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Metal Nanostructure-Multiexciton Interactions: From Emission Enhancement to Modification of Photon Statistics

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In the past decade, a tremendous amount of research efforts has been invested in the study of metal nanostructure (NM)-nanoemitter interactions. However, most of these studies have been conducted in the context of MNs interacting with single excitons. In contrast to these studies, we ventured into the realm of multi-exciton-MN interactions by performing low temperature photoluminescence (low-T PL) and photon-correlation spectroscopy studies on individual core/ultra-thick-shell NQDs ("giant"-NQDs or g-NQDs) deposited on nano-roughened silver films. Our low-T PL study show that (1) the multiexciton (MX) emissions in g-NQD coupled to silver films were enhanced mainly through the direct modification on the competition between the radiative and nonradiative recombination processes of MXs; and (2) strong enhancement in absorption is not necessary for a strong multiexciton emission.[1] Our room temperature photon-correlation spectroscopy studies reveal that the MN-g-NQD interaction can transform sub-Poissonian photon emission statistics of individual g-NQDs to strong super-Poissonian statistics (photon-bunching).[2] We further derived the conditions required for the manifestation of this phenomenon and show that it can also manifest in other nanoemitters such as epitaxially grown QDs and single walled carbon nanotubes. The understandings attained in this work could open a new plasmonic route for manipulation of important multiexciton processes such as optical amplification, lasing and entangled-photon-pair generation.

[1] *J. Phys. Chem. Letts.* **4**, 1465-1470, (2013).
[2] *Phys. Rev. Lett.* **110**, 117401, (2013).