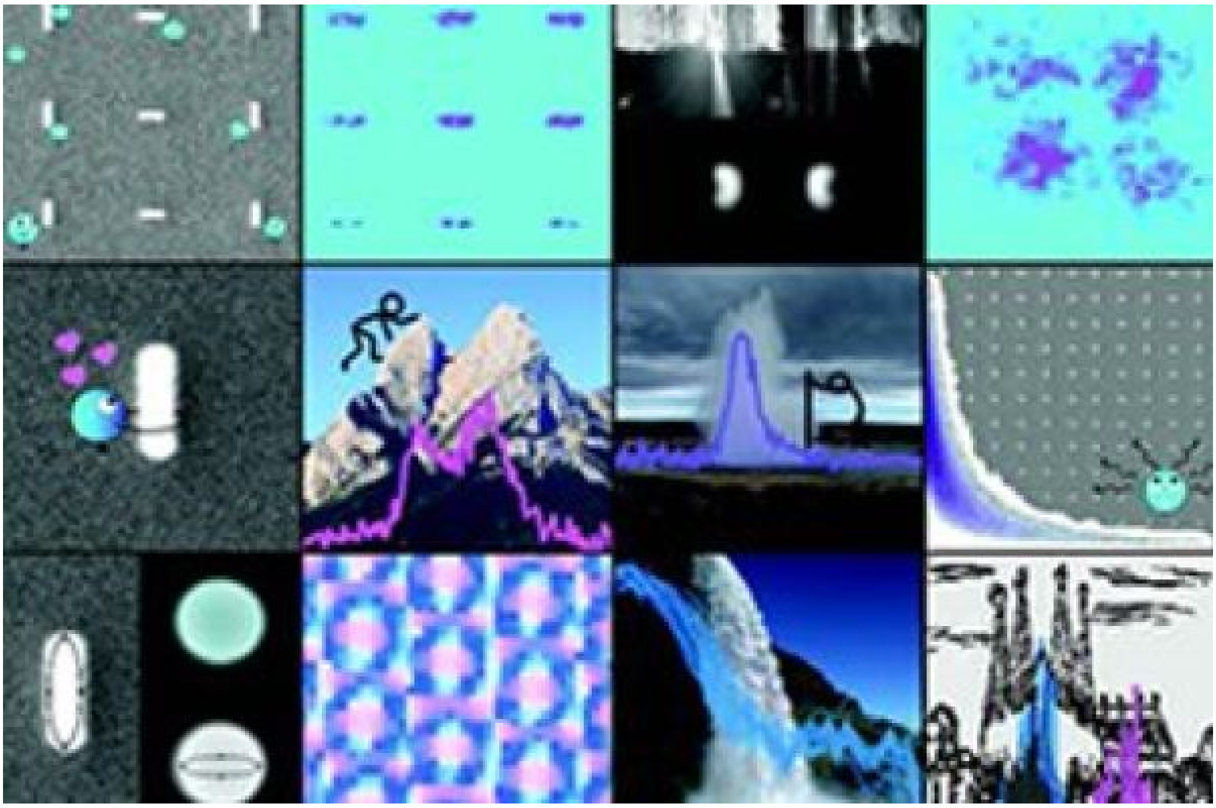


THESIS DEFENSE: Large-scale imaging of optical antennas and single molecules

LISA SAEMISCH

April 15, 2021

15:00

ICFO Auditorium and Online (Teams)

The interaction of light and matter is of crucial importance in fundamental science as well as in high-end technology.

Ultimately, this concerns the interaction between a photon and a single quantum system, e.g. the absorption or emission of a photon by a single molecule. At room temperature this interaction is very inefficient as the absorption cross-section of a molecule is small compared to the wavelength of light, which inhibits many photons from interacting and hence limits the absorption, emission and scattering of a photon. An equivalent problem, and its solution, is found in our daily lives: small electric circuits (as found e.g. in our smartphones), which radiate very poorly by themselves, are linked to (radio) antennas to radiate and transfer information efficiently. Analogously, antennas working in the visible, so-called nanoantennas,

are an effective tool to link matter and light. The strength of the coupling of a single molecule with a nanoantenna depends on many factors: the overlap of the antenna resonance and the molecular absorption/emission spectrum, the molecule's dipole orientation, the distance between molecule and nanoantenna, etc. Hence, strong interaction needs rather special conditions, which are hard to engineer. Moreover, to get a full interaction picture, a lot of single molecule encounters with different nanoantennas are needed - on one hand to make a statistically relevant statement including the many different factors and, on the other hand, to be able to observe the rare stronger interactions, that would have stayed hidden in experiments of only a few encounters. The central idea of this thesis is to statistically map and control the interactions of a very large number of single molecules
with different tailored nanoantennas, to cover the landscape of interaction factors and thus extend the current knowledge of the mutual interaction. For this purpose, a home-built wide-field microscope is combined with a large array of lithographically fabricated nanoantennas, which are all probed by freely diffusing molecules. Thus in time millions of encounters are recorded in parallel.

Chapter 2 introduces the necessary knowledge and methodology to understand the research work presented in chapters 3 to 5. Chapter 3 shows super-resolved nanoscale interaction maps of molecules and nanoantennas, linking the strength of interaction to the emission polarization and intensity of every encounter. Chapter 4 extends this approach by simultaneously recording the emission fluorescence and spectrum of every single molecule event, revealing strong spectral manipulation.

Here, a suppression as well as an extreme enhancement of the vibrational sideband of the used molecule is observed.

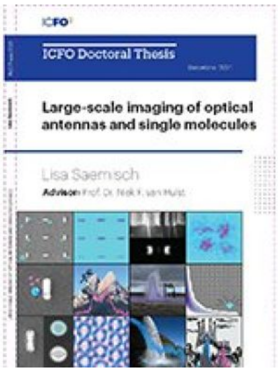
Additionally, the statistical mapping allows the freely diffusing molecules to encounter rare hotspots of extreme field intensities, enabling the observation of surface-enhanced Raman scattering.

Finally, chapter 5 takes the first step in the direction of characterizing the interaction of molecule and nanoantenna with high sensitivity via phase measurements. Here, an interferometric wide-field microscope enables the measurement of the absolute phase of nanoparticles and demonstrates the distinction of different plasmonic and dielectric particles via their phase behavior. Furthermore, we implement a novel two-color excitation method, capable of rapidly identifying two types of nanoparticles in a single-shot image.

Due to recommendations in place to contribute containing the spreading of COVID-19, th defence will be carried out semi presencial with a maximum of 66 Icfonians in t e Auditorium, and partly remotely via MS Teams. This is the link to follow the Thesis Defe se online Click here to join the meeting.?

If you are interested in attending in person, please address your request to mery.gil@icfo.eu **by Tuesday April 13th .**

Hosted by: Prof Dr Niek F. van Hulst



Lisa Saemisch's Thesi Cover