Excitonic Metamaterials for Enhanced Dark-Field Scattering: Comparative Analysis of Dielectric Nanoparticle Imaging on Tailored Substrates

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Organic molecular aggregates, particularly squaraine-based dyes, have garnered attention for their strong excitonic interactions and potential in optical applications. In this study, we explore the use of an excitonic film composed of 2,4-bis[8-hydroxy-1,1,7,7-tetramethyljulolidin-9-yl]squaraine (HTJSq), which exhibits both H- and J-aggregate characteristics, as a functional substrate to enhance light scattering in dark-field microscopy. Such molecular aggregation leads to unique optical modes, including surface exciton polariton (SEP) and epsilon-near-zero (ENZ) modes, which can localize and amplify electromagnetic fields near the film surface. [1,2]

To assess the efficacy of this approach, we fabricated two distinct substrates— 2,4-bis[4-(diethylamino)-2-hydroxyphenyl]squaraine (Sq1) and 2,4-bis[8-hydroxy-1,1,7,7-tetramethyljulolidin-9-yl]squaraine (Sq2)—each differing in film morphology and aggregation states due to variations in fabrication conditions. Dielectric nanobeads were deposited onto each substrate, and their scattering intensities were measured using dark-field microscopy. Our comparative analysis revealed that the degree of scattering enhancement varied significantly among the substrates, correlating with the specific excitonic properties induced by the differing aggregation states. Notably, substrates exhibiting a balanced hybridization of H- and J-aggregates demonstrated superior field enhancement, leading to markedly improved imaging contrast for nanoparticles that are otherwise challenging to detect on conventional glass substrates.

This study underscores the potential of tailoring organic molecular aggregate films to modulate optical properties for enhanced label-free imaging. The insights gained from the comparative analysis of Sq1 and Sq2 substrates provide a pathway for optimizing excitonic films in advanced optical microscopy applications.

References

1. Park, D. H. et al. Organic Exciton Polariton Structured Illumination Microscopy. *ACS Photonics* **12**, 981–988 (2025).
2. Choi, K. et al. Photoluminescence lifetime engineering via organic resonant films with molecular aggregates. *Nanophotonics* **13**, 1033–1037 (2024).