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Manipulating Energy Flow at the Nanoscale by Coupling Plasmons of Metal Nanostructures to Resonant Molecules

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Collective hybrid excitations resulting from the coupling of metal nanostructures with organic molecules present unique opportunities for manipulating light-matter interactions at the nanoscale. In this talk, I discuss recent studies that are examples of the breadth of phenomena that are possible. First, the interactions of coupled plasmonic nanostructures with azobenzene-based polymers are described, in which the spatial features of the plasmonic near-field can be used to manipulate molecular motion. The directional molecular transport that results is shown to be useful for imaging the spatial and polarization features of the optical near-field. The modeling of this effect is described. Second, the coupling of excitonic molecular aggregates to metal nanostructures produces coherent coupling that provides added structure to the optical extinction spectra of metal nanoparticles, thereby by providing a photonic handle with which to manipulate energy flow on an ultrafast timescale. Monitoring the rate of energy flow as a function of photon energy reveals important information about the energy dissipation channels and the structural interactions between molecule and metal. Third, the strongly enhanced optical nonlinearity resulting from coupled plasmonic nanorods is described. The closely spaced nanorod material exhibits nonlocality of the optical response that has an unusually strong nonlinear dependence on incident light intensity. Electromagnetic modeling confirms the nonlocal response of the plasmonic metamaterial. The broader impact of collective hybrid excitations on nanophotonics applications is described. Use of the Center for Nanoscale Materials was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Contract No. DE-AC02-06CH11357.