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Gold-Nanorod Based SPASER

Abstract

Upon resonant optical excitation noble-metal nanostructures exhibit spatially confined (localized) surface plasmons (LSP) that efficiently squeeze the electromagnetic field in nanoscale regions near the nanostructure surface and therewith, allowing for tailoring light-matter interactions at the nanoscale [1]. Optically excited excitonic systems, when located within 5 nm of the plasmonic surface, exhibit interfacial resonance excitation energy transfer. In case of an efficient feedback mechanism resonance energy transfer generates plasmons in the noble-metal nanostructure, and the resulting high local electromagnetic fields, in turn, excite the adjacent excitonic systems. Such plexcitonic nanostructures are suited for setting up a nanoscale laser, the so-called SPASER (i.e. surface plasmon amplification by stimulated emission of radiation). In the SPASER the plasmonic noble-metal nanostructure acts as a nanocavity, and incorporated or surface-coupled two-level emitters (i.e. excitonic systems) constitute the nanoscale gain medium. Resonant optical pumping of both, the gain medium and the plasmonic nanocavity, provides LSP stimulated excitation energy transfer between two-level emitters and the plasmonic nanostructure which enables the buildup of a macroscopic number of LSPs in a single mode and therewith, coherent local optical fields which drive the SPASER action.
In this contribution, we demonstrate that room-temperature SPASER emission can be achieved through amplifying longitudinal surface plasmon modes supported in gold nanorods and employing laser dyes or luminescent gold complexes and gold nanocluster that function as optical gain for compensation of plasmon losses. The central wavelength of the longitudinal surface plasmon resonance band was tuned between 650 to 690 nm via the aspect ratio that was adjusted through the synthesis procedure parameters. Suited laser dyes and properly synthesized gold complexes/nanocluster electrostatically attached within a 5 nm-distance at the gold nanorod surface at a high concentration provided an effective optical gain medium which facilitates SPASER emission in the red spectral range. These SPASER nanostructures were characterized using high-resolution transmission electron microscopy, uv/vis absorption spectroscopy, stationary and time resolved fluorescence spectroscopy and femtosecond transient absorption spectroscopy. Figure 1 demonstrates spaser emission at 716 nm observed as narrow-band stimulated emission band superimposed photo-induced bleach due to longitudinal surface plasmon resonance centered at 690 nm.

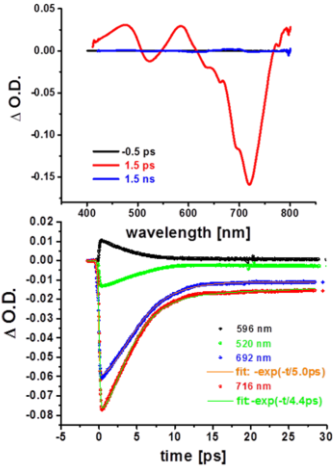

Figure 1: Time-evolution of the fs transient absorption spectra (top) and associated kinetic traces (bottom) of phthalocyanine tetrasulfonate coated gold nanorods upon excitation at 390 nm.

References

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[2] D. J. Bergman, M. I. Stockman, Phys. Rev. Lett. **90** (2003) 027402-1