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# Impact of the Nanoscale Gap Morphology on Plasmons in Doped Indium Oxide Nanostructure Dimers

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Doped indium oxide (In2O3) nanoparticles and thin films exhibit a unique combination of high electrical conductivity and optical transparency in the visible spectral range, which makes them attractive for applications in optoelectronics and light harvesting [1]. Further functionalities can be achieved by exploiting highly confined and tunable plasmons sustained by the In2O3 nanostructures in the infrared. We focus on plasmons in fluorine-doped In2O3 nanocubes [2] separated by nanometric gaps triggering electromagnetic coupling between the nanoparticles and thus producing hybridization of the plasmonic modes. We theoretically predict a non-singular transition, in which plasmon modes in separated dimers evolve in a continuous way to those of touching dimers. Our theoretical analysis is confirmed by experiments using electron energy-loss spectroscopy (EELS) in a scanning transmission electron microscope (STEM). Compared to traditional far-field spectroscopic techniques, STEM-EELS allows us to probe a complete set of plasmonic modes at the nanoscale, therefore enabling us to determine the precise correlation of the gap geometry with the spatial and spectral characteristics of the coupled plasmons, which is important for the detailed understanding of the studied system.

By further examining different types of geometries near touching conditions, as shown in Figure 1, we find that the presence of a finite two-dimensional touching area makes the transition non-singular, in agreement with our observations. On the contrary, a one-dimensional or zero-dimensional touching region produces a singular behavior characterized by the emergence of a dipole mode with a net charge in each of the dimer particles that becomes unphysical when they are separated.

Our results combining theory and experiment elucidate the influence of the gap geometry on the coupled plasmons and field concentration in doped In2O3 nanostructures, and suggest exciting applications in plasmonic sensing or surface-enhanced spectroscopies [3].

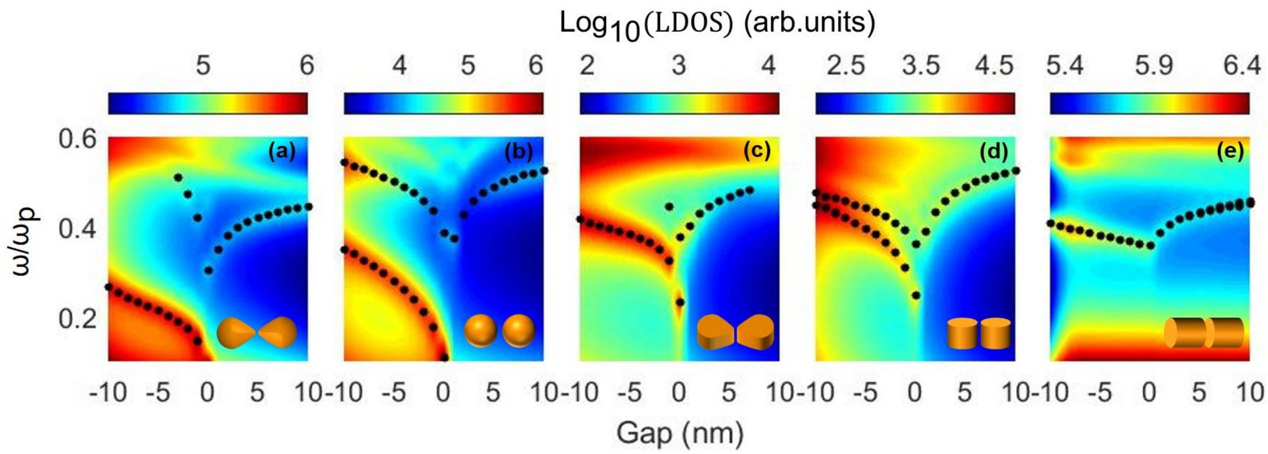


**Figure 1.** Local density of optical states in logarithmic color scale for nanoparticle dimers as a function of the gap size and photon energy. We consider (a, b) zero-, (c, d) one-, and (e) two-dimensionality of gap regions.

References:

[1] O. Bierwagen, Sci. Technol. **30**, 7 (2015), p. 024001. doi: 10.1088/0268-1242/30/2/024001

[2] S. H. Cho, et al. Chem. Mater. **31**, 7 (2019), p. 2661--2676. doi: 10.1021/acs.chemmater.9b00906

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