Plasmonic coloration of emissions from upconversion nanoparticles for imaging and encryption

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Upconversion nanoparticles (UCNPs) have attracted much attentions in the field of bio-imaging, three-dimensional displays and lasing for their low toxicity, photostability, and long lifetimes [1]. However, suffering from limited absorption cross-sections of rare earth irons, the low quantum efficiency of upconversion nanoparticles hinders practical applications in many respects. For example, cell imaging with UCNPs requires high excitation light intensities, which leads to phototoxicity against applications to living organisms.

For its extraordinary ability to augment local electric-fields, localized surface plasmon resonance (LSPR) [2] has already been applied in enhancing upconversion emission of UCNPs [3]. In this case, the photon radiative rate can be enhanced and upconversion luminescence intensity can be increased through the coupling between the LSPR and electron transitions in UCNPs. By tuning the LSPR to couple with the photon emission at the specific frequency, the color of upconversion luminescence can be preferably chosen, resulting in the coloration of emissions from UCNPs dependent on the LSPR wavelength.

In this paper, we present an upconversion emission augment and consequent image encryption approach through utilizing plasmonic enhancement to tune the color of upconversion emissions. Core shell UCNPs consisting of $NaYF_4$:Yb/Ho/Ce emitting red and green photons are used as the upconversion agents in the experiments. The resonance frequency of surface plasmon antenna is tuned by patterning metal-insulator-metal (MIM) structures with femtosecond laser beams at different laser parameters. To enable the coupling between the UCNPs and LSPR, UCNPs are spin coated on the MIM structures. Enhanced upconversion emission by the antenna with the LSPR wavelength at 670 nm is observed with decreased upconversion emission lifetimes at the red emission band from averaged 153 us to 80 us. Meanwhile, the antenna with the LSPR wavelength at 544 nm leads to reduced upconversion emission lifetimes at the green emission band from averaged 219 us to 116 us. The augment in the upconversion emission at different bands with different strength gives rise to the coloration of the emission of UCNPs, which is utilized for encoding images.

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References
[1] Renren. Deng et al. "Temporal full-colour tuning through non-steady-state upconversion," *Nature Nanotech.* 10, 237–242 (2015).
[2] Haoran Ren et al. "On-chip noninterference angular momentum multiplexing of broadband light," *Science*, 352, 6287, 805-809 (2016).
[3] Sanyang Han et al, "Enhancing Luminescence in Lanthanide-Doped Upconversion Nanoparticles," *Angew. Chem. Int. Ed.* 53, 11702 – 11715(2014).