Multi-wavelength Ultrafast Ptychography: A Flexible Beamline with a Compact High Harmonic Source

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Abstract: We report the design of a flexible beamline for ultrafast multiwavelength ptychography to image nano-to-mesoscale heterogeneity in nanostructures and interfaces, with quantitative amplitude and phase contrast, sub-50nm spatial and sub-50fs temporal resolutions. © 2022 The Author(s)

1. Introduction

With technology miniaturization, novel materials with functionality activated by light pulses are engineered in the deep nanoscale regime, where bulk macroscopic models can no longer accurately predict microscopic properties. To address this challenge, there is a critical need for new techniques that can visualize heterogeneity in energy conversion from light to charge carriers, and then into lattice heating, with high spatial and temporal resolution.

In this work, we present the design of a flexible beamline for Ultrafast Ptychography capable of full-field, quantitative, spectro-microscopy with a compact EUV High-order Harmonic Generation (HHG) source, supporting wavelength-scale spatial resolutions. This system will enable a better understanding of light-activated nanoscale function for the efficient engineering of next-generation devices.

2. Ultrafast Ptychography Experimental Setup

EUV to soft X-ray light from HHG has successfully been combined with Ptychographic Coherent Diffractive Imaging (CDI) [1], where multiple diffraction patterns from overlapping fields of view are processed by iterative algorithms [2] to reconstruct the real-space image of the specimen. The ptychography algorithm can solve for the amplitude and the phase of the sample - separately, plus the image-forming beam. The amplitude image exhibits quantitative sensitivity to material composition. The phase image is sensitive to both material composition and topography, and it can be used to extract two-dimensional transverse full-field images combined with 1-dimensional axial information [3]. Recently, ultrafast EUV (28.9nm) ptychography was demonstrated to visualize the impulsive response of an individual nanostructure irradiated by an ultrafast laser pulse [4]. Here, we report the design of an innovative Ultrafast Ptychography beamline with 13nm and 30nm HHG beams, characterized by higher spatial resolution, and higher throughput. The layout is shown in Fig. 1a. The pump and probe pulses are generated from the same Ti:sapphire regenerative amplifier system (KMLabs, 35fs, 785nm, 3mJ at 5kHz). The probe EUV light is generated via HHG: 13nm (10¹⁰ photons/sec, He at 400-700 torr) or 30nm (10¹² photons/sec, Ar at 30-60 torr). The fundamental driving frequency is eliminated before the sample with rejecter mirrors and metallic filters. The EUV light is focused and overlapped with the pump pulses nearly collinearly onto the sample. The time delay between pump photons and probing EUV light is controlled by a motorized delay-line. At each time delay, ptychography provides a complete description of the specimen in response to a laser-driven excitation, through amplitude and a phase images, in either a transmission or reflection geometry.

3. Beamline Design & Results

We designed the ultrafast multipurpose microscope by in depth analysis of EUV source size, divergence, and high-order aberrations effects through ray tracing simulations, using the open-source ShadowOui through the OrAnge SYNchrotron Suite (OASYS) [5]. Our microscope utilizes sets of multilayer mirrors in a z-fold geometry to filter the probe EUV beam to a narrow bandwidth and to focus it onto the sample (Fig. 1a). The combination of flat and curved (ROC=26cm) mirrors provides a geometry demagnification of 10. The transverse EUV beam profiles were simulated in a range of ±1mm of the tangential imaging plane of the spherical multilayer focusing mirror, extracting from each set the caustic horizontal and vertical traces of the EUV beam. A total of 64 caustic scans from ray tracing were obtained, with the EUV source modeled as a gaussian distribution of rays with 1/e² diameter between 10-150μm and divergence in the range 0.5-2mrad. Figure 1b displays the caustic curve for our 13nm HHG source, with 50μm size and 1.1mrad divergence. The insets display the EUV transverse beam profile changes across the focus.
In Fig. 1c we show the resulting relation between EUV source-size and divergence, and the estimated $1/e^2$ EUV focal spot at the sample in our experiment. The beamline is equipped with an in-line intensity monitor and a spectrometer capable of recording $I_0$ beam data and monitoring the spectral quality of the probe beam every 100 pulses at 5 kHz. The ultrafast microscope chamber is designed with full in-vacuum actuation of optics, sample, detector position and orientation, which allows for flexible selection of Numerical Apertures (NA) from 0.04-0.5, or diffraction-limited spatial resolutions from 150 nm down to 13 nm with 13 nm illumination. EUV light is scattered by the sample and it is collected on an in-vacuum EUV CMOS detector with $11 \mu$m pixel size (AXIS-SXR). Data can be collected at a maximum rate of 24 fps, with each ptychography scan requiring between 50-100 diffraction patterns. These specifications support recording raw data for a 100-images nanoscale movie in only 7 minutes, two orders of magnitude faster than the previous ultrafast ptychography tabletop system [4]. Fig. 1d shows a simulated reconstruction from the ultrafast microscope from a 20 nm-thick Siemens Star patterned onto a 200 nm Silicon membrane. This simulation was obtained from a 13 nm EUV beam focused down to $6 \mu$m $1/e^2$ in the circle of least confusion, obtained from a HHG source with 50 $\mu$m size and 1.1 mrad divergence. With a low NA = 0.087, we obtain images with a diffraction limited resolution of 75 nm. Reconstructions were performed with the matPty package, using 100 iterations of a parallel implementation of the RAA algorithm with the modulus enforced probe constraint [6]. With this system, the throughput of ultrafast ptychography is dramatically increased, relaxing the experimental requirements and sample analysis achievable using this method.

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5. References