Controlling the optical properties of ultranarrow plasmonic nanoparticle-on-mirror cavities

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Abstract

Plasmonic nanoparticles placed on a metallic substrate, separated by a dielectric layer that prevents conductive contact, offer an attractive option to produce ultranarrow nanogaps, owing to their relatively easy fabrication and reproducibility. When such structures are illuminated by a plane wave polarised normally to the substrate, the nanoparticle couples to its mirror image in the metallic film, and longitudinal plasmon modes equivalent to Bonding Dimer Plasmons (BDP) in dimer configurations can be excited. The electromagnetic field is strongly enhanced and localised within the dielectric spacer, forming narrow plasmonic nanocavities.[1] The thickness of the spacer is therefore crucial to control the optical response of the structure. Recent advances in fabrication of two-dimensional semicondutors have opened new pathways for the design of ultranarrow plasmonic cavities. Here we discuss several examples of such nanocomposites, and show how their optical response can be tailored by the appropriate choice of the spacer material or by engineering the nanoparticles. First we show that graphene layers can be exploited as spacers with a controllable, discrete thickness. Reducing the number of graphene layers of the spacer increases the interaction between the nanoparticle and the metallic substrate, leading to the excitation of higher-order BDP modes.[2] Additional control can be provided by laser irradiation of the system, which generates flat facets at the bottom of the nanoparticles and enlarges the width of the plasmonic cavities. For small-area facets the nanoparticle-film interaction is strongly amplified, leading to corresponding redshifts of the BDP modes.[3] As the facets become wider, additional, transverse modes, similar to those observed in optical patch antennas, can also be excited. The dependence of these modes on facet width and their interaction with the BDP modes provides a rich chart of modal interplay. Finally, self-assembled monolayers of organic molecules can be used as spacers to control the wavelength of the BDP modes. By mixing conductive and non-conductive molecules it is possible to create conductive junctions which allow charge transfer between the nanoparticle and the metallic film, leading to the screening and blueshift of the main BDP mode. This shift of the modes can be further controlled by the fraction of conductive molecules comprising the spacer.[4] The ability to manipulate light-matter interactions in narrow plasmonic gaps, as shown here, opens the path to probe complex optoelectronic processes and access atomic-scale photochemistry.

References

[1] J.J. Mock, R.T. Hill, A. Degiron, S. Zauscher, A. Chilkoti, and D.R. Smith, Nano Lett., **8** (2008) 2245.
[2] J. Mertens, A.L. Eiden, D.O Sigle, F. Huang, A. Lombardo, Z. Sun, R.S. Sundaram, A. Colli, C. Tserkezis, J. Aizpurua, S. Milana, A.C. Ferrari, and J.J. Baumberg, Nano Lett., **13** (2013) 5033.
[3] D.O. Sigle, J. Mertens, L.O. Herrmann, R.W. Bowman, S. Ithurria, B. Dubertret, Y. Shi, H.Y. Yang, C. Tserkezis, J. Aizpurua, and J.J. Baumberg, *submitted*.
[4] F. Benz, C. Tserkezis, L.O. Herrmann, B. de Nijs, A. Sanders, D.O. Sigle, L. Pukenas, S.D. Evans, J. Aizpurua, and J.J. Baumberg, *submitted*.

Figures

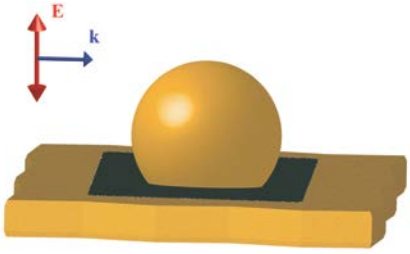

Figure 1. Schematic representation of a faceted gold nanoparticle on a gold film, separated by a semiconductor spacer. The structure is illuminated by a plane wave polarised normally to the substrate.