Full-Field Functional Imaging of Nanoscale Dynamics Using Tabletop High Harmonics

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Abstract: We report the first stroboscopic full-field EUV microscope using high harmonics. Specifically, we demonstrate nanoscale movies of thermal and acoustic wave propagation in nanostructures with ≈0.1nm axial resolution, 90nm lateral resolution, and 10fs time resolution.

OCIS codes: (180.7460) X-ray microscopy; (120.7440) X-ray imaging; (110.5120) Photoacoustic imaging; (190.2620) Harmonic generation and mixing; (240.6648) Surface dynamics

1. Introduction

Nanofabrication techniques continue to accelerate advances across a wide range of nano-enhanced materials for photovoltaics, electronics, thermoelectrics, data storage and medicine. However, at dimensions <100 nm, smart design of functional nanosystems is challenging because bulk macroscopic models can no longer accurately predict heat, charge or spin transport, or the mechanical properties of doped or nano-structured materials [1]. Moreover, many nano-structured materials are opaque to visible light. As a result, there is a critical need for new functional microscopes that can stroboscopically image with high spatial and temporal resolution, through opaque overlayers, with elemental, chemical and magnetic contrast.

Here we present the first full field dynamic imaging using tabletop high harmonics, where we directly visualize nanoscale thermal and acoustic wave propagation in real time with ≈0.1nm axial resolution, 90nm lateral resolution, and 10fs time resolution. To achieve this, we combine ptychography coherent diffractive imaging (CDI) [2,3] with a tabletop high harmonic (HHG) source. After exciting the nanosystem with a 23 fs laser pulse, we stroboscopically image the resulting thermal expansion and heat flow, as well as the impulsive longitudinal and transverse acoustic wave propagation of nickel nanostructures patterned on a silicon substrate. This work is a first step toward combining the ultra-high spatial resolution possible using EUV-CDI with the ultra-high femtosecond temporal resolution of HHG sources [4,5]. This advance can uncover new basic understanding of nanoscale materials, while also providing new tools for functional imaging to inform next-generation device design at dimensions <<100 nm.

2. Experimental methods

In our experiment shown in Fig. 1, the laser excitation pulse and the λ=30nm high harmonic probe illumination beam were both derived from a 23fs, 1.5mJ, 5kHz, Ti:Sapphire laser (KMLabs DRAGON). Bright, phase-matched, spatially coherent HHG beams, with duration < 10 fs, were produced by focusing most of the laser beam into a 5cm-long waveguide filled with 32 torr of argon gas. The residual laser light was rejected using a pair of super-polished silicon substrates set at Brewster's angle, together with a single 100nm Al filter. A single harmonic at a wavelength of 30nm (41eV) was then selected using a pair of multilayer mirrors set at 45 degrees. An ellipsoidal mirror at a 5° angle of incidence from the surface focused the HHG beam to a 21um x 21 diameter spot. The light diffracted from the sample was collected with an x-ray CCD, placed a distance of 36.5mm from the sample.

Our sample consisted of 20nm tall Ni nanostructures patterned on a Si wafer. To excite thermal and acoustic dynamics, we irradiated it with part of the 780 nm laser beam, at an incident fluence of 7.7mJ/cm^2 in a 500μm-diameter spot. Snapshots of the sample were then collected at different time-delays (Fig. 1). At each time delay, the sample was imaged using ptychographic CDI, which employs multiple diffraction patterns from overlapping fields of view. The transient response of the sample was retrieved from an analysis of the diffraction patterns from the Ni features at different time-delays between pump and probe pulses, which exhibit a sharp rise at zero delay (time zero) due to <1ps thermal expansion, followed by oscillations due to longitudinal acoustic waves and surface acoustic waves launched within the Ni lines and Si substrate. The dynamic diffraction efficiency of the Ni lines was calculated following [1], by computing the normalized difference in power in the 1st order diffraction peak and in the 0th order diffraction peak.

Figure 1c (yellow trace) plots the 1st order diffraction signal (where the rapid oscillations are due to longitudinal acoustic waves within the nanostructures, not random noise). To validate our measurement, we also extracted the signal from a single diffraction pattern from the ptychography scan (Fig.1c: blue triangles). The good

agreement between the two data sets indicates that the acoustic dynamics we are imaging are consistent with those extracted spectroscopically, as expected for this uniform sample.

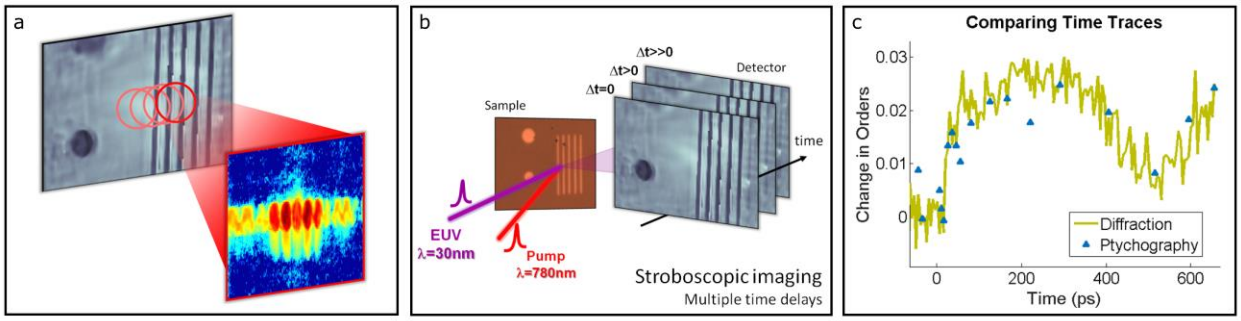

Fig. 1. Experimental setup for full field EUV dynamic imaging on a tabletop. (a) A single image formed by ptychographic scanning CDI. (b) A series of stroboscopic images are reconstructed for every pump probe delay. (c) Plot of change in the 1st diffraction order power for a single diffraction pattern (yellow line) and for the diffraction patterns from the ptychographic data set (blue triangles), to validate the consistency of our data.

3. Visualization of nanostructure dynamics

To demonstrate dynamic imaging of a nanostructure, we reconstructed amplitude and phase images of a 20 nm high, 16μm diameter, Ni nano pillar (Fig. 2). The phase images allow us to extract height information about the pillar. After thermal expansion and impulsive launching of longitudinal acoustic waves, the average height of the pillar increases by only 0.5nm. In Fig. 2, we show an AFM image of the static pillar (a) along with three reconstructions of the pillar (b-d) at different time delays between the laser pump and HHG probe beams. The image before time zero shows the baseline height of the pillar, in agreement with the AFM data. The image taken 5 ps after time zero (Fig. 2c) shows a slight reduction in the average height by 0.1nm, due to the phase of the longitudinal acoustic oscillation. In contrast, the image taken 65 ps after time zero shows a 0.5nm height increase. This demonstrates a nanoscale movie of picosecond thermal and acoustic wave propagation with ≈0.1nm axial resolution, 90nm laterial resolution, and 10fs time resolution.

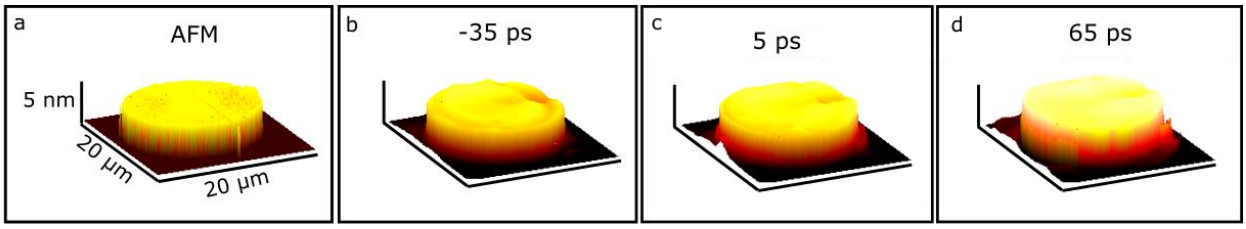

Fig.2. (a) AFM image of the Ni pillar, of diameter 16μm and height 20nm: we omit the bottom 16nm of the pillar to better emphasize the dynamics. (b-d) height images derived from ptychographic phase images, with ≈0.1nm axial resolution and 90nm transverse resolution. The scale bars in all four panels are the same.

4. Conclusions

We have presented the first full-field functional imaging of nanoscale dynamics using tabletop high harmonics. This work opens the door to dynamic imaging of energy, charge, and spin transport in opaque materials with nanometer and femtosecond resolution, enabling a direct view of the nano world at its intrinsic length- and time-scales.

5. Acknowledgments

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