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Starting from the beginning of 2019 the European consortium NARCISO has been forged to work on the study, development and exploitation of the **NAtuRal instability of semiConductors thIn SOlid films for sensing and photonic applications.** In about two years and a half and in spite of the long break imposed by the Covid-19 disease, we made several progresses on all the activities planned in the research program as described below.

The network is composed by 5 academic groups and 1 non-academic partner, each of them leading their own research program on different aspects of NARCISO. They were chosen to cover the needs of growth and fabrication, theoretical modeling, characterization, applicability and industrial scaling of the fabricated structures.
Members of the consortium:

-1 *CONSIGLIO NAZIONALE DELLE RICERCHE IT* (lead. Dr. Monica Bollani, monica.bollani@ifn.cnr.it, www.cnr.it). Activity: device fabrication (e-beam lithography, electric contact deposition), III-V integration on SiGe micro- and nano-structures.
-2 *UNIVERSITE D'AIX MARSEILLE FR* (lead. Prof. Marco Abbarchi, marco.abbarchi@im2np.fr, http://www.im2np.fr/doct/abbarchi.html). Activity: solid state dewetting of SiGe, sol-gel dip-coating and nano-imprint lithography.
-3 *TECHNISCHE UNIVERSITAET DRESDEN DE* (lead. Prof. Axel Voigt, axel.voigt@tu-dresden.de, http://www.tu-dresden.de/). Activity: phase field simulations of solid state dewetting and particle diffusion.
-4 *UNIVERSITE DE LAUSANNE CH* (lead Prof. Pietro De Anna, www.pietrodeanna.org). Activity: microfluidic, filtering and sensing with nano-imprinted micro- and nano-structures.
-5 *UNIVERSITA DEGLI STUDI DI FIRENZE* IT (lead. Prof. Francesca Intonti, intonti@lens.unifi.it, http://www.lens.unifi.it/nanostructures/index.php). Activity: optical spectroscopy (near- and far-field) of dielectric micro- and nano-structures.
-6 *OBDUCAT TECHNOLOGIES AB SE* (lead Dr. Kristian Thulin www.Obducat.com). Activity: scale up of nano-imprint lithography metal-oxides-based micro- and nano-structures.

The research that carried-on in NARCISO goes from fundamental aspects of semiconductor self-assembly (e.g. simulation of solid state dewetting), to industrial applications of the micro- and nano-architectures realized by nano-imprint lithography. More precisely, we are studying how spontaneous formation of complex patterns (e.g. via solid state dewetting) can lead to innovative functionalities in light and matter management.
To this end, we exploit a spontaneous phenomenon occurring in thin films (10-1000 nm thick) of SiGe alloys on silicon: exposing them to high-temperature annealing they special coarsening dynamic leading to spinodal-like structures (e.g. like those observed in binary mixtures undergoing phase separation, such as water and oil, as reported in Figure 1). Despite the apparent disorder in the arrangement of the islands or connected structures, the relative position of the objects in not random: the pattern formation dynamics results in hidden correlations, providing to the structures special properties that NARCISO aims at exploring and exploiting.
Patterns exhibiting correlated disorder are ubiquitous in nature and are typical of many phenomena ruled by far from-equilibrium processes. Prominent examples are morphogenesis in biological systems, thin-layer wrinkling, and the afore-mentioned phase-separation, that is for instance

commonly used for the purification of metals. These patterns are called “spinodal material” and a special property of these spinodal-like patterns obtained via solid state dewetting is “disordered hyperuniformity”. Disordered hyperuniform (dHU) materials, indeed, are an emerging class of amorphous systems at the edge between ordered crystals (as they do not display long-range density fluctuations) and liquids (as they do not have Bragg peaks in diffraction). This idea was theoretically introduced about 20 years ago by Torquato and Stillinger and its comprehension and exploitation in practical applications are still at their beginning. In the last years, dHU features have been highlighted in a variety of natural phenomena spanning from classical to quantum systems, from out-of-equilibrium at room temperature to in equilibrium including biological frameworks. Beyond fundamental understanding of natural phenomena, the interest in this peculiar class of correlated disorder springs from the potential applications it may have in anomalous light and matter transport.
Generally speaking, disordered materials are less prone to fabrication imperfections with respect to the ordered counterparts. As such, if obtained via self-assembly over large scales, they may be much less-expensive. In optics (e.g., for antireflection coatings), another advantage of disordered structures with respect to ordered arrays of objects (e.g., such as moth-eye structures for antireflection coatings) is the lack of diffraction from which neat colorization and angular dependencies can spring. Artificial structures, such as waveguide polarizers, lasers, metalenses, photonic band gaps for light propagation, plasmonic metasurfaces, and efficient light trapping in thin Si films are recent examples of dHU systems, typically obtained via top-down fabrication methods or hybrid self-assembly and top-down.

Thus, first of all, the work of NARCISO focused on the comprehension and engineering of spinodal dewetting of SiGe alloys showing its dHU character. From a theoretical point of view, after the work carried out during the first project’s years leading to the development of a theoretical (phase field) modeling reproducing 2D and 3D dewetting, two major advances have been achieved:
i) the application of the simulative approach to 3D, large patches (having size comparable to experiments) considering the faceting of the crystalline SiGe structures.
ii) the implementation of a machine learning algorithm, which, trained by previously rune simulations based on well-established methods, can predict the main features of spinodal dewetting in prototypical 2D systems.

Another important part of the work carried on is the exploitation of the complex architectures obtained in SiGe as hard-masters for sol-gel dip-coting and nano-imprint lithography to form hard-dielectric structures on glass and semiconductors (Figure 2). The advantage of our fabrication protocol is manifold. Most of the techniques (e.g. used for photonic applications at visible and near-infrared frequencies) exploit top-down methods (e.g., e-beam lithography and reactive ion etching). As such, they are hardly scalable, limiting the range of applicability of dHU materials. Solid-state dewetting is a valid alternative to build dHU architectures as it provides hard materials with a large dielectric constant with size tunability spanning more than 3 orders of magnitude. However, it is limited to few materials and requires epitaxial growth and expensive silicon on insulator wafers that, for instance, are not transparent at visible frequencies and are provided in a limited set of specifications. In addition to this, SiGe compounds feature strong absorption losses at near-UV frequencies, rendering them less adapted to applications at a short wavelength. All these issues limit the range of its exploitability.

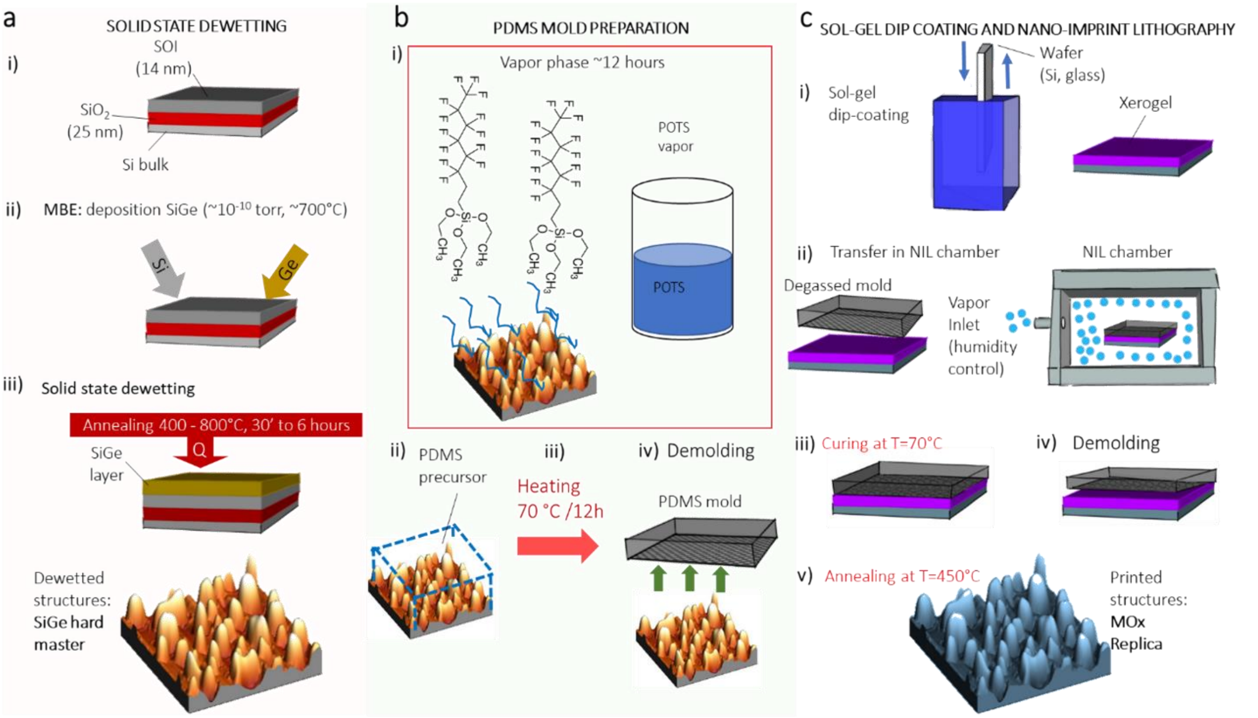

Figure 2. (a) Elaboration of hard masters by solid-state dewetting. (b) Preparation of the PDMS mold. (c) Dip-coating of a metal oxide sol−gel film and transfer of dewetted structures by nanoimprint lithography on the sol−gel film.

We further extended the use of SiGe dewetting: our method enjoys all the flexibility of sol−gel chemistry, allowing us to print a plethora of metal oxides, such as silica and titania, having a low and high refractive index, respectively, and sharing reduced absorption at near-UV frequencies. Nonetheless, it is possible to tune at will the refractive index (e.g., by mixing them or by changing their porosity) and introduce nanoparticles or organic moieties. It is possible to print on large surfaces up to 8 in., on metals, plastic, semiconductors, and glasses. Thus, provided an appropriate master with ad hoc morphologies, it is possible to reproduce it hundreds of times by framing several polymeric master molds. Note that, provided the sol−gel solutions and the mold, the overall process of soft-NIL can be performed in less than 30 min in a research laboratory, independently from the sample size. All these features render our solution for dHU metasurfaces scalable to large surfaces and appealing for realistic applications.

We also exploited our spinodal-like patterns for micro-fluidic. From a fundamental standpoint, the use of our micro-architectures in micro-fluidic channels allowed to study and understand the role of dead-end pores in particle transport. The ubiquity of complex patterns in porous systems (e.g. in soil and geologic reservoirs or in filtration devices), the fundamental properties in colloidal particle transport understood in NARCISO will impact a broad range of other colloidal-like particles, as bacteria and viruses. Furthermore, in view of an increasing need for fast and cheap bio-medical diagnostic tools, this fundamental knowledge and the device we are developing in NARCISO may be efficiently exploited in these fields.

From a more applied point of view, a manual 8-inch nano-imprint lithography machine (from OTAB) was modified and equipped with an imprinting chamber for environmental controlled atmosphere (Figure 3) to test Soft-NIL concept over large areas.

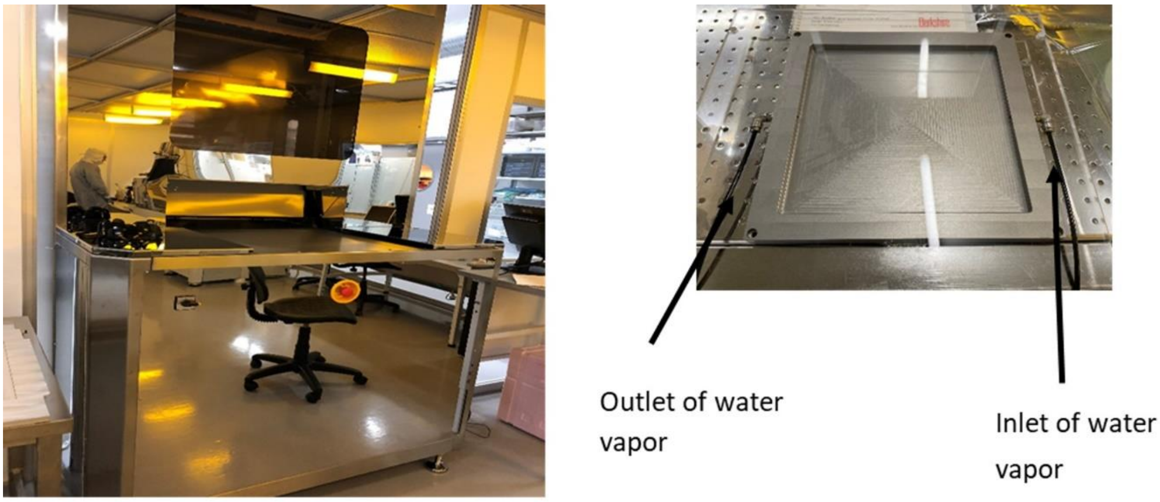

Figure 1: OTAB's NIL machine for 8 inches wafer. Left panel: full view. Right panel: detail of the NIL chamber with inlet and outlet of vapor

The process optimization was performed by imprinting porous SiO_2 and TiO_2 sol-gel precursors with mould bearing ~µm sized pillars onto 2-inch silicon substrates. Although there are only preliminary results, they already show the possibility to print high-quality patterns over relatively large scales (see Figure 4).

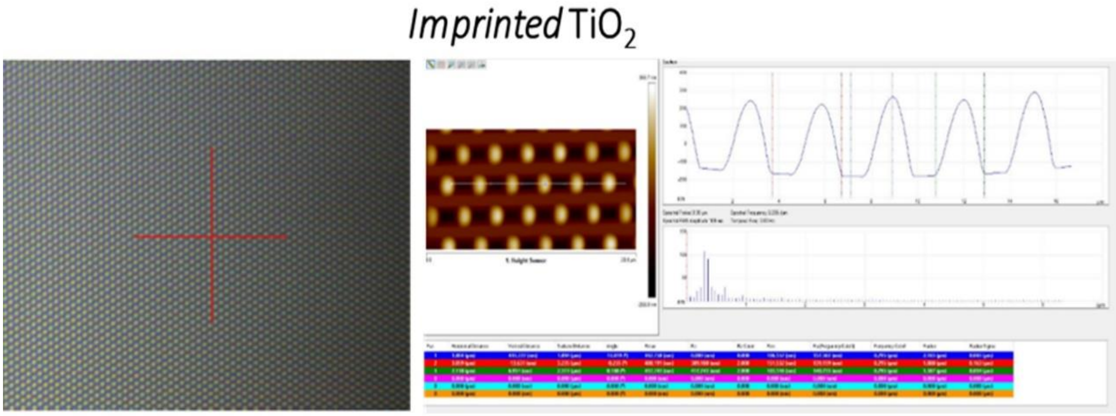

Fig. 4. Top panels: optical image (left) and AFM (central and right) showing porous TiO_2 pillars imprinted on a 2 inches Si wafer.

Finally, a relevant aspect of NARCISO's impact is the constitution of the start-up SOLNIL. The society has been funded in July 2020 and is now at work in securing a set of products, making contacts with potential customers and collaborators (bot academics and industrial). SOLNIL exploits the basic knowledge produced in NARCISO on sol-gel formulations and nano-imprint lithography techniques by extending their fabrication on larger surfaces and targeting specific applications in photonics (e.g.

anti-reflection coatings for high-power lasers, structural color, diffractive optics of augmented reality), bio-sensing (e.g. templates for micro-fluidic), anti-frosting and anti-wetting surfaces.

During the second year of the NARCISO project, several articles have been published (all articles are open access publications). The link to the published articles and the news about the project are visible at the following link: https://www2.mi.ifn.cnr.it/narciso/

Below the list of NARCISO network publications:

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