Ultrafast Gas Phase Spectroscopy using High Harmonic Generation

High Harmonic Generation (HHG)

The electric field from a femtosecond NIR laser pulse tunnel ionizes an electron away from the nucleus of an atom. Half an optical cycle later when the electric field of the laser reverses direction, the electron is accelerated back towards the nucleus, coherently emitting an XUV photon, with energy that is an odd-order harmonic of the original NIR.

Extension of the HHG source to the Carbon K-edge (~285 eV) has been achieved, opening up a new class of studies exploring fundamental hydration photocatalysis, non-Born-Oppenheimer dynamics, dissociative and bound wavepacket dynamics, and radical spectroscopy through femtosecond time-resolved NEXAFS.

Electron Motion in Atoms and Molecules

We use isolated attosecond pulses extending from 50 to 75 eV to probe the fastest charge migration dynamics that occur in nature. These dynamics are driven purely by many-body, electronic correlations and can give rise to the transport of charge across an entire molecule on the few femtosecond timescale.

We use an isolated attosecond pulse centered at 15 eV to create a coherent superposition of Rydberg states in nitrogen. Quantum beating between states in the superposition with periods as short as 1.3 fs is revealed by time-delayed NIR photons, that transfer population to neighboring levels. We will extend this technique to study coupled ultrafast electronic and nuclear dynamics in polyatomic molecules such as CO and H2O.

Femtosecond Molecular Dynamics

Transition state dynamics of prototypical alkyl iodide photodissociation reactions are investigated through the direct observation of the transient valence electronic structure in the vicinity of the iodine reporter atom.

Extension of the HHG source to the Carbon K-edge (~285 eV) has been achieved, opening up a new class of studies exploring fundamental hydration photocatalysis, non-Born-Oppenheimer dynamics, dissociative and bound wavepacket dynamics, and radical spectroscopy through femtosecond time-resolved NEXAFS.

Ultrafast Solid State Spectroscopy using High Harmonic Generation

Attosecond Dynamics in Solid State Materials

With attosecond resolution, we can observe electron and hole dynamics before lattice motion occurs.

By developing transient reflectivity, we hope to expand our studies to materials such as gallium arsenide and 2D transition metal dichalcogenides.

Transient absorption spectroscopy in Ge tracks both electron and hole dynamics simultaneously.

VO2 exhibits an ultrafast insulator to metal phase transition upon heating and ultrafast excitation. By separating electronic dynamics from lattice motion, we seek to understand the mechanism of the phase transition, which is still widely debated.

Ultrafast Charge Carrier Dynamics at Semiconductor Interfaces

In this experiment we investigate the ultrafast charge carrier dynamics at three different interfaces, i.e. semiconductor-gas phase, semiconductor-metal, and semiconductor-semiconductor, by monitoring the full suite of steps from charge excitation to charge separation and subsequent charge recombination. Here is presented an experiment in which the charge carrier dynamics at the n-TiO2/p-Si(100) interface is investigated.

We study the charge carrier dynamics in semiconductor materials after excitation by visible light. Transient XUV absorption spectroscopy has advantages of element/chemical valence specificity and low femtosecond temporal resolution, which is suitable to study ultrafast electron migration processes in semiconductor heterostructures.

Single Molecule Spectroscopy

Fluorescence Spectroscopy of Semiconductor Nanostructures

We investigate charge carrier dynamics in semiconductor nanostructures at the single molecule level. Transfer, trapping and recombination of charges can be studied in individual molecules using a combination of confocal fluorescence microscopy and time-correlated single photon counting. These techniques are being used to study emerging, all-inorganic perovskite materials.

Single CsPbBr3 perovskite nanowires exhibit low-threshold and wavelength-tunable lasing, which persists over one billion excitation cycles, even in ambient atmosphere, demonstrating unprecedented stability in perovskite nanowires.

Ultrafast Charge Carrier Dynamics by XUV Transient Absorption

Hole-transfer process from photo-excited Co3O4 to methanol is studied by the design of an in situ liquid flow cell. The kinetics show that CO2H acts as a hole scavenger after photoionization.

Based on the match between experimental results and charge transfer multiphoton simulation, the excited-state character of α-Fe2O3 after absorbing a 400 nm photon can be assigned to a LMCT state.