## Multimodal plasmonics in crystalline colloidal systems

Sviatlana Viarbitskaya<sup>1,2</sup>, Alexandre Teulle<sup>1</sup>, Upkar Kumar<sup>1</sup>, Jadab Sharma<sup>1</sup>, Kargal Gurunatha<sup>1</sup>, Aurélien Cuche<sup>1</sup>, Michel Bosman<sup>3</sup>, Alexandre Bouhelier<sup>2</sup>, Gérard Colas des Francs<sup>2</sup>, Arnaud Arbouet<sup>1</sup>, Christian Girard<sup>1</sup>, <u>Erik DUJARDIN</u><sup>1</sup>\*

 <sup>1</sup> CEMES CNRS UPR 8011 and Université Fédérale de Toulouse, 29 rue J. Marvig, 31055 Toulouse, France. Email: dujardin@cemes.fr
<sup>2</sup> Laboratoire Interdisciplinaire Carnot de Bourgogne, CNRS UMR 6303, Université de Bourgogne, 9 Avenue Alain Savary, Dijon, France.

<sup>3</sup> Institute of Materials Research and Engineering, A\*STAR (Agency for Science, Technology and Research), 3 Research Link, 117602 Singapore, Singapore.

Plasmonics has opened ways to tailor optical properties both at the macroscopic scale by allowing propagation, waveguiding and routing of plasmon polaritons but also at the nanometer-scale by taking advantage of the evanescent fields, strong confinement volumes and localized plasmonic resonances. While both regimes have been extensively studied and led to numerous applications, much less scrutiny has so far focused on the intermediate regime of micrometer-sized systems supporting a large number of confined higher order surface plasmon (SP) modes. Multimodal plasmonic systems open a new realm in which the modal behavior is better described by the SP local density of states (SP-LDOS), which is solely governed by the material properties and the boundary conditions set by the structure shape but is independent of the illumination parameters. The SP-LDOS can therefore be rationally designed to tailor the local spatial and spectral characteristics of the SP modes, while allowing information transfer over micrometer-sized distances. To reveal and exploit such spatio-modal engineering of plasmons, dissipation must be reduced by exploiting the enhanced performances of crystalline metal colloids.[1]



Figure 1: (a,b) SEM images of triangular and truncated triangular Au nanoprisms. Scale bars are 200 nm. (c, d) Confocal TPL images recorded with horizontally polarized, 700 nm excitation. (e, f) Corresponding simulated TPL images using the GDM method. [2,3] (g, h) Total SP-LDOS maps and (i, j) corresponding AFM images of prisms with modified surface induced by plasmonic hot printing. [7]

We will first present strategies to chemically tailor the plasmonic properties of anisotropic 1D and 2D plasmonic microstructures composed of either single crystalline Au colloids [2, 3] or self-assembled superstructures [4, 5] sustaining higher order plasmonic modes. We will then demonstrate that the SP-LDOS distribution of mesoscale 2D structure can be conveniently imaged by all optical technique such as two-photon luminescence (TPL) microscopy. [2, 3, 5] The influence of wavelength, excitation polarization, particle shape and interparticle coupling on the spatial and spectral characteristic of the SP-LDOS are explored

experimentally and fully confirmed by our new simulation tools based on the Green Dyadic Method (GDM). From the multimodal behavior of individual 2D colloids, we will derive a new approach of optical information processing by engineering the spatial and/or spectral distributions of higher order modes. Two routes will be presented: the near-field coupling between colloidal building blocks and the physical reshaping by focused ion beam. Our approach is applied to information propagation,[6] modal logic gates [2] and localized hot electron generation.[7]

Finally, electron probes, such as electron energy loss spectroscopy (EELS), will provide the required higher spatial resolution to investigate 10-nm wide crystalline plasmonic waveguides obtained by the self-assembly combined with electron-beam induced nanometerscale fusion.[8] The nanoparticle chain networks are thus converted into ultimately narrow plasmonic waveguides. Indeed, extremely low energy modes were identified in the 0.35-0.4 eV range of the EELS spectra that corresponds to SP modes delocalized over multimicrometer distances in spite of the 10-nm lateral confinement.

Perspectives on multimodal plasmonics in colloidal systems will conclude the presentation.



Figure 2: (a) TEM image of a branched and looped Au nanoparticle chain after in situ electron-beam-induced fusion. Scale bar is 50 nm. (b) EELS spectra recorded in positions I to IV shown in (a). (c) EELS map recorded from the resonance features at 0.79 eV with a 0.1 eV pass bandwidth.[8]

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