Service des Photons, Atomes et Molécules SÉMINAIRE

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CEA-Saclay ¡b¿¡font color ='red'¿SPAM¡/font¿¡/b¿ Bât 522, p 138

Following a chemical reaction using high harmonic homodyne interferometry

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A significant problem in femtochemistry is the difficulty in creating a large population in the excited state. If the probe step in a pump-probe experiment cannot distinguish the initial state, for example in x-ray diffraction, electron diffraction, or high harmonic spectroscopy, then it is difficult to observe the chemical reaction. In the case of high harmonic spectroscopy, we show that the strong emission from unexcited molecules acts as a local oscillator in a homodyne detection scheme, and permits the measurement of both amplitude and phase of the excited state emission. Using a transient grating excitation geometry in bromine, we can follow the dissociating molecules through two-center interference. When the products become atoms, we can further follow their dissociation through interference of electron trajectories. Because high harmonic spectroscopy is sensitive to the electronic state of a molecule, this technique shows promise to observe changes in electronic structure during a chemical reaction, for example at conical intersections.