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Upconversion nanophotonics : Photophysics, simulations, and applications

Nanophotonics localizes an optical phenomenon with small metallic particles. The effect is largest at a plasmon resonance. Plasmonics use resonances of the density of surface electrons with an incoming field to locally enhance the electric field strength. This increases the optical interaction in that small volume of space where the resonance is taking place. These plasmon resonances can be tuned by particle size and shape, or by gold coating thickness. A key manner in which nanophotonics can control an optical interaction is that the metal increases the local photon density of states (LDOS), so photon transition rates are sped up while phonon (non-radiative) rates remain fixed.

Rare earth ion doped upconverting nanoparticles are excited in the near infrared (NIR) and fluoresce via anti-Stokes emission in the visible energy range (400-650 nm). The NIR light provides large penetration depth of excitation, while the particles exhibit no blinking, and high signal-to-noise ratio due to zero tissue autofluorescence. In addition, since upconversion is a two-photon fluorescence process, it has the same ability as other 2-photon fluorescence microscopies to resolve the 3-dimensional structure of objects. In the co-doped rare earth ion upconverter system studied here, the Ytterbium and Erbium dopant couple, the upconversion occurs through an energy transfer upconversion (ETU) process, where the Yb^{3+} ion transfers its energy to the Er^{3+} ion. Despite using a real rather than virtual intermediate state, the brightness and upconversion efficiency of these nanoparticles is not comparable to that of semiconductor nanoparticles and dyes. The down-scaling of particle size also leads to a rapid loss of brightness. This has been attributed to the low absorption cross-section of the rare earth ion dopants. That is because transitions to the inner 4f-shell levels in rare earth ions are only very weakly allowed; hence their absorption coefficients are very small, limiting their maximum emission intensities. Although that shortcoming is partially compensated by its zero background fluorescence and its non-blinking and non-bleaching properties, we show that plasmonics lead to 1) local field enhancements that increase the absorption and emission efficiencies, and 2) a large anisotropy in the fluorescence yield if illuminated with polarized light. The emission is dependent on the particle orientation and is also polarized and directional.

Upconversion nanostructures are optimized with predictive finite element modelling (derivation of the LDOS) and correlated structural and optical single nanoparticle spectroscopy is performed to explore the influence of the nanostructure orientation, and geometry on the time scale of the optical transitions. Isolation at the single particle level allow for establishment of quantitative relationships between the crystal architecture and orientation that control emission properties, to enable direct comparisons with other lasing systems and allow for rational engineering. The single particle results are also more consistent with finite element calculations, without having to correct for anomalies generated by ensemble measurements.

The optimized nanostructures can potentially be applied in an array format in a display, quantum computing or in solar harvesting devices.

Speaker Biography

Following completion of a Ph.D. at the University of Cambridge, UK, 2004, Shuang Fang Lim served in a postdoctoral Research Position at Princeton University from 2004-2008. Her work there focused on upconverting nanoparticlels (UCNPs) and the synthesis, photophysics and bio-applications of nanoparicles. Following this assignment, she then served as in a postdoctoral position for one year at NC State University and then in a Research Assistant professor position for three years before accepting a Professor appointment staring in the fall of 2012.

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