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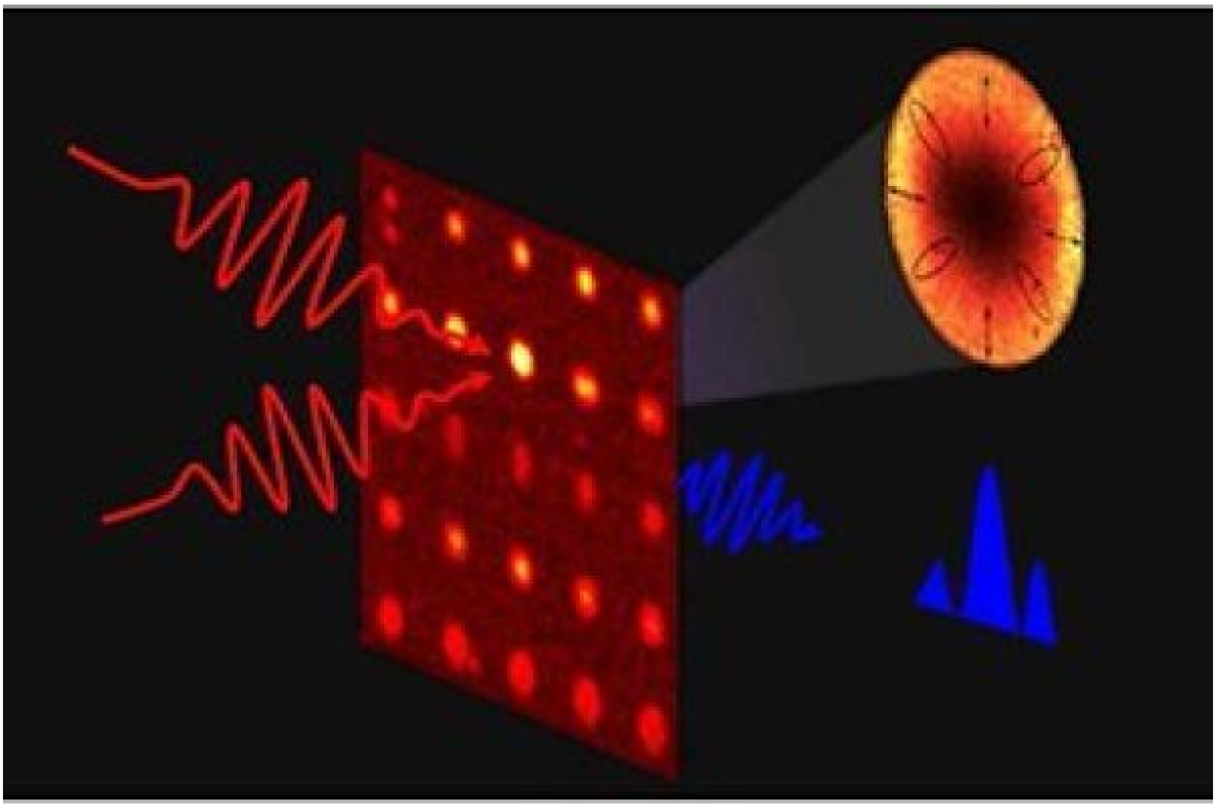

PhD THESIS DEFENSE: Spectral Phase Control of Nanoscale Nonlinear Optical Responses

VIKAS REMESH

June 08, 2020

15:00

ICFO Auditorium and Online (Teams)

PhD Thesis Defense, June 8, 2020, 15:00. Online

VIKAS REMESH

Molecular Nanophotonics

ICFO-The Institute of Photonic Sciences

The study of light-driven physical processes generated in nanomaterials require shorter (in time, or broader in frequency) laser pulses with techniques to tailor its phase and temporal/spectral characteristics at the nanoscopic interaction volume. Such studies can specifically address the aspect of coherence of excited states in molecular systems or of collective electronic oscillations in plasmonic nanoresonators. Addressing nonlinear coherent phenomena with broadband femtosecondpulses, usually involve achieving constructive or destructive interferences between multiple absorption pathways by spectral phase manipulation. Despite progress on in ultrafast spatiotemporal control of optical processes, a systematic, true optical coherent control on a fundamental dipole plasmon mode remained challenging due to the very short dephasing time of the plasmon oscillation in the 25 fs range. Unfortunately, luminescence is an incoherent process and therefore generally not explored for nanoscale coherent control of the antenna response. Firstly, we demonstrated that, in resonant gold nanoantennas, the two-photon absorption process can be coherent, provided that the excitation pulse duration is shorter than the dephasing time of plasmon mode oscillation. Exploiting this coherent response, we showed the pure spectral phase control of resonant gold nanoantennas, with effective read-out of the two-photon photoluminescence. High-index dielectric nanoantennas, for example, gallium phosphide (GaP) nanoantennas have recently emerged as promising alternatives to plasmonic nanoantennas displaying extremely low losses in the visible range and having high nonlinearities. They also support both multipolar electric and magnetic resonances in both visible and NIR frequency range. Especially, the low losses and large nonlinearities are promising for ultrafast optical switching and truly all-optical control of GaP nanodevices. Here first we used two-and three-photon excitation of GaP nanodisks to probe the size-dependent resonance enhancement of second-harmonic and bandgap emission. Next, we showed, by spectral phase control of broadband pulsed excitation, that GaP nanoantennas outperformed their metal counterparts in supporting nonlinear optical coherences. Next, our numerical studies on the SHG enhancement in GaP nanoantennas indicated that size-dependent SHG enhancement is the result of resonant electric field confinement in the nanoantenna volume. Study of angular emission pattern, combined with polarimetry of resonant nanoantennas revealed that SHG emission resulting from the excitation with linear polarization is predominantly radially polarized. The multipolar modal analysis at SHG frequency indicated the presence of an electric dipole oscillating along the disk axis, along with weak contributions of quadrupoles and octupoles. Finally, size-dependent SHG spectral shift confirmed the existence of multiple resonances in these nanodisks. Exploiting this feature, multipolar electromagnetic modes that resonate with relative phases, following a broadband laser illumination, can be made to interfere and the interference can be controlled by means of spectral phase modulation of the excitation field to achieve directionality. With an antisymmetric p step modulation, a switching in polarization state was observed. Our experiments pave way for all-optical control of directional radiation of nanoantennas by the control of multipolar interference in the nonlinear regime.

Monday, June 8, 2020, 15:00. Online

Thesis Advisor: Prof Dr Niek van Hulst

Hosted by: Prof. Dr. Niek van Hulst

Vikas Remesh's Thesi Cover