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Xu, Qing-Hua

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Single Particle Spectroscopy Studies on Plasmon Coupling Enhanced Two-photon Photoluminescence and Photo-thermal Responses

Qing-Hua Xu

Department of Chemistry, National University of Singapore, Singapore 117543

E-mail: chmxqh@nus.edu.sg

Noble metal nanoparticles (NPs) have been known to display many unique optical properties of surface plasmon resonance, which arises from collective oscillation of conduction band electrons induced by light radiations. Plasmon resonance can greatly modify the optical properties of nearby chromophores by enhancing their excitation efficiency, radiative and non-radiative decay rates [1, 2]. Aggregated metal NPs generally resulted in larger enhancement compared to monodispersed metal NPs [1, 3]. Our recent studies showed that aggregated metal NPs displayed significantly enhanced photo-thermal responses [4] and two-photon photoluminescence (2PPL) [1, 5-10]. Here I will present our recent effort on using single particle spectroscopy to investigate Plasmon enhanced optical responses.

We first studied single molecule fluorescence enhancement of chromophores by Au nanorod dimers with a tip-to-tip orientation [3]. It was found that the dimer with the smallest gap (6.1 nm) gives the highest enhancement (470-fold). The enhancement gradually decreases as the gap distance increases and eventually approaches that from a monomer (120-fold). Single particle spectroscopy studies on 2PPL properties of metal NPs [7-10] revealed that 2PPL of metal NPs and assemblies strongly depends on their morphologies and relative orientations. Plasmon coupling was found to play a critical role in enhancing the excitation efficiency in the aggregated metal NPs. 2PPL of metal NPs was found to be enhanced by several orders of magnitude on the single particle level.

Metal NPs were found to display enhanced near-infrared (NIR) photothermal effects upon formation of clusters. We have studied the photothermal properties of Au nanosphere clusters on the single-particle level using photothermal heterodyne imaging (PTHI) microscopy [4]. NIR photothermal responses of Au nanoparticle clusters were found to significantly increase from monomers to trimers. The averaged PTHI signal intensity of Au nanosphere dimers and trimers is ~10 and ~25 times that of monomers. The NIR photothermal effect of clustered nanospheres strongly correlates with their longitudinal plasmon mode. Clustered Au nanospheres were demonstrated to exhibit dual-capability NIR photothermal imaging and therapy of human prostate cancer cells with high efficiency and selectivity, which could be potentially utilized for simultaneous cancer imaging and therapy with 3D selectivity.

Keywords: Single particle spectroscopy, Metal Nanoparticles, Plasmon resonance, Two-photon excitation, Photoluminescence, Photothermal

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