Ultrafast imaging of nanoscale samples with intense XFEL pulses

C Bostedt

Laboratory for Synchrotron Radiation and Femtochemistry,
Paul Scherrer Institute, Villigen, 5232, Switzerland
LUXS Laboratory for Ultrafast X-ray Sciences, Inst. of Chem. Sciences and Engineering,
Ecole Polytechnique Fédérale de Lausanne (EPFL), Lausanne, 1015, Switzerland

Strong external light fields can induce significant electron excitation and redistribution within a sample and lead to strong changes in its interatomic potential energy landscape. In bulk systems, the external excitation can lead to new phenomena such as non-thermal melting or the opposite extreme of bond hardening. In nanoscale systems, the strong electronic excitation is typically intertwined with a strong structural response stemming from the finite size of the sample.

To observe and describe the light-induced processes in nanoscale systems a variety of approaches are available but the most powerful is visualizing the dynamics. However, the ability to directly image non-equilibrium processes in single isolated nanometer-sized samples is virtually impossible with the commonly available approaches. Electron microscopy requires samples deposited on a substrate, interfering with their dynamics, and is limited in temporal resolution. Optical light scattering can deliver high temporal but lacks spatial resolution.

Intense x-ray pulses from x-ray free-electron lasers (XFEL), containing on the order of 10^{12} photons in a 100 fs pulse, offer a new route for taking snapshot of single nanoparticles. We have used XFEL pulses to image clusters [1], to follow transient electronic excitations [2], or to observe quantum vortices in He droplets [3]. A novel holography approach with a cluster as reference scatterer next to the sample of interest allows reconstruction of the sample geometric information with simple inverse Fourier transform algorithms [4]. To push the frontiers in single-shot imaging we are working on a quantitative understanding of the interaction of x-rays with nanoscale matter, including the competition of electronic excitation vs. scattering cross section of bound and quasi-bound electrons.

For nanoparticles in strong fields, the electron excitation leads to strong structural responses. Combining ultrafast imaging and hard x-ray Bragg scattering, we were able to observe the sample dynamics on the surface and inside the nanoparticle core. Femtosecond time-resolved coherent x-ray diffractive imaging [5] revealed that the nanoparticle surface softens at the onset of the expansion. Hard x-ray scattering [6,7] experiments with van-der-Waals clusters showed different dynamics depending on the frequency of the pump. Optical excitation leads to a continuous shrinking of the otherwise undisturbed particle core. Isochoric heating with intense x-ray pulses induces an unexpected shrinking of the overall nanoparticle prior to expansion which is so far attributed to a transient metallization of the clusters. In the future, the new attosecond capabilities of the upcoming machines may enable imaging experiments that are sensitive to the transient electronic configurations.

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