Optical enhancement effect in metal-organic nanohybrids integrated with whispering-galery-mode microcavities

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Abstract

We report on enhanced optical effects in novel type of hybrid structures that combine high-Q spherical microcavities with Ag nanoparticles and organic dye molecules in a J-aggregate state. A layer-by-layer deposition technique provided controllable coating of the latex microspheres with a shell of close-packed Ag nanoparticles and J-nano-aggregates. Colloidal silver nanoparticles of 30 nm average size were synthesized by the conventional citrate reduction method. The formation of J-aggregates was promoted by electrostatic interaction between positively charged dye molecules and negatively charged polystyrenesulfonate, and confirmed using UV-VIS absorbance spectroscopy.

A periodic structure of narrow peaks was observed in the photoluminescence spectrum of the J-nanoaggregates (Fig.1), arising from the coupling between the emission of J-nanoaggregates and the whispering gallery modes (WGMs) of the microcavity, which are which are electromagnetic waves that circulate and are strongly confined within the microcavity [1,2].

The most striking result of our study is the observation of polarization sensitive mode damping caused by re-absorption of J-nanoaggregate emission (Fig.1). This effect manifests itself in dominating emission from the transverse magnetic (TM) modes in the spectral region of J-nanoaggregates absorption band where the transverse electric (TE) modes are strongly suppressed. In contrast, the TE modes totally dominate emission spectrum in the region where absorption is negligible. Polarization sensitive mode damping observed in the spectral region of high J-aggregate absorption can be used for suppression of unwanted modes in high Q optical whispering gallery resonators.

Our experiments also revealed that WGMs alone can be responsible for enhancement of optical responses from J-nanoaggregates. At the same time, coherent coupling between localized plasmons of the metalic nanoparticles and the exciton in molecular aggregates results in strongly plasmon-enhanced Raman signal. SEM analysis of surface of microcavity integrated with Ag nanoparticles revealed presence of fractal-like metallic aggregates which serve as so called “hot spots“. These spots are nanometer-scale spatial regions of high local electric field, and cause not only significant enhancement of Raman scattering, but also strong PL inhancement and shortening in PL lifetime of J-nanoaggregates observed in our lifetime-imaging experiments.

Owing to the concerted action of “photonic hot spots” (the locally enhanced electric fields due to WGM resonances in microcavities) and “plasmonic hot spots” in the Ag aggregates, we observe strongly increased photoluminescence intensity, intensified spontaneous emission rate and enhanced Raman scattering from the J-nanoaggregates (Fig.1). Coupling of the plasmonic fields supported by metal nanoparticles and excitonic states of J-aggregates to microcavity local fields might be employed to manipulate the density and quality of modes and to control spontaneous emission rate in coupled hybrid system.

References

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[2] Dzmitry Melnikau, Diana Savateeva, Andrey Chuvilin,Rainer Hillenbrand and Yury P. Rakovich Optics Express, **19** (2011) 22280.

Figures

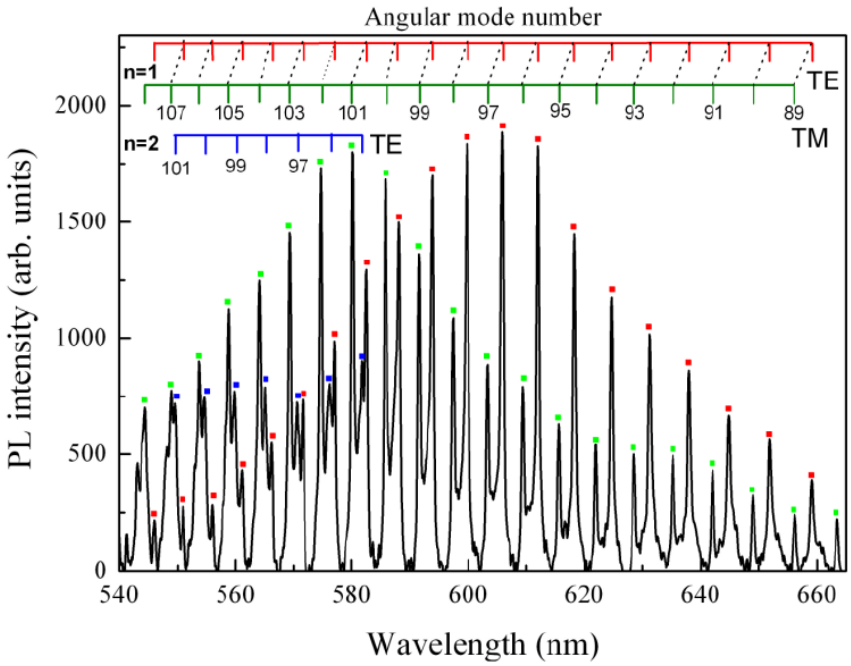

Figure 1. Room-temperature PL spectrum from single MF microsphere covered by monolayer of J-nanoaggregates. Inserts show the results of mode identification using Mie scattering theory. Green and red squares indicate TM and TE modes of first order, respectively. Blue squares indicate TE modes of second order.

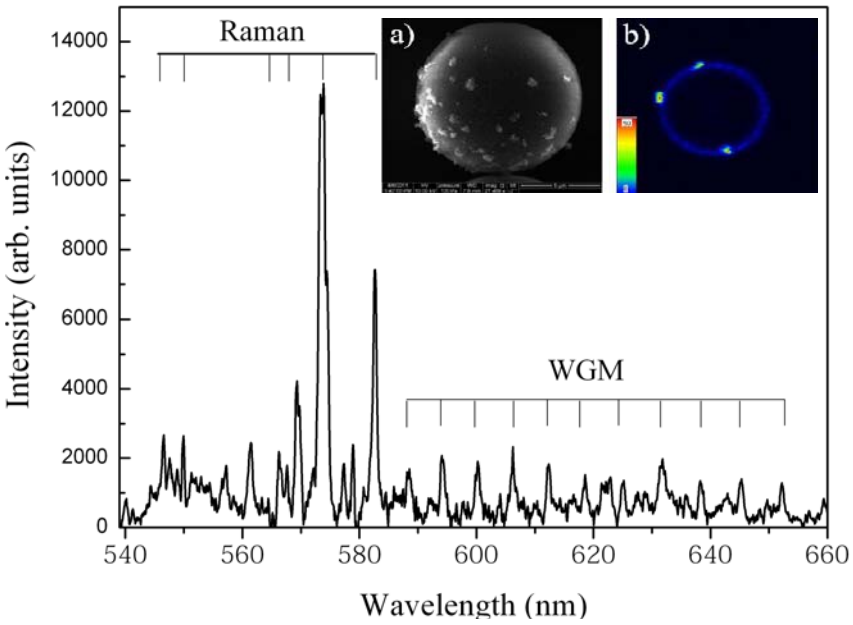

Figure 2. Micro-photoluminescence spectrum of single WGM microcavity with hybrid shell. Insert shows SEM image (a) and confocal photoluminescence image of microcavity/shell structure.