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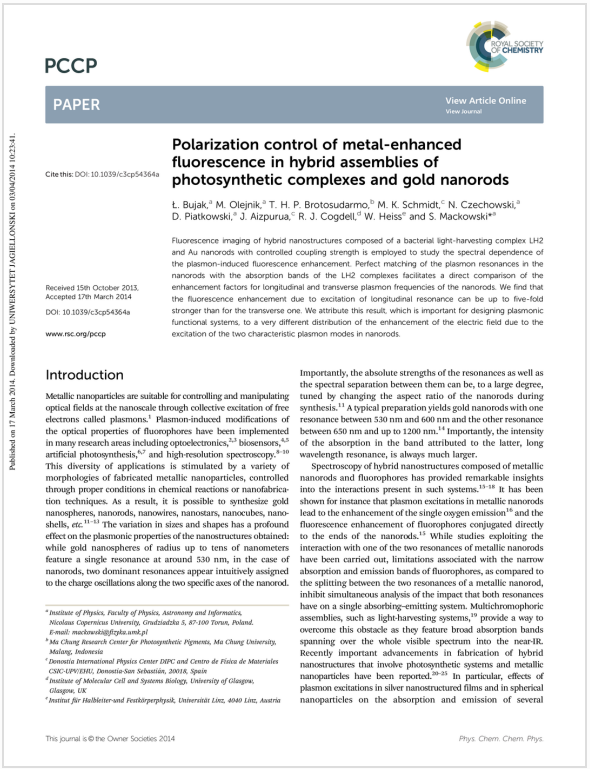

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Polarization control of metal-enhanced fluorescence in hybrid assemblies of photosynthetic complexes and gold nanorods

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Fluorescence imaging of hybrid nanostructures composed of a bacterial light-harvesting complex LH2 and Au nanorods with controlled coupling strength is employed to study the spectral dependence of the plasmon-induced fluorescence enhancement. Perfect matching of the plasmon resonances in the nanorods with the absorption bands of the LH2 complexes facilitates a direct comparison of the enhancement factors for longitudinal and transverse plasmon frequencies of the nanorods. We find that the fluorescence enhancement due to excitation of longitudinal resonance can be up to five-fold stronger than for the transverse one. We attribute this result, which is important for designing plasmonic functional systems, to a very different distribution of the enhancement of the electric field due to the excitation of the two characteristic plasmon modes in nanorods.

Introduction

Metallic nanoparticles are suitable for controlling and manipulating optical fields at the nanoscale through collective excitation of free electrons called plasmons.[1] Plasmon-induced modifications of the optical properties of fluorophores have been implemented in many research areas including optoelectronics,[2,3] biosensors,[4,5] artificial photosynthesis,[6,7] and high-resolution spectroscopy.[8–10] This diversity of applications is stimulated by a variety of morphologies of fabricated metallic nanoparticles, controlled through proper conditions in chemical reactions or nanofabrication techniques. As a result, it is possible to synthesize gold nanospheres, nanorods, nanowires, nanostars, nanocubes, nanoshells, *etc.*[11–13] The variation in sizes and shapes has a profound effect on the plasmonic properties of the nanostructures obtained: while gold nanospheres of radius up to tens of nanometers feature a single resonance at around 530 nm, in the case of nanorods, two dominant resonances appear intuitively assigned to the charge oscillations along the two specific axes of the nanorod. Importantly, the absolute strengths of the resonances as well as the spectral separation between them can be, to a large degree, tuned by changing the aspect ratio of the nanorods during synthesis.[13] A typical preparation yields gold nanorods with one resonance between 530 nm and 600 nm and the other resonance between 650 nm and up to 1200 nm.[14] Importantly, the intensity of the absorption in the band attributed to the latter, long wavelength resonance, is always much larger.

Spectroscopy of hybrid nanostructures composed of metallic nanorods and fluorophores has provided remarkable insights into the interactions present in such systems.[15–18] It has been shown for instance that plasmon excitations in metallic nanorods lead to the enhancement of the single oxygen emission[16] and the fluorescence enhancement of fluorophores conjugated directly to the ends of the nanorods.[15] While studies exploiting the interaction with one of the two resonances of metallic nanorods have been carried out, limitations associated with the narrow absorption and emission bands of fluorophores, as compared to the splitting between the two resonances of a metallic nanorod, inhibit simultaneous analysis of the impact that both resonances have on a single absorbing–emitting system. Multichromophoric assemblies, such as light-harvesting systems,[19] provide a way to overcome this obstacle as they feature broad absorption bands spanning over the whole visible spectrum into the near-IR. Recently important advancements in fabrication of hybrid nanostructures that involve photosynthetic systems and metallic nanoparticles have been reported.[20–25] In particular, effects of plasmon excitations in silver nanostructured films and in spherical nanoparticles on the absorption and emission of several

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