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Single-molecule imaging of LDOS modification by an array of plasmonic nanochimneys

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The direct measurement of a single emitter decay rate and the simultaneous knowledge of their position is a powerful tool for the study of light-matter interaction at the nanometer scale. In particular, the decay rate is directly related to the local density of states (LDOS) which measures the number of modes of the electromagnetic environment available for the decay of an emitter.

In this work we study the modification of fluorescence emission and decay rate of single fluorescent molecules in the near field of a periodic plasmonic nanostructure formed by a square lattice of Au nanochimneys (hollow conical pillars). Such nanostructure can be fabricated on a large area with a wafer-scale ($15cm^2$) fabrication method by sputter redeposition on a hybrid mask [1]. It behaves as a metamaterial in the near-IR and supports rich optical properties with tunable dipolar and quadrupolar plasmonic resonances in the visible spectral range with angular illumination dependence.

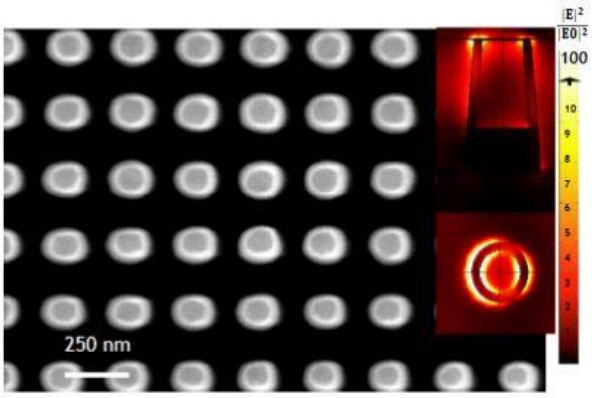

Fig. 1 Scanning electron microscopy image of the nanochimneys array and calculated field enhancement (Finite Element Method) for a Au nanochimneys square array with unitary cell as described at a 620 nm wavelength under the illumination with p-polarised plane wave at a 45°angle..

We perform nanometer-resolved imaging of the LDOS by simultaneously mapping the position and the decay rate of photoactivatable single-molecules with a novel super-resolved microscopy approach [2] which enables multiplexed and super-resolved fluorescence lifetime imaging at the single-molecule level (smFLIM) with a field of view of ~10 μm^2. Our method combines the use of an EMCCD camera for spatial measurements and a linear array of single-photon avalanche diodes (SPADs) for time-resolved measurements [3]. We observe the field enhancement and the LDOS modification of such optically rich material at different illumination conditions, for TM and TE polarization at different angles of illumination, and we measure a large Purcell factor enhancement which increases for oblique illumination of the nanostructure.

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