

Nanoparticles

NICOLO ACCANTO

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PhD Thesis Defense NICOLO ACCANTO 'Coherent Control of Nonlinear Optical Processes in Individual Nanoparticles'

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Friday, June 17, 11:00. ICFO Auditorium

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Molecular Nanophotonics

ICFO-The Institute of Photonic Sciences

Nanophotonics aims to understand and control the interaction of light-matter at the nanometer scale. Boosted by the development of nanotechnology and nanofabrication, nanophotonics is a thriving research field, with applications in areas as diverse as optical communications, biological imaging, super-resolution microscopy and photovoltaics. For the

design of specific structures and the development of new applications, it is vital to study and understand the basic ultrafast dynamical interactions of light with individual nanoparticles (NPs).

The practical realization of experiments in which single nanoparticles are addressed with temporal resolution on the femtosecond scale is very demanding. The first goal of this thesis is to develop a robust and easy to operate experimental scheme, based on the combination of ultrashort phase-controlled laser pulses and high-resolution optical microscopes, which allows the desired ultrashort laser pulses to be delivered to localized nanometric volumes.

Not only do we need to observe, but also to manipulate ultrafast light-matter interactions at the nanoscale by using precisely tailored laser fields, a concept that is often referred to as coherent control. The main goal of the thesis is to extend coherent control concepts, so far mainly applied to ensembles of systems, to the manipulation of ultrafast light-matter interactions in individual nanoparticles. Experimentally, this is realized by using ultrashort laser pulses, whose spectral phase can be precisely controlled, and measuring nonlinear optical responses from individual nanoparticles.

The NPs investigated in this thesis can be divided into two categories: coherent and incoherent NPs. Coherent NPs present an intrinsic resonant response, in amplitude and phase, which can be resolved and controlled by the laser field. Resonant plasmonic nanoantennas constitute the main example of coherent NPs studied in the thesis. Incoherent NPs can be either non-resonant, or characterized by a very broad (much broader than the laser spectrum) resonance. Dielectric nonlinear NPs and semiconductor quantum dots (QDs) with broad absorptions are the incoherent NPs addressed here. Incoherent NPs are of crucial importance in this thesis. Based on the second harmonic generation (SHG) from incoherent dielectric NPs, a novel method to obtain full control of ultrashort pulses on a sub-diffraction-limited area is demonstrated. Successively, by using single semiconductor QDs and a novel optimization algorithm, a closed loop coherent control scheme capable of addressing single molecules and quantum emitters is realized and tested.

Nonlinear interactions in plasmonic nanoantennas, depend both on the laser field and on their resonant response. Two different nonlinear responses, excited by ultrashort phase-controlled pulses, are studied: the SHG and the two-photon absorption (TPA). By studying the SHG, a precise measurement of the plasmon resonance in nanoantennas is performed, retrieving both spectral phase and amplitude. By using phase control, the possibility of using plasmonic nanoantennas for an improved imaging technique based on multicolor second harmonic labels is discussed and demonstrated. By investigating the TPA process, the presence of a coherent regime, only accessible by ultrashort pulses, is

demonstrated. Finally the realization of a highly sensitive closed loop coherent control experiment on single nanoantennas is presented.

The combined progress in nanotechnology and the ability discussed in this thesis to manipulate ultrafast nanoscale dynamics using light can lead the way towards new fascinating routes in the field of nanophotonics. The switching of light from one side to another of an asymmetric nanostructure, the selective investigation of different NPs at different times, the realization of super-resolution optical microscopy with femtosecond temporal resolution, are just few of the promising applications.

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