Towards enhanced absorption and emission with silicon

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Silicon quantum dots (SiQDs) are a promising alternative to replace toxic and rare material QDs that are researched for use in optoelectronics, photonics and bio-imaging. For competitive emission and absorption properties, however, the oscillator strength at the band-edge needs to be considerably improved as well as spectral tunability of emission that is typically limited by O- or N-based defect sites.

In our work, we focus on both aspects. Concerning the oscillator strength, we have in past suggested conversion of the bandgap structure by the use of charge transfer at the surface of SiQDs by electronegative ligands [1,2]. Competitive work by Kusova et al. [3] suggest that the same type of ligands leads to tensile strain, which further promotes direct band-edge transitions. In collaboration of both teams, we studied two types of such organic-capped SiQD materials (C:SiQDs) prepared by different approaches – top down by electrochemical etching and bottom up by wet chemical synthesis. In order to measure radiative rate directly, to be able to distinguish separate contribution of the non-radiative effects, we perform Drexhage-type experiment. C:SiQDs show enhanced radiative rates to 10^9 s^{-1} (Fig. 1), which is three to four orders of magnitude more than in other types of SiQD materials (hydrogen- or oxide-capped). However, despite radiative rates approaching those of direct bandgap QDs, the emission quantum yield (QY) in the visible range remains comparatively low (<20%).

Improved understanding of the processes underlying the ensemble photoluminescence (PL) and QY of these materials can be achieved from single QD spectroscopy, allowing study of the properties of individual emitters that are otherwise obscured in ensemble measurements. As generally in all quantum dot emitters, the QY is critically influenced by effects related to surface charge, local electric fields and trap states, leading to lower blinking duty cycle and enhanced non-radiative losses.

By comparison of the blinking duty cycle, internal quantum efficiency (radiative rate) and ensemble QY measurements, we identify the non-radiative mechanisms that prevent high emission yields.

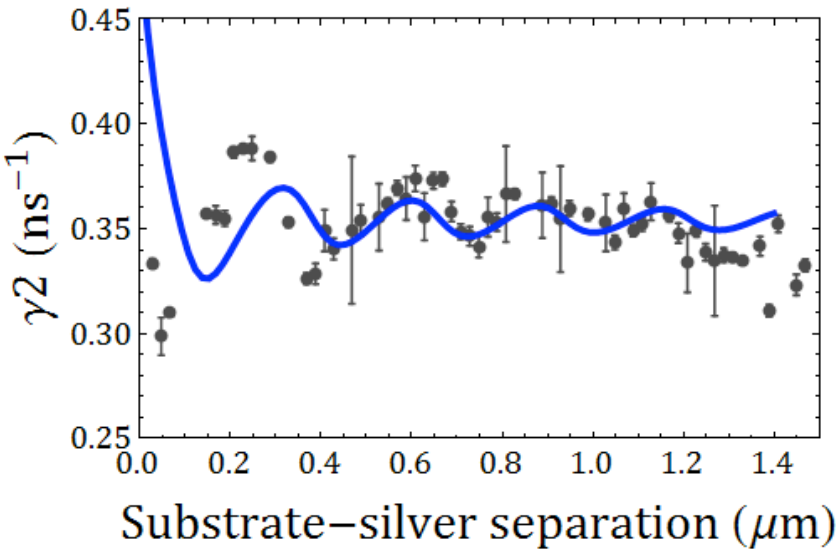

Fig. 1 – Drexhage experiment performed on alkyl capped SiQDs. Measurement suggests that radiative rate is of the order of 10^9 s^{-1}.

[1] K. Dohnalova et al., Light: Science and Applications 2 (2013) e47

[2] A. N. Poddubny and K. Dohnalova, Phys. Rev. B 90 (2014) 245439

[3] K. Kusova et al., Adv. Mater. Interf. 1 (2014) 1300042