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[L] Capturing the Fastest Dynamics in Materials using High Harmonic Sources

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Laser-like beams at very short wavelengths (1-50nm) can now be routinely generated using high harmonic up-conversion (HHG) of tabletop femtosecond lasers. These new quantum light sources are providing powerful new tools for probing and understanding nanoscale material properties and function. The short wavelength of HHG beams are well suited to advanced spectroscopies and imaging with high spatial resolution,[1-3] while the femtosecond-to-attosecond duration of HHG pulses is fast enough to capture the fastest spin-charge-phonon-elastic-thermal dynamics in materials.[4-8]

The high stability and spatial coherence of HHG sources made it possible to achieve record EUV imaging – demonstrating the first sub-wavelength imaging at short wavelength using any light source, small or large — achieving 12.6nm spatial resolution using 13.5nm HHG beams.

New materials behavior uncovered using HHG sources includes how thermal transport changes dramatically for nanoscale heat sources of dimensions on the order of the phonon mean free path, how the mechanical properties of doped materials dramatically differ from bulk properties, as well as how materials can change their electronic and magnetic state on surprisingly fast, < 20fs, timescales. In recent work, we captured intrinsic sub-femtosecond dynamics in materials for the first time. First, we measured the lifetime of a ~25eV excited state in the band structure of Ni - which represents the fastest lifetime ever measured, at 212±30 attoseconds. Essentially, this measurement shows that the photoelectron leaves the crystal before it can experience screening from the surrounding charges. Second, we use sequences of attosecond pulses to directly measure electron-electron interactions in different bands of different materials with both simple and complex Fermi surfaces. By extracting the time delays associated with photoemission, we show that the lifetime of photoelectrons from the d band of Cu are longer by ~100 attoseconds compared with those from the same band of Ni. We attribute this to the enhanced electron-electron scattering in the unfilled d band of Ni. This is the first way to directly probe the contribution of electron scattering and screening to low-energy excitations near the Fermi level.

1. Ultramicroscopy 158, 98 (2015).
2. Nano Letters 16, 5444 (2016).
3. Nature Photonics 11, 259 (2017).
4. Science 353, 62 (2016).
5. PNAS 114, E5300 (2017).
6. Science Advances 4, 9744 (2018).
7. Physical Review Letters 120, 093002 (2018).
8. PNAS 112, 4846 (2015).

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