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Appel à projets
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Title of the research project :

Emission of plasmonic antennas in high confinement regimes

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Name: Institut des NanoSciences de Paris

Code *UMR7588*

Doctorate School

Thesis supervisor's doctorate school Ecole Doctorale Physique en Ile de France

PhD students currently supervised by the thesis supervisor (number, year of the first inscription) :
2 students: first inscription: 2020, 2021

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Description of the research project *(ENGLISH):*

Plasmonics make it possible to confine and manipulate light at the nanoscale. When nanoemitters are coupled to plasmonic nanoantennas, near field emitted light is converted to far field. Such antennas govern emission properties like directivity, brightness, and may modify spectrum or emission statistics [1]. Properties of such light nanosources can be optimized depending of the foreseen applications.
At the single emitter scale, through its coupling to a single nanoantenna, and thanks to the high confinement in the tiny volume of the antenna, we obtain a quantum nanosource with unique properties opening to new opportunities in nanoptics and in quantum technologies. Such single quantum sources are basic bricks in quantum photonics [2].

Antenna geometries depend on the targeted properties. In plasmonics, antennas modal volume are very small compared to the wavelength dimensions. State density and resonant field intensity are huge. In weak coupling regime, very high Purcell factor can be achieved. Despite important losses in plasmonics, strong coupling and collective effects [3] can be achieved.
Most studies on antennas concern collective scale , for which a huge number of nanoemitters like molecules, nanocrystals, are coupled to single antennas. Very few realizations couple a single nanoemitter to a single nanostructure. Indeed, besides experimental complexity related to single object study, the conditions for maximum interaction between antenna and nanoemitter are stringent. The emitter has to be located at the maximum of the resonant electric field inside antenna and its orientation has to be orthogonal to the surface for plasmonic coupling.
Most of the time, experimental realization with single objects are non –deterministic and studies require a statistical treatment. Deposition of emitters at the surface and their orientation are random. After fabrication of antennas, a strignet selection has to done and the few best samples can be selected. Therefore deterministic approach at the single scale, controlling both position and orientation of the emitter inside antenna, make it possible not only to achieve a much better success rate in antenna fabrication, but as well to understand deeply the physics by controlling the different parameters.

We have developed new methods, of in-situ lithography [4,5], allowing the deterministic positioning of a single semi-conductor nanocrystal inside a patch plasmonic nanoantenna. We use single CdSe/cdS nanocrystals which are bright and stable nanoemitters, working at room temperature. We control their position inside the antenna with a lateral precision better than 50nm and an axial one smaller than a few nm. We have as well developed a polarimetric method [6,7,8] making it possible to control the orientation of the emitter inside the antenna, opening the way to a very efficient coupling antenna plasmon modes

Our plasmonic patch antennas, deterministically coupled to single CdSe/CdS nanocrystal present remarkable properties. Depending on the chosen parameters we obtained either single photon sources or high brightness nanosources. The dynamics of emission is accelerated. This acceleration is quantified by the so-called Purcell factor, reaching a value much larger than 200, its measured value being limited by the instrumental response function of the setup.
Thanks to the spontaneous emission high acceleration, multiexcitons can recombine radiatively, their recombination becoming quicker than the Auger non radiative processes. We have demonstrated that for each single laser excitation more than 5 radiating excitons were contributing to the emission, making the emission very bright. These antennas benefit from both very high optical and electronic confinements.
For these single emitter plasmonic nanoantenna, by high optical pumping, the emission statistics change even more dramatically. The dynamics of emission become extremely quick, spectrum of emission broadens up to white ones, emission intensity becomes non linear with optical pumping power.

Some of these effects are observed in a just few groups [9,10] but most of them remain not understood up to now.

During the PhD we want to understand the physical ingredients at the origin dramatic change of emission: spectral broadening, high brightness, nonlinear emission and emission dynamic. We want to understand if under high electronic confinement, and high optical excitation, the emission could be associated to excitons bath thermalization [11] inducing spectral broadening and acceleration of emission. The objective of the work will the realization of extremely efficient quantum nanosources of light.
In a second step by increasing optical confinement, we want to increase the non-linearity of emission. The realization of quantum nonlinear nanosources would be a breakthrough in quantum technology

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Implementation and methodolgy

This PhD will be divided in three parts, involving fabrication of the nanoantennas, experimental characterizations, interpretations and modelling.

1) Realization of the antenna with single nanocrystal by in-situ-optical-SEM lithography
2) Antennas under high excitation
    a. Experimental study of the antennas under increasing pumping intensity :emission spectrum and intensity, dynamics of emission
    b. Modeling of thermal emission. Thermalization of excitons
    c. Interpretating experimental datas
3) Increasing optical confinement
    a. Increasing the optical confinement by realization of small antennas
    b. study of nonlinear response of such nanoantennas under high excitation