

Video Article

Fabrication of Magnetic Nanostructures on Silicon Nitride Membranes for Magnetic Vortex Studies Using Transmission Microscopy Techniques

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Abstract

Electron and x-ray magnetic microscopies allow for high-resolution magnetic imaging down to tens of nanometers. However, the samples need to be prepared on transparent membranes which are very fragile and difficult to manipulate. We present processes for the fabrication of samples with magnetic micro- and nanostructures with spin configurations forming magnetic vortices suitable for Lorentz transmission electron microscopy and magnetic transmission x-ray microscopy studies. The samples are prepared on silicon nitride membranes and the fabrication consists of a spin coating, UV and electron-beam lithography, the chemical development of the resist, and the evaporation of the magnetic material followed by a lift-off process forming the final magnetic structures. The samples for the Lorentz transmission electron microscopy consist of magnetic nanodiscs prepared in a single lithography step. The samples for the magnetic x-ray transmission microscopy are used for time-resolved magnetization dynamic experiments, and magnetic nanodiscs are placed on a waveguide which is used for the generation of repeatable magnetic field pulses by passing an electric current through the waveguide. The waveguide is created in an extra lithography step.

Video Link

The video component of this article can be found at <https://www.jove.com/video/57817/>

Introduction

The magnetism of nanostructures was intensively studied in the last two decades following technological trends towards miniaturization. As the lateral dimensions of the structures become smaller and smaller, the magnetic properties of ferromagnetic structures start to be governed by the structure geometry in addition to the properties of the magnetic material. The behavior of different magnetic elements from bulk materials to microstructures has been reviewed in detail (e.g., by Hubert and Schäfer)¹. One of the most known examples of non-trivial magnetization ground state is magnetic vortices-curling magnetization structures occurring in micron- and submicron-sized thin magnetic discs and polygons. The magnetization here is curling in-plane around an out-of-plane vortex core^{2,3}. The magnetization reversal of magnetic vortices has been extensively studied in both static^{4,5,6} and dynamic^{7,8,9,10} regimes. The possible applications of magnetic vortices are, e.g., multi-bit memory cells¹¹, logic circuits¹², radio-frequency devices¹³, or spin-wave emitters¹⁴.

To image a magnetic vortex and especially the vortex core, the spatial resolution of the microscopy technique should be as close as possible to fundamental magnetic length scales (below 10 nm). Lorentz transmission electron microscopy¹⁵ (LTEM) and magnetic transmission x-ray microscopy¹⁶ (MTXM) are ideal candidates for the imaging of magnetic vortices as they offer a high spatial resolution and MTXM also offers a high temporal resolution for magnetization dynamics studies. The disadvantage of these techniques is the complicated sample preparation, which is the subject of the presented paper.

The processes presented here explain the fabrication of samples used for imaging magnetic vortices by TEM¹⁷ and MTXM^{10,11}. Both techniques are of transmission character, and because of that, it is necessary to fabricate the structures on thin membranes. The membranes are typically made from silicon nitride and their thickness ranges from tens of nanometers to a few hundreds of nanometers. Each of these two methods requires a different support frame geometry. In the case of MTXM, the frame is 5 x 5 mm² and the window is large, 2 x 2 mm². In the case of TEM, the frame geometry is a circle of 3 mm in diameter with the window size dependent on the experiment, typically 250 x 250 μm². The membranes bring additional challenges of more difficult sample handling with the risk of breaking the windows during all the lithography processes.

The fabrication of samples can be done by both positive and negative resist lithography techniques¹⁸. The positive resist lithography process uses a positive resist; the chemical structure of the resist changes upon irradiation and the exposed part will become soluble in the chemical developer. The exposed area will wash away while the unexposed area will remain on the substrate. In the case of a negative resist lithography process, the irradiation hardens the resist and the exposed area will remain on the substrate while the unexposed area will wash away in the chemical developer. Both techniques can be used for the fabrication of the samples, but we prefer positive resist lithography because it requires

fewer fabrication steps when compared to the negative resist lithography technique. It is also easier to handle, faster, and often provides better results.

Protocol

We demonstrate a method for the fabrication of samples for TEM and MTXM. The permalloy nanodiscs with diameters ranging from 250 - 4000 nm and thicknesses between 20 - 100 nm are fabricated on 30 nm thick SiN membranes for TEM and 200 nm thick SiN membranes for MTXM. Photographs of the SiN membranes are shown in **Figure 1**.

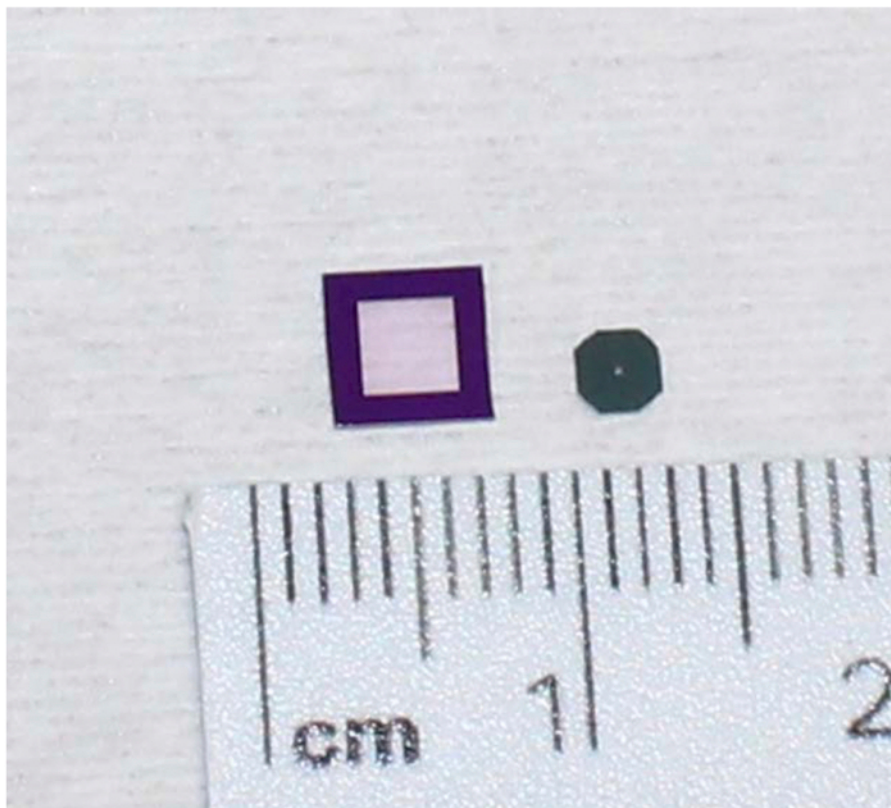


Figure 1: Photograph of SiN membranes used as a substrate for MTXM (left) and TEM (right) samples. The image shows size comparison to a ruler. The MTXM frame is a 5 x 5 mm rectangle with a window thickness of 200 nm and the TEM frame fits a 3 mm circle in diameter with a window thickness of 30 nm. [Please click here to view a larger version of this figure.](#)

1. Fabrication of the Samples for TEM

Note: In this section, we describe the fabrication of the samples for TEM which is used for the observation of the nucleation process of magnetic vortices¹⁷. The membranes are chosen as the substrates because they offer a solid support for the lithographical fabrication of magnetic structures. An important parameter is the membrane window thickness. A higher accelerating voltage allows penetrating thicker samples, but any unnecessary thickness will cause a loss of signal¹⁹. For that reason, we use the thinnest membranes available from our supplier (30 nm).

1. Substrate preparation and spin coating

Note: Spin coating is a widely used process to get a desired resist film on the substrate. A small amount of resist is dropped on the substrate which is then rotated at a very high speed to get the desired coating thickness. The spin coating of TEM membranes is rather peculiar because of the following reasons: (i) if the membrane is spun on the spin coater's axis, the resist will not be uniform due to the membrane's small diameter and (ii) vacuum holders cannot be used because they can break the membrane. For this purpose, we designed 3D printed adapters (see **Figure 2**) that hold the membrane off-axis and do not require a vacuum to hold the sample.

1. Prebake the SiN membranes on a hot plate at 180 °C for 15 min to remove any moisture.
2. Inserts adaptor onto spin coater and then places membranes in adapter.
3. Spin coat 950 K PMMA (poly-methyl-methacrylate) resist at 3,000 rpm for 1 min to produce a film thickness of approximately 200 nm.
4. Post-bake the samples on the hot plate at 180 °C for 3 min to harden the PMMA layer.



Figure 2: Photograph of 3D printed adapter, used to hold the TEM membrane off-axis during the spin coating. Multiple membranes can be coated at the same time. [Please click here to view a larger version of this figure.](#)

2. Electron-beam lithography (EBL)

1. Draw the desired pattern of discs in the Graphic Database System (GDS) format and upload it to the electron-beam (e-beam) lithography system.
2. Load the samples into the e-beam writer system, set the stage, and beam.
3. Expose the disc area to an electron dose of $260 \mu\text{C}/\text{cm}^2$ at the beam energy of 20 keV.
Note: The suitable parameters of the exposition process are a beam current of 250 pA and a step size of 10 nm. This dose is approximately 30% higher when compared to bulk substrates as the backscattering is highly reduced on the membranes.

3. Chemical development

1. After the exposure, develop the samples in a methyl isobutyl ketone (MIBK)-based developer for 2 min. Stop the development by using isopropyl alcohol (IPA) for 30 s.
2. Wash each sample in deionized water for 30 s and blow-dry it using nitrogen while holding with a tweezer.
3. Check the development of the samples using an optical microscope at a low magnification first (using a 5X objective) and then at a high magnification (using a 100X objective); the optical microscope image of a developed sample is shown in **Figure 3**.

4. Electron-beam evaporation

Note: Electron-beam evaporation²⁰ is a form of physical vapor deposition in which a target anode is bombarded with a high energy electron-beam produced by a charged tungsten filament under a high vacuum. The electron-beam causes atoms from the target to transform into a gaseous phase. These atoms precipitate into a solid form and coat everything in the vacuum chamber with a thin layer of the target material. It is better to use an e-beam evaporation system for a lift-off purpose as it gives a nice edge to the disc without depositing any extra material on the disc boundary.

1. Tape the membranes carefully using poly-oxydiphenylene-pyromellitimide (e.g., Kapton) onto the holder and transfer it into the deposition chamber of the e-beam evaporator via load lock.
2. Use the electron-beam evaporation system to deposit a thin layer of permalloy ($\text{Ni}_{80}\text{Fe}_{20}$) with thicknesses ranging from 20 to 100 nm at the deposition rate of approximately $1 \text{ \AA}/\text{s}$. Use the acceleration voltage of 8 kV and beam a current of approximately 120 mA.

5. Lift-off

1. Put the membranes for 1 h into a beaker with acetone (with at least 99.5% purity).
2. Now spray the membranes with acetone while holding each with a tweezer until the excess metal is removed.
3. If the excess metal remains on the sample, place the membranes back into the beaker and repeat the procedure.
Note: Optionally, a megasonic bath could be used to assist the lift-off procedure. Note that it is not possible to use a classical ultrasonic bath as it breaks the membranes.
4. Image the final array of the magnetic discs with a scanning electron microscope (SEM) at the accelerating voltage of 5 kV and a beam current of 100 pA for the final inspection. An image at the magnification of 100,000X is shown in **Figure 3b**.

6. LTEM imaging

1. Mount the sample into the TEM sample holder and insert it into the microscope.
2. Correct the sample height and align the microscope in the Lorentz mode at the desired accelerating voltage (in our case 300 kV) using the standard procedures of the microscope.

3. Introduce the magnetic signal by defocusing the Lorentz lens.
4. Carry on with the experiment. Tilt the sample in order to introduce the in-plane field component (e.g., the suitable angle is 30°, check the holder specification for the maximum tilt angle).
5. Apply the magnetic field by exciting the objective lens (normally turned off in the Lorentz mode).
Note: The field calibration curve should be provided by the TEM manufacturer.
6. Saturate the sample, gradually decrease the magnetic field by deexciting the objective lens and capture the images on camera.
Example results are shown in **Figure 3c**.

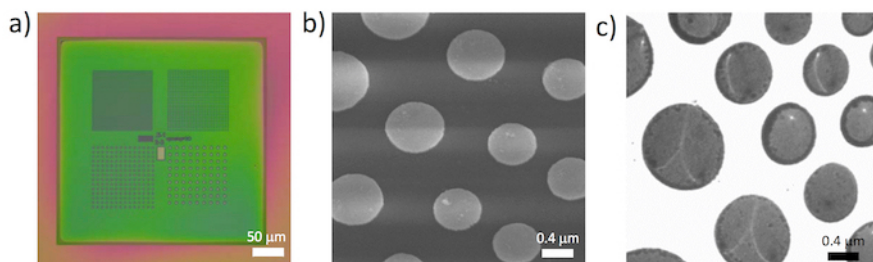


Figure 3: Final sample imaged by optical and electron microscopies. (a) This panel shows the silicon nitride membrane window with the arrays of discs in the resist after the electron-beam exposure and resist development. (b) This panel shows the final array of the magnetic discs imaged by the SEM. (c) This panel shows the LTEM image of the magnetic vortices nucleation states in an array of the magnetic nanodiscs. [Please click here to view a larger version of this figure.](#)

2. Fabrication of the Samples for MTXM

Note: In MTXM measurements, we can take advantage of the technique’s time resolution. In order to introduce a high-frequency excitation of the magnetic vortices, fabricate a gold waveguide in the first step and then place magnetic discs on top of the waveguide in the second lithography step. The whole structure is fabricated on a 200 nm thick SiN membrane which is transparent enough for soft x-rays²¹. The detailed steps are described in the following text and the schematic of the process is shown in **Figure 4**. The process of the MTXM sample fabrication goes through all the steps described above for the TEM samples fabrication but an additional lithography step is required to fabricate the waveguide.

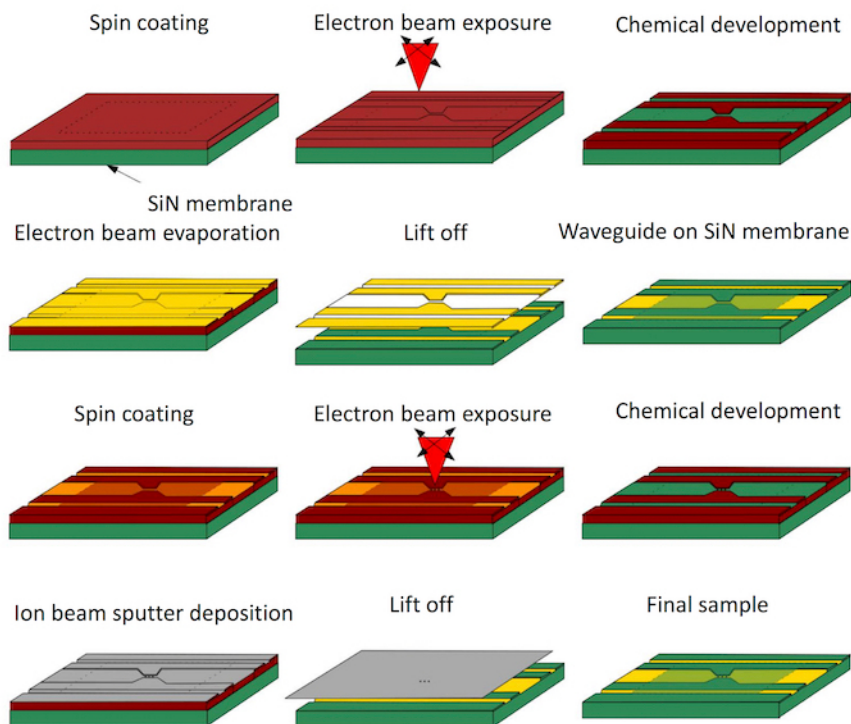


Figure 4: Schematics of preparation steps of a sample with discs and a waveguide on a silicon nitride membrane for MTXM time-resolved experiments. It involves a two-step lithography to obtain the final structure. [Please click here to view a larger version of this figure.](#)

1. Sample preparation and spin coating

Note: The membrane for MTXM is a 5 x 5 mm² frame with a 3 x 3 mm² wide and 200 nm thick central window. The membrane cannot be put on the spin coater vacuum chuck as it would break the membrane. In this case, we glued the membrane to a 10 x 10 mm² silicon wafer to make it easy to work with.

1. Prebake the SiN samples on a hot plate at 180 °C for 15 min to remove any moisture from the samples.
2. Spin coat the positive resist CSAR at 3,000 rpm for 1 min; the resulting film thickness is approximately 500 nm.
Note: This type of resist was chosen for its higher sensitivity resulting in faster writing times. The thickness-spin speed dependence can be found on the resist data sheet.
3. Post-bake the samples on a hot plate at 150 °C for 1 min to harden the resist layer.

2. Electron beam lithography of the waveguide

1. Create the desired pattern of the waveguide and alignment marks (for the second lithography steps) in GDS format and upload it to the e-beam lithography system.
2. Load the samples into the e-beam writer system, set the stage, and beam.
3. Expose the disc area to an electron dose of 65 μC/cm² at the beam energy of 20 keV, a beam current of 10 nA, and a step size of 200 nm.

Note: We used CSAR positive resist for exposing the waveguide and alignment marks. This resist has a higher sensitivity than PMMA with the electron dose of 65 μC/cm² at the beam energy of 20 keV and, therefore, is favorable to speed up the exposition.

3. Chemical development

1. After the exposure, develop the samples in the developer for 1 min and follow that with a stopper (IPA) for 30 s.
2. Wash the samples in deionized water for 30 s and blow-dry them with nitrogen while holding them with the tweezers.

4. Electron-beam evaporation

1. Use the e-beam evaporator to deposit the Ti(3nm)/Au(100nm) bilayer for the waveguide and alignment marks.
2. Rotate the samples continuously at a rate of 10 rpm to increase the layer homogeneity.
Note: The titanium layer of 3 - 5 nm makes an adhesive contact between the SiN sample and the Au layer. The thickness of the Au layer is typically between 80 - 120 nm. This range of thickness is suitable for the wire bonding of the sample to custom-made printed circuit boards, which we used to inject current pulses into the waveguide.
3. Use a deposition rate for Ti of 0.5 - 0.7 Å/s and for Au of ~2.5 Å/s. Maintain the base pressure of the e-beam system at approximately 10⁻⁷ mbar or better.
4. Alternatively, use Cu instead of Au for the fabrication of the waveguide for a better transparency for the soft x-rays.

5. Lift-off

1. After the deposition of the Ti/Au thin film, put the samples in acetone for 1 h.
2. Now, spray the membranes with acetone while holding them with tweezers until the excess metal is removed.
3. If the excess metal remains on the samples, place them back to the beaker with acetone and repeat the procedure.
Note: Optionally, a megasonic bath could be used to support the lift-off procedure. The samples with the Ti/Au waveguide structure and with alignment marks go through the same lithography steps again for the fabrication of the magnetic discs on the waveguide.

6. TiO₂ deposition

1. Insert the samples with the waveguide and alignment marks into the atomic layer deposition system and deposit 20 nm of the TiO₂ layer to make the insulating layer between the waveguide and the discs.
2. Use the Ti precursor Tetrakis(dimethylamido)titanium (TDMAT) and H₂O for the deposition of TiO₂ by oxygen plasma and grow it at a rate of 0.51 Å/cycle.

7. Spin coating of samples

Note: We used a double layer of resist to increase the edge quality of the discs. During the e-beam exposure, the bottom resist is overdosed, and after the development, it provides a refined undercut.

1. Prebake the samples on the hot plate at 180 °C for 15 min to remove the moisture.
2. Spin coat the copolymer resist at 4,000 rpm for 1 min.
Note: The resulting film thickness is approximately 30 nm.
3. Post-bake the samples on the hot plate at 180 °C for 3 min to harden the resist.
4. Spin coat the PMMA 950K resist at 4,000 rpm for 1 min. The resulting film thickness is approximately 270 nm.
5. Post-bake the samples at 180 °C for 3 min to harden the resist.

8. Electron-beam lithography of discs

1. Create the second lithographical pattern of discs in the GDS format and upload it to the e-beam lithography system.
2. Use the global marks to align the UV coordination system to the sample.
3. Use the local marks to align the write-field in order to calibrate the write-field's size, rotation, and shift to ensure a correct position of the discs on the waveguide.
4. Expose the disc area to an electron dose of 220 μC/cm² at a beam energy of 20 keV. Use a beam current of 200 - 300 pA and a step size of 10 nm for exposing the pattern.

9. Chemical development

1. Develop the samples in an MIBK-based developer for 1 min, and follow that with a stopper (IPA) for 30 s.
2. Then rinse the samples in deionized water for 30 s and blow-dry them with nitrogen while holding them with tweezers.

10. Ion-beam sputter deposition

1. Insert the samples into the ion-beam sputtering system.
2. Tilt the sample holder by 30° with respect to the direction of the sputtered material in order to taper the discs by the shadowing effect.
Note: The tapering is used to control the switching of the vortex circulation¹¹.

3. Deposit a 20 - 50 nm thick permalloy ($\text{Ni}_{80}\text{Fe}_{20}$) layer using a deposition rate of 0.5 - 0.7 $\text{\AA}/\text{s}$ at a working pressure of approximately 10^{-5} mbar.
Note: The base pressure should be 10^{-7} mbar or better.

11. Lift-off

1. Put the samples in acetone for 1 h.
2. Now, spray the membranes with acetone while holding them in a tweezer until the excess metal is removed.
3. If the excess metal remains on the sample, place the sample back to the beaker with acetone and repeat the procedure.
Note: Optionally, a megasonic bath could be used to support the lift-off procedure. We got the final structure of the permalloy discs over a Ti/Au waveguide on a SiN membrane as shown in **Figure 5**.

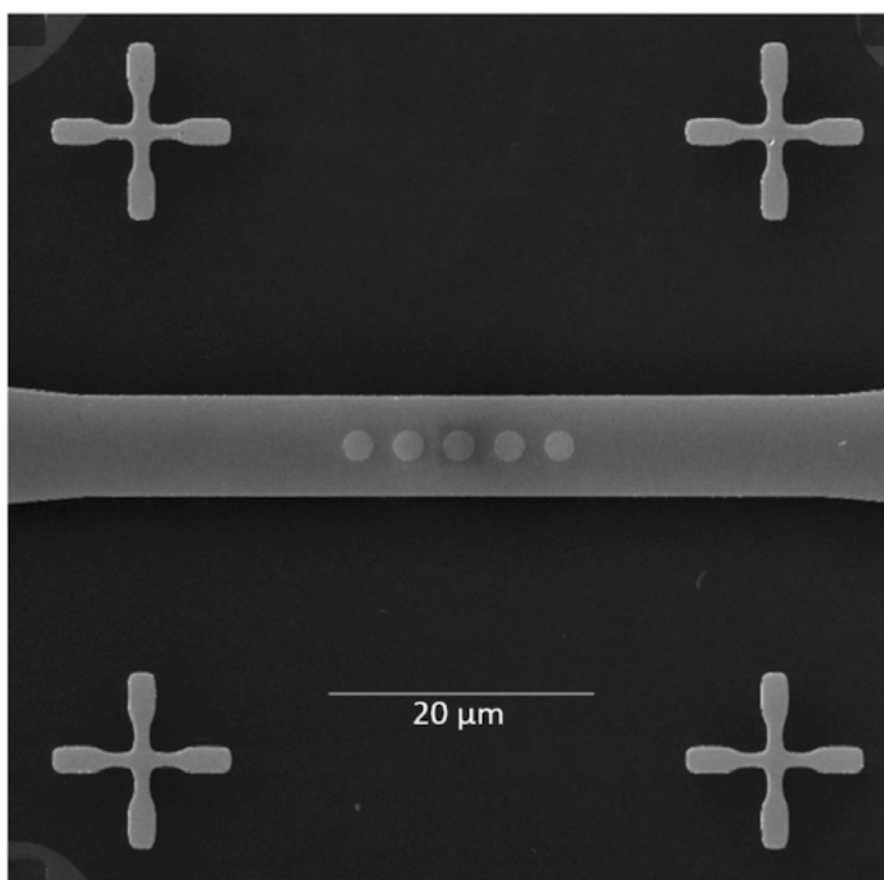


Figure 5: SEM image of the final structure of 30 nm thick and 2 μm wide permalloy discs on a gold waveguide with alignment marks. The samples are used further for time-resolved MTXM experiments. [Please click here to view a larger version of this figure.](#)

Representative Results

Figure 1 shows the photographs of the SiN frames and membranes used for the MTXM and LTEM microscopies. **Figure 2** shows the design of the 3D-printed adapters to hold the TEM membrane off-axis during the spin coating process. **Figure 3** shows the various steps of the LTEM sample preparation (after the resist development and after the lift-off procedure) and the final image observed by the LTEM. **Figure 4** shows the schematics of the preparation steps for the fabrication of the discs and the waveguide on the SiN membrane for the MTXM time-resolved experiments. **Figure 5** shows the final MTXM sample containing the discs placed on a waveguide.

Discussion

We have demonstrated the fabrication of samples for LTEM and MTXM magnetic microscopies. These samples need to be fabricated on thin SiN membranes so that the electrons, in the case of the LTEM, and the soft x-rays, in the case of the MTXM, can penetrate through the samples. These samples can be fabricated either by 1) a positive resist lithography or by 2) a negative resist lithography.

We used the positive resist lithography technique because it requires less sample preparation and fewer fabrication steps and allows easier processing. It also allows the researcher to use the shadowing effect, which we used for the precise disc shape control (a tapering of one side of the disc). This shape was used to control the circulation of the magnetic vortices during nucleation^{10,11}.

The disadvantage of this technique is the complicated lift-off procedure because the thin film material is sometimes deposited on the resist edge and then cannot be removed by a lift-off. We solved this problem by using a double resist layer. This slightly limits the resolution (approximately 20 nm) of the lithographical process but remains sufficient for the purposes of magnetic imaging.

The negative resist lithography technique offers a higher resolution as structures with a resolution down to 7 nm can be written into the resist. The material is then etched away either by wet etching or by ion beam etching. The problem with this approach is that the resist is difficult to remove after the etching. Commonly used oxygen plasma resist stripping is not possible in the case of thin permalloy structures, as they oxidize very easily. This fact, together with the need to use the shadowing technique, favors the positive lithography process which was used throughout this work.

We used the samples prepared by the methods described in this paper for the observation of the dynamics of magnetic vortices during a circulation switching by an MTXM^{10,11} and for the observation of various nucleation states¹⁷. This can be extended to more types of experiments requiring lithographically prepared structures on the membranes.

Disclosures

The authors have nothing to disclose.

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