PhD Thesis Defense ANSHUMAN SINGH 'Optical Nanoantennas as Cavities: Nanoscale Control of Coupling Strength and Single Photon Emission'

ANSHUMAN SINGH

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Friday, November 11, 2016, 11:00. ICFO Auditorium

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Molecular Nanophotonics

ICFO-The Institute of Photonic Sciences

Optical nanoantennas confine light on the nanoscale, enabling strong light-matter interactions with potential for ultra-compact optical devices. Apart from the direct

applications in optical nanoscopy and sensing, nanoantennas can strongly enhance the spontaneous emission rate of single photon emitters. A nanoantenna, acting as a nanocavity, enables high photon output due to its high radiative losses associated with plasmonic resonances and thus pave the way for compact, bright, and pure single photon sources with applications in quantum technologies. The sub-wavelength field localization at the nanoantenna causes a vectorial field distribution with non-zero field components in all dimensions. The efficiency of coupling between an emitter such as single molecule and nanoantenna will thus strongly depend on the spatial overlap of the emitter's dipole with the nanoantenna field. In order to achieve such coupling, careful nanopositioning of the emitter, in its location as well as the orientation, within the nanocavity field is required. Therefore, the full vectorial characterization of the emitter-nanoantenna is crucial to maximize the coupling strength. This thesis addresses the aforementioned issues and studies the controlled nanoscale interaction of a single molecule with a resonant nanoantenna.

To this end we first fabricate nanoantennas on the vertex of a fiber tip using focused-ion-beam milling and use a near-field technique for the nanopositioning control. In the first experiment, we investigate the excitation properties of a resonant dipole nanoantenna by mapping its vectorial near-field distribution with molecular resolution. The nanoantenna tip is scanned over specifically selected single molecules to map x-, y-, and z-field components. In addition to characterization of the vectorial field, we show the apparent position of the molecule shifts up to 20 nm depending on their orientation with important implications for localization microscopy.

Next, our unprecedented experimental approach allows us to examine an often overlooked, but important near-field concept of nanoantennas, local interference. The highly structured vectorial amplitude and phase distribution of the nanoantenna overlaps with the exciting far-field to create this local interference. We perform a detailed study through direct observation and show an example of exploiting this local interference to shape and control the near-field of the nanoantenna.

Lastly, we quantify the vectorial interaction of a molecule-nanoantenna cavity system by mapping the coupling strength g with 5 nm spatial resolution. We show that for a molecule's optimal position at the nanoantenna, the coupling rate reaches up to g_max= 206 GHz, much higher than for any conventional cavities. Such a large coupling provides ideal conditions for fast and pure non-classical photon emission, enabling a single photon source with an emission rate above 1 GHz at room-temperature.

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