Supplementary Information for:

Single-digit-resolution nanopatterning with extreme ultraviolet light for the 2.5 nm technology node and beyond

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S1. Mask fabrication

All EUV transmission-grating masks (Fig. 3) were fabricated on a 100-nm-thick Si₃N₄ membrane. The membranes were made by first depositing low-stress Si₃N₄ films on Si wafers using low-pressure chemical vapor deposition. Using photolithography and KOH etching from the backside these free-standing membranes were obtained in a 3x3 mm² area. On the membranes a Cr layer of 11 nm was evaporated, which improves the adhesion of the resist and serves as a conducting layer for EBL. The high-resolution Inpria IB photoresist was spin-coated with 1500 rpm on the membrane, which was pre-treated by oxygen plasma in order to render the surface hydrophilic and increase the adhesion of the resist. Before the exposure, the resist was post-applied backed (PAB) at 80°C for 180s. Line-space gratings were written by EBL (VISTEC EBPG 5000PLUS 100 KeV) in a dose range from 2000 up to 9000 μC/cm² depending on the periodicity and the desired linewidth. Afterwards the resist was post-exposure baked (PEB) at 80°C for 60s and developed in a tetramethylammonium hydroxide (TMAH) 25% solution for 30s, rinsed in water and dried with nitrogen blow. The line gratings obtained by this procedure had h=20 nm. The Cr between the gratings was then etched, using Cl-based reactive-ion etching (RIE) (see S5), which also improves diffraction efficiency of gratings. SEMs of the resolution capabilities of EBL before (top row of Fig. S1) and after (bottom row of Fig. S1) Cr etching illustrate high quality lines with very low LER, which results in better mask performance. The last step of the mask fabrication is making the photonstops that block the non-diffracted beams. In this step an overlay EBL exposure was made on a 100-nm-thick PMMA and after development a seed layer of Cr-Au was thermally evaporated. After successful PMMA liftoff in acetone, Au film of 400 nm thickness was electroplated.
Fig. S1 SEMs of gratings made of the Inpria resist on Si$_3$N$_4$ membranes with different periodicities. The top row shows the gratings before Cr etching and the bottom row shows the same gratings after Cr etching.
S2. Diffraction efficiency simulations

We used rigorous coupled-wave analysis (RCWA) method in order to characterize and optimize the diffraction properties of the gratings upon illumination with EUV light. These simulations provide a good idea of the required photoresist thickness and grating duty cycle (DC), defined as the ratio between the linewidth of the gratings to the periodicity. The light is coherent EUV at 13.5 nm wavelength, and the diffraction grating periods are over the range of 20 nm to 200 nm. The refractive indices for these simulated grating materials were obtained from the Berkeley X-Ray Optics database. Fig. S2 illustrates the calculated diffraction efficiencies of the 0th, 1st, and 2nd-order diffracted beams from HfO$_2$ gratings with $P_g=36$ and 28 nm as a function of the DC and grating height, $h$. The calculations show significantly higher efficiency of the 1st-order diffraction compared to the 2nd order, which is why in our experiments we rely on the interference of the 1st-order diffracted beams. In these simulations the used experimental mask configuration was modeled. This configuration consists of a 100 nm Si$_3$N$_4$ membrane, $h=25$ nm tall HfO$_2$ gratings on top of 11 nm of Cr. The highest 1st-order diffraction efficiency occurs when DC is about 0.5.
Fig. S2 Calculated field intensity of the the 0th, 1st, and 2nd order diffracted beams as a function of HfO$_2$ grating height ($h$) and duty cycle for (a) $P_g$=36 nm and (b) $P_g$=28 nm.
S3. Critical dimension (CD) as a function of dose for Inpria XE15IB (IB)

For a mask with resolution down to HP=16 nm we carried out a CD analysis and plotted its variation as a function of the dose on wafer.

**Fig. S3** Measured critical dimension as a function of dose on wafer.
S4. EUV lithography and photoresist preparation

We tested the patterning performance of two negative-tone inorganic resists (HSQ and Inpria IB) using EUV-IL. HSQ (XR-1541 2% from Dow Corning) was diluted with MIBK 1:1 and spincoated with 4000 rpm, resulting in a ≈25 nm thick layer. After EUV exposure it was developed for 120 s in a NaOH buffer solution (MICROPOSIT™ 351, Rohm and Hass), rinsed with water and dried with nitrogen blow. Inpria IB was spincoated with 2500 rpm for 45 s on the Si wafer pretreated with oxygen plasma. This resulted in a ≈25 nm thick layer, which was further PAB at 80°C for 180s. After the EUV exposure PEB was done at 80°C for 60s and development in a TMAH 25% solution for 30 s, followed by rinsing with water and drying with nitrogen blow. The wafers were then mounted on a laterally moving stage in the vacuum chamber of the beamline. The fabricated masks are mounted on a special mask holder that is positioned in front of the synchrotron beam. The axial distance of the mask is adjusted for highest overlap of the diffracted beams on the wafer. The dose-on-mask ($D_m$) is controlled by adjusting the opening time of a mechanical shutter that is placed in front of the beam and before the mask.
S5. Reactive-ion etching

Dry etching of silicon was done using a reactive-ion etching (RIE) machine with the induced coupled plasma (ICP) head (Oxford Instruments, Plasmalab System 100). The recipe contained C4H8/SF6 (15/10 sccm) under the pressure of 10 mTorr with RF and ICP power of 40 and 400 W respectively. This recipe showed very high selectivity in etching Si with respect to HfO2.

Dry etching of chromium was also done by RIE (BMP Plasma Etcher). The recipe contained Cl2 /CO2 (100/100 sccm) under the pressure of 100 mTorr. This recipe showed rather isotropic etching in Cr.