Photon-avalanche in single upconversion nanoparticle for STED super-resolution nanoscopy

Yujia Liu[1], Xusan Yang[1], Yiqing Lu[2], Dayong Jin[2*], Peng Xi[1*]
[1]Department of Biomedical Engineering, College of Engineering, Peking University, Beijing 100871, China
[2]Advanced Cytometry Laboratories, ARC Centre of Excellence for Nanoscale BioPhotonics (CNBP), Macquarie University, Sydney, NSW, 2109, Australia
[*]E-mail: xipeng@pku.edu.cn dayong.jin@uts.edu.au

KEY WORDS: STED, upconversion, rare-earth, nanoparticle

ABSTRACT:

Upconversion nanoparticles (UCNPs) can emit stable, bright visible luminescence with single-nanoparticle sensitivity [1, 2], making it suitable for STED super-resolution microscopy. Here we show that UCNPs doped with high concentrations of Tm^{3+} ions, termed super dots, can easily establish population inversion on their intermediate metastable energy level. The reduced inter-emitter distance at high Tm^{3+} doping concentration leads to intense cross-relaxation, inducing photon avalanche effect that rapidly populates metastable $^3H_4$ levels resulting in population inversion to the $^3H_6$ ground level within a single nanoparticle. As a result, the onset of a laser at 808 nm, matching the upconversion band of $^3H_4$-$^3H_6$, can trigger amplified stimulated emission to discharge the $^3H_4$ intermediate level, so that the further upconversion pathway to generate blue luminescence can be optically inhibited with very low intensity. We employ this approach to realize low-power super-resolution stimulated emission depletion (STED) microscopy and achieve 28 nm optical resolution (λ/36) to image the single UCNPs. These engineered nanocrystals in the photon avalanche regime offer saturation intensity 2 orders-of-magnitude lower than those of fluorescent probes currently employed in STED microscopy, suggesting a new route to alleviate the square root law in STED super resolution [3, 4]. Long term STED imaging of over 200 minutes has been demonstrated to show the photostability of UCNPs. It has also been employed in the low intensity STED imaging of the actin fiber bundles with super-resolution capability [5].

References:

1. Lu, Y., et al., *Tunable lifetime multiplexing using luminescent nanocrystals.* Nature Photonics, 2014. **8**(1): p. 32-36.
2. Zhao, J., et al., *Single-nanocrystal sensitivity achieved by enhanced upconversion luminescence.* Nature Nanotechnology, 2013. **8**(10): p. 729-734.
3. Liu, Y., et al., *Amplified stimulated emission in upconversion nanoparticles for super-resolution nanoscopy.* Nature, 2017. **543**(7644): p. 229-233.
4. Liang, L. and X. Liu, *STED Nanoscopy Goes Low Power.* Chem, 2017. **2**(3): p. 331-333.
5. Zhan, Q., et al., *Achieving high-efficiency emission depletion nanoscopy by employing cross relaxation in upconversion nanoparticles.* Nature Communications, 2017. **8**(1): p. 1058.