

Charge oscillation controlled molecular excitation

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Direct manipulation of charge oscillations has emerged as a new perspective in chemical reaction control. Here we demonstrate in a joint experimental and theoretical study, that the electron dynamics of a molecule is efficiently steered by controlling the interplay of a driving femtosecond laser pulse with the photo-induced charge oscillation on the sub-cycle time scale [1]. Specifically, we show how precision pulse shaping is used to manipulate the coupled electron and nuclear dynamics in order to address different bound electronic target states in a molecule.

We present a resonant coherent control scheme based on precisely shaped intense femtosecond laser pulses where the initial part of the laser pulse creates an oscillating charge distribution with maximum amplitude in a molecule. After evolution on coherently coupled potential energy surfaces the later part of the laser pulse timed with sub-cycle precision switches the population to different final states [2]. The scenario allows to steer the coupled electron and nuclear motion of the molecule to selectively populate different target channels, each of which entails different subsequent nuclear dynamics. Initial theoretical studies on selective population of a single target channel employing pulse sequences used the potassium dimer (K_2) as the molecular prototype system [3]. Now we report on the first experimental implementation of this control mechanism in molecules. The underlying control mechanism can be understood in terms of a simple classical picture and is at the same time verified by solving the time-dependent Schrödinger Equation [3]. This scenario is universally applicable and opens a wide spectrum of applications in reaction control of complex systems [4].

References:

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