Intensity dependence of strong field double ionization mechanisms of diatomic molecules: from field-assisted electron recollision to recollision-assisted field ionization

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Given the state of the art in computational capabilities addressing the double ionization of strongly driven diatomic molecules with three-dimensional (3-d) firstprinciple techniques, namely quantum mechanical ones, is an immense task. We study the correlated electron dynamics in the double ionization of diatomic molecules with "frozen" nuclei in the non-sequential regime as a function of the intensity of the laser pulse [1]. We do so, using a 3-d quasiclassical technique that we have first developed for conservative systems, namely, the multiple ionization of atomic systems such as Li, Be, by single photon absorption [2]. We have very recently extended this technique to non-conservative systems treating the correlated electron dynamics of the He atom when driven by strong laser fields [3]. Here we further build on this technique by tackling more than one atomic centers. The advantage of this technique is that it is numerically very efficient. In addition, the method treats the Coulomb singularity with no approximation in contrast to techniques that use "soft-core" potentials. Accounting for the Coulomb singularity will be in addition very important in pump-probe set-ups where VUV or XUV pulses are used to pump the process.

For intensities corresponding to the tunneling regime, we find that the recollision time is close to a zero of the field—as expected from the recollision model. We identify the main ionization mechanisms in this regime



FIG. 1: Form left to right the laser intensity is 10^{14} W/cm², 1.5×10^{14} W/cm², 3×10^{14} W/cm² and 4×10^{14} W/cm² for a 3-cycle laser pulse. Top panel: correlated momenta parallel to the polarization axis; Medium panel: the distribution of the inter-electronic angles of escape binned in 14 intervals, $180^{\circ}(l-1)/14 < \theta < 180^{\circ}/14 \times l$ with l = 1, ..., 14; Bottom panel: sum of the parallel momenta as a function of the inter-electronic angle of escape.

and show that they have distinct traces when considering the sum of the momenta parallel to the laser field as a function of the inter-electronic angle of escape, see Fig. 2. However, when the laser intensity is increased above the barrier we find an unexpected concentration of the correlated momenta in the second and fourth quadrant in contrast to the concentration in the first and third quadrant for the tunneling regime. We discuss the mechanism responsible for this shift of the correlated momenta, see Fig. 1, [1]. This surprising shift of the correlated momenta is a very nice demonstration of both the strong IR field and the Coulomb potential contributing to double ionization with the IR field playing an important role even before the recollision time—thus the name recollision-assisted field ionization.



FIG. 2: For 10^{14} W/cm² we identify four different double ionization mechanisms depending on each electron's time of ionization: the different columns refer to: a) the SE, b) the RESI , c) the NSE2 and d) the DE mechanisms. First row: the sum of the momenta of the two electrons as a function of the inter-electronic angle of escape; second row: the correlated momenta of the two electrons; and third row: the ionization time of each electron in units of laser cycles.

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