

The role of attosecond electronic coherence in molecular dissociation

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In molecules electronic and nuclear degrees of freedom are always correlated, but due to different timescales of their evolution, effects of electronic coherence on nuclear motion are usually not observed. However, we show that electronic coherence can significantly influence nuclear dynamics when interference of paths associated with vibrational motion along different potential energy surfaces becomes important.

We consider attosecond XUV pump – IR probe setup. A N_2 molecule is first ionized by an XUV pump pulse. Ionization creates a coherent superposition of two excited electronic states in N_2^+ ion. The dissociation of the molecular ion then takes place over two different potential energy surfaces. Interaction with IR field ensures that both of them lead to the same dissociation limit. The fragment spectrum obtained depends significantly on how the two paths interfere.

Interference is observed if the continuum electron wavepackets correlated with each of the paths have an overlap. This condition is set during ionization, which happens in a fraction of a femtosecond and is given by the length of the XUV pump pulse. However, it determines the appearance of molecular dissociation spectrum hundreds of femtoseconds later. Therefore the final spectrum observed is an interplay between attosecond electronic and femtosecond molecular dynamics.

When one of the pathways involves a direct and the other a sequential transition to a dissociative state, the phase acquired along the propagation on sequential path creates a chirp in the interference pattern and introduces additional phase in a RABBITT type measurement on molecules. It can be identified in an XUV-IR time delay scan and can potentially encode information on the initial wavepacket and the potential energy surface.

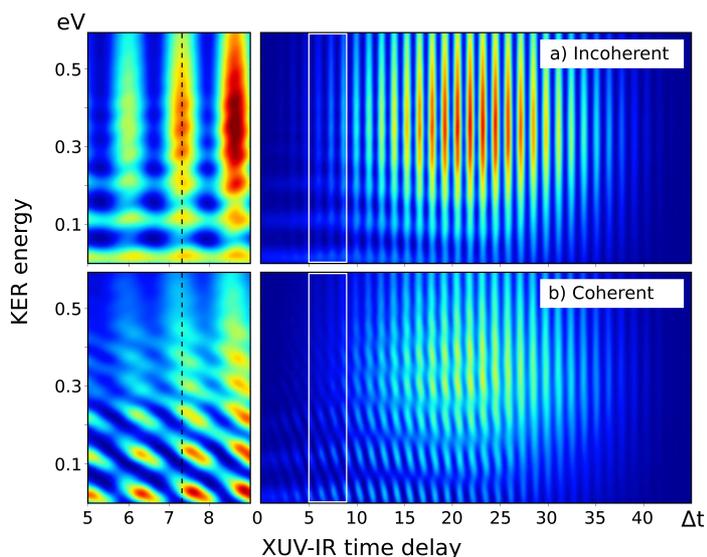


Figure 1: The Kinetic Energy Release spectrum of an attosecond XUV-IR time delay scan for the case of incoherent (a) and coherent (b) interference between direct and sequential dissociation channels. Zero time delay corresponds to XUV pulse coming before the IR pulse.