

Energy migration and collective fluorescence effects from self-assembled nanoparticles

Keywords: nanosciences, fluorescence, optical microscopy, exciton interactions, quantum optics

The group *Nanostructures and Optics* at INSP studies light emission and propagation at the nanoscale: fluorescent nano-sources, emission control by photonic crystals or nano-antennas, chirality of fluorophores and antennas, bio-inspired photonic crystals etc. Among our interests, semi-conductor nanoparticles are very bright, stable and versatile light sources with more and more applications in bio-imaging, lighting and TV displays, and possibly for photovoltaics. When a *single* nanoparticle is examined by fluorescence microscopy, its emission often displays purely quantum-optical properties such as single photon emission (photons are emitted one by one) which can be used for quantum information.

While fluorescence from isolated emitters is now well known, most opto-electronic applications (LEDs, solar cells...) involve nanoparticles packed in a dense layer, where they should behave very differently because of interactions, charge transport and energy diffusion between neighboring particles. By using adequate solvent and ligands, the group of B. Abécassis in ENS Lyon has managed to assemble chains of hundreds of semiconductor nanoplatelets (fig. 1(a-c)) which constitute a good model system for nanoparticles interactions.

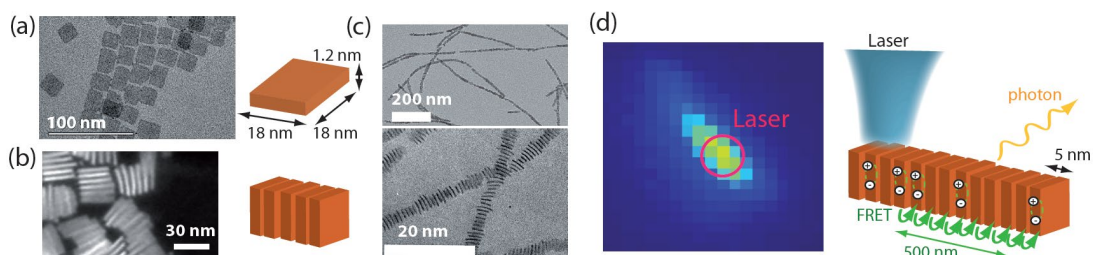


Figure 1 : TEM images of (a) CdSe nanoplatelets, (b) aggregated platelets and (c) self-assembled stacked nanoplatelets [S. Jana et al., Science Advances 2017]. (d) A laser excites a spot on the chain and energy propagates by FRET between around 100 platelets so that a 1- μ m portion radiates light [1]

We have shown that, due to near-field dipole-dipole Förster-type interactions (FRET), energy migrates between platelets over record distances of 500 nm (fig. 1(d)) [1] corresponding to around 100 platelets, with typical rate of $(1 \text{ ps})^{-1}$. As a result of this exceptional level of FRET coupling, one defect platelet is sufficient to quench the emission from many neighboring platelets, leading to collective quenching and collective fluorescence intermittency effects [2].

The aim of the PhD work will be to explore further coupling mechanisms among the nanoplatelets, in particular at higher excitation powers :

- when several excitons are created simultaneously in different platelets, an exciton can migrate by FRET and may encounter another exciton, which would enhance exciton-exciton interactions. Such FRET-mediated multi-excitonic effect can manifest in various properties (spectral, dynamics etc). The most remarkable would be the observation of single-photon emission from a large ensemble of nanoplatelets (simultaneous excitons annihilate each other by Auger coulombian interaction so that only one exciton remains and emits a photon).
- if all nanoplatelets are in their excited state, they can also experience the quantum-optical mechanism of superradiance leading them to interfere constructively and emit at a highly accelerated rate. This effect has been analyzed on ensembles of trapped atoms and ions, but for the moment few reports have demonstrated superradiance for nanoparticles and the role of phonons and inhomogeneities remains to be understood.

The PhD work will be mostly experimental (fluorescence microscopy, single-photon counting) but should also include some analytical or numerical modelling.

References :

[1] Jiawen Liu, Lilian Guillemeney, Benjamin Abécassis and Laurent Coolen, *Long range energy transfer in self-assembled stacks of semiconducting nanoplatelets*, Nano Lett. 20, 3465 (2020),

[2] Zakarya Ouzit, Jiawen Liu, Juan Pintor, Benoît Wagnon, Lilian Guillemeney, Benjamin Abécassis and Laurent Coolen, *FRET-mediated collective blinking of self-assembled stacks of CdSe semiconducting nanoplatelets*, ACS Photonics 10, 421 (2023),

Techniques/methods in use: Photoluminescence microscopy, spectroscopy, polarization analysis, single-photon detection

Applicant skills: Motivation for experimental work, organization, careful treatment of results, basic knowledge in optics

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<http://www.insp.jussieu.fr/-Themes-de-recherche,104-.html>

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