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### **Ultrafast Nonlinear Plasmonics of Single Nano-Objects**

NATALIA DEL FATTI, Institut Lumiere Matiere, CNRS - Universite Lyon 1

Investigating, understanding and modeling the physical properties of nano-objects are intense fields of research. Of particular interest are metal-based nano-objects, where their morphology and environment dependent surface plasmon resonances (SPR) have been extensively exploited to design new optical systems. As a SPR is associated to electromagnetic local field enhancement in the nano-object, it also leads to enhancement of its optical nonlinearity, opening many possibilities for investigating fundamental processes at nanoscale [1]. Most of these studies were performed on large ensembles of nano-objects, providing mean information which impedes detailed comparison between experimental data and theoretical models. With the advance of single nanoparticle spectroscopy methods, the linear and nonlinear responses of a single nano-object can now be addressed, which, associated to determination of its morphology by electron microscopy, opens the way to their quantitative modeling [2]. In this context we discuss experimental and theoretical investigations of the ultrafast response of individual model nano-objects, either formed by a single particle (gold nanorod) or by two particles at a nanometric distance (gold-silver nano-dimer). Results obtained in gold nanorods are in excellent quantitative agreement with a model computing the change of the metal dielectric function due to ultrafast electron heating and relaxation. This shows that the nonlinear response of a metal nano-object can be fully described as that of the bulk metal enhanced by plasmonic effects. Extension of these studies to more complex nano-objects, as nano-dimers formed by two different materials, permits analysis of the impact of their interaction. We demonstrate here the existence of Fano effect in the absorption of a single Ag-Au dimer, experimentally proving previous theoretical predictions [3]. Furthermore, we show that ultrafast pump-probe nonlinear spectroscopy permits to selectively address at nanoscale only one of the components of a dimer, paving the way toward quantitative investigations of energy and charge exchanges in multi-material nano-objects. [1] F. Vallée and N. Del Fatti, in “Plasmonics: theory and applications”, eds. T.Shahbazyan & M.Stockman, Springer, p. 167 (2013) [2] A. Crut, P. Maioli, N. Del Fatti and F. Vallée, Chem. Soc. Rev. 43, 3921 (2014) [3] G. Bachelier, I. Russier-Antoine, E. Benichou, C. Jonin, N. Del Fatti, F. Vallée, P.F. Brevet, Phys. Rev. Lett. 101, 197401 (2008).