

Ultrafast nanoscopy via optically controlled high-harmonic generation from solids

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The highly complex collective electron-dynamics in strongly correlated materials mandates experimental techniques with sub-fs temporal and nanometer spatial resolution. While a plethora of techniques exists, true nanometer imaging on fs or even sub-fs timescales remains elusive. Such techniques are required to follow the collective dynamics of electrons in strongly correlated materials in real time, which drive ultrafast phase transitions that are accompanied by technologically relevant order-of-magnitude resistivity switches.

High-harmonic generation (HHG) in solids has emerged 14 years ago and found many applications, including as a probe in ultrafast spectroscopy experiments. We recently conducted a thorough analysis of current literature [1], which suggests that optical excitation during or prior to HHG generally suppresses HHG – if optimized with near-100% efficiency. This constitutes an unparalleled control mechanisms over light emission.

In this talk, I will present the mechanisms behind this optical suppression of solid-state HHG, and our ongoing research to use this light switch for sub-fs super-resolution nanoscale imaging by HHG from correlated materials. In particular, I will present a number of key experiments on our roadmap towards this goal.

On the femtosecond time scale, we used the sensitivity of HHG to electronic band structure to follow ultrafast phase transitions in strongly correlated materials [2], and photocarrier dynamics in perovskites [3]. Similarly, we studied a number of other semiconducting and insulating materials which now provides a general understanding of the microscopic mechanisms behind suppression of HHG by optical control pulses.

We use this general suppression mechanisms to spatially confine HHG from solids by pre-exciting materials with an orbital-angular-momentum carrying pulse, which thus spatially confines HHG to below the diffraction limit. This enables HArmonic DEactivation microScopy (HADES) - a label-free super-resolution microscopy below the diffraction limit of light. [4]

Thinking ahead, the development of these techniques may enable resolution on the nanometer and femto- to attosecond scale fitted into a regular microscopy setting, with application potential ranging from strongly correlated materials to semiconductor metrology, photosynthetic processes, and medical imaging.

References:

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