Full-Field Stroboscopic Imaging of Acoustic and Thermal Dynamics in Isolated Nanostructures Using Tabletop EUV Coherent Imaging

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Understanding the mechanical properties of nanostructured systems is critical for accelerating advances in material, nano and bio sciences, as well as various nanostructured electronic, photovoltaic and thermoelectric devices. However, macroscopic models cannot accurately predict nanoscale (<100 nm) transport or mechanical properties [1]. Moreover, many nanostructured systems are opaque to visible light. This creates a critical demand for functional microscopes that can image with elemental, chemical and magnetic contrast through opaque materials, as well as with high spatial and temporal resolution. Here we report the first full-field stroboscopic imaging using tabletop high harmonics, where we directly visualize nanoscale thermal and acoustic dynamics in an isolated nanostructure with <0.1nm axial precision, 90nm lateral resolution, and 10fs time resolution.

In our experiment, we combine ptychographic coherent diffractive imaging (CDI) [2,3] with a tabletop high harmonic generation (HHG) source [4,5], which in combinations allows us to achieve diffraction-limited spatial resolution and femtosecond temporal resolution. An ultrafast near-infrared laser pump pulse (800nm, 23fs, 1.5mJ, 5kHz, KMLabs DRAGON) was used to excite complex thermal and acoustic dynamics in the nanoantenna (see Fig. 1a). Bright, phase-matched, spatially coherent EUV probe beams, with pulse duration <10 fs, were generated through high harmonic generation by focusing the driving laser beam into an argon-filled waveguide. The residual driving laser was filtered and a single harmonic at 28.9nm (43eV) was selected. The HHG probe beam was focused onto the sample using an ellipsoidal mirror and the diffracted light was collected by an x-ray CCD. The structure studied here was a 20nm tall Ni nanoantenna fabricated on a Si wafer, with a length of 53 μ m and a maximum width of 5 μ m. Snapshots of the laser-excited sample were then collected at different time delays between laser pump and EUV probe pulses, for separations ranging from -40ps to 650ps. At each time delay, the sample was imaged using ptychographic CDI, which employs multiple diffraction patterns from overlapping fields of view. Figure 1b shows the quantitative CDI phase map in one of the ptychographic images, from which a height map of the nanostructure can be extracted.

The transient response of the sample consisted of an initial rapid thermal expansion just after excitation (<1ps), which launched acoustic waves at the edges of the structure, followed by oscillations due to transverse and longitudinal acoustic waves propagating within the Ni triangle and Si substrate. A lineout of the ptychographic images across the Ni triangle is shown in Fig. 1c. Due to the large aspect ratio of this feature, simulating the dynamics of the full 3D structure is computationally too intensive. Instead, we simulated a uniform 2.2um cross-section bar, where we see the same qualitative dynamics at similar time scales (Fig. 1d). The significant differences between the experimental data and the simulation primarily resulted from the shape of the triangle, which causes the acoustic waves launched by two edges to be at a slight angle relative to one another. This leads to spatially varying oscillations that are not shown in the

simplified 1D simulation. Furthermore, using the time-resolved measurement, we were able to extracted the oscillation frequency and the spatial frequency for 5 different orders of acoustic waves. From these, we extracted the dispersion of the surface acoustic waves and determined an acoustic wave velocity of 2970 ± 240 m/s, which agrees very well with the result of finite-element modal simulation, as shown in Fig. 1e [6].

We have demonstrated the first full-field stroboscopic imaging of thermal and acoustic dynamics using tabletop high harmonics. This work provides new tools for dynamic imaging of energy, charge, and spin transport in nanoscale materials with nanometer- and femtosecond-scale resolution, enabling a direct visualization of the nano-world at its intrinsic length- and time-scales [7,8].

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Figure 1. (a) Experimental setup for full field EUV dynamic imaging on a tabletop. (b) Phase image of a nanostructured nickel triangle taken using coherent diffractive imaging. (c) Lineout of the height of the nanostructure as a function of pump probe delay. (d) Simulation of the time evolution of a 2.2um cross-section of the nickel triangle after impulsive heating. (e) The dispersion relation of the acoustic waves launched by the pump pulse.