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Field-level characterization of strong coupling between excitons and surface plasmon polaritons in J-aggregate/metal hybrid nanostructures

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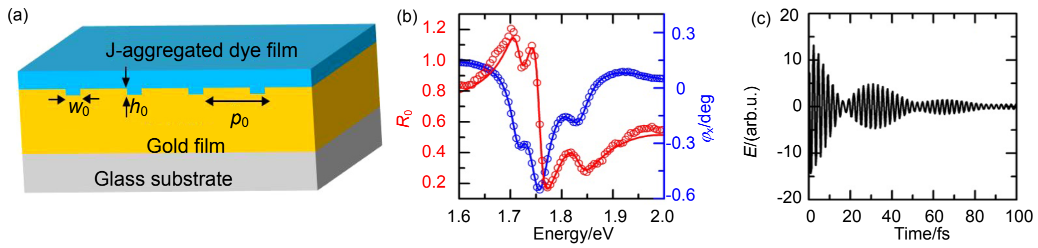

(a) Scheme of the hybrid nanostructure consisting of a gold nanogroove array. (b) Experimental (circles) and simulated (solid lines) reflectivity spectra (blue) and spectral phases (red). (c) Time structure of the electric field emitted by the hybrid polariton modes.

Abstract: Metallic nanostructures have highly interesting optical properties. When illuminating light on them, surface plasmon polaritons (SPP) can be induced due to the coupling of the electromagnetic fields to collective charge density oscillations near the metal surface. SPPs have recently been used in a variety of new applications due to their abilities to guide light on the scale of nanometer. However, most of these emerging applications are limited by the ultrashort lifetime of SPP and the corresponding short propagation length caused by the strong ohmic loss of metal and radiative damping within the nanostructures. Moreover, SPP is generally a photon-like optical excitation showing intrinsically weak nonlinearities, which hinders active nanoplasmonic device fabrication, such as all-optical switching or information processing.

A promising way to compensate losses and provide missing nonlinearity of SPPs is to couple SPPs to nonlinear optical resonances, such as excitons (Xs) in molecular or semiconducting nanostructures. Consequently, hybrid nanostructures containing J-aggregate molecules and metallic nanostructures have attracted considerable interest. Strong coupling between Xs and SPPs enables an efficient transfer of the strong optical nonlinearities of the excitonic emitters to the passive plasmonic nanostructures on the ultrashort time scale of femtosecond.

Here, we demonstrate a field-level characterization of the optical response of J-aggregate/metal hybrid nanostructures by white-light broadband chirp-compensated spectral interferometry. We show that both the amplitude and spectral phase of the strongly coupled X-SPP system can be measured with high precision by compensating the chirp in both arms of the interferometer. A quantitative description of both the excitonic resonance and the hybrid X-SPP polariton response is obtained by fitting the measured amplitudes and spectral phases simultaneously to a Fano lineshape model. We find that the resonance of the majority of J-aggregated molecules which are not coupled to SPPs is homogeneously broadened. We also demonstrate accurate reconstruction of the time structure of the electric field emitted by the hybrid nanostructures, corresponding to polarization oscillations with short damping time shorter than 100 fs.

Keywords: surface plasmon polaritons; spectral interferometry; subwavelength nanostructures; strong coupling

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