Site-specific and state-resolved coherent quantum control of atoms and molecules

Using intense ultrashort laser pulses with a duration across the femtosecond and attosecond timescale, it is possible to couple and control multi-electron transitions which involve short-lived states in atoms and molecules. Their extreme-ultraviolet (XUV) and x-ray absorption spectra hereby encode the time-resolved dynamics with state-specific spectroscopic information about the relevant quantum states. In this talk, I will first provide an overview how we extract quantum-dynamics information from spectral absorption line shapes. We will discuss for instance the laser-controlled transformation of Fano to Lorentzian spectral line shapes of a correlated two-electron transition in helium, and how its absorption profile coherently builds up on the femtosecond timescale. We will further apply these concepts to nonlinear absorption spectroscopy with free-electron lasers and discuss XUV-induced energy shifts of strongly coupled states, e.g., in helium and neon atoms. Finally, we will also look at time-resolved measurements with XUV-pump – XUV-probe absorption spectroscopy to resolve the state-specific dynamics in small molecules, accessing structural dynamics from the perspective of individual electronic states. With all these experiments, we explore new methods of nonlinear light-matter interaction for the quantum control of atoms and molecules down to the natural attosecond timescale of the electron motion and coherently addressing specific transitions of individual constituents within the larger quantum system.