Brana Jelenkovic

Four way mixing in hot pottasium vapor – slow light with a gain, and intensity squeezing of probe and conjugate

Bojan Zlatkovic, Aleksandar Krmpot, Milan Radonjic and Brana Jelenkovic Institute of physics, University of Belgrade, Serbia

We have investigated reduction of group velocity due to nondegenerate four-wave mixing (FWM) in hot potassium vapor in the presence of gain of both probe and conjugate. Using an amplifying medium to alleviate the absorption and distortion of propagating pulses are important for ideal slow-light propagation, which can happen if (1) probe and conjugate are not be attenuated, (2) fractional delays, defined as the ratio of the delay to the duration of the pulse, for probe and conjugate, are larger than 1, and (3) fractional broadening of probe and conjugate are very small, ideally 1. When these three requirements are met, the probe and conjugate pulses can be delayed by an arbitrary amount, simply stacking the systems.

Figure 1 shows schematic of the experiment and atomic level scheme. Pump and probe are colinear and orthogonally polarized. Diagram of atomic levels consists of three hyperfine levels and a double Λ scheme in D1 line in ³⁹K. There are not previous studies of FWM with co-propagating pump and probe. Important feature of this set up and the amplification in the 4WM process is the conjugate pulse coupled to the probe and propagating alongside the probe.



Figure 1. FWM in pottasium vapor.. Schematic of the experiment and atomic level scheme.

We demonstrate [1] high-efficiency of optical phase conjugatin in potassium, with gains of ~ 80 for pump and ~ 60 for congugate, with modest intensities of the pump beam, near ~10 W cm–2 for specific atomi denisty, one photon and two photon detuning. Exceptionally high gains in K, in comparisom to other alkaly atoms under similar experimentla ocnditions, might be due to smallest ground state hyperfine splitting in potassium among alkalies: cross-susceptibilities between probe and conjugate that give rise to FWM process are higher for smaller hyperfine splitting [2].

We have observed the ultraslow propagation of Gaussian like probe pulses as short as 20 ns, resulting in a fractional delay of the probe and conjugate of ~ 5. During the propagation a probe pulse is amplified and, depending on the gain, generates a conjugate pulse which is faster and separates from the probe pulse before exiting the K cell at a fixed delay. There is broadening or distortion of pulses resulting in fractional broadening ~1.5.. We observed that the gain and the delay are in the trade-off relation, i.e., the higher the gain, the smaller the delay and vice versa. The presence of delay with the gain makes this system very interesting in the context of all-optical information processing.

Due to their high gains in Potassium, the probe and the conjugate beams may be suitable for quantum correlation and relative intensity squeezing experiments. We are exploiting capability of this system to generate twin beams with intensity difference squeezing between the probe and

conjugate beams. It was found recently [3] that FWM in Rb atomic vapor can produce entangled twin beams with intensity-difference squeezing levels approaching the highest quadrature squeezing levels demonstrated with downconversion crystals in an OPO.

[1] B. Zlatković et al Laser Phys. Lett. 13 015205 (2016)

- [2] M T Turnbull et al Phys. Rev. A 88 033845 (2013)
- [3] C. F. McCormick et al Phys. Rev. A 78, 043816 (2008).

Jerome Wenger

Enhanced dipole-dipole energy transfer in plasmonic nanogap antenna

P. Ghenuche,¹ M. Mivelle,² J. de Torres,¹ S. B. Moparthi,¹ H. Rigneault,¹ N. F. van Hulst,² M. F. Garcia-Parajo,² J. Wenger ^{1*}

¹ CNRS, Aix-Marseille Université, Institut Fresnel, Campus St Jérôme, Marseille 13013, France ² ICFO-Institut de Ciencies Fotoniques, Mediterranean Technology Park, 08860 Castelldefels, Spain

* jerome.wenger@fresnel.fr

Plasmonic optical antennas are receiving a large interest to interface light with molecules on dimensions much beyond the optical wavelength. A specific design called antenna-in-box has been developed for the enhanced detection of single fluorescent molecules in solutions at high concentrations, reaching detection volumes down to 58 zL (four orders of magnitude smaller that the diffraction limit) and large enhancement of the single molecule fluorescence, up to 1100-fold [1].

Thanks to their ability to control and manipulate optical fields down to the nanometre scale, it is appealing to use plasmonic antennas to enhance the energy transfer between single quantum emitters. Energy transfer between molecules is an essential phenomenon for photosynthesis, photovoltaics and biotechnology.

Our group has recently demonstrated enhanced energy transfer within single donor-acceptor fluorophore pairs confined in single gold nanoapertures [2]. Here we investigate the nanogap antenna between two metal nanoparticles to reach strong intensity and LDOS enhancement [3]. Our experiments monitor both the donor and the acceptor emission photodynamics on single donor and acceptor quantum emitters attached on a double stranded DNA linker. Our results establish that nanophotonics can be used to intensify and control the near-field energy transfer.



Fig. 1. Förster resonance energy transfer (FRET) in a nanogap antenna and enhancement of the donor decay rate as the LDOS is increased.

References

[1] D. Punj, M. Mivelle, S. B. Moparthi, T. van Zanten, H. Rigneault, N. F. van Hulst, M. F. Garcia-Parajo, J. Wenger, A plasmonic 'antenna-in-box' platform for enhanced single-molecule analysis at micromolar concentrations, Nature Nanotech. 8, 512-516 (2013).

[2] P. Ghenuche, J. de Torres, S. B. Moparthi, V. Grigoriev, J. Wenger, Nanophotonic Enhancement of the Förster Resonance Energy-Transfer Rate with Single Nanoapertures, Nano Lett 14, 4707-4714 (2014).

[3] P. Ghenuche, M. Mivelle, J. de Torres, S. B. Moparthi, H. Rigneault, N. F. Van Hulst, M. F. García-Parajó, J. Wenger, Matching Nanoantenna Field Confinement to FRET Distances Enhances Förster Energy Transfer Rates, Nano Lett 15, 6193-6201 (2015).

Xavier Zambrana-Puyalto

Purcell factor and chirality control at the nanoscale

Xavier Zambrana-Puyalto¹, and Nicolas Bonod¹

¹Aix-Marseille Université, CNRS, Centrale Marseille, Institut Fresnel UMR 7249, 13013 Marseille email: xavislow@protonmail.com

Mie resonators made of dielectric materials exhibit electric and magnetic resonances. These resonances are promising to study light matter interactions at the nanoscale and to design silicon based light cavities. In this context, quantifying the Purcell factor and the effective volume of such cavities is of high interest. In order to do that, we use the latest advances on this field of research [1,2] to derive the Purcell factor $F(\omega)$ and the effective volume of a normal mode of spherical resonators [3]. It will be shown that subwavelength silicon spherical cavities can give rise to enhancement of decay rates as high as 10^4 .

At the same time, we will also show that silicon resonators over-perform those made of plasmonic materials to tailor the chirality of single emitters. Indeed, it will be seen that the scattered field of a spherical antenna can yield values of chirality which practically encompass all possible values. In particular, the chirality of the field emitted by the antenna can exceed the initial chirality emitted by the molecule. Thanks to the Mie theory formalism in the helicity basis [4], we show the conditions under which this phenomenon takes place. It will be seen that the interplay between electric and magnetic modes of the resonator is required to tailor the chirality of light [5].

REFERENCES

[1] E.A. Muljarov, M.B. Doost, W. Langbein, arXiv:1408.6877v2 (2014).

- [2] C. Sauvan, J.P. Hugonin, I.S. Maksymov, P. Lalanne, Phys. Rev. Lett. **110** (23), 237401(2013).
- [3] X. Zambrana-Puyalto, and N. Bonod, Phys. Rev. B 91, 195422 (2015).
- [4] X. Zambrana-Puyalto, PhD Thesis (2014).
- [5] X. Zambrana-Puyalto, and N. Bonod, Nanoscale 8, 10441 10452 (2016).

Evelyne Salançon - Aix-Marseille Université - LUMINY

Electron point source : A study of the wave coherence with a low energy electron projection holographic microscope

E. Salançon, L. Lapena, A. Degiovanni, R. Morin

Holography is commonly known because of 3D imaging... but, this is also a mean to test the physical characteristics of a source and beyond the nature of the particles. Here, an electron projection microscope set up - currently used to produce holograms - is used to study the coherence of the wave produced by a new kind of electron source. This source illuminates an opened object and the physical shadow - from a wave, like an interferogram - obtained further, is detected with a very high sensitivity and resolution screen. By this way, a new demonstration of single-electron buildup of an interference pattern can be made, but, this time with low energy electrons (figure 1). I'll show how the high resolution obtained by the one by one detection, allows us to identify the source size precisely (Φ <1,8nm).



Figure 1 : Interferogram obtained by accumulating low energy electrons along one night in a projection holographic microscope.

Irene D' Amico

(Dynamics) of topologically protected states in spin chains.

Topologically protected localised states in spin chains and their dynamics

M. P. Estarellas, I. D'Amico, and T. P. Spiller Information Centre and Department of Physics, University of York, York, UK

The topological confinement of quantum states has engaged the condensed matter community for a few decades. Now this field is receiving increasing interest due to its potential applications for topological quantum computation, quantum state transfer and quantum memories.

Here we consider spin chain systems analogous to the Su, Schrieffer and Hegger (SSH) model, which was first presented to describe soliton formation in polyacetylene [1]. Recently, an analogy of this model has been implemented with a set of identical, coupled dielectric resonators placed in a microwave cavity [2], inducing spatially confined states. Here we present a model inspired by these systems and explore the presence of topological localisation in finite one-dimensional spin chains.

Spin chains are of particular interest due to their versatility to be engineered. By tuning the couplings, we can construct our chains to be topologically analogous to the SSH model. Experimentally, this can be done for any system where it is possible to engineer the couplings between the sites, e.g. using electrons and excitons trapped in nanostructures.

We demonstrate explicitly the topologically induced spatial localisation of quantum states, analyse their dynamics, and present detailed investigations of the effects of random noise, showing that these topologically protected states are very robust against this type of perturbation. Systems with such topological robustness are good candidates for quantum information tasks.

[1] A. J. H. W. P. Su, J. R. Schrieffer, Phys. Rev. Lett. 42, 1698 (1979).
[2] U. K. F. M. C. Poli, M. Bellec and H. Schomerus, Nat. Commun. 6 (2015).
[3] M. P. Estarellas, I. D'Amico, and T. P. Spiller, submitted (2015)

Niek F. van Hulst ICFO Barcelona

Coherent feedback control of optical antennas and single photon emitters

Animesh Datta, Warwick Univ. UK,

Efficient energy transfer in quantum molecular networks.

The nature and role of quantum correlations in efficient energy transport (EET) in molecular systems is a problem of wide interest. We consider coupled excitonic models, which include naturally occurring light harvesting complexes such as the FMO (Fenna-Matthews-Olson) complex. We also study randomly generated molecular aggregates to study their transport properties and the quantumness allied with this process. Quantumness in excitonic networks using can be measured using several measures such as entanglement, coherence measurements, Leggett-Garg inequalities, which we argue are the same in the single excitation regime.

These aggregates can be classed in to equivalence classes depending on the number of chromophores states per site etc., allowing a systematic analysis of EET in molecular systems. The dynamics are studied using Markovian noise models.

One of the aims of the study is to garner a better understanding of the role of quantumness in EET by studying a large class of systems - natural as well as artificial, in a unified manner. The study should also clarify whether natural light harvesting complexes are special in particular way as far as EET is concerned. Finally, our work should provide insights into experimental methods of observing and confirming quantumness in EET in light harvesting complexes.

Erich Runge

Long-lived modes in nano-porous gold sponges

Felix Schwarz,¹ David Leipold,¹ Jan Vogelsang,²Germann Hergert,² Martin Silies,² Dong Wang,³ Peter Schaaf,³ Christoph Lienau,² and Erich Runge^{1,*}

¹ Institut für Physik, Technische Universität Ilmenau, 98693 Ilmenau, Germany
 ² Institut für Physik, Carl von Ossietzky Universität, 26129 Oldenburg, Germany
 ³ Institut für Werkstofftechnik, Technische Universität Ilmenau, 98693 Ilmenau, Germany

Controlling and localizing light on ultrafast time- and nanometer length-scales is a key requirement for future optical technologies. Localization of light using nanostructured materials with taylored disorder for electromagnetic engineering has some conceptional advantages: They are cheap to produce and, more importantly, inherently unaffected by perturbations, e.g., temperature variations. The highly enhanced near-fields of localized light modes imply nonlinear effects due to strong light-matter interaction.

In this contribution, we discuss the localization of light in disordered dielectric and hybrid metal-dielectric nanostructures. As prototypical examples, we consider (i) randomly distributed zinc-oxide nanoneedles with [1] and without an ultrathin gold-coating as well as (ii) irregular nanoporous gold sponges [2]. In all cases, we find surprisingly long-lived modes with greatly enhanced near-field intensities, making the samples particularly interesting for the study of nonlinear phenomena. We present calculated scattering spectra and near-field dynamics Results compare favourably with recent experimental results [2]. Our findings indicate that such systems provide interesting novel functionality for next-generation data-processing and sensing applications.

* erich.runge@tu-ilmenau.de

- [1] M. Mascheck et al., Nat. Photon. 6, 293 (2012)
- [2] C. Vidal et al., ACS Photonics 2, 1436 (2015)

Figure 1. Squared electric field intensity on a nanoporous metal-nanoparticle after excitation by an ultrashort laser pulse $(/E)^2$ in a.u., slice through the particle, logarithmic plot). Long-lived resonances can occur at specific frequencies, strongly localized even within the nano-particle.



Javier Aizpurua

Quantum approaches to electrons and photons in nonlinear and active nanoplasmonics

Materials Physics Center (CSIC-UPV/EHU) and DIPC, Donostia-San Sebastián, Spain

The optical response of metallic nanoparticles has been successfully addressed within classical electrodynamics. However, there are situations where the quantum nature of the electrons that build the collective response becomes relevant. In small metallic nanoparticles as well as in narrow plasmonic nanogaps, quantum size effects, strong nonlocal interactions, the electron spill out, or electron tunneling are quantum effects that can be correctly included when a quantum treatment of the response is implemented. We develop a quantum description of the electrons within time-dependent density functional theory (TDDFT) to address quantum effects in the optical response of a metallic system, and compare the results with those from classical theories.

Within the TDDFT, we adopt an atomistic quantum approach of the optical response that considers explicitly the atomic positions in the metallic nanoparticles, and find important differences with respect to simpler approaches based on a description of smooth interfaces. This allows to confine and enhance light at ultra-confined volumes in the atomic scale.

Furthermore, based on the strong dependence of the intensity of the gap plasmon on the tunneling current across the gap, we explore the quantum regime of metallic gaps to obtain an active control of the plasmonic response which can be modulated with the application of a DC external bias. Other quantum effects in metallic nanoparticles, such as the dynamical screening in charged clusters, as well as the nonlinear quantum optical response of plasmonic nanosystems will be also covered.

Another important aspect of quantum plasmonics is connected with the quantization of the electromagnetic field associated with the plasmonic modes, in the spirit of quantum electrodynamics. When plasmonic cavities with low population of plasmons are coupled to vibrational modes of a molecule, new properties derived from the fully coherent nature of the interaction emerge, driving the dynamics of photons, plasmons and vibrations to a novel nonlinear regime. The spectral features of the Raman signals, as well as the correlations of the photons emitted reveal the importance of a quantum treatment in single plasmon nanophotonics.

Both quantum aspects of nanoplasmonics will thus be covered in this presentation.

References

[1] M. Barbry, P. Koval, F. Marchesin, R. Esteban, A. G. Borisov, J. Aizpurua, and D. Sánchez-Portal, *"Atomistic Near-Field Nanoplasmonics: Reaching Atomic-Scale Resolution in Nanooptics"*,

Nano Lett. 15, 3410 (2015)

[2] F. Marchesin, P. Koval, M. Barbry, J. Aizpurua, and D. Sánchez -Portal, "*Plasmonic Response of Metallic Nanojunctions Driven by Single Atom Motion: Quantum Transport Revealed in Optics*", ACS Photonics (2016), DOI:10.1021/acsphotonics.5b00609

[3] D. C. Marinica, M. Zapata, P. Nordlander, A. K. Kazansky, P. M. Echenique, J. Aizpurua, and A. G. Borisov, *"Quantum active plasmonics"*, Science Advances 1, e1501095 (2015).

[4] M. Schmidt, R. Esteban, A. González-Tudela, G. Giedke, and J. Aizpurua, "QED description of Raman scattering from molecules in plasmonic cavities", (2016) arXiv:1509.03851

Paivi Torma Towards quantum fluids in plasmonic systems; strong coupling studies

We have proposed the concept of *quantum plasmonic lattices*, that is, arrays of metal nanoparticles combined with emitters, as a platform to study quantum many-body physics, especially quantum fluids [1]. Here we present the concept and our experimental work towards its realization. We have studied strong coupling in metal nanoparticle arrays combined with organic molecules. We show strong coupling involving three different types of resonances in plasmonic nanoarrays: surface lattice resonances (SLRs), localized surface plasmon resonances on single nanoparticles, and excitations of organic dye molecules [2]. The delocalized nature of the collective SLR modes suggests that, in the strong coupling regime, molecules near distant nanoparticles are coherently coupled. Furthermore, we study spatial coherence properties of a plasmonic nanoarray covered with a fluorescent organic molecule film by a double slit experiment [3]. A continuous evolution of coherence from the weak to the strong coupling regime is observed, with the strong coupling features clearly visible in the interference fringes. We outline future studies on strong coupling phenomena with emphasis on identifying purely quantum features [4]. Finally, we show with magnetic nanoparticles how the intrinsic spin-orbit coupling of the material interplays with the symmetries of the nanoparticle array [5].

References

[1] J.-P. Martikainen, M.O.J. Heikkinen, and P. Törmä, Condensation phenomena in plasmonics, Phys. Rev. A 90, 053604 (2014)

[2] A. I. Väkeväinen, R. J. Moerland, H. T. Rekola, A.-P. Eskelinen, J.-P. Martikainen, D.-H. Kim, and P. Törmä, Plasmonic surface lattice resonances at the strong coupling regime, Nano Lett. 14, 1721 (2014)

[3] L. Shi, T. K. Hakala, H. T. Rekola, J.-P. Martikainen, R. J. Moerland, and P. Törmä, Spatial coherence properties of organic molecules coupled to plasmonic surface lattice resonances in the weak and strong coupling regime, Phys. Rev. Lett. 112, 153002 (2014)

[4] P. Törmä and W. L. Barnes, Strong coupling between surface plasmon polaritons and emitters: a review, Rep. Prog. Phys. 78, 013901 (2015)

[5] M. Kataja, T. K. Hakala, A. Julku, M. J. Huttunen, S. van Dijken, and P. Törmä, Nat. Comm. 6, 7072 (2015)

François Marquier Quantum optics with surface plasmons

Laboratoire Charles Fabry, Institut d'Optique Graduate School, CNRS, Université Paris-Saclay, 91127 Palaiseau cedex, France francois.marquier@institutoptique.fr

Surface plasmon-polaritons (SPPs) result from collective oscillations of free electrons coupled to an electromagnetic field at a plane interface between the metal and a dielectric medium. As photons, SPPs can be considered either as waves or as particles [1,2] and they can experience striking quantum interferences such as Hong-Ou-Mandel effect [3], entanglement [4] and one of the most remarkable one: wave-particle duality.

Although demonstrated for guided plasmons supported by a nanorod [5], to our knowledge, wave-particle duality has not been tested yet on SPPs propagating on a simple plane metallic interface. We report an experiment showing this dual behavior. The SPPs are created and detected by coupling single photons to nanostructured couplers etched on an optically thick gold film. As a first step, the particle-like behavior is tested with Hanbury-Brown-Twiss experiment by sending a single SPP onto a splitter separating SPPs in two orthogonal in-plane directions. We observe a strong anticorrelation between the two output ports, demonstrating the particle behavior. We use then this single SPP source in a Mach-Zehnder interferometer. We observe fringes with a visibility of $60\% \pm 10\%$, demonstrating the wave behavior of the single SPP. In these experiments, the plasmonic interferometer layout is similar to the usual free-space experiments with single photons which makes it easy to compare the results with the fundamental wave-particle duality of photons experiments [6].

Experimental demonstration of this quantum property is essential to further validate the quantum description of SPPs and for future applications in quantum communication mediated by SPP. Another experiment will also be presented to highlight the role SPP can play to change the properties of single-photon emitters [7].

References

[1] JM Elson and R.H. Ritchie, *Phys. Rev. B* 4, 4129 (1971)

- [2] A. Archambault et al., *Phys. Rev. B* 82, 035411 (2010)
- [3] J.S. Fakonas et al., Nat. Phot. 8, 317 (2014)
- [4] E. Altewischer et al., *Nature* **418**, 304 (2002), Chang, Nature Phy., (2007)
- [5] R. Kolesov et al., Nat. Phys. 5, 470 (2009)
- [6] P. Grangier et al., *EuroPhys. Lett.* 1, 173 (1986)
- [7] B. Ji et al., Nat. Nanotech. 10, 170 (2015)

Walter Pfeiffer Long-range cavity-assisted coherent energy transfer between plasmonic nanoantennas

Faculty of Physics, Bielefeld University, 33615 Bielefeld, Germany

Martin Aeschlimann¹, Tobias Brixner², Benjamin Frisch¹, Bert Hecht³, Bernhard Huber², Matthias Hensen⁴, Christian Kramer², Enno Krauss³, Thomas Löber⁵, Walter Pfeiffer⁴, Martin Piecuch¹, and Philip Thielen¹

¹ Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, 67663 Kaiserslautern, Germany

² Department of Physical and Theoretical Chemistry, Würzburg University, 97074 Würzburg, Germany

³ Experimental Physics 5, Würzburg University, 97074 Würzburg, Germany

⁴ Faculty of Physics, Bielefeld University, 33615 Bielefeld, Germany

⁵ Nano-Structuring-Centre, TU Kaiserslautern, 67663 Kaiserslautern, Germany

Rather diverse phenomena such as light harvesting in photosynthesis, nanoscale quantum entanglement, or photonic mode hybridization rely on efficient photonic coupling mediated via the optical near-field. Hence, without further tailoring the near field distribution these coupling phenomena are efficient only for separations much shorter than the optical wavelength and long-range energy or signal transfer is difficult to achieve. This limits the application of nanophotonic devices for example for signal transfer or nanoscale quantum optical applications that require selective optical addressing of individual nanoscale qubits. Plasmonic waveguide structures and photonic cavities can overcome this limitation. Here we demonstrate efficient cavity assisted coherent energy transfer between plasmonic nanoantennas separated by more than twice the wavelength. Whispering gallery mode antennas placed in the two foci of an elliptic plasmonic arena cavity exhibit periodic energy exchange. Both, time-resolved two-photon photoemission electron microscopy experiments and time-domain electromagnetic field calculations match closely and are quantitatively described using the plasmonic analogue of the quantum mechanical Tavis-Cummings model. Based on this it is shown that the cavity damping critically determines the character of the cooperative behavior of the coupled antennas. Whereas, high contrast energy exchange between both antennas occurs if the dissipation of cavity and antenna modes are equal, energy equipartition is rapidly established for a high-quality cavity. Hence, besides enabling long-range plasmonic coupling in nanophotonic applications the demonstrated cavity design could serve as a versatile model system for investigating the role of dissipation in coherent energy transfer processes and might be implemented in nanoscale quantum optical devices.

Martti Kauranen

Tailoring nonlinear optical responses of plasmonic metasurfaces

Martti Kauranen,¹ Robert Czaplicki,¹ Jouni Mäkitalo,¹ Kalle Koskinen,¹ Joonas Lehtolahti,² Janne Laukkanen,² and Markku Kuittinen²

¹ Department of Physics, Tampere University of Technology, Tampere, Finland

² Department of Physics and Mathematics, University of Eastern Finland, Joensuu, Finland

<u>martti.kauranen@tut.fi</u>

The plasmonic resonances of metal nanoparticles can give rise to strong local fields ("hot spots"), which can enhance optical interactions. This is particularly important for nonlinear optical effects, which scale with a high power of the optical field. Here, we summarize our work on second-harmonic generation (SHG) from metasurfaces consisting of arrays of metal nanoparticles and discuss future prospects for this line of research.

SHG relies on non-centrosymmetric structures and is also otherwise sensitive to structural properties of the sample. In our case, the symmetry is broken using either L or T-shaped nanoparticles. However, particle de-fects support their own hot spots and can play a large role in the SHG response. The resulting effects can be interpreted in terms of higher-multipolar (magnetic, quadrupolar) contributions to the nonlinear signals. The quality of contemporary nanostructures can greatly suppress such effects, resulting in electric-dipole limit of the nonlinear response and opening the path towards true designer nonlinear metamaterials. We have subse-quently pursued several concepts to tailor the SHG response.

We first investigated concepts from molecular nonlinear optics by varying the ordering of the particles in the array. We showed that, not only orientational distribution, but other subtle details of ordering can greatly af-fect the efficiency of SHG. These effects arise from diffractive coupling between the particles.

We then demonstrated the use of nanoantennas in nonlinear optics by fabricating arrays combining non-centrosymmetric SHG-active particles with centrosymmetric SHG-passive particles. The passive particles can enhance the SHG response through lattice interactions between the two types of particles.

The nonlinear responses are strongly dependent on having the fundamental wavelength close to the plasmon resonance wavelength of the sample, but this as such is not sufficient for strong SHG response. In particular, the local-field distributions depend on particle geometry and can favor or suppress selected nonlinear signals.

In summary, we have explored fundamental concepts that affect SHG from metasurfaces. None of the con-cepts has yet been optimized, providing future opportunities for even stronger nonlinear responses.

Ruth Oulton Bristol Univ. U.K. Gender Balance Session 1. "Unconscious bias amongst scientists".

Gender Balance Session 2. "Sexual harassment in science: discussion forum".

The idea here is that we will outline a specific issue (a well-known case of serial sexual harassment by a famous scientist), and have about 45mins for discussion of the topic (i.e. the impact on the people involved, how it could have been handled better etc).

Pierre-François Brevet Nonlinear Optics with Single Metallic Nanoparticles: Fundamentals and Applications

Institut Lumière Matière, UMR CNRS 5306, Université Claude Bernard Lyon 1

Plasmonics has emerged as an important research field in nanoscience and nanotechnology. Recently, significant attention has been devoted to the observation and the understanding of nonlinear optical processes in plasmonic nanostructures, giving rise to the field of nonlinear plasmonics. A comprehensive insight into the physical mechanisms of these nonlinear optical processes is now within reach. In particular, second and third order processes are of deep interest because the parity is different and therefore they obey different selection rules. As such, they exhibit different features, in particular a marked difference for volume and surface effects. In this presentation, second harmonic generation and Kerr effect will be presented as examples of these processes with an emphasis on experiments performed at the single nanoparticle level.

Besides fundamental aspects, many applications are now regularly emerging. These propositions range from the nonlinear optical characterization of nanostructures to the optimization of laser beams or sensing. These future directions and developments will also be addressed and discussed.

References

 J. Butet, P.F. Brevet, O.J.F. Martin, Optical Second Harmonic Generation in Plasmonic Nanostructures: From Fundamental Principles to Advanced Applications, ACS Nano, 9 (2015) 10545-10562

Anatoly Zayats

Ultrafast Nonlinear Plasmonics.

Tobias Brixner

Coherent Multidimensional Space–Time-Resolved Spectroscopy

Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany, brixner@phys-chemie.uni-wuerzburg.de

The experimental investigation of nanoscale quantum-optical phenomena is often hampered by (1) the small relevant spatial scale of investigated systems and associated wavefunctions that usually lie below the diffraction limit of light, as well as (2) the ultrafast timescale of many time-dependent processes. Thus special techniques are required that can provide simultaneously both ultrahigh spatial and ultrahigh temporal resolution.

Combining coherent two-dimensional (2D) spectroscopy and time-resolved photoemission electron microscopy (TR-PEEM), we have developed coherent 2D nanoscopy [1] that provides access to nm and fs spatiotemporal phenomena. Thus it is possible to obtain subdiffraction information on nonlinear optical response functions. For that purpose, sequences of ultrashort laser pulses are irradiated on the sample, and emitted photoelectrons are measured with spatial resolution as a function of inter-pulse time delays and relative phases. Fourier transformation then provides 2D correlations of excitation versus detection frequency for each spatial position. Suitable modeling of the signals in turn leads to mechanistic insight into various aspects of spatiotemporal dynamics.

In this presentation the principle of the method will be described and its applicability illustrated on a number of examples.

Specifically, we apply the technique to study amorphous Si thin-film solar cells that are nanostructured for enhanced light-collection efficiency. We demonstrate the existence of strongly (Anderson) localized photonic states that are responsible for perfect absorption in the long-wavelength region [2]. This principle offers interesting opportunities for the future design of efficient absorber layers.

Next we move from such coherent, but classical, nanooptical systems that can still be described by Maxwell's equations, toward quantum systems. Specifically, we use the technique to study a monomolecular molecular film of Alq3 on a metal surface. This leads to hybrid interface states whose dynamics are observed again via 2D nanoscopy, but with an additional initial pump pulse that prepares coherences in the electronic excited-state manifold. The signatures of the excited states are imprinted on the resulting 2D spectra [to be published].

Coherent 2D nanoscopy is a unique method that can provide both spatial and temporal information on nanoscale quantum-optical systems.

[1] M. Aeschlimann, T. Brixner, A. Fischer, C. Kramer, P. Melchior, W. Pfeiffer, C. Schneider, C. Strüber, P. Tuchscherer, and D.V. Voronine, *Coherent two-dimensional nanoscopy*, Science **333**, 1723 (2011).

[2] M. Aeschlimann, T. Brixner, D. Differt, U. Heinzmann, M. Hensen, C. Kramer, F. Lükermann, P. Melchior, W. Pfeiffer, M. Piecuch, C. Schneider, H. Stiebig, C. Strüber and P. Thielen, *Perfect absorption in nanotextured thin films via Anderson-localized photon modes,* Nature Photonics **9**, 663 (2015).

Sébastien Bidault

Picosecond radiative lifetimes from bright single-photon emitting nanostructures at room temperature

ESPCI ParisTech, PSL Research University, CNRS, Institut Langevin, Paris, France

Because of homogeneous broadening effects, single organic molecules exhibit weak absorption cross-sections at room temperature even though they feature large dipolar transition moments for their electronic excited states associated to nanosecond radiative lifetimes. In order to enhance their excitation probability and further increase their decay rates, we assemble them in the center of a plasmonic cavity using a short DNA doublestrand.

In practice, gold nanoparticle (AuNP) dimers are obtained in large scale as a purified colloidal suspension. Confocal luminescence lifetime measurements in microfluidic conditions demonstrate that these nanostructures are single photon-emitters with picosecond lifetimes [1,2]. A conjunction of time-resolved luminescence and fluorescence correlation spectroscopy allows us to fully characterize the photophysical properties of these hybrid emitters that feature excitation cross-sections and decay rates enhanced by more than one order of magnitude with respect to isolated organic dyes [3]. To optimize the fluorescence quantum yield, we increase the size of the AuNPs and reach an average 44 times enhancement of the fluorescence count rate with picosecond lifetimes. These values correspond to unprecedented effective dipolar transition moments for isolated quantum emitters at room temperature.

[1] M. P. Busson, B. Rolly, B. Stout, N. Bonod and S. Bidault, Nature Commun. 3, 962 (2012)

[2] M. P. Busson and S. Bidault, Nano Lett. 14, 284 (2014)

[3] M. P. Busson, B. Rolly, B. Stout, N. Bonod, J. Wenger and S. Bidault, Angew. Chem. Int. Ed. 51, 11083 (2012)

Andre Stefanov

Broadband energy-entangled photon for high resolution temporal sensing and spectroscopy

Broadband energy-time entangled photon pairs are produced by pumping a non-linear crystal with a cw laser. Because of their quantum nature, they exhibit at the same time narrowband and short time features. Indeed the sum energy of both photons is equal to the well defined energy of the pump photon, whereas the correlation time between the two photons is of the order of few tens of femtoseconds. Those properties can be used for measurements beyond the capabilities of classical devices. Here we show how to make use of those features to study the temporal properties of photons through various media. The propagation of the entangled two-photon quantum states is described by a temporal wavefunction which is comparable for certain aspects to the one of coherent ultrashort laser pulses. However, because this light is in a continuous way regime, femtosecond timing can be performed without relying on high intensities which could destroy the investigated sample, like with short pulses. As application, we show a proof of principle experiment where ultrafast optical coincidences of the photon pairs allow selecting only the ballistic photons. Imaging through a scattering medium can therefore be performed. Using techniques from the ultrafast optics, we are able to manipulate the two-photon wave function with the help of a pulse shaper, which combines dispersive elements and a spatial light modulator. In that way, the temporal shape of the twophoton states before and after being transmitted through a sample can be reconstructed. leading to the dispersion properties of the sample. Ultimately the precise temporal control of the two photon wavefunction can lead to the implementation of proposals for two-photon spectroscopy with entangled photons, allowing to reveal new properties of the investigated molecules.

Bilge Can Yildiz

All-plasmonic enhancement of second harmonic generation via interference of conversion paths

<u>Bilge Can Yildiz^{1,2,3}</u>, Mehmet Emre Tasgin⁴ and Husnu Emrah Unalan^{2,6} Alpan Bek^{1.2.5}

¹Department of Physics, Middle East Technical University, Ankara, Turkey
 ² Center for Solar Energy Research and Applications, Ankara, Turkey
 ³Department of Applied Physics, Atilim University, Ankara, Turkey
 ⁴Institute of Nuclear Sciences, Hacettepe University, Ankara, Turkey
 ⁵Department of Micro and Nano Technology, Middle East Technical University, Ankara, Turkey
 ⁶Department of Metallurgical and Materials Engineering, Middle East Technical University, Ankara, Turkey
 ^email: bilge.vildiz@metu.edu.tr

This study focuses on exploring nonlinear optical conversion by metal nanostructures with plasmonic resonance properties. Second harmonic generation (SHG) can be enhanced when a plasmonic nonlinear converter is coupled with a higher quality single mode metal nanostructure. This phenomenon emerges due to interferences in conversion paths. In former studies [1, 2], we show that coupling of plasmonic converters with quantum emitters can highly enhance the nonlinear conversion efficiency. In this work, we demonstrate that SHG enhancement in hybrid structures can be obtained even in the absence of coupled quantum emitters. This is an important simplification for facilitating the use of purely metal nanoparticles with appropriate experimental function. Strongly localized nature of optical conversion can be utilized to address single or few molecules in a background-free fashion. We introduce a theoretical model examining the SHG response of two coupled metal nanostructures, which is developed to interpret the results of an experiment on a similar structure [3]. The theoretical model well predicts the experimental enhancement factor, ~30 in the SHG from a coupled Silver nanowire – Silver nanoparticle system.

REFERENCES

D. Turkpence, M. E. Tasgin, G. B. Akguc, A. Bek, Journal of Optics **16** 105009 (2014).
 M. E. Tasgin , I. Salakhutdinov, D. Kendziora, M. K. Abak, D. Turkpence, L. Piantanida, F. Fruk, M. Lazzarino, A. Bek, arXiv:1402.5244v3 (2014).
 B. C. Yildiz, M. E. Tasgin, M. K. Abak, S. Coskun, H. E. Unalan, A. Bek Journal of Optics **17** 125005 (2015).

Aurelien Drezet Quantum plasmonics using near-field optical microscope <u>Aurélien Drezet¹</u>, Martin Berthel¹, Quanbo Jiang¹, Aline Pham¹Serge Huant¹

¹ Institut Neel-CNRS, 25 rue des Martyrs 38042 Grenoble Aurelien.drezet@neel.cnrs.fr

Quantum plasmonics, i.e., the study of surface plasmon polariton (SPP) interactions in the quantum regime, has generated a growing interest in recent years due to the huge potentialities it offers to quantum information processing and integrated nano-photonics. In the recent years we developed a systematic approach based on near-field scanning optical microscope (NSOM) to study this regime. For this purpose we attached a nanodiamond hosting a single fluorescent Nitrogen Vacancy center at the apex of a NSOM tip [1]. This provided us a quantum NSOM and paved the way for quantum optical studies where single photons coupled to single plasmons are deterministically launched in a complex plasmonic environment at the nanoscale [2]. Over the year we adapted this method to the study of leaky SPP on a thin metal film using leakage radiation microscopy [3,4,5] (see Figure). Here, we review this original method and show its potentialities for quantum plasmonics. We demonstrate single plasmon launching [2,6], wave-particle duality [6] and second order coherence measurements [6,7]. We will discuss recent progresses obtained in the domain of NSOM imaging with classical and quantum sources [8] and show how the methods allows us to study local density of states (LDOS) associated with SPPs in various environments such a planar cavities.



Figure : A quantum NSOM tip launching SPPs on a thin gold film and detected using LRM. (a) image in the direct plane (b) image in the Fourier plane [8].

[1] A. Cuche et al., *Near-field optical microscopy with a nanodiamond-based single-photon tip* Opt. Express **17**, 19969 (2009).

[2] A. Cuche, O. Mollet, A. Drezet, S. Huant, "Deterministic" Quantum Plasmonics Nano Letters **10**, 4566 (2010).

[3] B. Hecht, et al., *Local Excitation, Scattering, and Interference of Surface Plasmons* Phys. Rev. Lett. **77**, 1889 (1996).

[4] A. Drezet et al., *Leakage radiation microscopy of surface plasmon polaritons* Mater. Sci. Eng. B **149**, 220 (2008).

[5] A. Drezet, C. Genet, *Imaging Surface Plasmons: From LeakyWaves to Far-Field Radiation* Phys. Rev. Lett. **110**, 213901 (2013).

[6] O. Mollet, S. Huant, G. Dantelle, T. Gacoin, A.Drezet, *Quantum plasmonics: Second-order coherence of surface plasmons launched by quantum emitters into a metallic film* Phys. Rev. B **86**, 045401 (2012).

[7] M. Berthel, S. Huant, A. Drezet, *Spatio-temporal second-order quantum correlations of surface plasmon polaritons* Optics Lett. **41**,37 (2016).

[8] M. Berthel, Q. Jiang, C. Chartrand, J. Bellessa, S. Huant, A. Drezet, *Coherence and aberration effects in surface plasmon polariton imaging* Phys Rev E **92**,033202 (2015).

Thomas Durt.

Quantum coherence at the nano and mesoscale, and non-linear modifications of Schroedinger equation.

It is not clear today whether the superposition principle is valid at all scales, in particular whether macroscopic superpositions are possible in the classical regime. Several attempts were made during the last decade, theoretical as well as experimental, in order to detect a possible transition between the classical and quantum regime at the nano/meso scale. Some models involve a non-linear modification of Schroedinger equation due to self-gravity, the Schroedinger-Newton (S-N) equation. We shall give an overview of the present state of the art in relation with the S-N equation and its still hypothetical experimental validation.

Mohamed E. Yahia

Rogue waves lead to the instability in GaN semiconductors.

Stephane Barland.

Particle-like behavior of light in semiconductor lasers.

Solitary waves are usually considered in conservative settings such as surface water waves or optical fibers. In dissipative systems, localized nonlinear waves have been analyzed as "dissipative solitons" since they share some of the properties of their conservative counterparts. However, due to dissipation these localized waves are robust attractors of the dynamics, which confers them many additional properties. In this contribution, we analyze (some of) the interactions between localized waves in semiconductor lasers and show intriguing experimental observations of particle-like behavior of light.

Alexander Matzkin

The emergence of classical dynamics in quantum systems... and why we don't see any Bohmian dynamics there.

Emmanuel Fort, Institut Langevin, ESPCI Paris "Dualité onde-particule à l'œil nu"

Il y a près de trois siècles, Newton suggéra que des particules de lumière produisaient des ondes dans un milieu éthéré, comme une pierre ricochant génère des ondes dans l'eau, leurs mouvements étant alors affectés par ces ondes. Newton tentait ainsi de réconcilier dans une même entité les propriétés ondulatoires et corpusculaires de la lumière, a priori incompatibles. Avec l'avènement de la mécanique quantique au XXème siècle, il est apparu que toutes les particules à l'échelle microscopique, de lumière aussi bien que de matière, possédaient cette nature duale, se comportant tantôt comme une onde, tantôt comme une particule. Néanmoins, toutes les tentatives reprenant les caractéristiques de la métaphore de Newton où les particules sont guidées par leur propre onde n'ont pu aboutir. Une des difficultés de ces théories réside dans l'absence d'équivalent macroscopique sur lequel s'appuyer.

Nanoscale Contact as a Source of Plasmons for Plasmonic Nanocircuitries

<u>A. V. Uskov^{1,2}</u>, I. V. Smetanin¹, I. E. Protsenko¹, J. B. Khurgin³, M. Buret⁴ and A. Bouhelier⁴

 ¹ P. N. Lebedev Physical Institute, Moscow, Russia
 ² ITMO University, Sankt-Petersburg, Russia
 ³ Department of Electrical & Computer Engineering, John Hopkins University, Baltimore, Maryland, USA
 ⁴ Laboratoire Interdisciplinaire Carnot de Bourgogne, CNRS-UMR 6303, Université Bourgogne Franche-Comte, 21078 Dijon, France email: alexusk@lebedev.ru

We show that nanoscale metal contacts (constrictions) can serve as an efficient sources of plasmons for future nanoplasmonic integrated circuits. Electron, passing ballistically through nanoscale contact, can emit plasmons with the probability ~0.1 due to multiple collisions with walls of the constriction.

Fig. 1 illustrates an electron passing through a nanoscale metal constriction with a width W(x). The constriction is positioned inside a localized surface plasmon polariton (SPP) mode of the metallic structure. The passage of the electron through the constriction is assumed *ballistic*: the electron does not experience electron-electron and electron-phonon collisions inside nanoscale constriction; the electron's motion is governed only by the potential walls and the field of the mode. If the voltage *V* is applied to the structure, the electron can get the energy eV during its passage through the constriction, and correspondingly, may emit plasmons with the energy $\hbar \omega < eV$. The excitation of SPP mode occurs when electron collides with the walls of the constriction, so that one can talk on *bremsstrahlung*. Collisions are dominating mechanism of emission of plasmons. Our calculations show the probability of the plasmon emission can reach ~10%.



Fig. 1. Electron passage through a nanoscale contact and interaction with a plasmonic field with a modal volume outlined by the dashed line.

I.V. Smetanin

I. Nanoresotron – novel concept of optical nanoantenna excitation through the dissipative instability of DC electric current

I. V. Smetanin¹, A. Bouhelier², I. E. Protsenko¹, and <u>A. V. Uskov^{1,3}</u>

 ¹ P. N. Lebedev Physical Institute, Moscow, Russia
 ² Laboratoire Interdisciplinaire Carnot de Bourgogne, CNRS-UMR 6303, Université Bourgogne Franche-Comte, 21078 Dijon, France
 ³ ITMO University, Sankt-Petersburg, Russia email: <u>smetanin@sci.lebedev.ru</u>

In this report, we propose a novel physical mechanism for excitation of optical nanoantennas, which utilizes the dissipative instability of DC electric current in the quantum well situated near the nanoantenna. Realization of this approach in nanoplasmonics can lead to a new device – *nanoresotron*.

The simplest nanoresotron geometry is shown in the Fig.1. We demonstrate that the 2D DC electric current in the vicinity of surface of absorbing material exhibits the collective instability in the frequency domain where the imaginary part of the material's dielectric permittivity is non-vanishing (Im $\epsilon_m>0$). Light amplification thus emerges as a result of coupling of 2D electron plasma self-consistent oscillations with the slow surface electromagnetic mode. Instability is absent at frequencies at which the material is transparent (Im $\epsilon_m\rightarrow0$).

Self-consistent solution of Maxwell equations for the electromagnetic mode and Euler hydrodynamics equations for the current-carrying 2D electron liquid leads to the dispersion equation which has two kinds of roots, one of which represents conventional rapidly-decaying surface plasmon mode. The presence of DC current leads to appearance of another pair of much more slower solutions with the wavenumber $h\rightarrow\omega/v_0$. The amplification effect Im h<0 arises if the 2D electrons drift velocity v_0 exceeds the sound velocity *s*. Assuming for estimates GaAs QW with the 2D electron density $n_0=10^{12}$ cm⁻², and the drift velocity of the order of Fermi velocity $v_F\approx4.2\cdot10^7$ cm/s, we find the increment is rather large, of the order Im $h\sim10^{5}$ - 10^{6} cm⁻¹, which makes the proposed scheme extremely attractive.



Fig. 1. Simplified scheme of the resotron in which plasma oscillations in carrying DC current QW result in surface wave amplification near metal film.

Nonlinear Optics with surface plasmons and metamaterials

A. Baron^{1,2}, and D. R. Smith²,

¹ University of Bordeaux, CNRS, CRPP, UPR 8641, 33600 Pessac, France ² Center for Metamaterials and Integrated PLasmonics, Duke University, Durham, North Carolina 27708, USA

Nonlinear optics plays a major role in modern photonics applications, because it has enabled the development of ultrafast lasers, optical frequency converters, nonlinear microscopy and optical switches. Surface plasmons, which are coupled oscillations of electromagnetic radiation and charge density at metal/dielectric interfaces, are of particular interest in this context because they carry highly concentrated fields well below the diffraction limit. Since enhancing power densities lowers the nonlinear threshold, surface plasmons may hold the potential for designing efficient or highly sensitive nonlinear devices. The metamaterials and nanophotonics communities have both taken an interest in nonlinear plasmonics recently, though often by designing situations where the dielectric is considered to be the nonlinear material. However metals are known to have high nonlinear optical susceptibilities. The nonlinear physics of metals is rich as it has different origins depending on the time-scale over which the nonlinearity is probed. I will present our work on ultrafast nonlinear plasmonics. We focus on situations where both the metal and the dielectric materials are nonlinear and derive a metric that enables us to determine which of the two materials is the dominant contributor to nonlinearity [1]. We also investigate experimentally the nonlinear response of plasmonic waveguides composed of a single air/gold interface to intense 100 fs pulses. These measurements reveal that the surface plasmon undergoes strong self-action in the form of self-induced absorption [2]. Finally, I will present designs of metasurfaces that present optical bistability and may serve as low-operating power all-optical switches [3].

Bibliographie

[1] A. Baron, S. Larouche, D. J. Gauthier, and D. R. Smith, "Scaling of the nonlinear response of the surface plasmon polariton at a metal/dielectric interface", J. Opt. Soc. Am. B **1**, 9-14 (2015)

[2] A. Baron, T. B. Hoang, C. Fang, M. H. Mikkelsen, and D. R. Smith, "Ultrafast self-action of surface-plasmon polaritons at an air/metal interface", Phys. Rev. B **91**, 195412 (2015)

[2] Z. Huang, A. Baron, S. Larouche, C. Argyropoulos, and D. R. Smith, "Optical bistability with film-coupled metasurfaces", Opt. Lett. 40, 5638-5641 (2015)

Nonlinear optics in double resonant hybrid plasmonic nanoantenna structures

<u>N. Chauvet</u>¹, M. Ethis de Corny¹, G. Laurent¹, M. Jeannin¹, S. Huant¹, A. Drezet¹, G. Nogues¹, G. Dantelle¹, T. Gacoin² and G. Bachelier¹ ¹Institut Néel, CNRS – Université Grenoble Alpes, France ²Laboratoire Charles Fabry de l'Institut d'Optique, CNRS, Orsay, France email: guillaume.bachelier@neel.cnrs.fr

Combining the optical nonlinear efficiency of dielectric media with the electromagnetic field enhancement of double resonant plasmonic nanostructures [1] is a promising way towards nanosized nonlinear photonic devices, with potential applications in quantum optics. However, previous studies on nanostructure arrays [2, 3] have not yet shown significant nonlinear efficiency increase. In this work, we characterize the Second Harmonic Generation (SHG) response of double resonant aluminum antennas. Finite Element Method simulations of SHG [4, 5], extended by

taking into account focusing and collection by large NA objectives [6] as well as substrate effects, quantitatively accounts for the experimental data allowing to infer the origin of the response. These simulations have been applied to the case of aluminum nanoantennas / KTP nonlinear nanocrystal hybrid structures and predict a strong enhancement of SHG. Such structures have been realized with monocrystalline KTP [7] and investigated at the single particle level. Comparison with simulations underlines the crucial role of crystalline orientation.



SHG simulated cartography of (top) a single aluminum antenna and (down) the same antenna with a nonlinear crystal at its apex.

REFERENCES

- [1] K. Thyagarajan et al., Optics Express 20 12860 (2012).
- [2] H. Linnenbank et al., Light: Science & Applications 5 e16013 (2016).
- [3] B. Metzger et al., Nanoletters 14 2867-2872 (2014).
- [4] G. Bachelier et al., J. Opt. Soc. Am. B, 25, 955 (2008).
- [5] G. Bachelier et al., PRB 82 235403 (2010).
- [6] A. Gloppe et al., Nature Nanotechnology 9 920-926 (2014).
- [7] L. Mayer et al., Nanoscale 5 8466 (2013).

SHG from plasmonic nanostructures and metasurfaces: How to design the nonlinear conversion at the nanoscale?

Jérémy Butet, Kuang-Yu Yang, Gabriel D. Bernasconi, and Olivier J. F. Martin Nanophotonics and Metrology Laboratory, EPFL email: jeremy.butet@epfl.ch

In this presentation, we will discuss different methods proposed for the evaluation of the SHG from plasmonic nanostructures. SHG from gold split-ring resonators has been computed using different theoretical methods, namely, Miller's rule, the nonlinear effective susceptibility method, and full-wave computation based on a surface integral equation method [1, 2]. The results confirm that Miller's rule is, in general, not well suited for the description of SHG from plasmonic structures. On the other hand, the comparison of the nonlinear effective susceptibility method with full-wave computations shows that this method permits to

evaluate the SH emission patterns from noncentrosymmetric nanoparticles with good accuracy. However, the nonlinear effective susceptibility method fails to reproduce the multipolar nature of the SH emission from centrosymmetric nanoparticles. This shortcoming is attributed to the intrinsic nature of the nonlinear effective susceptibility method, which neglects the exact positions of the nonlinear sources at the nanoparticle surface.



A split ring resonator

To overcome this limitation, we have established a clear link between SHG and linear absorption. Considering the simple case of spherical nanoparticles, we have clearly identified the most efficient channel for SHG thanks to the generalized Mie theory. This channel corresponds to the excitation of electric dipolar modes at the fundamental wavelength and a quadrupolar second harmonic emission. Interestingly, it is observed that the SH intensity is directly related to the square of the absorbed power which reproduces both the electric field enhancement and the specific size dependence of SHG in the small particle limit. Additionally, the absorbed power can be optimized by controlling the nanoparticle size. These results demonstrate that the optimization of the fundamental electric field is not sufficient for reaching the highest nonlinear conversion in plasmonic nanosystems.

REFERENCES

[1] J. Butet and O. J. F. Martin, J. Opt. Soc. Am. B 33, A8-A15 (2016).

[2] G. D. Bernasconi, J. Butet, and O. J. F. Martin, J. Opt. Soc. Am. B (2016), accepted.

Modal engineering in plasmonic crystalline colloidal systems.

Upkar Kumar¹, Sviatlana Viarbitskaya^{1,2}, Aurélien Cuche¹, Alexandre Bouhelier², Gérard Colas des Francs², Jadab Sharma¹, Christian Girard¹, Erik Dujardin¹*

¹ CEMES CNRS UPR 8011 and Université Fédérale de Toulouse, 29 rue J. Marvig, 31055 Toulouse, France. ² LICB, CNRS UMR 6303, Université de Bourgogne, 9 Av. A. Savary, Dijon, France. ^{*}corresponding author: dujardin@cemes.fr

Abstract- We report on the engineering of the spatial and spectral distributions of plasmon modes in mesoscale 2D crystalline colloids by physical reshaping and dimer assembly. This approach allows us to match the plasmonic characteristics of the complex metallic structure to the specifications of emitters, for example, in order to optimize the resulting in hybrid plasmonics device.

Plasmonics research has traditionally focused on the macroscopic metallic structures that facilitate propagation, waveguiding and routing of surface plasmon polaritons (SPP) or on nanometer-scale particles producing evanescent field, strong confinement volumes by means of localized surface plasmon (LSP) resonances. Both regimes have been exploited in a broad range of technological areas and led to numerous applications, yet much less scrutiny has so far been focused on the intermediate regime of mesoscopic scale systems. These systems sustain higher order plasmonic modes in the visible/near-IR spectral window and combine together the advantages of both LSP and SPP. Multimodal plasmonic systems open a new realm in which the modal behavior is better described by the Surface Plasmon local density of states (SP-LDOS). SP-LDOS is solely governed by the material properties and the boundary conditions set by the structure shape, but is independent of the illumination parameters. The SP-LDOS can therefore be rationally designed to tailor the local spatial and spectral characteristics of the SP modes, while allowing information transfer over micrometer-sized distances. To reveal and exploit such spatio-modal engineering of plasmons, dissipation can be reduced by exploiting the enhanced performances of crystalline metal colloids [1].

We will start with the demonstration that the SP-LDOS distribution of mesoscale 2D nanoprisms (Fig. 1a,b) can be conveniently mapped by two-photon luminescence (TPL) microscopy (Fig. 1c,d) [2, 3, 4]. We will present results revealing the dependence of the spatial and spectral characteristics of the SP-LDOS on excitation wavelength, polarization, particle shape and interparticle coupling. Interestingly, when the excitation power is increased, non-linear thermal effects lead to extremely localized particle melting, which is shown to provide a well-resolved mapping of the SP-LDOS (Fig. 1i-j). [5] All results will be supported by theoretical analysis with our simulation tools based on the Green Dyadic Method (GDM) as shown in Figs. 1e-h. From the multimodal behavior of individual 2D colloids, we will derive two new ways to tailor the spatial/spectral behavior of the higher order SP modes. First, physical reshaping of the colloidal nanoprisms is performed by creating a hole defect by focused ion beam. [6] Depending on the diameter and location of the aperture, the SP-LDOS spectrum and in-plane distribution is moderately perturbated or significantly modified. This allows to selectively modify plasmonic features to be enhanced or suppressed for tailoring hybrid plasmonic systems. Alternatively, when individual colloidal nanoprisms are brought in the near-field coupling regime superstructures are formed that

exhibit new plasmonic modes. In particular, we show that the enhanced field in the gap region can be tuned independently from the higher order modes borne by the metallic prisms, therefore opening a new way to design multimodal antennas [7]. Time allowing, we will consider applying this approach to the tailoring of multimodal information transfer devices [8] as a first step to implement our proposed concept of modal plasmonic circuits. [2]



Figure 1: (a,b) SEM images of triangular and truncated triangular Au nanoprisms. Scale bars are 200 nm. (c, d) Confocal TPL images recorded with horizontally polarized, 700 nm excitation. (e, f) Corresponding simulated TPL images using the GDM [2,3] (g, h) Total SP-LDOS maps and (i, j) corresponding AFM images of prisms with modified surface induced by plasmonic hot printing. [5]

Acknowledgements

The authors acknowledge the financial support of the European Research Council (ERC) (contract number ERC-2 2007-StG Nr 203872 COMOSYEL), Agence Nationale de la Recherche (ANR) (Grant ANR-13-BS10-0007-PlaCoRe), and the computing center CALMIP in Toulouse.

REFERENCES

- 1. C. Girard, E. Dujardin, G. Baffou and R. Quidant. Shaping and manipulation of light fields with bottom-up plasmonic structures. *New J. Phys.* 10, 105016 (2008)
- 2. S. Viarbitskaya, A. Teulle, R. Marty, J. Sharma, C. Girard, A. Arbouet and E. Dujardin. Tailoring and imaging the plasmonic local density of states in crystalline nanoprisms. *Nature Materials* 12, 426 (2013)
- S. Viarbitskaya, A. Teulle, A. Cuche, J. Sharma, C. Girard, E. Dujardin, A. Arbouet. Morphology-induced redistribution of surface plasmon modes in 2D crystalline gold platelets. *Appl. Phys. Lett.* 103, 131112 (2013)
- 4. T. Hoheisen, J. Cordeiro, O. Lecarme, V. Paillard, C. Girard, E. Dujardin, D. Peyrade, A Arbouet. Plasmonic Shaping in Gold Nanoparticle 3D Assemblies. J. Phys. Chem. C, 117, 23126 (2013)
- 5. S. Viarbitskaya, A. Cuche, A. Teulle, J. Sharma, C. Girard, A. Arbouet, E. Dujardin. Plasmonic hot printing in gold nanoprisms. *ACS Photonics*, 2, 744 (2015)
- 6. A. Cuche, S.Viarbitskaya1, J. Sharma, A. Arbouet, C. Girard, E. Dujardin. Modal engineering of Surface Plasmons in apertured Au Nanoprisms, *Scientific Report*, 5:16635 (2015).
- 7. A Cuche et al, in prep
- 8. U. Kumar, S. Viarbitskaya, A. Cuche, A. Bouhelier, G. Colas des Francs, C. Girard, R. Mezzenga, E. Dujardin. in prep