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Electrothermal control of upconversion emissions in rare-earth doped NaY(Gd)F4 nanoparticles by coupling with silver nanowires networks: A versatile platform for in situ thermal experiments

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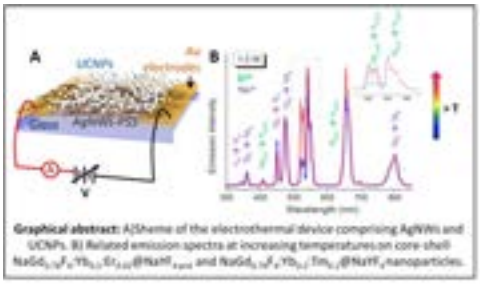
Photon upconversion (UC) is a non-linear optical anti-Stokes process by which low energy photons stimulate the emission of higher energy photons. The hexagonal phase (P63/m) -NaYF4 doped with rare-earths elements stands as one of the most efficient UC materials, finding applications in bioimaging, solar-cells and displays. However, many of these applications operate in fluctuating temperature conditions affecting each emission line of the UC spectra in a different manner. In this work, we develop a functional device to study in situ the thermal effects on UC nanoparticles (UCNPs) of different size and composition by using a percolating network of silver nanowires (AgNWs) as a highly transparent heating element. The electrical power dissipated by Joule effect allows for the electrothermal control. This device was successfully applied to characterize the thermal dependence of UC in large (>100 nm) -NaYF4:Yb:Er(Tm,Ce-Ho) and small (<20 nm) core-shell -NaGd4:Yb:Er(Tm,Ce-Ho)@NaYF4 UCNPs in the 20 °C-140 °C interval. Just the presence of AgNWs was enough to produce an enhancement of 20-30% in the intensity of UC emissions. We find that an increment in temperature can enhance or partially quench the emission lines selectively on each UCNPs system. The most temperature-sensitive case was that of Er doped UCNPs, in which the optical transitions 2H11/2→4H15/2 (H transition) and 4S3/2→4H15/2 (S transition), were found to reversible change in a different manner. For the case of bigger NaYF4 UCNPs, the S transition is quenched while the H transition was barely constant. For the small-sized UCNPs, the S transition remained unaffected while the H transition was sturdily enhanced. Time-resolved spectroscopy at different temperatures revealed further insights on the mechanisms involved. A rate-equation model was proposed to unravel the underlying mechanism. Finally, we take advantage of the electrothermal device to analyze in-situ other relevant chemical processes.



Graphical abstract: A)Scheme of the electrothermal device comprising AgNWs and UCNPs. B) Related emission spectra at increasing temperatures on core-shell $NaGd_{0.78}F_4:Yb_{0.2}:Er_{0.02}@NaYF_{4\,wet}$ and $NaGd_{0.78}F_4:Yb_{0.2}:Tm_{0.02}@NaYF_4$ nanoparticles.

Biography

Eduardo D. Martínez is an assistant researcher of CONICET working at the Low Physics Division of the Bariloche Atomic Center (CAB), a national research facility located in the city of San Carlos de Bariloche, Río Negro, Argentina. He has a materials engineering degree with a PhD in chemistry at the University of San Martín, Argentina. He performed a postdoctoral research at CAB-Argentina developing nanocomposite materials for microfabrication. At the moment, he is running a postdoc project in the physics department at the Universidad Estadual de Campinas, (UNICAMP), Brazil. His expertise is in the field of nanomaterials and nanocomposites, mostly applied to plasmonics and photonics. Specifically, he works in the chemical synthesis of nanoparticles and their assembly into nanostructures and devices by applying techniques of self-assembly and combining bottom-up and top-down methods.

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