Third-order nonlinear optical properties of 2D plasmonic nanoprism arrays: spectral and dichroic effects

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The coupling of light to surface plasmons in metallic nanostructures with tailored shapes and/or composition induces intense enhancements of the local electromagnetic field at the nanostructures, which can be further increased by exploiting the interaction among the plasmonic nano-building blocks. These properties are of great interest for nanophotonics and nonlinear optics (NLO) applications since they can be effectively exploited to design plasmonic nanosystems with tunable and amplified nonlinear optical response [1]. In this presentation, we will report on the study of the spectral and polarization dependence of the third-order nonlinear optical properties of bi-dimensional ordered arrays of plasmonic (Ag and Au) triangular nanoprisms, synthesized by nanosphere lithography on silica substrates (Fig. 1). The peculiar topological configuration makes these nanostructures very promising for nonlinear optics applications [2]. Due to their capability of confining electric fields at the sharp tips, indeed, the triangular nanoprisms produce very intense hot-spots of the local-field which are further enhanced by near-field coupling when the nanoprisms are arranged at distances comparable to the surface plasmon resonance decay length of the isolated nanoprism.
The NLO characterization is performed by means of the z-scan technique using a ps laser at 10 Hz repetition rate to excite the ultrafast, electronic component of the nonlinear response of the nanoarrays, inducing minimal thermal load to the samples. An optical parametric amplifier allows the wavelength tuning of the laser source onto the linear absorption resonances of the samples in the visible and near-IR range. The nonlinear parameters (nonlinear refractive index and nonlinear absorption coefficient) are measured as a function of the excitation wavelength, the pump intensity as well as the nanoprisms composition. The nonlinear response is also investigated as a function of the polarization of the laser source and dichroism effects in the nonlinear absorption properties of the systems are reported. The results demonstrate a dramatic enhancement of the nonlinear optical response for the investigated nanosystems and the possibility to obtain the fine tuning of their nonlinear properties by acting on the different degrees of freedom available. The experimental findings are also compared and interpreted on the basis of the results of finite elements methods (FEM) simulations of the plasmonic properties (near-field and far-field) of the nanosystems.

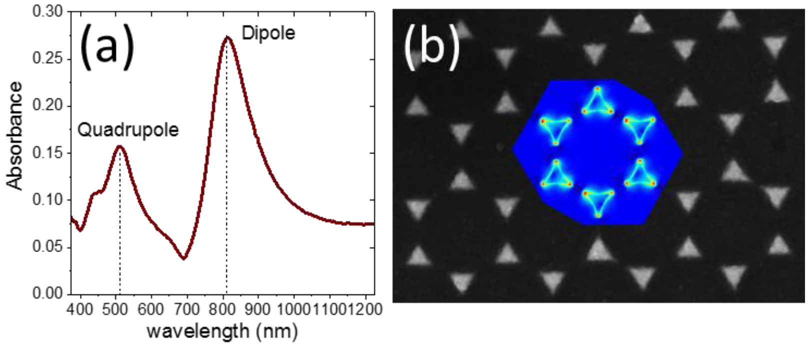

Fig. 1: *(a) Optical extinction spectrum and (b) SEM image of a Ag nanoprism array. Inset: simulated map of local-field enhancement at the dipolar resonance of the array.*

References
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