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Non-sequential double ionization of CO_2 molecule and Kr atom in linearly polarized laser fields

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Abstract. Using two-dimensional classical ensemble method, a theoretical study of non-sequential double ionization (NSDI) with Krypton (Kr) and carbon dioxide (CO_2) is presented at different laser intensities. The numerical results show that the probability for NSDI of Kr atom is higher than that of CO_2 molecule. Moreover, for the same laser intensity, the momentum correlation spectrum of CO_2 molecule is drastically different from Kr atom. For example, for the laser intensities I = 0.065 PW/cm^2 and I =0.15 PW/cm^2, the correlation spectrum of CO_2 molecule tends to distribute in the first and third quadrants, and presents a "finger-like" structure. However, for Kr atom at I = 0.065 PW/cm^2, the emitted electrons pairs tend to distribute in the second and fourth quadrants; When the laser intensity increases to 0.15PW/cm^2, the two electrons mainly distribute in the first and third quadrants and along two distinct lines being paralleled to the diagonal. In addition, our numerical calculations reveal that this different phenomenon is closely related to the Coulomb focusing effect: Coulomb potential will attract the returning electron more dramatically when it moves near the atomic or molecular core. For CO_2 molecule, the returning electron is dramatically attracted by three cores, so the returned electron of CO_2 molecule possesses higher energy than Kr atom does.

1 Introduction

Double ionization (DI) is the fundamental and important process when atoms and molecules are exposed to ultra short laser pulses, which has been attracting much attention during the past ten years, because it provides a particularly clear manner to study the electron-electron correlation [1–3]. Recently, the recollision model [4] is widely accepted to describe the ionization events that one electron ionizes first and revisits the core to let the second electron free by collision. According to the recollision mechanism, atomic and molecular NSDI process in strong laser fields can occur either by directly ionizing of the second electron up on collision with turning first electron (recollision impact ionization, RII), or by excitation of the second electron to an excited state to be ionized in the laser field at a later time (recollision excitation with subsequent ionization, RESI) [5]. Comparing to atoms, due to diverse molecular structure and additional nuclear degree of freedom, molecules exhibit much more complicated processes in strong-field ionization [6]. Fortunately, previous studies have shown that many aspects of strong-field ionization of molecules are similar with those in atoms. For example, the “knee” structure has been observed in simple diatomic [7–10] and linearly triatomic molecules [11–13], and even more complicated polyatomic molecules [14]. Moreover, the presence of revisiting electrons, which are at the basis of atomic NSDI, was reported for molecules [13, 15, 16]. Thus, electron correlation in a rescattering event is also at the basis of molecular NSDI in general. Furthermore, the investigations [17–20] have shown that electronic structure plays a key role in influencing nonsequential processes. For example, a closed-shell molecule will behave like a rare gas atom [17].

In this paper, we investigate the double ionization mechanism of CO_2 molecule and Kr atom in linearly polarized laser fields by the classical ensemble method, and make a comparative study. The numerical results show that the rate for NSDI of Kr atom is higher than that of CO_2 molecule. The momentum distribution illustrates this phenomenon. In addition, our numerical calculations of angular and energy distributions reveal that this different phenomenon is closely related to the Coulomb focusing effect: Coulomb potential will attract the returning electron more dramatically when it moves near the atomic or molecular core.

2 Theoretical Method

In this paper, we use the classical ensemble method proposed by Haan and Eberly et al [21, 22] which has previously been used successfully [8–10, 13] to explore the ionization dynamics of CO_2 molecule and Kr atom in intense laser fields. In our calculation, the CO_2 molecular axis is along the x axis. The classical Hamiltonian of CO_2 molecule and Kr atom in an intense laser field can be given by:

$$\mathrm{H}(\mathrm{r}_1,\mathrm{r}_2;\mathrm{P}_1,\mathrm{P}_2;\mathrm{t}) = \mathrm{T}(\mathrm{p}) + \mathrm{V}(\mathrm{q},\mathrm{t}), \quad (1)$$

where the CO_2 molecular and Kr atomic kinetic energy T are given by:

$$\boldsymbol{T}_{\mathbf{CO_2}}(\boldsymbol{P}) = \frac{\boldsymbol{P}_1^2}{2} + \frac{\boldsymbol{P}_2^2}{2},$$
$$\boldsymbol{T}_{\boldsymbol{Kr}}(\boldsymbol{P}) = \frac{\boldsymbol{P}_1^2}{2} + \frac{\boldsymbol{P}_2^2}{2}, \quad (2)$$

respectively.

The CO2 molecular and Kr atomic potential energy V are given:

$$\boldsymbol{V}_{\boldsymbol{co_2}}(\boldsymbol{q},\boldsymbol{t}) = -\sum_{i=1}^{2}\frac{\boldsymbol{Z_C}}{\sqrt{\boldsymbol{x}_i^2+\boldsymbol{y}_i^2}} - \sum_{i=1}^{2}\frac{\boldsymbol{Z_O}}{\sqrt{(\boldsymbol{x}_i-\boldsymbol{R})^2+\boldsymbol{y}_i^2}} - \sum_{i=1}^{2}\frac{\boldsymbol{Z_O}}{\sqrt{(\boldsymbol{x}_i-\boldsymbol{R})^2+\boldsymbol{y}_i^2}} + \frac{1}{|\boldsymbol{r_1}-\boldsymbol{r_2}|} + (\boldsymbol{r_1}+\boldsymbol{r_2})\boldsymbol{E}(\boldsymbol{t}), \quad (3)$$

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$$V_{Kr}(q,t) = -\sum_{i=1}^{2}\frac{2}{\sqrt{r_i^2}} + \frac{1}{|r_1-r_2|} + (r_1+r_2)E(t), \quad (4)$$

respectively.

In the above equations, q = (r_1 ,r_2) stands for the positions of the two electrons, x_1 , x_2 , y_1 , y_2 presents the x-axis, y-axis coordinate of two electrons, respectively. p = (P_1, P_2) is the corresponding conjugate momenta, E(t) is the laser field. Z_C =2/3 and Z_O =2/3 are effective nuclear charge. R = 2.19a.u. is the C–O bond length. Soft-core Coulomb potential is used to avoid auto-ionization and remove the singularity in exact Coulomb potential

$$V_{CO_2}(q,t) = -\sum_{i=1}^{2}\frac{Z_C}{\sqrt{x_i^2+y_i^2+a_c^2}} - \sum_{i=1}^{2}\frac{Z_O}{\sqrt{(x_i-R)^2+y_i^2+a_s^2}} - \sum_{i=1}^{2}\frac{Z_O}{\sqrt{(x_i+R)^2+y_i^2+a_s^2}} + \frac{1}{\sqrt{(x_1-x_2)^2+(y_1-y_2)^2+q_e^2}} + (r_1+r_2)E(t), \quad (5)$$

$$V_{Kr}(q,t) = -\sum_{i=1}^{2}\frac{2}{\sqrt{x_i^2+y_i^2+a^2}} + \frac{1}{\sqrt{(x_1-x_2)^2+(y_1-y_2)^2+b^2}} + (r_1+r_2)E(t), \quad (6)$$

where a_s =0.8, a_c =1.0 and q_e =0.05, a=1.7, b=0.1; the canonical system of equations for CO_2 molecule and Kr atom is

$$\frac{dp}{dt} = -\frac{\partial V(q,t)}{\partial q}, \quad \frac{dq}{dt} = -\frac{\partial T(P)}{\partial P}, \quad (7)$$

The symplectic method is different and preserves the symplectic structure especially suitable for the long-time many-step calculations. We choose a set of initial stable states {$r_i(0)$,$P_i(0)$} i=1,2 and solved the above canonical equations numerically in order to obtain the time evolution of the electron positions and the corresponding momenta {$r_i(t)$,$P_i(t)$} i=1,2. Since the Hamiltonian system(1) is a separable Hamiltonian system in the sense that q and p are contained separately in V(q, t) and T(p). Meanwhile, the Hamiltonian function contains the time variable. We may use the four-stage fourth-order explicit symplectic scheme to solve it so that we can obtain the classical trajectories of CO_2 molecule or Kr atom in an intense laser field [25]. In our calculation, we assume the initial condition has the same energy approximately equal to the sum of first and second ionization energy. CO_2 molecule and Kr atom have similar single and double ionization potentials (CO_2, 13.778 eV and 23.3 eV; Kr, 14 eV and 24.36 eV). Once the initial conditions for the participating pairs are obtained, the field is turned on and all trajectories are propagated in time. In this work, we utilize a micro-canonical ensemble consisting of 5×10^6 two-electron "trajectories". The electric field of the linearly polarized laser pulse $E(t) = E_0 f(t)\cos(\omega_o t)$, where ω_o is the laser frequency; E_o is the maximum field strength of the linearly polarized electric field; $f(t) = \sin^2(\pi t/T_0)$ is the laser envelope; the pulse duration is 4 optical cycle.

3 Results and discussion

Figure 1 shows the double ionization probability of CO_2 molecule and Kr atom as a function of the laser intensity for 800 nm field. For both gases, the characteristic "knee" structure of NSDI can be clearly seen. Besides, the double ionization probability of Kr atom is higher than that of CO_2 molecule. For triatomic molecule, the electronic structures play a key role in NSDI [17, 18], thus we first take into account the valence electron orbits of CO_2 molecule (...$(4\sigma_g)2(3\sigma_u)2(1\pi_u)4(1\pi_g)4$)[23]. It should be noticed that CO_2 molecule are closed-shell structure with the outermost $1\pi_g$ orbital fully occupied with 4 electrons, like the rare gas atom. The closed-shell molecule will behave like a rare-gas atom (e.g. N_2 and Ar) [17]. However, why the rate for NSDI of Kr and CO_2 show a different behavior? Due to CO_2 molecule, the removal of $1\pi_g$ electrons has two chances: (i) the two electrons can be both removed from one of the pairs of degenerate orbits; (ii) the two electrons can each be removed from a separate $1\pi_g$ orbital [18, 23]. The rate for NSDI is higher when two electrons are removed from the same orbital but lower when two electrons are removed from two different orbits [18, 23].That is to say, in theory, when the two electrons are each removed from a different $^1\pi_g$ orbital, the doubly ionized CO_2^{2+} is in either the $^3\sum_g^-$ or $^1\Delta_g$ state. When the two electrons are both removed from a same $^1\pi_g$ orbital, the doubly ionized CO_2^{2+} is in the $^1\sum_g^+$ state, and this state is about 2.0 eV above the $^1\sum_g^-$ state and 0.8 eV above the $^1\Delta_g$ state [20]. Thus, the different behavior of NSDI of Kr and CO_2 may be due to the different second ionization energy of Kr and CO_2.

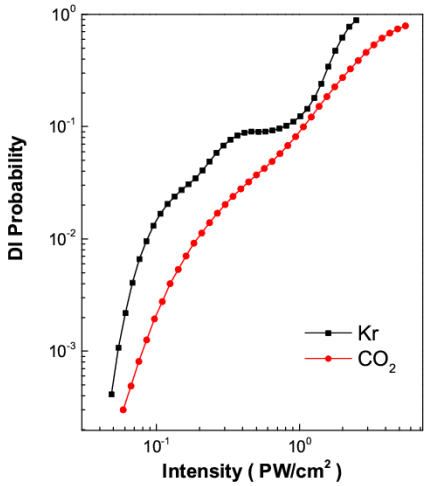

Figure 1: (Color online) Probabilities of CO_2 molecular (black line with circles) and Kr atomic (red line with squares) double ionization as a function of the laser peak intensities in linearly polarized laser fields.

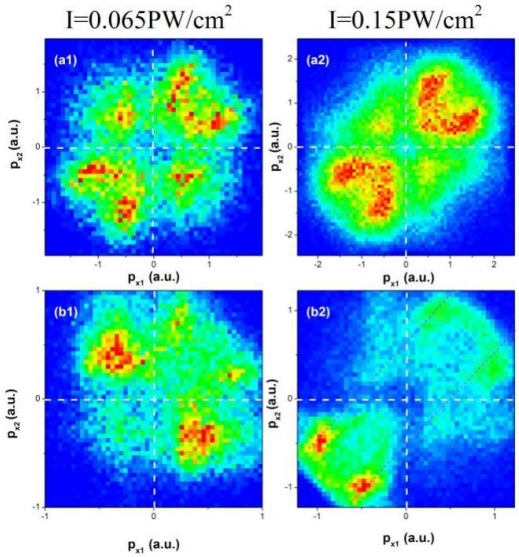

Figure 2: Electron momentum correlation spectra of CO_2 molecule (a) and Kr (b) atom by 800-nm linearly polarized laser pulses, the intensities are 0.065 PW/cm^2 (left column (1)) and 0.15 PW/cm^2 (right column (2)), respectively.