[Invited Talk]

Second Harmonic Generation from Plasmonic Nanostructures: Toward New Applications

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Abstract– In this presentation, second harmonic generation (SHG) from plasmonic systems supporting Fano resonances is discussed. Second harmonic generation from silver heptamers is addressed in order to demonstrate that a proper design of their optical properties increases their nonlinear efficiency. It is also shown that Fano resonances permit to control the fundamental near-field distribution over metallic nanostructures. This ability is used to design 3D nonlinear plasmon rulers which are more sensitive than their linear counterparts.

1. Introduction

It is well known that metallic nanoantennas are able to enhance and control light-matter interactions down to the nanoscale. Indeed, optical antennas have the ability to concentrate the electric field inside their nanogap beating the diffraction limit. The enhancement of the electric field enables the observation of nonlinear optical processes. For instance, second harmonic generation (SHG) from metallic nanoantennas, the process thereby two photons at the fundamental frequency are converted into one photon at the second harmonic (SH), has been experimentally reported recently [1]. Nevertheless, SHG is forbidden in centrosymmetric media in the dipolar approximation. For this reason, the SH cross section is predicted to be weak in the case of centrosymmetric nanoantennas despite the high electric field enhancement in the nanogap. On the other hand, the fabrication of regular metallic nanostructures is quite challenging and defects can affect their nonlinear optical response. For practical applications, as nonlinear plasmonics sensing [2], it is important to understand how SHG is modified by shape variation. Futhermore, new strategies must be developed to increase the nonlinear conversion at the nanoscale.

2. Shape Characterization

In this presentation, we will discuss results obtained using a surface integral formulation [3] extended to the case of surface SHG. Our method allows efficient evaluations of the SH near-field and far-field distributions. Calculations were performed for idealized (rectangular arms) and realistic (mesh adapted from a scanning electron microscope image) gold nanoantennas. As previously reported in the case of symmetric antennas, the SH electric field at both sides of the idealized nanogap is found oscillating out of phase indicating a non radiative behavior (SH dark mode). This behaviour is no longer observed considering a realistic gold nanoantenna. Due to the shape asymmetry of the arms, the SH near-field distribution is more complex and the SH cross section increases because of symmetry breaking at the nanoscale. Interestingly, the dissymmetry is also clearly revealed by far-field analysis demonstrating that SHG is a promising tool for sensitive optical characterization of plasmonic nanoantennas [4].

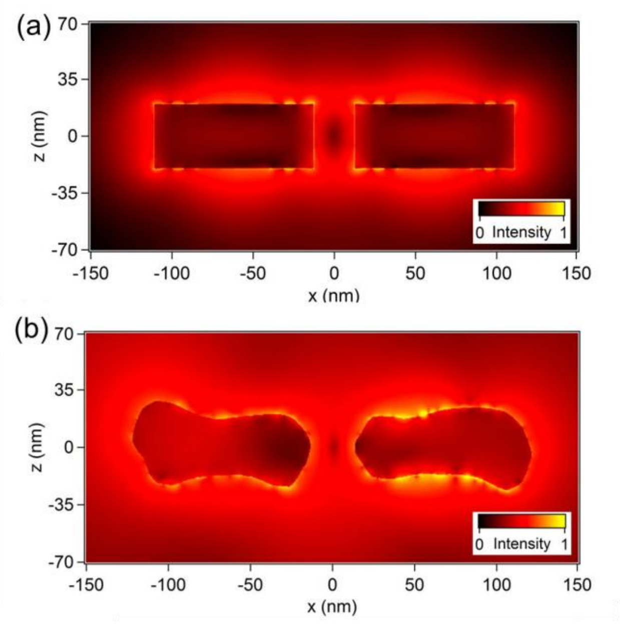

Fig. 1 : Near-field distribution of the second harmonic intensity close to (a) an idealized gold nanoantenna and (b) realistic gold nanoantenna.

3. Nonlinear Fano Resonances

We will discuss a new strategy that we recently developed to increase nonlinear optical processes in plasmonic systems. This strategy is based on Fano resonances which stem from the coupling between a dark mode and a bright mode. Dark modes are weakly coupled to far-field radiations, resulting in a strong localization in the near-field, but need to be coupled with an optically active mode

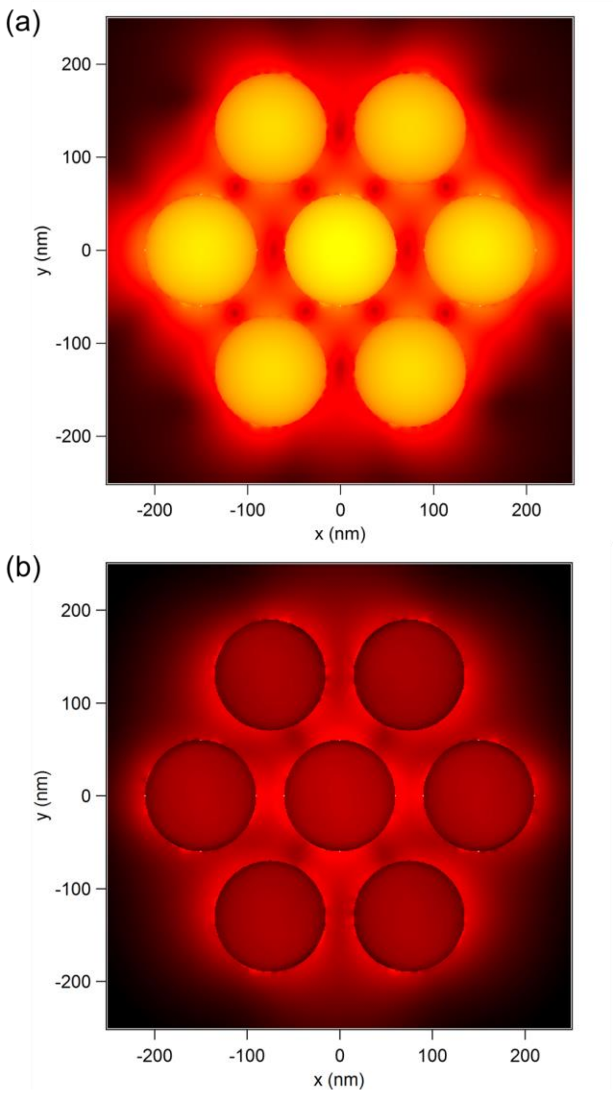

Fig. 2 : Near-field distribution of the second harmonic intensity close to a silver heptamer. The fundamental wavelength is (a) λ = 800 nm and (b) λ = 950 nm.

to be effectively excited. This coupling can be mediated by Fano resonances in order to increase the near-field at the fundamental wavelength. The optical properties of silver heptamers were tailored in order to observe simultaneously a Fano dip at the fundamental wavelength (800 nm) and a high order scattering peak at the second harmonic wavelength (400 nm) [5]. The observation of a Fano dip at the fundamental wavelength ensures that the dark mode is effectively excited. This strategy effectively increases second harmonic generation. We also propose a new measurement method of displacement at the nanoscale based on the control of the nonlinear optical response of plasmonic nanostructures by means of Fano resonances [6]. In the case of second harmonic generation from gold nanodolmens, the different nonlinear sources distributions induced by the different coupling regimes are clearly revealed in the far-field distribution. Hence, the configuration of the nanostructure can be accurately determined in 3-dimensions by recording the wave scattered at the second harmonic wavelength. We show that 3-dimensions plasmonic nanorulers can be implemented with simpler geometries than in the linear regime whilst providing complete information on the structure conformation [6].

Acknowledgments

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References

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