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The importance of Rydberg orbitals in dissociative ionization of small hydrocarbon molecules in intense few-cycle laser pulses

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Synopsis We demonstrate the importance of ionization from Rydberg orbitals via experimental and theoretical work focusing on the strong-field dissociative single ionization of small hydrocarbons. Our findings suggest that Rydberg states should be routinely considered when studying polyatomic molecules in intense laser fields.

Much of our intuition about strong-field processes is built upon studies of diatomic molecules, which have electronic states that are typically well separated in energy. In polyatomic molecules, however, the electronic states are closer together, leading to the involvement of more states. We studied a specific process initiated by intense few-cycle laser pulses in C₂D₂, C₂D₄, and C₂D₆, namely single ionization of the parent molecule followed by hydrogen elimination. We measure the momentum of the remaining C₂D_{n-1}⁺ fragment in each case using velocity map imaging (VMI). Our measurements, along with angle-dependent ionization probability calculations [1], demonstrate that Rydberg orbitals are readily populated for certain orientations of the molecules relative to the laser polarization. Moreover, these commonly-neglected Rydberg orbitals make a key contribution to strong-field ionization.

As an example, the comparison between measured and calculated C₂D₃⁺ angular distributions from C₂D₄, shown in Fig. 1, clearly illustrates the need to include contributions from Rydberg states. The general four-lobed structure of C₂D₃⁺ from dissociative single ionization is independent of pulse intensity and duration (at least up to 45 fs [2]). Calculations that do not include ionization from Rydberg orbitals result in an approximately isotropic angular distribution, as illustrated by Fig. 1(c), while including contributions from Rydberg orbitals yields the four-lobed structure shown in Fig. 1(d), which qualitatively matches the experimental results. This improved understanding of the links between photofragment images and the electronic dynamics in the molecule can improve endeavors towards image-

based strong-field coherent control.

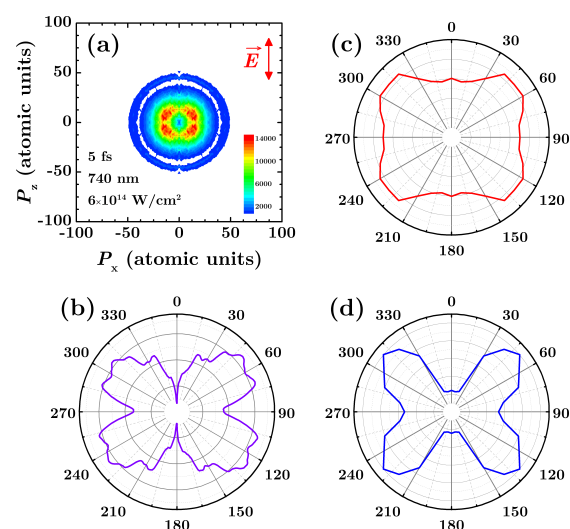


Figure 1. (a) Momentum distribution of C₂D₃⁺. The inner lobes come from dissociative single ionization of C₂D₄, and the weaker outer ring is due to dissociative double ionization. (b) Measured yield of the inner lobes as a function of the angle between the C₂D₃⁺ and the laser polarization. Calculated angular distribution for C₂D₃⁺ (c) without including Rydberg orbitals and (d) with ionization from Rydberg orbitals included.

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