Supplementary methods

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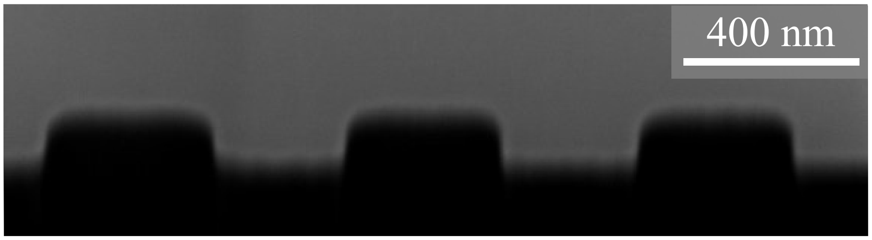
ABSTRACT

1 Methods

1.1 Nanostructured samples

Each scatterometry target consists of two nanoscale line gratings with a rectangular profile etched into a Silicon substrate, as shown in Supp. Figure 1. The samples are designed and fabricated in-house in the NanoLab Amsterdam facilities, via Electron Beam Lithography (Voyager, Raith), followed by dry etching (Cobra, Oxford Instruments). The substrate consists of Czochralski grown, Prime Grade Silicon wafers, with $\{111\}$ crystal orientation (Siegert Wafers gmbh). After the etching process and resist stripping procedures, the samples were cleaned in an Acid Piranha solution at $135°C$ for 10 minutes, followed by an O_2 plasma ashing process. The work presented in the following was carried out on a set of 36 different targets with 700 nm pitch. The structures were made with 18 different designs, including six different groove heights between 80 and 130 nm and three different CDs. For every designed we have manufactured 2 replicas. The morphology **p** of each target was characterized with both Atomic Force Microscopy (AFM) and Scanning Electron Microscopy (SEM).

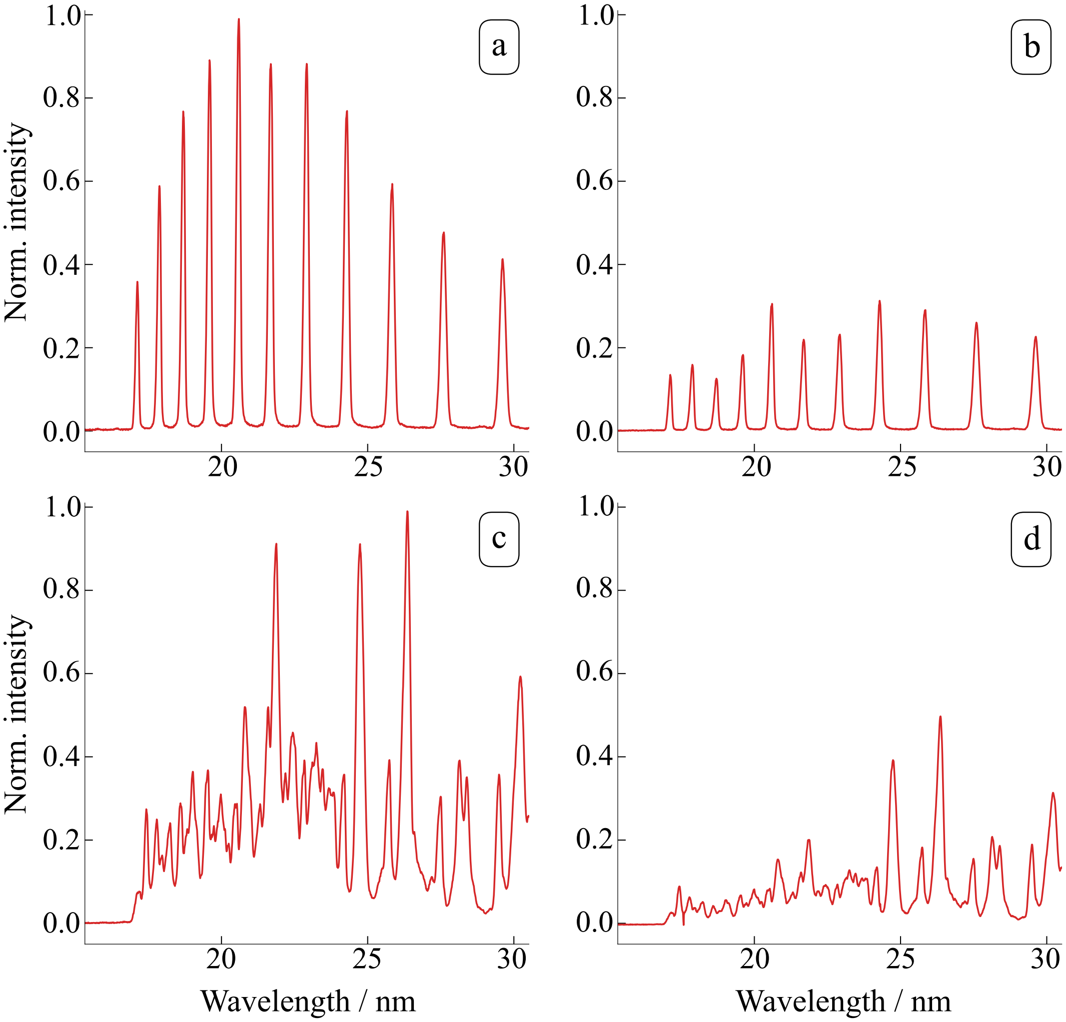


Supp. Figure 1. Grating profile acquired with a SEM imaging. The cross section was obtained by capping a portion of the sample with Ion-Beam-Induced Deposition followed by Ion Milling.

After the fabrication process, the samples were characterized via X-ray Photoelectron Spectroscopy which measured a 1.2 ± 0.1 nm thick native oxide layer on the surface. The evaluation was conducted by comparing the atomic concentrations of the silicon oxide and silicon peaks in the Si-2p region within the binding energy range of $96 - 110$ eV.

1.2 Experimental setup

The primary source is a Ti:Sapphire laser system (Solstice ACE, Spectra Physics), which offers 40 fs pulse duration and an operating repetition rate of 2 kHz. Approximately 3 W are utilized for High Harmonic Generation. The beam is focused using a 250 mm focal length lens into a 3 mm long gas cell reaching $\sim 10^{15}\,\mathrm{W/cm^2}$ peak intensity. The cell was wrapped to obtain the highest pressure difference with respect to the surrounding chamber, and pressurized with gas supplied by a mass-flow controller that ensures a stable flow and pressure. The high-harmonics generation apparatus can be switched between two configurations to allow some tunability over the XUV spectrum: one dedicated to an ordinary HHG regime, characterized by a discrete harmonics spectrum, and one where the generation conditions are tweaked to obtain a continuum-like spectrum. An example of the spectral coverage is shown in Figure 3. The switching between these two generation conditions was achieved by adjusting the focusing position inside the gas cell along the beam propagation direction and by altering the intensity of the focus beam tuning the aperture of an iris placed before the focusing lens.

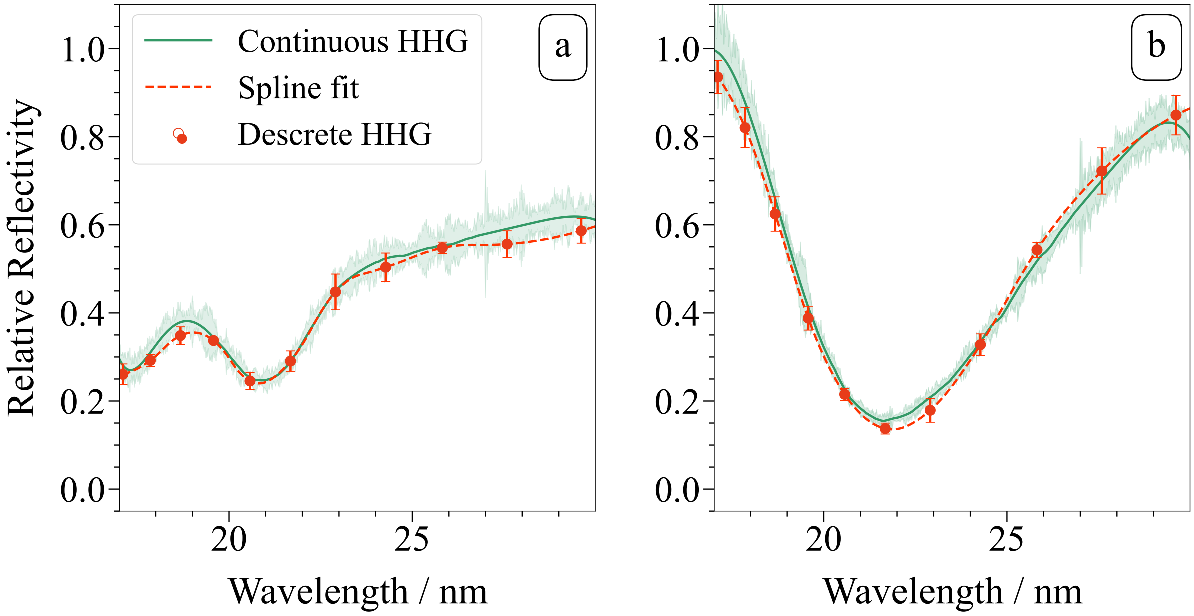


Supp. Figure 2. XUV reflectivity spectra measured under different experimental conditions. a) Discrete spectrum HHG regime, measured on a flat Silicon surface. b) Discrete spectrum HHG regime, measured on a nanostructured sample. c) Nearly continuous spectrum HHG regime, measured on a flat Silicon surface. d) Nearly continuous spectrum HHG regime, measured on a nanostructured sample. b) and d) report the date from 0^{th} order spectra measured on a target with 350 nm CD and 108 nm groove height, in planar diffraction conditions ($\phi = 0°$.

2 Experimental results and discussion

2.1 Data Processing

For quasi-continuous XUV spectra, the relative reflectivity was directly derived by dividing the measured zeroth-order spectrum by that obtained from a pristine silicon reference surface. A narrow Gaussian low-pass filter was applied in the Fourier space to minimize artifacts caused by rapid signal oscillations while preserving the overall signal integrity. It is important to note that the beyond phase-matching regime usually presents spectral fluctuations that are significantly larger than those of the optimal HHG regime. For this reason, we chose to discuss and show our morphology reconstruction results for the optimal HHG regime, while we only use data acquired with a nearly continuous illumination spectrum to choose the most fitting and interpolation routine that best describes the signal measured with a continuous illumination spectrum. After the exploration of different fitting methods, such as polynomial and sum of trigonometric functions, we have concluded that the most suitable fitting procedure for the hereby observed relative reflectivity signal is the piecewise polynomial interpolation, also known as the spline interpolation routine, as shown in Figure 3. It emerges that the curve fitted to the measured relative reflectivity with discrete HHG correctly maps the signal measured with nearly continuous spectrum within the $1-\sigma$ uncertainty interval.



Supp. Figure 3. Comparison of the measured relative reflectivities probed with XUV light generated in both discreet and continuum-like HHG regime. a) Planar diffraction configuration ($\phi = 0°$). b) Conical diffraction configuration ($\phi = 90°$).