Strong Enhancement of Two-Photon Absorption and Emergence of Unusual Extinction Saturation in Silver Sulfide Quantum Dots Integrated with Gold and Silica Nanostructures

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Streszczenie

Hybrid nanosystems, such as those combining plasmonic, dielectric, and quantum-confined nanostructures, have long been of interest for enhancing and tailoring diverse light–matter interactions. Here, we present a series of hybrid nanomaterials exhibiting strongly enhanced nonlinear optical (NLO) properties, fabricated by combining silver sulfide quantum dots (Ag_2S QDs) with silica and gold nanostructures. We studied their NLO properties (two-photon absorption and saturable absorption) in colloidal solutions over a wide spectral range (500–1600 nm) using the femtosecond Z-scan technique. Embedding Ag_2S QDs into silica nanospheres gives rise to remarkable enhancement of two-photon absorption (up to a factor of 16 increase in the merit factor σ_2/M compared to bare QDs), whereas covering such QD-doped silica nanospheres with gold nanoparticles or attaching the QDs to the surface of gold nanoshells (NSs) leads to even further enhancement (up to 73-fold increase in σ_2/M), accompanied by a competing effect of saturable absorption. Furthermore, in the case of QD-doped silica spheres covered with a continuous gold layer, we observe a previously unreported saturation of extinction in the near-infrared region that follows an unusual intensity dependence, suggesting the involvement of two-photon absorption as the pumping mechanism. In addition to the experimental studies, we have performed numerical simulations, revealing the plasmonic origin of the observed spectral dependences of the NLO properties, with the underlying enhancement mechanisms involving local field enhancement and, possibly, also coupling between plasmon modes and QD excitons, giving rise to a double peak in the σ_2 spectrum. Our findings demonstrate the unique potential of hybrid NLO nanomaterials combining quantum-confined, plasmonic, and dielectric components.

Słowa kluczowe

Z-scan, nanomaterials, quantum dots, silver sulfide, Ag2S, nanoshells, plasmon resonance, gold nanoparticles, two-photon absorption, saturable absorption

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