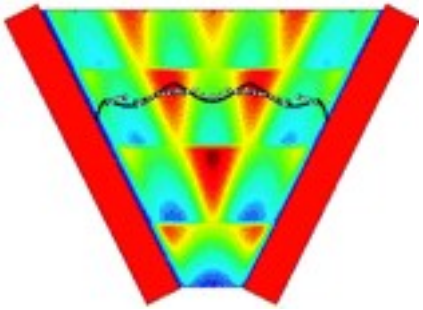
Unique single molecule probing of cavity mode density by ICFO in Nanoletters.



Spatial single molecule
lifetime variation inside a
tapered metal nanocavity.Cavities are found everywhere to control the emission of light: in lasers, parametric oscillators and many quantum-optics experiments. In nanocavities, due to proximity of the cavity edges, the near field becomes important. Now ICFO researchers Jacob Hoogenboom, Gabriel Sanchez-Mosteiro and Dominique Heinis, all members of Prof. Niek van Hulst's group, have looked inside a basic nanocavity. In doing so, they probed simultaneously the position, orientation and fluorescence lifetime of single molecules, and by using single molecule superresolution microscopy they managed to actually map with nm-precision both spatial and orientational spontaneous emission distribution inside metal nanocavities on a surface. For dipoles perpendicular to the edge, even up to 150 nm distance, the decay rate is enhanced due to strong coupling to surface plasmon polariton modes. In contrast, for dipoles parallel to the edge, coupling to the cavity modes results in an oscillatory behaviour up to a proximity of about 30 nm. These experiments concur nicely with the vectorial mode density calculations made by Gerard Colas des Francs of the Institut Carnot de Bourgogne in France. So far, a factor 3 in lifetime variation is observed. This first demonstration of single molecule "LDOS mapping" is encouraging for research in photonic crystals, nanoantennas and other plasmonic nanostructures, where much stronger mode density effects take place. Finally, the concept of superresolution single-molecule fluorescence lifetime and polarization imaging has exciting prospects for nanoscopy imaging of biological systems.