Simulation of laser-induced quantum dynamics of the electronic and nuclear motion in the ozone molecule on the attosecond time scale[1]

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The nonadiabatically coupled dynamics of electrons and nuclei is investigated for the ozone molecule on the attosecond time scale[2,3]. A coherent superposition of nuclear wave pakets located on different electronic states in the Chappuis and in the Hartley bands is created by pump pulses[2,3]. The multiconfiguration time-dependent Hartree method is used to solve the coupled nuclear quantum dynamics in the framework of the adiabatic separation of the time-dependent Schrödinger equation including nonadiabatic couplings. Our nuclear wave-packet calculations demonstrate that the coherence between Hartely state B and one of the Chappuis states (Chappuis 1) is significantly large, while it is almost negligible for the other two cases (between Hartley B and Chappuis 2 or between Chappuis 1 and Chappuis 2) (FIG. 2.). At present we limited our description of the electronic motion to the Franck-Condon region only due to the localization of the nuclear wave packets around this point during the first 5-6fs (FIG. 1.).

It is now an important challenge to extend attosecond spectroscopy techniques to molecules. We are collaborating with the group of Prof. F. Krausz where such experiments are being carried out on ozone.

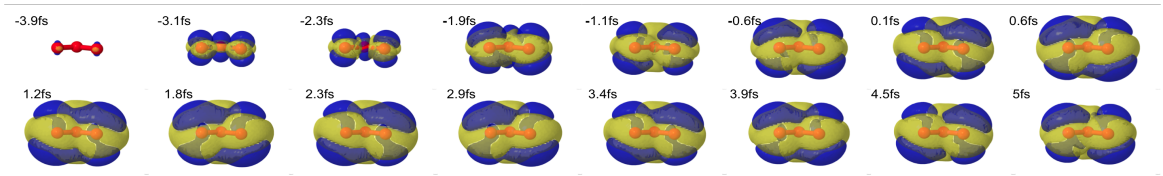

FIG. 1. Time evolution of the excited differential electronic charge density[4], at the FC geometry (side view). Dark gray: electron, light gray: hole.

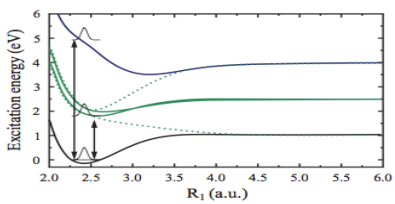

FIG. 2. The diabatic (solid line) and adiabatic (dashed line) potential energy surfaces of ozone as functions of the dissociation coordinate: Ground state G (bottom line), both Chappuis states (C1 and C2; middle line), and Hartley state B (top line). The arrows denote the coherent excitation of the superposition of the C1,C2, and B states.

References:

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