SU-NTU joint thesis proposal

Förster Non-Radiative Energy Transfer in a Strong Coupling Light-Matter Interaction System

Agnès Maître [1]/ Cuong Dang [2]/ Renaud Bachelot[2]

[1] Sorbonne Université / Institut des Nanosciences de Paris (INSP), Acoustique et optique pour les nanosciences et le quantique (ACONIQ)
[2] CNRS-International-NTU-Thales Research Alliance (CINTRA)

The emission of nanoemitters embedded in optical cavities, can be coupled to resonant ones, either dielectric microscopic ones or nanoscopic plasmonic ones [Ge2020]. In the regime of weak coupling the radiative recombination of nano-emitter exciton is affected by the cavity, leading to deexcitation acceleration, directional emission, spectral filtering and fluorescence enhancement. On the other hand, under strong coupling, the behavior of the system becomes completely different, with periodical exchange of energy between the exciton and the optical mode of the cavity, so that both can no more be considered separately. This resulting exciton-photon particle is called polariton. This leads to a splitting of frequency resonances of the system [Yu2020, Yuan2022]. This very specific coupling regime plays an important role in the quantum information toolbox.

Förster Energy Transfer (FRET) between to emitters, donor and acceptor, is a process allowing a non-radiative energy transfer from an excited donor to the acceptor. For semi-conductors, this non radiative energy transfer between donor and acceptor excitons, allows optical excitation of the donor and emission of the acceptor after radiative recombination of the exciton. Usually, FRET occurs on very short length, typically <10 nm.

The objective of the PhD is to explore the modification of the FRET when donors and acceptors are embedded within a dielectric or plasmonic cavity and they are resonant with the cavity modes. We hypothesise that the strong coupling between donor's exciton and acceptor's exciton with the photonic mode of an optical cavity will enhance the FRET significantly and alter its dynamic and length propagation.

Single emitters like colloidal CdSe/CdS quantum dots or nanoplatelets are single photons sources at room temperature with a very good brightness and stability. Within cavities, their emission can be controlled. SU/ACONIQ has already demonstrated the deterministic coupling of a single colloidal quantum dot to a plasmonic nanoantenna, with a large increase of brightness, large acceleration of spontaneous emission and directional emission [Dhawan2020]. Emission cross section of single quantum dots within the antenna has been increased by 3 orders of magnitude [Dhawan2022]. With many nanoplatelets at the hotspot of a nanocavity made of a Silver nanocube (Fig. 1a), NTU team has demonstrated the strong coupling with a giant Rabi splitting energy up to 400 meV and cooperativity exceeding 11 [Yu2020]. With appropriate synthesis and solvent control, NTU team has stacked CdSe/CdS nanoplatelets by their surface, constituting long chains of tens to hundreds of nanometers, even though the thickness of each nanoplatelet individually is just about 1 nm. Neighbors nanoplatelets are connected by ligands of a few nm length. We have proved that exciton energy can be transferred non radiatively from one platelet to the next ones within the chain. Such efficient homo-FRET allows excitons to freely move along the chain. By inserting lower bandgap nanoplatelets or a mid-gap dopant nanoplatelets into the chain, we create donors for chain's exciton emission [Yu2022]. The density of donors and chain length determine the emission properties (Fig. 1b).

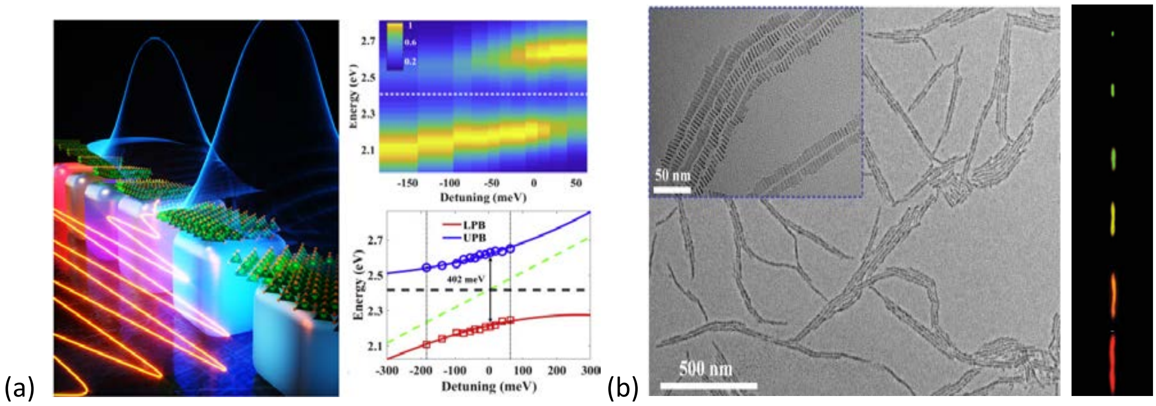

Fig. 1: (a) Strong coupling of nanoplatelets with a Silver nanocube. **Left**: Illustration of nanoplatelets at the hot spot of the nano plasmonic cavity. **Right**: The anti-crossing characteristics of strong coupling with giant Rabi splitting energy [Yu2020]. (b)**:** Long chain of nanoplatelets (**left**) and their length-dependent emission (**right**).

In the strong coupling regime within the cavity, unlike conventional FRET, the dipole-to-dipole coupling from donors to acceptors is now modified by exciton-photon cavity mode interaction. The FRET rate and distance will be quantified to provide the evidence of FRET enhancement. One of the objectives is to identify the role of polaritonic states in mediating energy transfer, and explore the influence of parameters such as coupling strength, detuning, and geometric configuration on FRET.

The candidate will join both teams of physicists and engineers to conduct the research with analytical modelling, numerical simulations, and experimental validations. The NTU team will provide nanoplatelets of different thickness with or without Cu dopants. Thin nanoplatelets will be used as donors while acceptors can be the thicker nanoplatelets or the ones with dopants. Strong coupling experiments will be done in both places, in NTU with dielectric cavities and in SU with plasmonic ones. Numerical simulation and theoretical study will be done at SU. The program will conclude with 6 months in SU for writing and publication. Advanced fabrication techniques in cleanrooms in NTU and SU will be utilized to fabricate micro/nano-cavities with embedded semiconductor nanoplatelets. The structures will be characterized by microscopy techniques optical and or electronic ones. Strong coupling characteristics will be measured by the back-focal plane imaging technique, while FRET characterization will be done with time-resolved spectroscopic techniques.

The outcomes of this research are expected to provide new insights into the control of energy transfer in nanostructured materials with a novel fundamental physics of polariton states. The potential applications are broad, from advanced photonic devices, energy harvesting systems to quantum information processing technologies.

References

[Ge2020] Ge, D., et al., *Hybrid plasmonic nano-emitters with controlled single quantum emitter positioning on the local excitation field.* Nature Communications, 2020. **11**(1): p. 3414.

[Yu2020] Yu, J., et al., *Strong Plasmon-Wannier Mott Exciton Interaction with High Aspect Ratio Colloidal Quantum Wells.* Matter, 2020. **2**(6): p. 1550-1563.

[Yu2022] Yu, J., et al., *Modulating Emission Properties in a Host–Guest Colloidal Quantum Well Superlattice.* Advanced Optical Materials, 2022. **10**(4): p. 2101756.

[Yuan2022] Yuan, Z., et al., *Direct Imaging of Weak-to-Strong-Coupling Dynamics in Biological Plasmon–Exciton Systems.* Laser & Photonics Reviews, 2022. **16**(8): p. 2200016.

[Dhawan2020] A.R. Dhawan, et al, *Extreme multiexciton emission from deterministically assembled single emitter subwavelength plasmonic patch using spatially modulated light,* Advanced materials, 2022. **34**, 2108120

[Dhawan2022] A.R. Dhawan, et al, *Efficient single-emitter plasmonic patch antenna fabrication by deterministic in situ optical lithography antennas*, Light: science and application, 2020, **9**, 3.