Coexistence of classical and quantum plasmonics in large plasmonic structures with subnanometer gaps

Large metal nanostructures with subnanometer interparticle separations (gaps) can provide extremely high local fields and are of particular interest in surface enhanced spectroscopy, as well as for basic understanding of plasmonics. In this experimental electron energy loss study, we monitor the transition of plasmonic dimers from a classical to a quantum system by decreasing gaps to dimensions when tunneling occurs and a conductive nanobridge evolves. Our studies show that silver dimers with atomic scale gaps can exhibit a regime, in which charge transfer plasmon modes, as a hallmark of a quantum nature, and "classical" bright and dark dipolar plasmon modes can be seen simultaneously.