Monitoring Ultrafast Core Dynamics Using Rydberg Electrons

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Attosecond XUV pulses combined with infrared (IR) laser pulses have led to impressive advancements in probing and controlling electron dynamics in atoms and molecules. Here were apply XUV-IR pump probe spectroscopy using the complementary techniques of attosecond transient absorption spectroscopy (ATAS) and photoelectron interferometry. The objective of our work is to study the correlated and coupled non-adiabatic dynamics in the core ion through interaction of light with the Rydberg electron. Two cases will be presented: 1) Study of photoionization to the spin-orbit split continuum of Argon, 2) Study of conical intersection in CO2 ion by observing the time evolution of strong-field ATAS signals produced from the Rydberg series. In the former case, the phases of quantum beat in the two channels capture the effect of short-range electron-core interactions. This phase difference between the two channels is shown in the Fig. 1. In the latter, we show that electron hole dynamics in the core is manifest in the transient changes in the absorption.

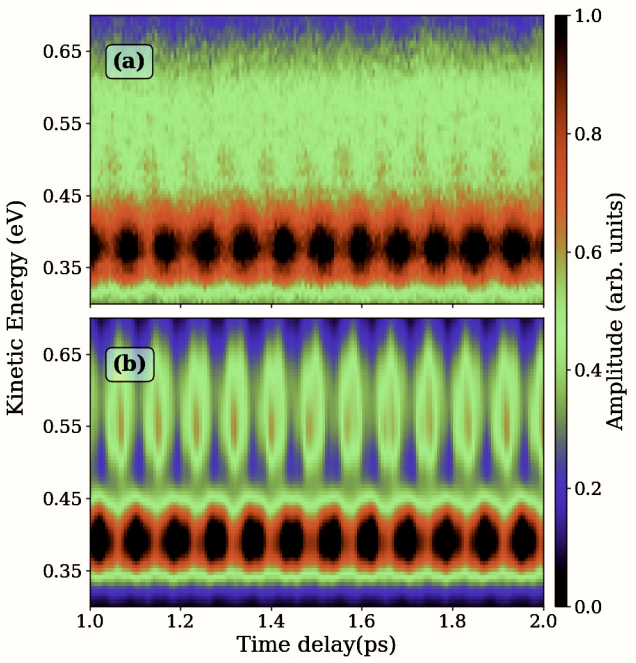

Figure 1: (a) Experimental photoelectron spectrogram showing the beating of 3d-5s electronic wave packet in the two channels of spin-orbit split continuum. (b) Theoretical modeling showing good agreement in amplitudes and phases of two channels