First year PhD report

Student: Elena Ghidorsi

Tutor: Andrea Toma

Academic year: 2022-2023, XXXVIII cycle

Research activity

In the first year of my PhD I focused on the fabrication of photonic nanocavities for their possibility to interact with semiconductor nanocrystals (NCs) [1]. The realization of polaritonic architectures for photocatalytic applications [2,3] is indeed among the main objectives of the ERC project in which I am involved.

Since the nanoparticle-on-mirror (NPoM) geometry is a valid and viable configuration for the investigation of photocatalytic light-matter interactions [4], as a first approach, I started working on the deposition of gold (Au) and silver (Ag) thin films to implement Fabry-Pérot (FP) nanocavities with silica (SiO_2) and titania (TiO_2) as dielectric spacers.

For the growth of metallic and metal oxide layers, I have acquired hands-on experience with different deposition techniques available in the Clean Room Facility of the Istituto Italiano di Tecnologia (CCT), such as electron beam evaporation and atomic layer deposition (ALD). In particular, I have worked on the optimization of the optical quality of the films within the FP cavities in terms of contamination, roughness and thickness. This with the purpose of sharpening the FP resonances and improving their related Q-factors. In fact, having photonic resonators with reduced losses, or equivalently longer lifetimes, can foster a stronger interaction between the confined photons and the elementary excitations of the extra material - NCs/organic dyes - occupying the cavities.

To verify the thickness and the refractive index of the dielectric spacer I have performed spectroscopic ellipsometry while the optical response of the photonic architectures has been fully characterized recurring to UV-Vis micro-spectroscopy. The experimental results have been also compared with numerical computations (rigorous coupled wave analysis - Synopsys' Optical Solutions, RSoft).

I also fabricated distributed Bragg reflectors (DBRs), as a viable alternative to FP metallic films, due to their high-quality reflectivity in the visible range.

The integration of the optical resonators with organic molecular aggregates has been the following step. Within this context, J-aggregates represent the ideal test-bed to promote and investigate light-matter interaction under strong coupling regime. Due to their narrow exciton linewidth and high oscillator strength these molecular aggregates can lead to the formation of "new quasi-particles", known as exciton-polaritons, endowed with reshaped physico-chemical properties [5]. In line with this, I spin coated J-aggregate molecules dispersed in polyvinyl acetate (PVA) solutions inside the cavities in order to observe coupling effects between cavity modes and J-aggregate excited states. However, since the interest of my research activity is centered in the NCs-cavity interaction, I took into account Cadmium Selenide (CdSe) quantum dots solutions too, whose control of thickness and homogeneity is the critical point in fabricating the systems here described to maintain a good optical quality of the fabricated FP cavity. This is why I also dedicated significant efforts in the optimization of different spin coating recipes, dealing with various CdSe solutions, concentrations, etc.

Therefore, I characterized both the optical cavities and the semiconductor NCs in their uncoupled states with the steady-state micro-spectroscopic setup.

I obtained preliminary results of CdSe- and J-aggregate-integrated FP cavities, that are still under investigation in terms of hybrid states formation and anticrossing behavior. Thus, I partially contributed to implement the analysis script realized to estimate quantitatively the coupling strength parameters.

I took into account different NCs-based cavity configurations depending on the position of the NCs film referring to the cavity size, in order to place it in correspondence of the maximum of the confined radiation field.

I also characterized the planarization of polymethyl methacrylate (PMMA) films, after being processed with oxygen plasma for the future fabrication of vertical nanocavities. This configuration requires indeed the intercalation of a polymeric matrix into the NCs layers, which in turn allows for the correct exposure of quantum dots apexes to a subsequent metal cocatalyst deposition.

Finally, I spent effort to learn the principles of transient absorption spectroscopy technique. In collaboration with post-doc colleagues and team members I also started the use of the available setup in order to start the dynamic investigation of the fabricated samples in the ultra-fast regime. This will support us to characterize the photophysics of the coupled systems in terms of energy transfer rates between hybrid states, radiative process enhancements of the emitters, investigation of predominant characters from lifetimes comparisons, anticrossing behaviour, relaxation processes and eventual population transfers from coupled states and excitonic reservoir.

Other activities

Nanophotonic resonators fabricated and/or investigated:

- Planar “nano-gears” for chiral dichroic studies.

Experimental methods:

- Electron beam lithography (EBL), thermal evaporation, dichroic optical setup.
- Ellipsometer and ALD cross calibration for the measurements of the effective thickness of the deposited layers (in collaboration with Dr. Tamagnone’s research group at IIT).

Exams:

- Nanophotonics and nanofabrication (attended course, exam to be given at the end of September 2023)
- Quantum optics (exam given)
- Optoelectronics of nanomaterials, IIT (exam given)

Workshops:

- How to write a scientific paper, IIT 27th-28th March 2023

References:

1. Bitton O. *et al.*, Quantum dot plasmonics: from weak to strong coupling. Nanophotonics **2019** 8 (4), 559-575. DOI: 10.1515/nanoph-2018-0218
2. He Ren *et al.*, Core–Shell–Satellite Plasmonic Photocatalyst for Broad-Spectrum Photocatalytic Water Splitting. ACS Materials Letters **2021** 3 (1), 69-76. DOI: 10.1021/acsmaterialsleH.0c00479
3. Peng Zeng *et al.*, Photoinduced Electron Transfer in the Strong Coupling Regime: Waveguide – Plasmon Polaritons. Nano Letters **2016** 16 (4), 2651-2656. DOI: 10.1021/acs.nanolett.6b00310
4. Rocío Sáez-Blázquez *et al.*, Plexcitonic Quantum Light Emission from Nanoparticle-on-Mirror Cavities. Nano Letters **2022** 22 (6), 2365–2373. DOI: 10.1021/acs.nanolett.1c04872
5. Vasa P. *et al.*, Ultrafast Manipulation of Strong Coupling in Metal – Molecular Aggregate Hybrid Nanostructures. ACS Nano **2010** 8 (12), 7559-7565. DOI: 10.1021/nn101973p