Electron beam lithography and pattern transfer on membrane grids for transmission electron microscopy

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We describe a fabrication process for electron beam lithography (EBL) and the following pattern transfer steps on transmission electron microscope (TEM) grids. For demonstration purposes, we use commercial off-the-shelf TEM grids consisting of a thin suspended silicon nitride membrane on a silicon frame supporting substrate. For the pattern transfer, we demonstrate both an additive patterning technique with metal deposition and lift-off [1], and a subtractive patterning technique with reactive ion etching [2]. This process could enable direct nanofabrication on TEM grids for electron microscopy and spectroscopy, optical elements for electron and charged particle beams, as well as suspended or membrane-based nanoscale devices.

The transmission electron microscope (TEM) provides ultra-high spatial resolution microscopy and spectroscopy techniques for nanomaterials. Besides high resolution imaging, there has been a growing interest in using TEM to investigate the mechanical, electrical, and optical properties of artificially fabricated nanostructures via top-down lithography. However, nanofabrication processes that usually work at the wafer-level and die-level are quite challenging when applied to TEM grids (standard TEM sample holders) due to difficulties in handling the grid, which typically consists of a thin, electron-transparent membrane (few to tens of nanometers thick) on a supporting frame with 2-3 mm diameter. In this report, we outline a fabrication process of electron beam lithography (EBL) and pattern transfer on silicon nitride (SiN hereafter) TEM grids. This process has enabled several experiments, including aluminum nanodisks for electron energy loss spectroscopy [1] and SiN mesh gratings for electron diffraction [2]. Beyond these examples, the reported fabrication process could also be used in stencil (shadow mask) lithography, high-resolution backscattering-free charged particle lithography, and suspended or membrane-based nanoscale devices.

EBL ON TEM GRIDS

For the demonstration of the fabrication process, we use SiN TEM grids from *SiMpore Inc.* with a membrane thickness of 5 nm and 10 nm on a supporting silicon (Si) frame with opening windows. Before fabrication, the TEM grid substrate is cleaned. Due to the fragile membrane on the TEM grid, sonication should be avoided. A gentle rinse with acetone and isopropanol (IPA) is sufficient for most applications. After cleaning, oxygen plasma ashing is used, both to remove the residual solvents and organic contamination, and to promote resist adhesion. The ashing time and plasma power should be carefully controlled to avoid deteriorating the SiN membrane. Figure 1 shows a long ashing time (6 min) leads to film corrugations in the SiN membrane and the spin-coated resist film. The optimal ashing time depends on several factors, including the SiN membrane thickness, the window size, and the film stress. Typically, the ashing time should be kept below 1 min.

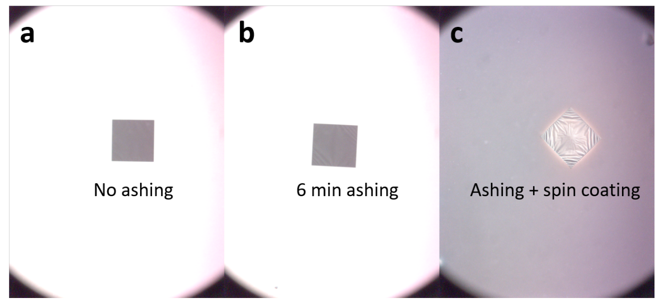

Figure 1. **SiN film corrugations induced by oxygen plasma ashing**. (a) SiN membrane before oxygen plasma ashing. (b) SiN membrane after 6 min oxygen plasma ashing. (c) PMMA spin-coated on SiN membrane after 6 min oxygen plasma ashing.

After ashing, EBL resist is spin-coated onto the TEM grid. Typical spin coaters use vacuum to hold the wafer or die. The TEM grid should not be directly placed on the vacuum chuck, as vacuum will break the membrane. Instead,the TEM grid should be taped (backside) to a dummy silicon die, and the dummy die is put onto the spin chuck. Regular spin coating procedure is then followed to spin-coat $\sim$ 70-nm-thick polymethyl methacrylate (PMMA), a positive tone EBL resist, on the SiN membrane. After spin coating, the TEM grid is detached from the dummy die, and soft baking is performed at 180 °C for 2 min. When taping and removing the grid, care should be taken not to break the membrane.

After spin coating, the resist film is inspected with an optical microscope to determine whether one can proceed with EBL on the film. Figure 2 shows the film quality of 4 commonly observed resist films spin-coated on a 5-10 nm SiN membrane. In Figure 2a, a uniform resist film is obtained. In Figure 2b, corrugations in the film is observed, most likely due to long time oxygen plasma ashing. Figure 2c shows a thick and non-uniform resist film, as evidenced by the optical interference fringes

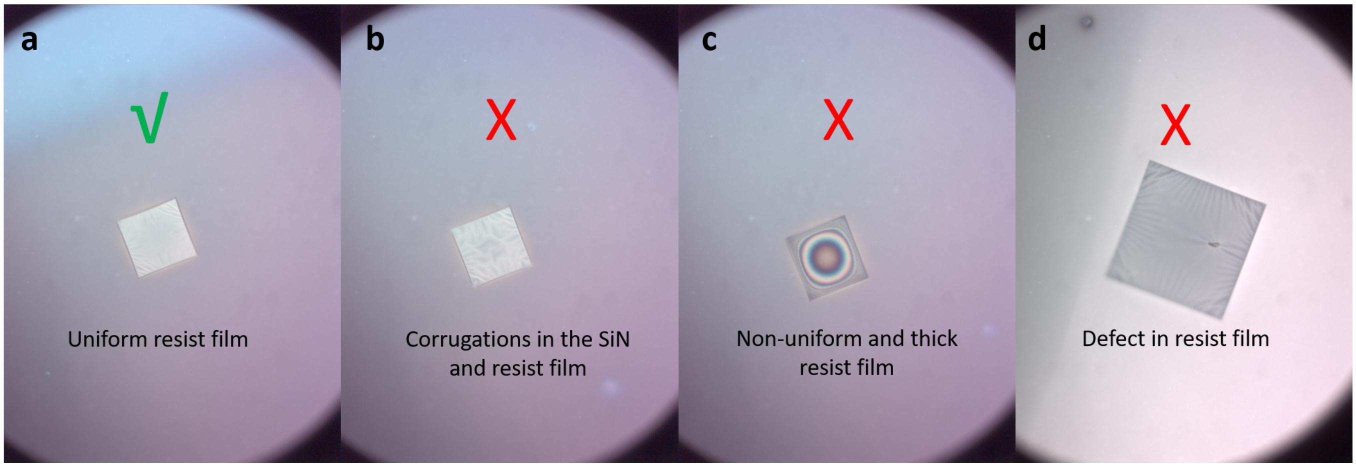

Figure 2. **PMMA resist film quality after spin coating**. (a) A uniform resist film. (b) Corrugations in the SiN and the resist film. (c) A non-uniform and thick resist film. (d) A defect in the resist film. Out of the 4 resist films, electron-beam lithography should only be performed with the uniform film (a).

(Newton's rings). In Figure 2d, a defect is observed in the resist film, causing non-uniformity around it. EBL should only be performed with a uniform resist film as shown in Figure 2a.

Exposure is performed with the Elionix-F125 EBL system with a typical areal dose of 4800-6400 μC/cm^2 (300-400 e^-/nm^2). Due to insufficient optical reflection, the interferometric height sensor does not function properly and is turned off, and the EBL is performed with a constant stage height. The proper stage height is determined before exposure by calibrating the working distance from the scanning electron microscope (SEM) images of either the Si frame of the TEM grid or an exposed spot in PMMA created by spot exposure near the target patterning region. If more accurate stage height and active correction are required during exposure, automatic stage movement and correction can be enabled according to the membrane surface height and tilt determined from height and position measurements at three or more points. After exposure, development is performed at 0 °C in 3:1 IPA:MIBK for 30 sec.

ADDITIVE PATTERN TRANSFER

For additive pattern transfer, we demonstrate the fabrication of sub-10-nm aluminum nanodisks with a lift-off process. In the EBL process, dot exposure with varying doses is used to create nanodisk patterns with different diameters. After resist development, we deposit aluminum with a thickness of 15-20 nm via electron beam evaporation. Lift-off is done in N-methyl-2-pyrrolidone (NMP) at 50-60 °C for 60-120 min. After lift-off, the sample is rinsed with acetone and IPA. As a last step, a gentle oxygen plasma ashing is used to remove the residual resist and solvents. A reverse action tweezer is used to hold the sample to facilitate ashing of both sides of the grid. Figure 3a shows the SEM image of an array of nanodisks with a large diameter created with a high exposure dose, and Figure 3b shows the TEM image of another array created with a low exposure dose. More details on relevant experiments can be found in ref. [1].

SUBTRACTIVE PATTERN TRANSFER

For subtractive pattern transfer, we demonstrate the fabrication of large-area mesh gratings in SiN membranes. EBL generates patterns of mesh gratings (square arrays of circles) over an area of 100 × 100 μm^2 on the SiN membrane. CF_4 reactive-ion-etching (RIE) transfers the pattern into the membrane by etching through-holes. After etching, residual PMMA resist is stripped with oxygen plasma ashing. An optional final metallization step can be performed via evaporation. Figure 4a shows a TEM grid with several mesh gratings fabricated in multiple windows. Figure 4b & c show one large-

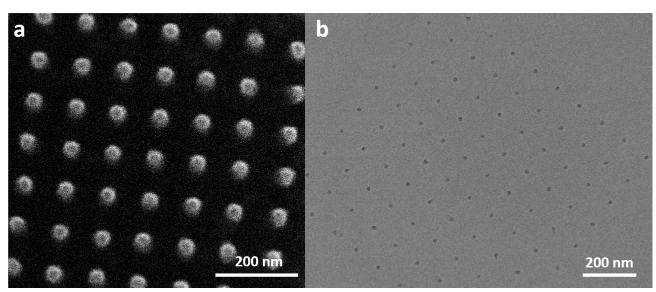

Figure 3. **Aluminum nanodisks fabricated on a 5-nm-thick SiN membrane**. (a) SEM image of an array of nanodisks with a large diameter created with a high exposure dose. (b) Bright-field TEM image of an array of nanodisks with a small diameter created with a low exposure dose.

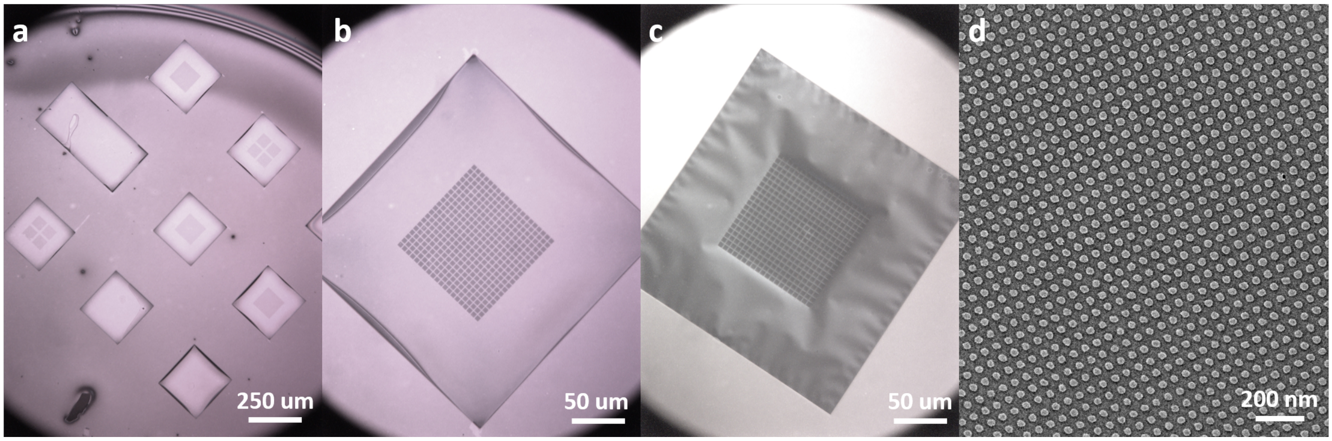

Figure 4. **Mesh gratings (hole arrays) fabricated in suspended SiN membranes**. (a) Optical image of the TEM grid with several mesh gratings fabricated in multiple windows. (b) Optical image of one window on the TEM grid with a large-area mesh grating. Only the supporting bars [2] can be seen, and the grating mesh is too small to be resolved. (c) The same as (b), but after coating with a 10-nm-thick aluminum film. (d) Bright-field TEM image of the fabricated mesh grating. The grating half-pitch is 50 nm.

area mesh grating with and without metallization (10-nm-thick aluminum), respectively. Figure 4d shows a bright-field TEM image of the mesh grating with a 25 nm half-pitch. More details on relevant experiments can be found in ref. [2].

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