Phys. **67**, 623 (2004)
39. H. Schröder, C.J. Uiterwaal, K.L. Kompa, Laser Phys. **10**, 749 (2000)
40. G.L. Kamta, A.D. Bandrauk, Phys. Rev. A **75**, 041401 (2007)
41. H.Z. Lu, A.D. Bandrauk, J. Molec. Str. (TheoChem) **547**, 97 (2001)
42. H. He et al, Phys. Rev. A. **84**, 033418 (2011)
43. H. Sabzyan, M. Vafae, Phys. Rev. A. **71**, 063404 (2005)
44. M. Schmidt, D. Normand, C. Cornaggia, Phys. Rev. A. **50**, 5037 (1994); *ibid*, **53**, 1958 (1996)
45. G.N. Gibson, M. Li, C. Guo, J. Neira, Phys. Rev. Lett. **79**, 2022 (1994)
46. K. Yamanouchi, Science **295**, 1659 (2002)
47. G.L. Kamta, A.D. Bandrauk, Phys. Rev. Lett. **94**, 203003 (2005); Phys. Rev. A **76**, 053409 (2007)
48. X.B. Bian, A.D. Bandrauk, Phys. Rev. Lett. **105**, 093903 (2010)
49. Y.J. Jiu, X.M. Tong, N. Toshima, Phys. Rev. A. **81**, 013408 (2010)
50. A.D. Bandrauk, H.Z. Lu, Phys. Rev. A. **62**, 053406 (2000)
51. D. Pavicic, A. Kiess, T.W. Hänsch, H. Figger, Phys. Rev. Lett. **94**, 163002 (2005)
52. K. Sandig, H. Figger, T.W. Hänsch, Phys. Rev. Lett. **85**, 4876 (2000)
53. V. Serov et al., Phys. Rev. A. **72**, 033413 (2005)
54. E. Constant, H. Stapelfeldt, P.B. Corkum, Phys. Rev. Lett. **76**, 4140 (1996)
55. J.N. Gibson, M. Li, C. Zuo, J. Neira, Phys. Rev. Lett. **79**, 2002 (1997)
56. A. Hishikawa, A. Iwamae, K. Yamanouchi, Phys. Rev. Lett. **83**, 1127 (1999)
57. C. Siedschlag, J.M. Rost, Phys. Rev. A **60**, 2215 (1999); Phys Rev Lett. **89**, 173401 (2002); Few-Body Systems, **31**, 211 (2002)
58. I. Last, J. Jortner, Phys. Rev. A. **60**, 2215 (1999)

59. A.D. Bandrauk, G. Paramonov, AIP Conf. Proc. **1209**, 7 (2010)
60. Y. Sato, H. Kono, S. Koseki, Y. Fujimura, J. Am. Chem. Soc. **125**, 8019 (2003)
61. Y. Nagata et al., Phys. Rev. Lett. **71**, 3774 (1993)
62. L.N. Gaier, J. Mod. Opt. **52**, 1019 (2005)
63. A.D. Bandrauk, J. Ruel, Phys. Rev. A. **59**, 2153 (1999)
64. H. Yu, T. Zuo, A.D. Bandrauk, J. Phys. B. **31**, 1533 (1998)
65. I. Ben-Itzhak et al., Phys. Rev. A. **78**, 063419 (2008)
66. M. Ergler et al., Phys. Rev. Lett. **95**, 093001 (2005)
67. A.D. Bandrauk, S. Chelkowski, I. Kawata, Phys. Rev. A. **67**, 013407 (2003)
68. A. Staudte et al., Phys. Rev. Lett. **98**, 073003 (2007)
69. S. Chelkowski et al., Phys. Rev. A. **76**, 013405 (2007)
70. F. He, A. Becker, U. Thumm, Phys. Rev. Lett. **101**, 213002 (2008)
71. N. Takemoto, A. Becker, Phys. Rev. Lett. **105**, 203004 (2010), Phys Rev A **84**, 023401 (2011)
72. T. Zuo, A.D. Bandrauk, P. Corkum, Chem. Phys Lett. **259**, 313 (1996)
73. D. N. Fittinghoff, P.R. Bolton, B. Chang, R.C. Kulander, Phys. Rev. Lett. **69**, 2642
74. A.D. Bandrauk, S. Chelkowski, H. Yu, E. Constant, Phys. Rev. A. **56**, 2537 (1997)
75. P. Moreno, L. Playa, L. Roso, Phys. Rev. A. **55**, 1593 (1997)
76. A.D. Bandrauk, S. Barmaki, G.L. Kamta, Phys. Rev. Lett. **98**, 013001 (2007)
77. H. Yu, T. Zuo, A.D. Bandrauk, Phys. Rev. A **54**, 3290 (1996); J Phys B **31**, 1533 (1998)
78. I. Kawata et al., Phys. Rev. A **62**, 031401 (2000); **66**, 043403 (2002)
79. A. Saenz, Phys. Rev. A **61**, 051402 (2000); **66**, 063407 (2002)
80. I. Kawata, H. Kono, A.D. Bandrauk, Phys. Rev. A. **64**, 043411 (2001)
81. D. Tchitchekova, S.C. Chelkowski, A.D. Bandrauk, J. Phys. B. **44**, 065601 (2011)
82. S. Saugaut et al., Phys. Rev. Lett. **98**, 253003 (2007)
83. S. Sukiasyan et al., Phys. Rev. Lett. **102**, 223002 (2009)
84. E. Dehghanian, A.D. Bandrauk, G.L. Kamta, Phys. Rev. A. **81**, 061403 (2010)
85. A. Tong et al., Opt. Exp. **18**, 9064 (2010)
86. S.A. Trushin, W.E. Schmid, W. Fuss, J. Phys. B. **44**, 165602 (2011)
87. G.N. Gibson, M. Li, C. Guo, J.P. Nibarger, Phys. Rev. A. **58**, 4123 (1998)
88. I.V. Litvinyuk et al., Phys. Rev. Lett. **94**, 033003 (2005)
89. Y. Liu et al., Phys. Rev. Lett. **106**, 073004 (2011)
90. J.C. Slater, *Quantum Theory of Molecules and Solids*, vol I (McGraw-Hill, NY, 1963)
91. S. Chelkowski, A.D. Bandrauk, Phys. Rev. Lett. **101**, 153901 (2008)
92. E. Dehghanian, A.D. Bandrauk, G.L. Kamta, Phys. Rev. A **81**, 061403(R) (2010)
93. T.A. Green et al., J. Chem. Phys. **61**, 5156 (1974)
94. A. Etches, L.B. Madsen, J. Phys. B. **43**, 155602 (2005)
95. F. Légaré et al., Phys. Rev. A **72**, 052717 (2005); **71**, 013415 (2005)
96. A. Hishikawa et al., Phys. Rev. Lett. **99**, 258302 (2007)
97. I. Bocharova et al., Phys. Rev. Lett. **107**, 063201 (2011)
98. A. Hishikawa, A. Iwamae, K. Yamanouchi, Phys. Rev. Lett. **83**, 1127 (1999)
99. J.P. Brichta, S.J. Walker, R. Helsten, J.H. Sanderson, J. Phys. B. **40**, 117 (2007)
100. E.P. Fowe, A.D. Bandrauk, Phys. Rev. A. **81**, 023411 (2010)
101. F. Légaré et al., Phys. Rev. Lett. **91**, 093002 (2003)
102. J.P. Brichta, A.N. Seaman, J.H. Sanderson, Comput. Phys. Commun. **180**, 197 (2009)
103. M. Okunishi et al., J. Chem. Phys. **127**, 064310 (2007)
104. M. Meckel et al., Science **320**, 1478 (2008)
105. Y. Chen, J. Chen, J. Liu, Phys. Rev. A. **74**, 063405 (2006)
106. Y. Huismans et al., Science **331**, 61 (2011)
107. M. Abel et al., Laser Photonics Rev. **5**, 352 (2011)

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