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Near-field Mediated Plexcitonic Coupling and Giant Rabi Splitting in Individual Metallic Dimers[1] ANDREA SCHLATHER, NICOLAS LARGE, ALEXANDER URBAN, PETER NORDLANDER, NAOMI HALAS, Rice University — We investigate hybrid metallic dimer – J-aggregate nanostructures that show coherent coupling between the localized surface plasmon (LSP) of the metallic disks and the exciton of the J-aggregate molecular complex. This hybrid nanostructure, combining both bottom-up and top-down approaches, is designed to probe the limitations of coherent coupling detection. Indeed, this allows us to report, for the first time, an experimental investigation of a plexcitonic coupling mechanism at the single-particle regime. By varying the diameter of the nanodisks, the LSP energies of the dimers can be systematically modified and tuned across the exciton energy of the J-aggregate. This allows for the direct measurement of the plexcitonic coupling energy of the dimer – J-aggregate hybrid nanostructures. In this work, using single particle dark-field scattering spectroscopy as well as Finite-Difference Time-Domain (FDTD) calculations, we report giant Rabi splitting energies up to 400 meV resulting in a plexcitonic-induced transparency window observed both experimentally and theoretically at the overlap energy of the individual excitations. Furthermore, through a rigorous study of the polarization dependence and of the tunable geometric parameter effects (gap, diameter), the plexcitonic coupling mechanism has been investigated in these hybrid nanostructures, leading to the determination of the crucial role played by the plasmonic hot spots in the formation of the hybrid plexcitonic modes.

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