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DELIVERABLE REPORT

WPN WP13 - JA3 Nano-engineering and pattern transfer methods

D13.2

High resolution lithography methods: performance and figure of merit

Due date

M24

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Prof. Ivan Maximov (LUND)

DELIVERABLE DETAILS

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D – 13.2	"High resolution lithography methods: performance and figure of merit".

DELIVERABLE DESCRIPTION

The deliverable aims at developing methods and materials (new resists) for ultrahigh-resolution and precision lithography by collaborative efforts of all four participants of the WP and including both "top-down" and "bottom-up" lithographic approaches (Talbot displacement lithography, thermal scanning probe lithography, He+ ion beams and block copolymers).

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AUTHORS

Andrea Cattoni (C2N-CNRS)

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PERSON RESPONSIBLE FOR THE DELIVERABLE

Ivan Maximov (LUND)

NATURE

☒ R – Report

☐ P - Prototype

☐ DEC - Websites, Patent filing, Press & media actions, Videos, etc

☐ O – Other

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☐ RE - Restricted to a group (Specify)

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FOR MORE INFO PLEASE CONTACT

Andrea Cattoni,
C2N-CNRS
10, boulevard Thomas Gobert,
91120 Palaiseau, France

email: andrea.cattoni@c2n.upsaclay.fr

Currently: Department of Physics,
Politecnico di Milano
Via Giuseppe Colombo, 81
20133 Milano

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CONTENTS

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Outline

The Joint Activity JA3 (WP13) is aiming at development of methods of high-resolution lithography and pattern transfer by collaborative efforts of all four participants of the WP. Among the goals of the JA3 one can mention extended applicability of the tools offered in TA and new functionalities in lithographic and patterning techniques to be available among the TA providers. Both "top-down" and "bottom-up" lithographic approaches that include high-resolution optical Talbot displacement lithography, thermal scanning probe lithography, He^+ ion beams and block copolymers are being used within the JA. Those lithographic methods are combined with high-resolution patterning, such as reactive ion etching and atomic layer etching. We focus our activity to provide relevant packages of nano-engineering methods combined with optimum protocols and know-how. The results generated within the current JA will be used in other work packages, such as WP14 and WP 15.

The **Deliverable D13.2** is related to the **Task 13.1**: «Methods of ultrahigh-resolution and precision lithography», which includes the following subtasks:

Subtask 13.1.1: «Seamless thermal scanning probe (t-SPL) and direct laser write (DLW) lithography» (EPFL, LUND, CSIC)

t-SPL is a new low-damage lithography method using heatable nanoprobes to directly pattern thermal sensitive materials. Modern t-SPL systems have an integrated DLW extension to speed up patterning of coarse features, that allow nano- and microfeatures to be written in a single step. t-SPL/DLW based processes will be optimized towards process integration with the new pattern transfer concepts (ALE and lift-off) developed in Task 13.2.

Subtask 13.1.2: «Directed self-assembly (DSA) of block-copolymers at 5 nm accuracy» (CSIC, LUND, EPFL, C2N-CNRS)

DSA is considered the main alternative for <10 nm lithography at high throughput. We will develop processes based on DSA, using high-chi block-copolymers (BCP) (with LUND) to obtain the highest resolution, and to improve control over the self-assembly process. We will combine DSA with the other methods, t-SPL (EPFL), DTL (LUND) and He-ion (C2N-CNRS). The processes will be used to define high accuracy masks compatible with many substrates and etching processes (Task 13.2).

Subtask 13.1.3: «He-ion lithography with ultra-high resolution» (C2N-CNRS, LUND)

Atomically defined He+ ion source can produce a beam with a spot size ≈ 0.5 nm. This sharp beam gives no proximity effects, allowing He+ ion lithography to achieve <10 nm resolution. As the interaction of He+ (or Ne+) ions with matter is very strong, one can use direct milling of mask layers and successive pattern transfer. Other resist materials with strong etching resilience and high resolution e.g., sol-gel derived metal oxides can be used with ALE-based resist processing (Task 13.2).

Subtask 13.1.4: «Displacement Talbot Lithography (DTL)» (LUND, C2N-CNRS, CSIC)

DTL is a novel optical interference technique, suitable for mass-fabrication of periodic high-resolution structures. However, the requirement of an anti-reflective layer (BARC) complicates further process steps, e.g., lift-off. This sub-task will focus on development of DTL to provide a set of stable processes, that include both the optimum exposure conditions and methods of patterning. We will also study the HSQ-based resists or metal oxide sol-gel derived materials to avoid the need of a subtractive pattern transfer.

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Subtask 13.1.1: Seamless thermal scanning probe (t-SPL) and direct laser write (DLW) lithography (EPFL, LUND, CSIC)

T-SPL is an emerging technique for high-resolution grayscale nanolithography. Sub-10 nm lateral and sub-2 nm vertical resolution is achievable for the patterning of grayscale nanostructures by scanning probes having a tip with a diameter of 20 nm. We used T-SPL for sinusoidal nanostructuring of thin film oxide as shown in Figure 1.

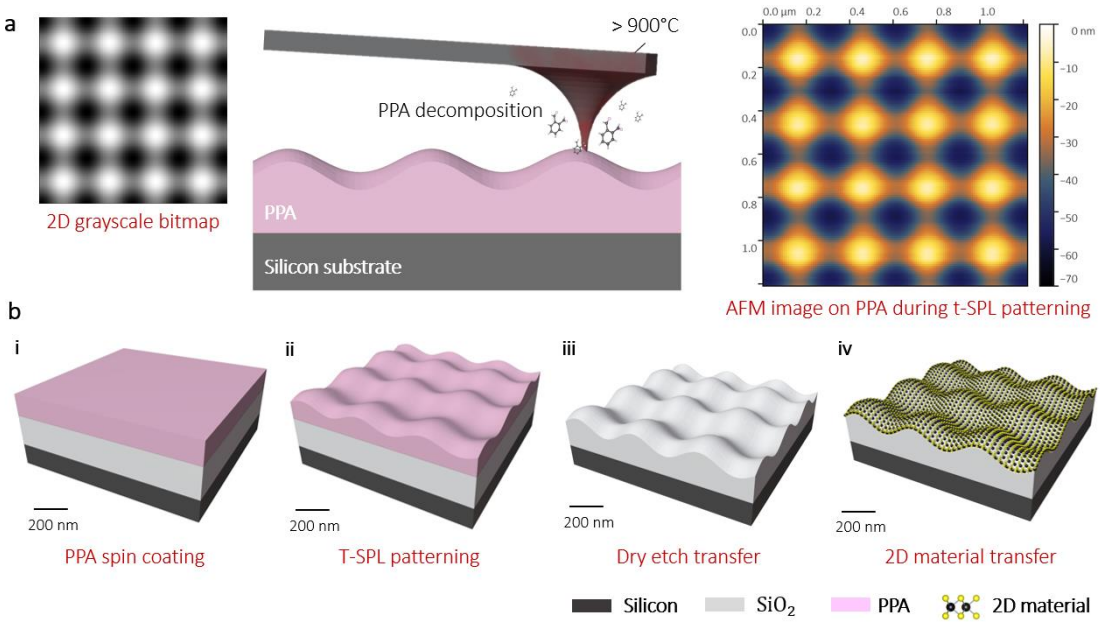

Figure 1: (a) Working principle of T-SPL-based nanopatterning on the thermosensitive resist. The heated nanotip locally removes materials depending on the depths defined for each pixel on a 2D grayscale bitmap (left). Since the thermal resist PPA directly sublimates upon exposure to heat without the need of any development step, the measurement of the cantilever deflection during T-SPL patterning provides simultaneous in-situ AFM imaging of the created patterns (right). (b) Fabrication process flow for 2D material straining device. Scheme of the processes after (i) PPA spin coating, (ii) T-SPL patterning, (iii) dry etch transfer, and (iv) 2D materials transfer on top of the nanostructures SiO_2.

However, the speed of grayscale tip patterning is limited to approximately 6000 μm^2/min. On the other hand, the patterning speed of DLW is almost 20 times faster. Therefore, the markerless overlay of T-SPL and DLW with sub-1 µm accuracy decreases lithography time. Figure 2 shows the schematic of this combination. We benefited from this approach, and the combination of tip patterning with laser sublimation is used to create corrugated nanostructures, which strain 2D semiconductor materials transferred on top of them (Figure 3). Later, markerless overlay ability of T-SPL is used for next lithography steps. First Au metal deposition and lift-off processes are done to fabricate source and drain electrodes. Finally, relatively larger microscale contact path are created by DLW, and Cr/Au metal deposition followed by lift-off process are done to fabricate 2D material-based field-effect transistor (FET).

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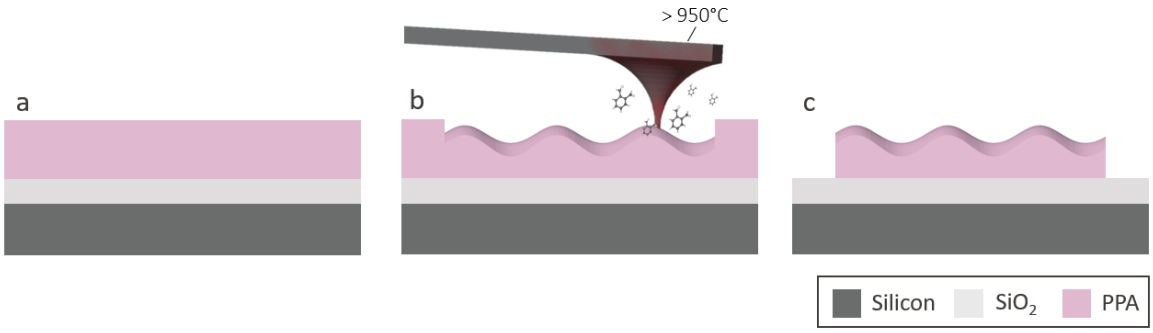

Figure 2: Schematic of process flow after (a)thermal resist, PPA, spin coating on silicon/SiO_2 substrate, (b) tip patterning with scanning probe, (c) direct laser sublimation.

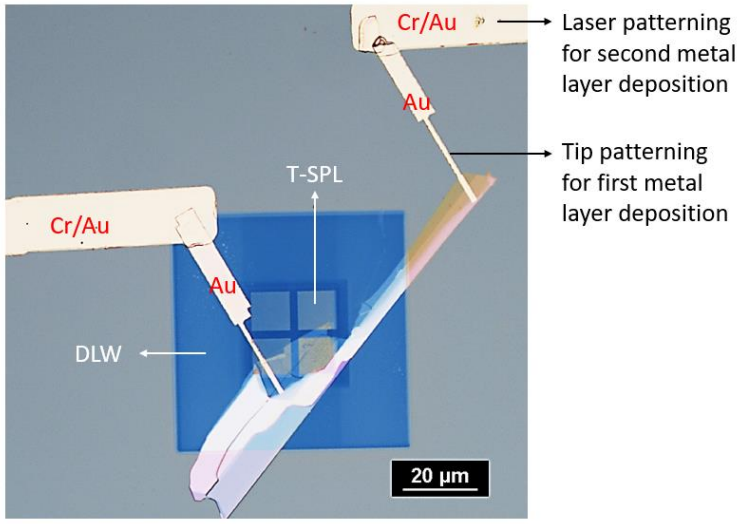

Figure 3: Optical microscope image of 2D material-based FET fabricated by combination of T-SPL and DLW.

Subtask 13.1.2: Directed self-assembly (DSA) of block-copolymers at 5 nm accuracy (CSIC, LUND, EPFL, C2N-CNRS)

Subtask 13.1.2 is linked to milestone MS18: DSA of high-chi BCP in combination with high resolution lithography. In September 2022 we submitted the request to delay the milestone, which reflected the situation and plans for next months:

Although there has been some activity regarding DSA by some of the partners, the main purpose of the deliverable (to combine DSA with a high-resolution method) has not been addressed yet, partially because there has been a lack of resources (personnel) at CSIC, but mainly because the specific focus of the task was not properly defined in the workplan document. In consequence, MS18 has been delayed to Month 32. The new plan to fulfill MS18 is the following: a combination of thermal-SPL with DSA will allow to facilitate the realization of dense arrays of contacted electrostatic gates for semiconductor qubits. It requires the definition of the arrays with sub-10 nm

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accuracy (DSA) and the opening of contacts on top of the electrodes with sub-5 nm accuracy (t-SPL).

During the JA3 consortium meeting at EPFL (January 25-26, 2023) the plan for achieving Milestone MS18 at Month 38 was updated, according to the results obtained up to that moment.

EPFL has been optimizing the definition of guiding patterns for DSA using t-SPL. It is found not obvious to properly define a circle with t-SPL on PPA that will fulfill the requirements for pattern transfer into silicon oxide. However, a significant progress has been reported attending to optimizing the t-SPL conditions and also, in pattern transfer. This route will continue the next months.

In parallel, two additional routes will be explored shortly:

1) Definition of specific pattern transfer stacks by CSIC that will facilitate the definition of the guiding patterns after t-SPL
2) Use of templates formed by the new resists developed by C2N-CNRS as guiding patterns for contact shrink.

Subtask 13.1.3: He-ion lithography with ultra-high resolution (C2N-CNRS, LUND)

In the previous NFFA project (WP7, Subtask 7.1.1: «Novel resist materials and processing») we have developed a hybrid organic-inorganic alumina-based a negative-resist for e-beam and He-FIB lithography with resolution and low Line Edge Roughness comparable to commercial HSQ resist but with high selectivity for fluorine-based dry etching of silicon.[1] The resist was synthesized using a sol-gel method and the main drawback was the relatively low stability of the solution as aluminum-tri-sec-butoxide (very reactive to oxygen) and 2-methoxyethanol solvent (highly hygroscopic) were used.

In the framework of the NEP project, we decided to explore a new chemistry for the synthesis of the resist with two main goals:

i) Synthetize a high resolution, inorganic positive-tone resist that could complement negative-tone resists previously developed (Alumina-based)[1] or commercially available (Hydrogen Silsesquioxane, Silica-based). Today, inorganic positive-tone resists are not available. The developed resist may serve for the direct fabrication of dielectric masks for selective area growth of nanowires, in connection with the subtask 13.3.1.
ii) Achieve superior stability of the resist solution (a drawback of the Alumina-based resist developed previously) by avoiding precursors and solvents sensitive to moisture.

Due to the fact that the He-FIB system at C2N has been out of order for several months (we requested an extension of the MS19: «High resolution (10 nm) process of He-FIB patterning using direct hard mask and/or resist-based patterning of sol-gel derived metal oxides»), the optimization of the resists is being performed by electron beam lithography. However, high-resolution tests with the new resists will also be made with the He-FIB as soon as it is available again. We also plan to test these resists with displacement Talbot lithography (DTL) in Lund.

[1] A. Cattoni et al., Microelectronic Engineering 193, 18 (2018) – 28 citation already

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First off, we explored the use of a relatively new chemistry method called Polymer Assisted Deposition (PAD),[2] which uses water-based mixture of metal precursor - typically cheap nitrates salts - and a soluble polymer - branched polyethylenimine (PEI) possibly assisted by Ethylenediaminetetraacetic acid (EDTA) - that actively binds the metal cations thus creating solutions that are stable for months and allow for an even distribution of the metals or the formation of undesired phases during calcination and decomposes without traces. With these method in the last 10 years were demonstrated the synthesis of epitaxially grown complex multicationic metal-oxide films Ge, Nitrides, Carbides. The key step for the stability of the solution is the ultra-filtration step performed after the synthesis to remove unbonded metal cations and PEI fragments with low molecular weight responsible for the instability of the solutions (Figure 4a). We have successfully synthetized Al-based hybrid resist using different PEI molecular weights (1.800, 10.000, 25.000, 60.000 Dalton and precursors ($Cu(NO_3)_2$*$3H_2O$, $Al(NO_3)_3$ hydrate, $Al(NO_3)_3$ *$9H_2O$). We used electron beam lithography for the first tests and the He-FIB system at C2N has been out of order for more than 9 months (a part from the ultimate resolution, the two techniques can be interchanged during the resist optimization). So far, we were able to pattern features as small as 80 nm (Figure 4b, top) in the hybrid positive-tone resist (Al-based and Cu-based) developed in acetic acid and using relatively low doses comparable to PMMA positive-tone resist (1200-2000 μm/cm^2). Calcination at 500 °C to recover a totally inorganic Al_2O_3 nanostructured film tends to destroy the nanostructuring and for the moment we were able to maintain it using a post-developing UV treatment before the calcination process (Figure 4b, bottom). We are currently optimizing the process in order to increase the resolution. That being said, this is the first ever demonstration of the direct patterning of films synthetized by PAD and the outcome of this work can potentially go beyond the planned scope of this WP as it opens the possibility to directly pattern complex multicationic metal-oxide films Ge, Nitrides, Carbides.

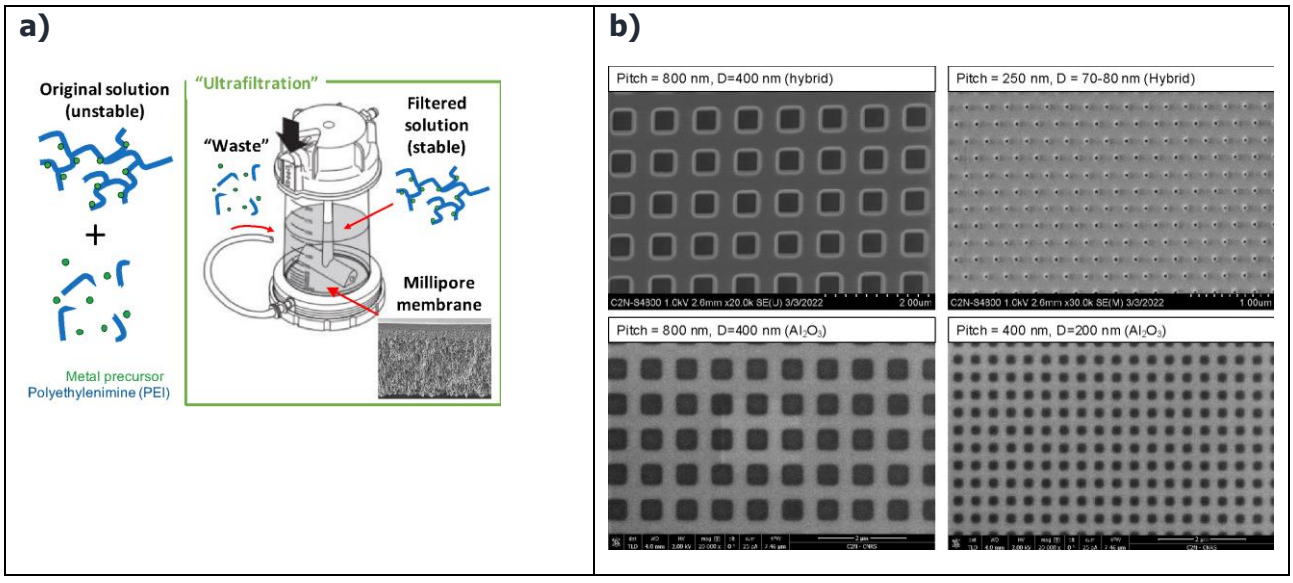

Figure 4: **a)** Schematics of the Polymer Assisted Deposition (PAD) method: unwanted anions or cations not bonded to the polymer and polymer fragments are filtered through an Amicon ultrafiltration unit. **b)** SEM images of the positive-tone hybrid resist (Al-based) after development and inorganic Al2O3 patterned films (bottom) after UV light curing and post-backing annealing.

[2] US6589457 (2003), Nature Materials 3, 529 (2004), US7365118 (2008), Angew. Chem. 122, 1826 (2010)

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More recently, we explored the use of a more conventional sol-gel chemistry in order to improve the resolution and the simplicity of the process. We used a combination of TetraethylOrthosilicate (TEOS) and TriethoxyMethylsilane precursors in ethanol, adjusting the PH with HCl and using Cetrimonium bromide (CTAB) as surfactant to improve the spin-coating of the films.

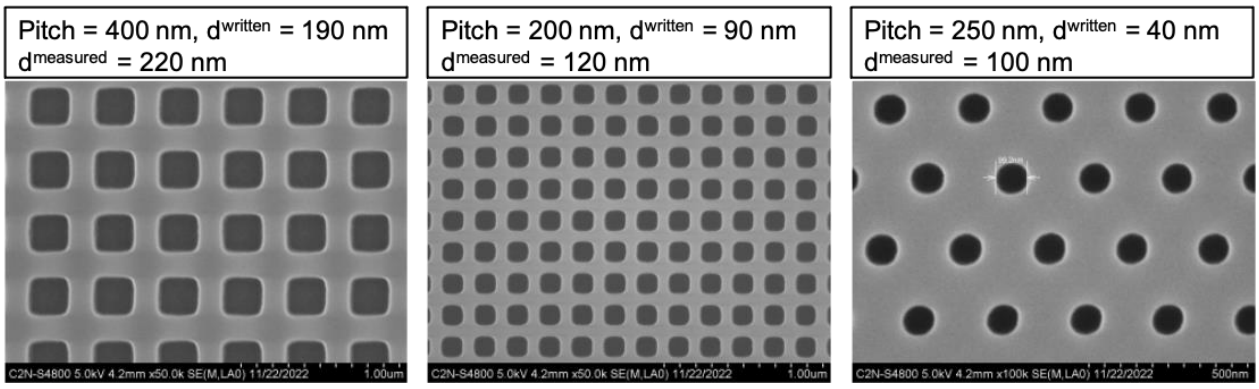
Table 1. Typical chemical composition of the tested resist.

ETHANOL	5.5 G
HCl 0.1M	0.5 g
TetraEthylOrthoSilicate	0.5 g
TriethoxyMethylsilane	0.5 g
Surfactant	Cetrimonium bromide

By adjusting the molar ratio of the two precursors we were able to obtain a resist sensitive to the electron beam exposure that behaves as a positive-tone resist when developed in HF 1% (Figure 5a). Once developed the remaining resist is an inorganic SiO_2 without need of calcination. A relatively high resolution (Figure 5b) was achieved for 100 nm-thick films with isolated features of 38 nm (period 100 nm). Although the electron beam doses used are relatively important (anyway comparable with the doses used for commercially available HSQ negative-tone resist), we expect this can be further reduced by optimizing the process (pre-bake, HF concentration (%), developing time). We also expect to have higher sensitivity (and resolution) when used in combination with He-FIB (which will be soon operational again).

Finally, we have preliminary results (not shown) that suggest that this resist can be made sensitive to UV-light (365 nm with high doses) and at 193 nm with lower doses (see Subtask 13.1.4) when TriethoxyMethylsilane is replaced with TriETHOXYphenylsilane. We are currently working in collaboration with the partner Lund University in order to combine this resist technology with Talbot Lithography at 193 nm.

a)



b)

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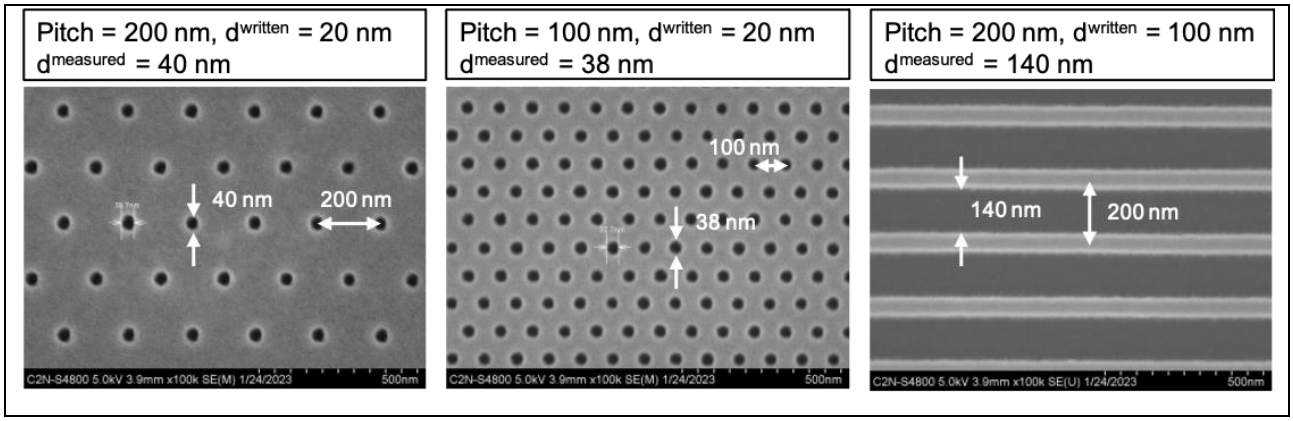

Figure 5: **a)** SEM images of 220 nm-thick SiO2 resist exposed by EBL at 100kV using (dose = 15000 µC/cm2) and developed 15 sec in HF1%. **b)** SEM images of 100 nm-thick SiO2 resist exposed by EBL at 100kV using (dose = 9000 µC/cm2) and developed 15 sec in HF1%.

Subtask 13.1.4: Displacement Talbot Lithography (DTL) (LUND, C2N-CNRS, CSIC)

Displacement Talbot lithography (DTL) is a relatively novel optical lithographic technique that allows formation of regular arrays of nanostructures with sub-100 nm resolution. The subtask deals with the development of a modified DTL that allows patterning on 4-inch wafers with feature sizes below 90 nm after etching or lift-off. The formal milestone MS7 due on month 12 has been successfully fulfilled in February 2022 after acceptance of the technical report.

The approach we explored to optimise the DTL process was based on a post-processing resist step that allowed us to reduce the feature sizes to about 70 nm in etched Si or in metal lift-off dots. The motivation behind this approach was a limited resolution of the 193 nm DTL tool, that does not allow easy lithographic patterning below 100 nm. Control of the positive resist openings was realised by a dedicated baking step that result of hole shrinkage using a "mix-bake" (swelling) process. Change of baking temperature gives sufficient control to adjust the feature sizes in the resist. We found that both lift-off and reactive ion etching in F-chemistry can be used in combination with DTL to make structures well below 90 nm. The developed process can be used for structures around 70 nm in diameter, while smaller features can be produced with some loss of reproducibility.

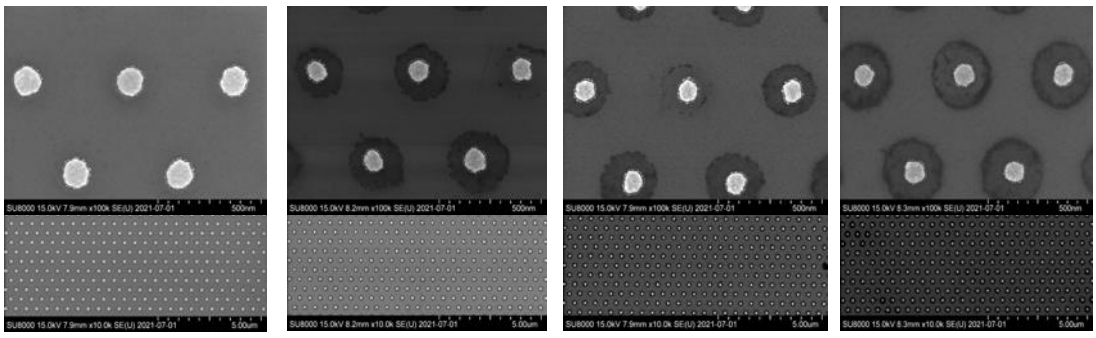

Figure 6. SEM top view of 30 nm thick Au dots produced by an exposure in DTL, thermal evaporation and lift-off. To control the size of the lift-off dots, different "mix-bake" temperatures

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were used (from left to right): no baking,125 nm diameter, 90° C, 90nm, 110° C, ≈85 nm and 130° C, 90 nm.

This subtask involves a cooperation between LUND and C2N-CNRS where we planned to try novel resists with DTL. However, in spring 2022, the DTL tool in LUND broke down and could not be quickly repaired due to problems with delivery of spare parts. It resulted in some delays of our experiments, that resumed at the end of 2022.

The experiments to characterise the new silica resists developed at C2N-CNRS with DTL at 193 nm continued in December 2022. We used two versions of the resist with different concentration of organic precursors, see the Table 2 below. The resists were spin-coated to obtain a film thickness of about 200 nm on 4" Si wafers and baked at 130, 170 or 210 °C. No bottom anti-reflection coating (BARC) was used in our experiments, since one of attractive applications of the resists would be selective epitaxial growth of III-V nanowires and the presence of the BARC layer would complicate processing steps.

Table 2. Chemical composition of the tested resists.

	SiO2 with 30% TriEthoxyPhenylSilane	SiO2 with 20% TriEthoxyPhenylSilane
Ethanol	5 g	5 g
HCl 0.1M	1 g	1 g
TetraEthylOrthoSilicate	1.15 g	1.0 g
TriEthoxyPhenylSilane	0.35 g	0.5 g
Surfactant	Cetrimonium bromide	Cetrimonium bromide

The DTL exposure experiments were performed using a mask with feature sizes of 200 nm and the pitch of 500 nm. In order to find the optimum exposure doses, the resists were exposed with doses from 2 to 50 mJ/cm^2. After exposure and development in 1% HF water solution, optical and scanning electron microscopy revealed the expected patterns at some areas with a thinner resist only, Figure 7. This observation may indicate that an optimum thickness of the resists is needed for a correct exposure due to an interference effect in the resist film.

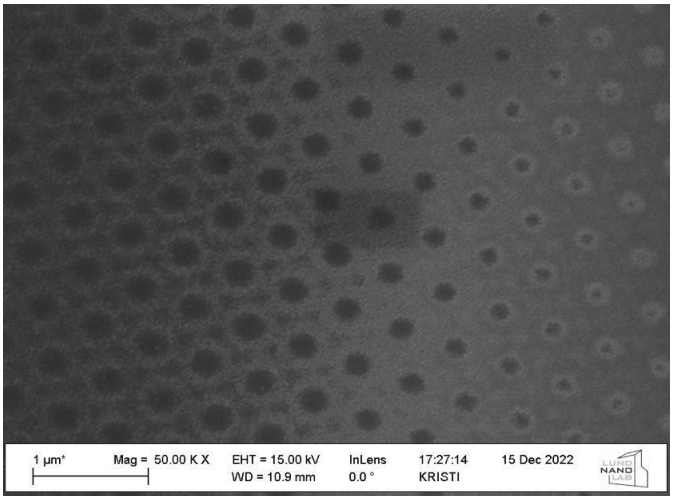

Figure 7. Top SEM image of SiO_2 resist with 30% precursor after DTL exposure with 50 mJ/cm^2 and development in 1% HF. Different hole sizes are obviously due to variation of the resist film thickness.

The experiments with a new batch of the silica resists are underway at the moment.

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Performance and figure of merit of lithographic methods

Subtask 13.1.1

Applications to device/structure fabrication: Strain has been induced in 2D materials using a variety of techniques, including the bulk stretching/bending of 2D materials on flexible polymer substrates,[3] straining on pre-patterned structures,[4,5] the use of probes impinging directly on the 2D material,[6] and the pressure-induced bulging of suspended 2D membranes.[7] Integrated macroscale and microscale actuators produce changeable strain in 2D materials. However, their bulky structures are not applicable for compact and scalable nanoscale transistors. Here, we report grayscale nanostructuring of thin film dielectrics by combining t-SPL and DLW to induce uniform strain in 2D materials. In comparison to nanopillar array substrates fabricated by E-beam lithography,[8] t-SPL nanopatterning results in the conformal attachment of 2D materials to the dielectric beneath by minimizing the wrinkling and suspension of 2D materials, hence providing us with a mechanically stable device. By adjusting the pitch and depth of nanopatterns, grayscale nanolithography might also produce varied strain rates and directions within the same 2D material-based nanodevice.

Subtask 13.1.3

Resolution: the only commercially available SiO2-based resist, is the **negative**-tone Hydrogen Silsesquioxane (HSQ) resist. Sub 10 nm half-pitch resolution was demonstrated using e-beam lithography and specific NaCl-NaOH-based development and doses exceeding 2500 μm/cm^2, although we can question how these patterning can be used for patten transfer (a more convincing 14-nm-pitch structures were fully resolved).[9] Similar results were obtained using He-FIB with lower doses (1/30) given the higher sensitivity of the HSQ resist for He-ions with low proximity effects, degraded Line Edge Roughness (LER) but improved resolution.[10] In the previous NFFA project, we developed an alumina-based negative-tone resist for e-beam and He-FIB with fully resolved 10 nm half-pitch resolution (He-FIB) with excellent Line Edge Roughness (LER) and demonstrating a pattern transfer in Silicon using fluorine-based dry etching.[1] In this WP we developed a **positive**-tone SiO2-based resist that doesn't have a commercially available equivalent (as far as we know). Alumina-based **positive**-tone resist were demonstrated in literature, but with relatively poor resolutions: 200 nm diameter dots with a pitch of 1 μm, using e-beam lithography at 20 kV and doses of 50-1500 μC/cm^2).[11] Using our newly developed SiO2-based resist, we demonstrated sub-40 nm isolated features (38 nm diameter dots with a pitch of 100 nm) using e-beam lithography at 100 kV and doses comparable to the ones used for HSQ resist. We expect to be able to increase the resist resolution (even for e-beam lithography) by optimizing the pre-baking and the development process.

[3] Z. Li et al., Nature Communications 11, 1-8 (2020)
[4] R. Maiti et al., Nature Photonics 14, 578-584 (2020)
[5] T. Liu et al., Nature Nanotechnology 14, 223-226 (2019)
[6] X. Liu et al., Nano Letters 20, 8250-8257 (2020)
[7] D. Lloyd et al., Nano Letters 16, 5836-5841 (2016)
[8] J. Chaste et al., ACS Nano 12, 3235-3242 (2018)
[9] J. K. W. Yang et al., J. Vac. Sci. Technol. B 27, 2622 (2009)
[10] V. Sidorkin et al., J. Vac. Sci. Technol. B 27, L18 (2009)
[11] G. Grenci et al., Applied Materials Today 1, 13–19 (2015)

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Throughput: High resolution HSQ resist pattering typically requires doses exceeding 4000 μC/cm^2 when used in combination with 100 KV e-beam. Even higher doses (tens of mC/cm^2) are required for other high-resolution inorganic negative-tone resists based for example on tungsten-trioxide[12] or the recently developed one based on cadmium-(II) ethylxanthate complexed with pyridine,[13] restricting their applicability to relatively small surface areas. Typical doses used for our SiO2-based resist range from 3000 to 15000 μC/cm^2. Although these doses are still relatively high, we expect to be able to increase the resist sensitivity by playing with the chemistry of the resist (molar ratio of the two precursors), the processing (pre-baking, %development). Certainly, given our expertise in e-beam/He-FIB patterning using AlOx resist and results from literature,[4] we expect to increase the sensitivity of the resist of at least one order of magnitude when used in combination with He-FIB.

Applications to device/structure fabrication: Negative-tone HSQ resist has been already used for the direct fabrication of SiO_2 structures that does not require any patten transfer, for example for the direct fabrication of high-resolution of SiO_2/Si master stamp for Nanoimprint lithography[14],[15] or for the direct fabrication of two-dimensional photonic crystals.[16] The availability of a SiO_2 positive-tone resist open the possibility to extend this technology to complementary patterns without the need to expose large surface areas. Our current application goal is to develop SiO_2-based masks for selective area growth of nanowires. These SiO_2 masks (nanohole diameters: 30-80 nm, periods: 100-500 nm) are typically obtain by dry etching of a thin (20-40 nm) PECVD SiO_2 film using a positive-tone PMMA resist mask exposed by e-beam lithography (C2N) or Talbot Lithography (LUND). In this process, the dry etching step needs to be carefully controlled in order to avoid the direct contact of the CHF_3/SF_6 gases with the Si interface (where the nanowires will be epitaxially grown). For this reason, the SiO_2 mask is partially etched, and the last few nm of SiO_2 (inside the nanoholes) isotropic wet-etched by 1% HF solution (thus increasing also the nanohole diameter). We expect that using our new SiO2-based positive tone resist this will whole process will be greatly simplified.

Subtask 13.1.4

Resolution: Deep UV (193 nm) Displacement Talbot Lithography (DTL) with a phase-shift mask can produce an array of holes down to approximately 120 nm using a standard combination of DUV resist and bottom anti-reflective coating (BARC) layers. Our previous work[17] has demonstrated that replacement of the BARC layer with polymethylglutarimide (PMGI) may push the resolution of DTL slighly below 100 nm due to a simplified, more reliable process, that does not need an oxygen plasma step to etch the BARC layer. Instead, a negative slope in the resist is created by a partial dissolution of PMGI layer during development. The double-layer resist may also be used for pattern transfer into substrate using reactive ion etching. Further steps to improve the resolution of DTL included a post-development step of resist shrinkage (a "mix-bake" process) that decrease the size of the holes thus improving the resolution even further (Milestone MS7, *Process of Displacement Talbot Lithography with lift-off or etch pattern transfer with sub-90 nm features on 4-inch wafer*). The size decrease depends on the "mix-bake" temperature, the higher baking temperature leads to smaller features and may result in the hole openings as small as 70 nm. The 70 nm holes in the resists, that can be served as the figure of merit, were reliably transferred to Au lift-off patterns and etched SiO_2 features. The overall performance of the developed "mix-bake" process can be

[12] F. Carcenac et al., J. Vac. Sci. Technol. B 14, 4283 (1996)
[13] M. S. M. Saifullah et al., Nano Lett. 22, 7432−7440 (2022)
[14] A. Cattoni et al., Microelectronic Engineering 87, 1015–1018 (2010)
[15] M. Graczyk et al., Microelectronic Engineering 190, 73–78 (2018)
[16] B. Hamza et al., Microelectronic Engineering 91, 70-74 (2012)
[17] V. Gomez, et al., Nanotechnology 31, 1–9 (2020)

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characterised as sufficiently reliable to 70 nm, although another DTL mask with feature sizes of 100 nm and higher "mix-bake" temperatures produced holes down to 50 nm, although with some loss of reproducibility.

Throughput: The DTL is a parallel lithographic technique providing very high throughput: a typical wafer exposure takes several minutes only. However, its main restriction is a limited number of patterns, the Talbot effect, the basis of the DTL, yields a regular periodic pattern that can be used for a low-cost and fast nanofabrication. The recently developed "mix-bake" process does not affect the whole throughput of the DTL, as additional steps of spin-coating of the shrink resist, its baking and removal are not very time-consuming. Compared to t-SPL, the He-ion beam lithography or BCP, the DTL is very fast and can be used on large areas, e.g., 2 or 4" wafers.

Applications to device/structure fabrication: Applications of DTL are focused on formation of regular arrays of Au dots for epitaxial growth of III-V nanowires, where the "mix-bake" process can be used for producing Au seeds with size well below 100 nm. Another application is test exposures of positive-tone novel silica-based resists developed by C2N-CNRS, this is ongoing work at the moment.

Dissemination activities (LUND, C2N-CNRS, EPFL, CSIC)

- Collaboration in the organization of the NEP session at e-MRS symposium (Workshop on advanced nanolithography and nanopatterning) May 30-31, 2022. (LUND, CSIC)
- Webinars for the Nanoscience and Nanotechnology master at the autonomous University of Barcelona (February-April, 2023). (Lund, EPFL, C2N-CNRS, CSIC)
- Presentations at MNE 2022. September 2022. (CSIC)

Article submitted to Micro and Nano Engineering Journal. CSIC. (Jordi Llobet, David Bricio-Blazquez, Jordi Antoja-Lleonart, Alberto del Moral, Xavier Borrisé, J Francesc Pérez-Murano, Joan Bausells). Mix and match fabrication of silicon nanowire devices with tunable doping profile. Submitted December 2022.

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