Controlling the emission of colloidal quantum dots: from the single photon manipulation to collective effects for the generation of quantum states of light

In the field of quantum information science, photons play a crucial role due to their ability to propagate through long distances with very small loss of coherence. In this context, the generation and the manipulation of quantum states of light can benefit from the numerous nanophotonics tools and devices. Modifying the electromagnetic environment of a single photon emitter using cavities [1] enables to fully control the spontaneous emission of a single photon source such as a quantum dot to achieve bright single photon sources (SPSs). In addition, the interplay between an assembly of nanoemitters and plasmonic structures allows to design new types of light sources and to develop light-emitting quantum devices based on strong coupling and superradiance.

The PhD project will take place in the Quantum Nanophotonics (QNP) group, which has an expertise in colloidal semiconductor quantum dots (QDs) emission and their coupling to photonic structures based on metallic or SU8 polymer structures, the latter in collaboration with team of Ngoc Diep Lai from (LuMIn laboratory, ENS Paris-Saclay)t.

Quantum integrated photonics with deterministic hybrid SU8 photonic structures

LuMIn has implemented 10 years ago direct laser writing (DLW) method, which is based on a photosensitive polymer (SU8) exhibiting an ultralow absorption coefficient at the writing laser wavelength (532 nm continuous-wave laser). Through a successful collaboration between LuMIn and QNP groups, we demonstrated that the incorporation of a single QD into SU8 is very useful for suppressing the residual QD fluorescence blinking, to preserve QDs from oxidation, and to enhance the photon detection rate [2]. We have also shown that the insertion of single NC at a well-defined position into a polymer dielectric antenna results in a directional radiation pattern [3]. Finally, we manipulated also the single photon propagation (wave guiding, beam splitting/combination) by relying on submicron micropillars [4].



Figure 1: DLSPPW configuration [18].

The first objective of the PhD project is to take advantage of the SU8 structures flexibility to develop new polymerbased photonic and plasmonic structures of increasing complexity, deterministically coupled to a single QD as building blocks towards full on-chip integration. We will explore the possibility to realize a Mach-Zehnder interferometer with a phase shifter; which is an important basic element for quantum integrated photonics (QIP). We also plan to achieve a high level of

integration by waveguiding single photons on subwavelength scale with a dielectric-loaded surface plasmon polariton waveguide (DLSPPW) [5, 6], which consists in a SU8 dielectric stripe on a metallic surface. The dimensions of the stripes (height ~ 100 nm, width~200 nm, see Figure 1) will be optimized in order to achieve a propagation length of the DLSPPW plasmonic mode greater than 10 μ m.

Generation of original quantum states of light

The second part of the thesis will be devoted to the generation of quantum states of light by coupling high-ordered colloidal architectures of QDs to plasmonic nanostructures. Recently, in the context of an ANR project, we studied the emission properties of a few thousands of closed packed QDs encapsulated into a gold shell. We observed photon bunching at short time related to QDs coherent emission, i.e. superradiance [7]. During the same project, the case of colloidal nanopatelets deposited on gold films or inserted in 1 or 2 dimensional grating of the same metal were investigated. The strong coupling between a thermalized mesoscopic ensemble of nanoplatelets and the surface plasmon mode as well as the emission directivity were also both theoretically and experimentally studied [8, 9].

During the thesis, the objective is to use a new design of supraparticles (SPs) as building blocks:



Figure 2: Assembly of gold nanoparticles obtained by AFM manipulation in our team. The gap between the particles is lower than 50 nm. spherical gold particles with diameters around 50 nm encapsulated in a silica shell with a thickness lower than 10 nm and a single layer of colloidal QDs. When compared to the previous SPs, such approach will enable to synthesize non-porous gold with a high crystallinity and higher quality plasmon resonances. A small number of SPs (<10) will be manipulated with an AFM and elementary structures will be assembled (Figure 2). The goal of this step is to optimize the diameter of the SPs by FDTD simulations and observe collective modes. More interestingly, we will try to achieve ultra-strong coupling regime as was observed with organic molecules coupled to a metallic nanoshell. [10]; Such a regime, for which the interaction rate is of the order of the bare frequency of the uncoupled systems could be also obtained by depositing the SP on a flat gold film due to the

small gap between the SP and the film [11]. In the long term, such systems in the ultrastrong coupling regime could be used for quantum information processing or quantum metrology [12], high-resolution spectroscopy or squeezed light generation [13].

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