Ref. Ares(2020)2596638 - 18/05/2020

Project title: **NAtuRal instability of semiConductors thIn SOlid films for sensing and photonic applications (NARCISO)**
Grant Agreement: 828890
Call identifier: H2020-FETOPEN-2018-2020/H2020-FETOPEN-2018-2019-2020-01
Start Date of Project: 01/03/2019
Duration: 36 months

Deliverable 2.5 – V2.0

Title: Assessment of the optical properties of SiGe ordered and spinodal dewetted structures

Leader partner	UNIFI
Author(s)	Francesca Intonti
Estimated delivery	29/2/2020
Actual delivery	
Revised version	
Work Package	WP2- Spontaneous spinodal dewetting
Task	2.2, 1.4, 5.1
Dissemination level	Public
Version	2.0

Contents

1. Introduction

This document is a deliverable of the NARCISO project which is funded by the European Union's H2020 Programme under Grant Agreement (GA) No. 828890.

In this deliverable we describe the progress in the spectroscopic investigation of individual dewetted islands (WP1) and the approach to face study of spinodal dewetted structures with emerging disordered hyperuniformity (WP2). In order to address the collective optical behavior of complex nano-architectures implemented via solid state dewetting (SSD), that will be studied in WP5, it is essential to understand first the optical behavior of a single dewetted structures. For this purpose, we exploited our expertise of high spatial resolution spectroscopy and imaging that allow us to investigate the spatial distribution of the electric field intensity confined in micro- and nano-resonators using scanning near-field optical microscopy (SNOM). The preliminary results achieved on dewetted silicon islands are very promising, demonstrating that the SNOM technique combined with resonant scattering spectroscopy allows accessing the light confined in single dewetted islands. But we also understood that to better understand the light confinement within the dewetted islands, in the future we need to compare the near-field results with the far-field properties of the scattered light. We expect that the intricacies in the interpretation of the data collected in resonant scattering spectroscopy will be particularly evident in the study of the optical properties of spinodal dewetted structures (materials from WP2). These studies will be fundamental to demonstrate the photonic applications to be achieved in WP5. The content of this deliverable has been developed within the tasks principally included in WP1 but it has been already applied for preliminary optical characterizations in WP2.

2. Description of the optical experiments

Scanning near-field optical microscopy (SNOM) is proven to be a powerful technique for mapping the electric local density of states (LDOS) of resonators with subwavelength dimensions [1]. In particular, in Ref. 1 we demonstrated, in the case of photonic crystal nano-cavities in the absence of absorption, that the spectrum locally collected by the near-field probe directly corresponds to the LDOS of the cavity. The key feature of a SNOM is the near-field probe, a tapered single mode optical fiber, with apex dimensions smaller than 100 nm. The probe is maintained at a distance of a few nanometer from the sample surface collecting the near-field component of the optical signal under investigation. In this way the spatial resolution of the microscope is not more diffraction limited but is defined by the size of the probe. By raster scanning the probe on the sample surface it is then possible to reconstruct the optical map of the investigated structure.

Two different spectral-imaging approaches can be followed to detect the LDOS of a resonance, depending on the presence or absence of internal light source inside the resonator: (i) for optically active systems the SNOM probe collects the photoluminescence (PL) signal of the embedded light sources coupled with the resonator optical modes [2]; (ii) in the absence of internal light emitter the SNOM is combined with resonant scattering (RS) spectroscopy accessing the spectral and spatial distribution of the confined optical modes [3].

3. Preliminary results on dewetted islands

The first investigated structures, dewetted Si islands on a thin SiO_2 layer optimized by WP1 are optically inactive, therefore we implemented the SNOM with resonant scattering geometry. Testing the spectroscopy setup for optically non active systems is also essential for the characterization of the samples that will be investigated in WP5. This SNOM imaging technique involves the interference between two distinct scattering pathways: light diffused by the surface through a continuum of extended states, which is not coupled to the cavity mode, and light resonantly coupled to the cavity mode and subsequently diffused in free space. Owing to the phase retardation of the light coupled within the resonant modes interference build up. From this interference phenomenon originate Fano-like resonances. To ensures the hyperspectral imaging (the collection of the entire spectrum at each tip position) in the full optical range of interest, the sample is excited with a supercontinuum laser, using dielectric tapered SNOM probes in the illumination/collection geometry. Both the excitation and the signal collection occur through the probe (illumination-collection configuration). We performed crossed-polarization detection to cut most of the reflected light, which has the same polarization of the incident beam.

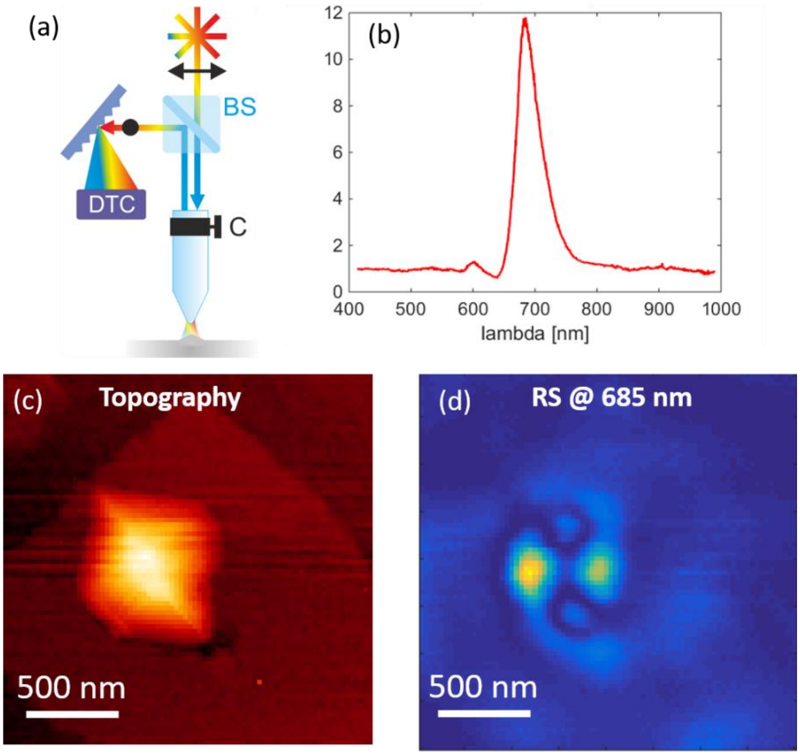

Figure 1: (a) Schematic of the experimental setup. (b) Typical resonant spectrum collected on a single Si dewetted island. (c) Topography of the dewetted island. (d) Resonant spectrum map at λ = 685 nm of the island reported in (c).

In Fig. 1(a) is reported the schematic of the SNOM experimental setup in an illumination-collection geometry: light from a supercontinuum laser transmitted through a polarizing beam-splitter cube (BS) is s-polarized and coupled to an optical fiber that ends with a SNOM dielectric tip. The backward scattered light collected by the probe is filtered in cross-polarization configuration by the BS, dispersed by a spectrometer and finally detected by a Si detector (DTC). Fig. 1(b) shows a typical

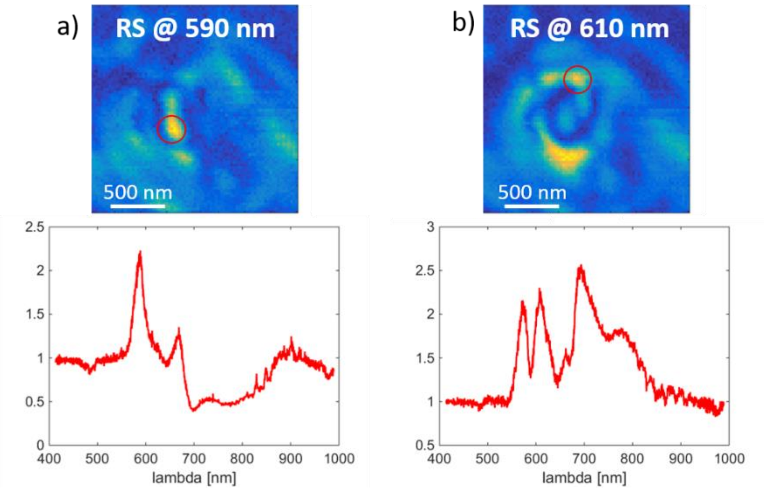

Figure 2 (a) Resonant spectrum map at λ = 590 nm of the island reported in Fig. 1(c) together with the resonant spectrum collected with the tip at the position indicated in the red circle. (b) Resonant spectrum map at λ = 610 nm of the island reported in Fig. 1(c) together with the resonant spectrum collected with the tip at the position indicated in the red circle.

resonant spectrum collected on a single Si dewetted island (developed in WP1), whose topography is reported in Fig. 1(c). The island has a pyramidal shape characterized by a width of about 600 nm and a height of 170 nm. The resonance is centered at 685 nm and has a full width at half maximum smaller than 60 nm. The spatial distribution of the RS signal at the peak wavelength is reported in Fig. 1(d) and shows a clear localization in the Si island, with a maximum of intensity on the lateral facets of the pyramid. Beside the main resonance at 690 nm, the SNOM scan shows others less intense peaks, that are also localized in the island region, as shown in Fig.2. The bright spot in the RS map of Fig. 2a) is located on one of the lateral facets of the pyramid, and the spectrum collected in this position is reported on the bottom of Fig. 2a, showing a peak centered at 590 nm. The optical map shows the spatial distribution of the resonant scattering signal at 590 nm. Similarly, the peak at 610 nm of the spectrum reported on the bottom of Fig. 2b) is spatially localized around the Si island. The red circles in the optical maps indicate the position where the spectra, reported on the lower part of the figure, are collected.

4. Implementation of SNOM and RS spectroscopies on dewetted islands

First of all, we realized that the blazing of the spectrograph grating cut the signal below 570 nm. For solving this issue, we acquired and mounted in our setup a new grating with the correct blazing, that allows to collect signal from 450nm to 800nm. Then, to better understand the light confinement within the dewetted islands we are going to compare the near-field results with the far-field properties of the scattered light. To this end, we are implementing, in our commercial SNOM, a dark-field spectroscopy setup with a spatial resolution of the order of 1 um. In this way, for sufficiently sparse dewetted islands, we will be able to firstly characterize the far-field scattering of individual island, and then investigate with the SNOM the spatial distribution of the modes of the same island. In order to be able to measure the same island, first in dark field and then in near-field, have been realized samples with marker that allows to deterministically identify each single dewetted structure. These kind samples have been produced within WP1 by AMU and are available for pure Si and pure Ge in arrays of islands with variable size.

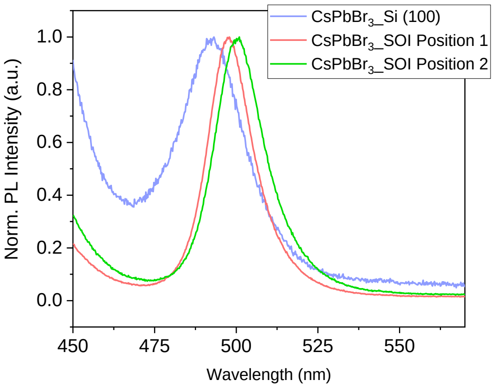

Figure 3 PL spectra a film of CsPbBr$_3$ perovskite deposited on two different substrates, Si with <100> orientation and silicon on insulator (SOI)

The above reported measurements on a single dewetted island confirmed us also what we already knew from our experience with photonic crystal cavities: the interpretation of data collected with optically active systems is much more straightforward and precise than the understanding of RS spectra collected in structures lacking of internal light sources. Therefore, we also investigate a way to place light emitter on the dewetted samples. One promising approach is the use of inorganic perovskite that show a brilliant PL signal in the visible range, that can be tuned in a controlled way by varying their composition. To study the feasibility of this approach we grew a thin layer of CsPbBr$_3$ perovskite on two different substrates as Si with <100> orientation and silicon on insulator (SOI). Fig. 3 shows the PL collected on the two substrates exciting at 405 nm. The peak around 500 nm is clearly related to the perovskite PL, and its spectral position is shifted to the blue with respect to the data in literature, probably due to strain effects. Following these positive results, the next step is to deposit the CsPbBr$_3$ perovskite on samples with dewetted structures that are expected to show resonances in the green.

5. Preliminary results on hyperuniform structures

It has been recently demonstrated that spinodal-like structures realized by solid state dewetting in WP2 show an effective disordered hyperuniform character [4]. Therefore, since the optical properties of spinodal structures are not yet deeply explored, we decide to follow an approach that allows us to go step by step: (i) first get familiar with the behavior of disordered hyperuniform structures realized on optically active platform, (ii) then face the optically properties of spinodal dewetted structures with disordered hyperuniform features.

In collaboration with other European teams, different disordered hyperuniform patterns have been designed like the ones showed in Fig.4. The white circles, positioned following a hyperuniform geometry, represent the pores that will be drilled on a 220 nm thick GaAs membrane in which emitting InAs quantum dots are

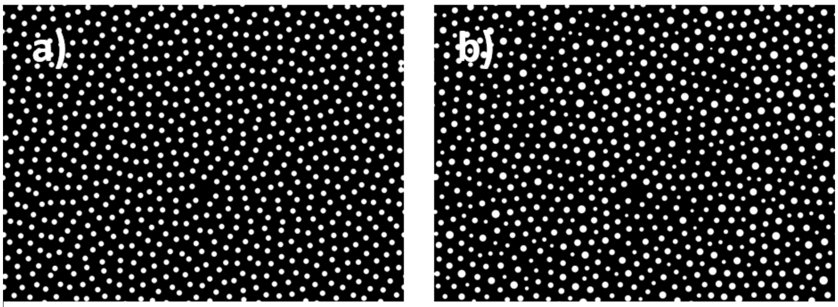

Figure 4: disordered hyperuniform patterns, the white circles represent the pores that will be drilled on a 220 nm thick GaAs membrane. (a) pores with the same diameter (b) pores with different diameter.

embedded, thus acting as internal light sources in the near-infrared.

Once established a reliable comprehension of the near-field features of light-emitting disordered hyperuniform structures obtained via top-down approaches, bottom-up disordered hyperuniform nano-architectures obtained via spinodal dewetting will be addressed. This information will be used within NARCISO to study the optical properties (WP2) and photonic applications of hyperuniform materials developed within WP5.

[1] Pellegrino et al. Phys. Rev. Lett. **124**, 123902 (2020)
[2] Intonti et al. Phys. Rev. B **78**, 041401(R) (2008)
[3] Caselli et al. 4, e326(2015)
[4] M. Salvalaglio, M. Bouabdellaoui, M. Bollani, A. Benali, L. Favre, J.B. Claude, J. Wenger, P.de Anna, F. Intonti, A. Voigt, M. Abbarchi, “Hyperuniform monocrystalline structures by spinodal solid-state dewetting”, arXiv preprint arXiv:1912.02952