



Nanotechnology Seminar, Wednesday 30.3.2016, 12:00

IKI Auditorium, Building 51, room 015

From Molecules to Meta-Molecules for Nonlinear Optics: Physics, Symmetry and Nanotechnologies

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Abstract:

Molecular engineering for NLO has experienced a first revival from the early nineties to this day, when its scope was enlarged from a more restricted earlier blueprint of donor-acceptor dipolar entities, to encompass the broader pool of multipolar molecules, with octupoles as a new tensor symmetry guided template for quadratic effects (1,2). In the wake of the development of nanotechnologies and nano-sciences, the field of nanoplasmonics appeared, more recently as a further enlarged playground for molecular engineering, with the early proposition of octupolar associations of nanoparticles showing the way towards up-scaled and enhanced nonlinear optical entities, in both 2-D and 3-D arrangements (3). Over recent years, the concept of « meta-molecules » has emerged, by way of associating « meta-atomic » building blocks in the form of either metallic nano-cavities or nanoparticles down to a typical 200nm and less. This second revival has been guided, like the previous one, by combining fundamental group symmetry and tensor arguments, applied now in the different physical frame of plasmonics, entailing the additional challenge of non-local interactions. The interplay between localized plasmons and propagative ones has led to interesting associations from nano-particles (4) and nano-cavities (5), onto meta-molecular associations of these (6,7), all the way to meta-arrays (8). The implementation of chirality is further leading to interesting possibilities towards high-density data encryption (9). Polarization resolved nonlinear confocal microscopy stands-out in the toolbox that we have conceived and developed over the last decade towards nonlinear optical characterization down to single nano-scale nonlinear entities (10). We will review the different steps of this ongoing story, from theoretical, experimental and technological viewpoints, covering basics all the way to current advances in our laboratory.

1. J. Zyss, Nonlinear Optics: from Dipolar to Octupolar Molecules and Materials, J. Chem. Phys. 98, 6583 (1993)
2. C. Dhenaut, I. Ledoux, I. D. W. Samuel, J. Zyss, M. Bourgault, H. Le Bozec, Chiral Metal Complexes with Large Octupolar Optical Nonlinearities. Nature, 374, 339–342 (1995).
3. M. I. Stockman, K. Li, S. Brasselet, J. Zyss, Octupolar metal nanoparticles as optically driven, coherently controlled nanorotors. Chem. Phys. Lett. 433, 130–135 (2006).
4. R.Hou, V.Shynkar, C.Lafargue, J.Zyss, Lagugné Labarthe JPPC (2016)

5. A. Salomon, M. Zielinski, R. Kolkowski, J. Zyss, Y. Prior, Size and Shape Resonances in Second Harmonic Generation from Silver Nanocavities. *J. Phys. Chem. C* 117, 22377–22382 (2013).
6. A. Salomon, Y. Prior, M. Fedoruk, J. Feldmann, R. Kolkowski, J. Zyss, Plasmonic coupling between metallic nanocavities, *J. Opt.* 16, 114012 (2014).
7. R. Kolkowski, J. Szeszko, B. Dwir, E. Kapon, J. Zyss, Effects of surface plasmon polariton-mediated interactions on second harmonic generation from assemblies of pyramidal metallic nanocavities, *Opt. Express* 22, 30592-30606 (2014).
8. R.Kolkowski, J.Szesko, B.Dwir, E.Kapon, J.Zyss, Non-centrosymmetric plasmonic crystals for second-harmonic generation with controlled anisotropy enhancement, *Laser Photonics Review*, to appear (2016)
9. R. Kolkowski, L. Petti, M. Rippa, C. Lafargue, J. Zyss, Octupolar plasmonic meta-molecules for nonlinear chiral watermarking at subwavelength scale, *ACS Photonics* 2, 899–906 (2015)
10. S. Brasselet, V. Le Floch, F. Treussart, J. F. Roch, A. Ibanez, J. Zyss, In-situ diagnostics of the crystalline nature of single organic nanocrystals by nonlinear microscopy, *Phys. Rev. Lett.* 92(20), 207401 (2004)