

**Tuesday, December 10<sup>th</sup>**

**Session : NANO-OPTICS & NANOPHOTONICS 1**

*Santenay-Chablis room*

**10h45 - 12h45**

**Keynote speaker: Stéphane BERCIAUD**

*Atomically thin heterostructures made from graphene and  
transition metal dichalcogenides*

**14h15 - 17h45**

**Keynote speaker: Alexandre BOUHELIER**

*Transducing electrons and photons in atomic-scale optical feed*

## Abstracts

**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** Supercontinuum, mid-infrared, dispersion trimming, chalcogenide

## Dispersion trimming for mid-infrared supercontinuum via chalcogenide deposition on a silicon-germanium waveguide

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On-chip mid-infrared (from 3 to 20  $\mu\text{m}$ ) supercontinuum (SC) generation is a technological challenge that is promising to have a strong impact in many different fields such as bio imaging, environmental sensors and security [1]. On-chip SC can be generated via nonlinear effects that broaden the spectrum of a pulse injected in an optical waveguide. Recently, the prediction of great nonlinear properties, wide transparency window from 3 to 15  $\mu\text{m}$  and CMOS compatibility of germanium have attracted a growing interest toward germanium-based platforms [2]. Octave spanning supercontinuum generation up to 8.5  $\mu\text{m}$  has been already demonstrated by our group in a SiGe on Si waveguide [3]. The span and the spectrum of the generated SC are determined by the waveguide's dispersion profile. In general, the dispersive properties are set at the design stage and cannot be adjusted once the device has been fabricated. However, fabrication inaccuracies, surface roughness, surface contamination and the presence of defects may result in a deviation from the targeted dispersion profile. Post-process tuning mechanisms are therefore of great interest to adjust or correct "a-posteriori" the waveguide dispersion to match the target value. Here, we show that it is possible to fine tune the dispersion profile a-posteriori by adding a chalcogenide cladding layer on top of a highly nonlinear SiGe waveguide, introducing a simple post processing tool to control the supercontinuum dynamics and its properties [4]. We experimentally show that an anomalous-to-normal dispersion shift takes place when a chalcogenide top cladding is added.

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**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** chirality; plasmon resonance; layer-by-layer assembly; silver nanowires.

### Assembled hybrid chiral plasmonic metasurfaces

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Recently there has been great interest in developing metamaterials that can control the flow of electromagnetic waves in unprecedented ways. These metamaterials need to have subwavelength dimensions, i.e. in the range of tens of nm for optical applications, and have mostly been manufactured by top-down technologies that have the disadvantage of being quite expensive and slow. Obtaining large areas and regular (3D) ordering is difficult, and nanoparticle self-assembly appears to be a promising alternative. A big challenge, however, still resides in the hierarchical organization of these nanoscale building blocks into two- or three-dimensional structures with well-controlled location, orientation, and spacing across multiple length scales. In particular, chiral assemblies of plasmonic nanoparticles are proposed as an alternate route for the fabrication of optical metamaterials[1].

We will present a novel technique, called Grazing Incidence Spraying, that we have developed for the self-assembly of anisotropic nanoparticles as mono- and multilayer thin films.[2-4] It allows aligning anisotropic nano-objects on large areas with tunable particle density and orientation, and the combination with the Layer-by-Layer assembly[5] is used to build helical (and thus chiral) multilayer thin films in which the composition and orientation can be controlled independently in each layer. The optical properties as function of the thin film geometry will be detailed, with a special emphasis on oriented mono- and multilayers and on helical plasmonic superstructures, which display very high chiroptical activity.

Furthermore, the versatility of the Layer-by-Layer assembly allows combining various kinds of materials with our chiral structures. Here we present the insertion of layers of cyanine dyes, TDBC for instance, into the multilayer films and the investigation of optical phenomena induced by the coupling between the electronic transition of TDBC J-aggregates and the plasmonic resonance of silver nanowires.

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**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** Nonlinear photonics, graphene, guided-wave optics, Silicon nitride

### Saturable absorption of nonlinear graphene coated $\text{Si}_3\text{N}_4$ waveguides

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The unique properties of graphene make it an attractive candidate for optoelectronic and photonic devices [1]. Its planar geometry facilitates its integration onto chip-based photonic devices, which is key to enhance the otherwise poor light-matter interaction with this monolayer-thick material. Among its relevant properties, the graphene nonlinear optical response includes both photo-refractive processes and saturable absorption. Most graphene nonlinear studies combined with integrated optics used the mature silicon-on-insulator platform. However, both high values of the silicon nonlinearity and its undesirable two-photon absorption make it difficult to efficiently harness the graphene nonlinear response and to unambiguously extract it from measurements. Here, we study graphene integrated onto chip-based  $\text{Si}_3\text{N}_4$  waveguides. This very robust material platform exhibits low propagation losses while the large  $\text{Si}_3\text{N}_4$  electronic bandgap (5eV) forbids two-photon absorption and free carrier generation under optical excitation at telecom wavelengths.

We probed the nonlinear response of these hybrid waveguides via measuring the transmission of sub-picosecond pulses at  $1.5\mu\text{m}$ . We focused here on graphene saturable absorption, which is linked to Pauli blocking of photo-excited carriers in the upper band of the Dirac cone. We indirectly probed the excited carrier dynamics by varying the pulse duration and comparing the transmission measurements with simulation results. We indeed developed a model through adapting the nonlinear Schrödinger equation so that it accounts for a power dependent absorption loss, i.e. graphene saturable absorption. This allows us to extract the relaxation lifetime and saturation carrier density values that phenomenologically describe graphene saturable absorption in our hybrid waveguides [2].

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**Thematic Session:** Surface & interface at the nanoscale

**Keywords:** Hybrid nanomaterials, nanostructured silicon, rare earth, optical emission, photoluminescence

### Surface functionalization of silicon with rare earth ions for optical applications

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Because of his indirect gap, crystalline silicon is a poor light emitter. It is challenging for the optoelectronic industry and optical telecommunications to obtain optical properties, including emission, from this material. A possible way to enhance these properties is to modify the surface to become optically active. In this work, complexes containing luminescent elements as lanthanides have been grafted to the silicon surface. These elements are very interesting for optical applications because the wavelength of their emission peaks is almost independent of the environment and an emission from the blue to the near infrared can be obtained, depending on the rare earth. To fix the rare earth based complexes to the silicon surface, the oxidation of the surface is needed to generate reactive groups like silanols. A second required step is the functionalization of the surface by an aminosilane which allows us to link the silica surface and the complexes. To fix the optically active ions, it is necessary to complex the lanthanide ions with a ligand which can react with the ammine group to create a covalent bound of the complex. In this work, we show that the synthesized lanthanide complexes (Tb, Eu, Ce, Yb and Nd) are optically actives and that after grafting on the silicon surface, Tb, Eu and Ce based complexes have a strong luminescence while Yb and Nd based complexes are weakly actives. Excitation and emission processes will be discussed. Moreover, the environment's effect on these molecules has been studied and will be exposed.

**Thematic Session:** Nanophotonics & nano-optics.

**Keywords:** Nanorods, backfocal plane, confocal microscopy, magnetic dipole transitions.

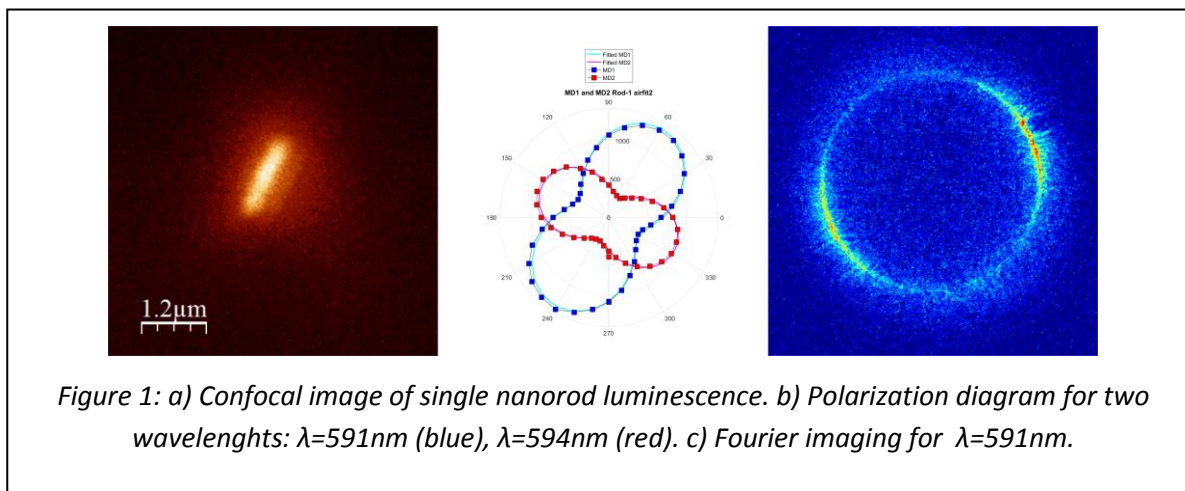
## Quantifying single nanorod magnetic dipole emission by Fourier microscopy.

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Light-matter interaction is principally of electric nature because the magnetic contribution is order of magnitude lower in the optical regime. Engineering optical magnetic dipole (MD) opens new class of applications such as metamaterials, considering artificial MDs [1] or magnetic optical nano-antennas, considering natural magnetic transitions observed in the trivalent lanthanide ions [2].

We studied the MD transition  ${}^5D_0 \rightarrow {}^7F_1$  of  $\text{Eu}^{3+}$  doped  $\text{NaYF}_4$  single nanorods. Using confocal microscopy, we measured two magnetic dipoles emissions, induced by the crystal-field fine structure, polarized parallel and perpendicular to the rod axis (Fig 1.a and b) [3]. Afterwards, following the work of Taminiau *et al.* [4] we quantified for each single nanorod the magnetic and electric contributions to the emission line by Fourier microscopy (Fig 1.c).



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**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** Quantum dots, indium phosphide, nanocrystals, semiconductors, hot-injection synthesis

### Synthesis and characterization of III-V based quantum dots for light emission

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In the race of miniaturization of optical systems like display devices, the development of nanometer sized particles with high photoluminescence quantum yield (PLQY) and covering a wide color range constitutes a tremendous issue. Colloidal semiconductor nanocrystals, commonly called quantum dots (QDs) seem to meet all the conditions for displays application. QDs have generally a diameter lower than 10nm, a high PLQY and are able to cover a wide spectral range from UV to IR. QDs having currently the highest optical performances and the longest stability under irradiation (UV) contain cadmium (e.g. CdSe), a strongly toxic element prohibited in Europe (RoHS and REACH directives). The purpose of this work is to develop Cd-free QDs by thermolysis, a method allowing an accurate control of the stoichiometry and QDs' final diameter. To this end, we have focused our attention on indium phosphide (InP) QDs. Recently, aminophosphines were integrated as precursors in the synthesis to provide an economic alternative to synthesize InP in presence of indium halides [1]. Three types of shells were grown onto InP cores, ZnS [1], ZnSe [1,2] and ZnSe/ZnS (this work), in order to enhance their photostability and their optical efficiency. By modifying the InP core growth time or the nature of halide precursors, QDs covering wavelength from 480 to 680nm were synthesized. QDs have been characterized structurally (XRD, SAXS), morphologically (HAADF-STEM), chemically (ICP-OES...) and optically (PLQY, excitation/emission spectra, fluorescence decays). Finally, the surface chemistry has been analyzed by several technical tools (<sup>31</sup>P, <sup>1</sup>H, <sup>13</sup>C MAS NMR, IR).

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**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** Helical traveling-wave nanoantenna, plasmonics, subwavelength polarization optics, optical chirality, spin-orbit interaction.

## Subwavelength Polarization Optics Using Individual and Coupled Helical Travelling-Wave Nanoantennas

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A wide variety of optical applications and techniques demand control of light polarization. Manipulation of light polarization has recently experienced extraordinary advances with the emergence of plasmonics, leading to the concepts of polarization meta-optics [1]-[4]. However, all the structures developed so far rely on collective optical effects on arrays of nano-elements. They are therefore restricted to areas much larger than the wavelength of light, which limits design strategies in polarization control. Tailoring light with individual subwavelength devices would overcome limits but it remains a challenge [5], [6].

On the basis of the spin-orbit interaction of light, we demonstrate a helical travelling-wave nanoantenna to produce directional light beam of tunable polarization through a swirling plasmonic effect. Our optical nanoantenna differs from existing helical plasmonic structures by its non-resonant nature, thus extending the concept of helical travelling-wave antenna to optics [7]. Four closely packed HTNs are shown to locally convert an incoming light beam into four beams of tunable polarizations and intensities. Moreover, by coupling HTNs of opposite handedness, we demonstrated a subwavelength waveplate-like structure providing a degree of freedom in polarization control that is unachievable with usual polarization optics and metamaterials [8].

Taken as individual or coupled nanostructures, such nanoantennas lay a basis for subwavelength polarization optics, thus opening new perspectives in photonic information processing, polarimetry, miniaturized displays, optomagnetic data storage, microscopy, sensing and communications, etc.

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**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** Nanolaser, Photonic Crystal, Excitability, III-V on SOI

### Proposal for a spiking nanolaser

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Artificial neural networks proved to be efficient for signal recognition and especially image recognition. Although their software implementation through conventional Von-Neumann processors has been extremely successful, efficiency considerations in training have recently motivated direct hardware approaches taking direct inspiration from the mode of operation of the brain to process information.

We focus on a possible photonic implementation of a spiking neuron. The response is based on the excitability that is possible in semiconductor lasers when they are configured with a saturable absorber [1]. This has been demonstrated experimentally in a vertical-cavity surface-emitting laser [2]. To implement an all photonic network equivalent to the one of biological neurons the issue of interconnectivity has to be addressed.

This is why in the work presented here, we numerically study the spiking mechanisms in a new technology of hybrid InP on SOI photonic crystal nanodiode laser [3] which are extremely interesting with their compact size and low energy consumption to develop efficient optical neural networks. This technology is modified by creating a gain and a saturable absorber section by selectively pumping a specific region of the photonic crystal wire cavity. We adapt the Yamada model [4] for a bisection nanolaser and predict an excitable dynamics mimicking a neuromorphic response. The energy efficient operation (fJ excitation energy,  $\mu\text{W}$  pump power), compactness (10  $\mu\text{m}$ ) and speed (100 ps) of the technology considered here would allow the creation of a highly scalable neuromorphic photonic system.

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**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** plasmonic, hot electrons, Multi-Photon Luminescence, Schottky barrier, sub-bandgap photodetector

## Remote Generation of Hot-Electron assisted by Surface Plasmon Polaritons

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Hot-electrons within metallic devices are quite promising to set off tremendous physico-chemical reactions and photocatalysis [1]. Indeed, the high energy of hot-electrons remaining within nanostructures yields high local temperature at their boundaries. For this purpose, controlling their generation at the nanoscale and their dynamics within plasmonic devices is a key for the future development of hybrid hot-electrons technologies [1]. However, the efficiency of hot-electrons generation is quite low and Surface Plasmons Polaritons (SPPs) are generally used to enhance it through a mechanism called plasmon-induced hot-electron generation. Indeed, nanoantennas, which allow for localized SPP resonances, are known to produce efficiently hot-electrons under femtosecond laser pulse excitation in the near IR [2].

Here we present the first remote generation of hot-electrons assisted by propagative SPPs [3]. For that a plasmonic waveguide efficiently launch SPPs which carry enough energy to generate hot-electrons at distance up to 10 microns. More precisely, surface plasmons loose energy towards the medium leading to hot-electrons generation which relax emitting a multi-photon photoluminescence (MPPL). The spatial distribution of plasmon-assisted hot-electrons is achieved by recording the MPPL signal by hyperspectral near-field microscopy techniques [4]. We find that their spatial and energy distribution are mapping by the surface plasmon field intensity and that such hot-electrons do not diffuse on distance higher than few nanometers. Following this work, we are now studying the transfer between hot-electrons generated by plasmonic gratings and a thin Silicon semiconductor layer with the ultimate goal to fabricate an ultrafast photodetector with a bandwidth driven by the hot-electron lifetime in the picosecond range [2]. Our preliminary results will be presented in this communication.

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**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** nanoplasmonic, nanofabrication, near-field photopolymerization, gold nanoparticles

### Plasmon-triggered living photopolymerization for elaboration of hybrid polymer/metal nanoparticles

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Over the past ten years, localized surface plasmons have been exploited for triggering and controlling photochemical and photophysical reactions. Achieving hybrid nanosystems coupling metal nanostructures with enhanced effect is a major challenge for applications in nanophotonics, sensors or spectroscopy. In our case, surface plasmon dipolar resonance of individual metal nanoparticles was used as an optical near-field source to locally trigger free radical polymerization of an acrylic monomer: metal nanoparticles embedded in a photopolymer sensitive at the resonance wavelength are irradiated under the polymerization threshold but close enough to this threshold to initiate the free-radical polymerization thanks to electromagnetic near-field enhancement. We will report on the physicochemical and optical parameters controlling the photopolymerization process in near-field. Kinetic parameters were shown to be of paramount importance to control the polymerization in highly confined space. Different photopolymers were used in this context to finely tune the final properties of the nanoparticles.

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**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** plasmon; waveguide; optical interconnect; polymer ; SU-8

## Engineering of gold-polymer long-range plasmonic waveguides as single-mode optical interface chips

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We have developed two different concepts of plasmonic optical interfaces. Potential applications include single-mode low-loss interconnectors in high speed data communication systems like e.g. in data centers or high performance computing, in particular for interfacing photonic chips with optical printed circuit boards or different boards together, where perpendicular coupling is often needed. This work was performed in the framework of European project PhoxTroT. Both concepts are based on long-range surface plasmon polariton structures and both comprise a  $\sim 15$ -nm thin gold strip core embedded into polymer claddings [1]. The typical propagation loss measured is often less than 1 dB/mm.

The first development is a mechanically flexible waveguide array, an elastomeric ribbon which can be bent with radius of curvature down to the millimeter making it attractive for the facile adaptation to different geometrical interfacing configurations [2]. The second is an on-chip waveguide array, rigid but integrating a  $45^\circ$  plasmon mirror based on total internal reflection, providing perpendicular wave re-direction [3]. For both, we have developed original fabrication methods and have explored their optical performances at  $1.55 \mu\text{m}$  with TM-polarized light and butt-coupled single-mode fibers. For applications, we also coupled them with optical PCBs and VCSELs.

In the rigid integrated mirror concept, reflection is obtained by a prismatic cavity aligned with the waveguide  $90^\circ$  turn point. This is realized by lithography in both upper and lower SU-8 claddings. To obtain a flat, efficiently reflecting plane, we developed a self-alignment method for the two cladding lithography steps.

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[2] C. Vernoux, et al. Opt. Mater. Express, 8 (2018) 469–484.

[3] L. Markey, et al. paper submitted to Opex



**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** Metallic nanoparticle gratings; Organic emitters; Plasmonics; Near and Far field effects; Organic Light Emitting Diodes.

## Controlling the coupling between organic emitters and surface modes of metallic nanoparticles gratings for efficient organic light emitting diodes

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Understanding the influence of ordered metallic nanoparticles on the optical responses of organic emitters is crucial to develop efficient and new organic light emitting diodes (OLED). Moreover, a deep understanding of the phenomena is required in order to efficiently control their emission properties.

This work concerns a study of integrated periodic metallic nanostructures on the optical and electrical responses of organic light emitting diodes. Various Ag nanoparticle arrays with periods ranging from 180nm to 500nm have been realized. The obtained structures have been experimentally characterized by performing lifetime measurements and radiative emission pattern characterizations. Two coupling regimes can be distinguished: the so-called short period regime ( $p < 280\text{nm}$ ), dominated by highly localized plasmonic modes (LSPR), showing significant lifetime reductions and homogeneous emission patterns; and a so called long-period regime ( $p > 280\text{nm}$ ) dominated by collective hybrid lattice modes (SLR), showing slight lifetime reductions and highly directive emission patterns. Therefore, tuning the array parameters offers a possibility to select a specific coupling effect at near or far field zones. Afterward, two plasmonic organic light emitting diodes using gratings of 180nm and 480nm have been investigated. *IVL* measurements and angular characterizations of the emitted power have been performed. In addition to the electrical and the optical enhancements, the angular patterns show directional improvements of the emission for plasmonic OLEDs compared to the regular one. The obtained features demonstrate that metallic nanoparticles arrays act as an integrated nano-tool that allows a high control of the emission properties in terms of intensity, directivity and efficiency.



**Thematic Session:** (Nanophotonics&nano-optics, nanomaterials, Surface & interface at the nanoscale)

**Keywords:** (Raman, Resonance, Scattering, Nanostructures, Nano manipulations)

### Resonance Raman spectroscopy of Nano-designed assembly

**Bayle Maxime<sup>1</sup>, D'Orlando Angéline<sup>1</sup>, Louarn Guy<sup>1</sup>, Moussaoui Saïd<sup>2</sup>, Winterauer Dominik<sup>3</sup>, Batten Tim<sup>3</sup>, Humbert Bernard<sup>1</sup> (Calibri 11, Bold, black) ,**

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Our group is developing an experimental approach combining Atomic Force Microscopy and confocal-Raman micro-spectroscopy [1], where AFM microscope is used to image and to manipulate nano-particles [2] under the confocal optical microscope coupled at the Raman spectrometer. Our optical device allows us on the one hand to scan the resonance effects by tuning the wavelengths of excitation and on the other to spatially scan sample with nano-step to super-resolve numerically the hyperspectral mappings [3].

This talk will show the results obtained with some symmetric structures of assembling of metallic nano-particles (Nps), in the vicinity of a single and isolated carbon nanotube (CNT). We will investigate the different possibilities of coupling between Nps structures and CNTs by studying the Resonance Raman profiles[2].

In order to spatially resolve the optical near field effects in the inelastic scattering process by these nano-assemblies, we will show our experimental tentative to resolve at the subwavelength scale the recorded hyperspectral images. We will discuss our super-resolved Raman results as a function of the different sub-wavelength-scale geometries of aggregates. We will discuss the different interactions between metallic NPs aggregates and the nano-carbon objects, including their impact on resonance effects. In particular, we will focus on the enhancement of the local electrical field by metallic nano-structures to probe single objects

[1] A. D'orlando Thesis (2015) Nantes.

[2] A. D'Orlando, M. Bayle , G. Louarn and B. Humbert (2019) Materials 12(9) 1372.

[3] D.J. Winterauer, D. Funes-Hernando, J.L. Duvail, S. Moussaoui, T. Batten and B. Humbert (2019) Applied Spectrosc. 73(8) 902-909.



**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** Micro and nanosystems, MEMS, NEMS, SNOM, THz

## Sensors for near-field microscopes based on micro–nano-systems and their application to THz imaging at the nanoscale

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Nano-characterization with custom setups based on Atomic Force Microscope (AFM) and Scanning Near-Field Optical Microscope (SNOM) have today a limited choice and availability of probes. For instance, quartz sensors used in AFM ([1]) have high stiffness constants, no standardized tips, and signal chains limited by their intrinsic low frequencies. Moreover, commercial cantilevers technologies don't allow tip heights above 15  $\mu\text{m}$ , which limits optical access and apertureless SNOM techniques beyond mid-IR wavelengths.

Here we present our technological research making use of modern Micro-nano-Electro-Mechanical-Systems (MEMS/NEMS) [2]. Two sensors architectures, vertical and lateral, enable to use microresonators or cantilevers with integrated detection and high solid angle around the tip. To make possible SNOM imaging up to THz frequencies, we achieved high tip length up to 300 $\mu\text{m}$ . The stiffness constants are in the range 70 N/m to 2000N/m for working frequencies from 160 kHz to 4.2 MHz. We will present the first imaging results on a SNOM microscope using a 10 $\mu\text{m}$  IR source, and in a second configuration with a setup operating a 2.5THz source.

### References

[1] F. J. Giessibl, "Atomic Resolution of the Silicon (111)-(7x7) Surface by Atomic Force Microscopy," *Science* (80-. ), vol. 267, no. 5194, pp. 68–71, 1995.

[2] B. Walter, E. Mairiaux, M. Faucher, " Atomic Force Microscope based on vertical silicon probes, vol. 110, no. 24, 243101, 2017.



**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** nonlinear nanophotonics, nano-antennas, open systems, modal formalism

### Quasinormal mode expansion for nonlinear nano-optics

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We propose an alternative to the multipolar expansion for studying nonlinear processes in optical nanoantennas. Subwavelength resonators can be studied as non-Hermitian open cavities, whose eigenstates are called quasinormal modes (QNMs). Their expansion allows to better explain the different contributions from the modes involved in three-wave-mixing processes. For any external optical excitation, the QNM formalism enables one to reconstruct the total and scattered fields as a linear combination of the modal fields, whose excitation coefficients are known analytically.

Here this method is generalized to nonlinear processes, where the usual external excitation is replaced by a local nonlinearly generated current source. Our approach differs from other methods since the involved modal contributions can be studied separately, in the near and far fields, and enable to quantitatively establish the mode matching.

To highlight the features of this model, we consider the case of second harmonic generation (SHG) in an AlGaAs nanocylinder with 240 nm radius and 400 nm height, excited by a plane wave (1.55  $\mu\text{m}$  wavelength, 1  $\text{GW}/\text{cm}^2$  intensity) at normal incidence.

The generalization of QNM expansion to nonlinear processes offers at least two main advantages: (a) the opportunity to separately study the resonances, in order to gain physical insight on the origin of the nonlinear generation, which can provide design guidelines for the development of novel nanoscale devices; (b) an advantageous tool in terms of computational cost, as the eigenvalue problem is solved once for a given structure, and the method quickly retrieves analytically the excitation coefficients.





**Thematic Session: Nano for imaging, diagnosis & theranostics**

**Keywords: Multi-modal Raman & Enhanced Darkfield Hyperspectral Microscopy**

## **Improving Nano-Theranostic Studies with Multi-Modal Raman and Enhanced Darkfield Hyperspectral Microscopy**

**Byron J. Cheatham<sup>1</sup>**

1. CytoViva, Inc., Auburn Research Park, Auburn, AL USA

### **Abstract**

Effective nano-theranostic studies require that researchers can effectively observe and measure the interactions of nanoparticles and the cell or tissue environment where they are targeted. HORIBA Scientific and CytoViva, Inc. now provide a new multi-modal Raman and darkfield hyperspectral microscopy system that is highly effective for nano-theranostic research. This system includes patented enhanced darkfield optics that enable observation of nanoparticles as small as 10nm in diameter when isolated in solution and in cells and tissue. The optical hyperspectral imaging capabilities of the system enable large spatial area spectral measurements and spectral mapping of these nanoparticles or their drug load based on their surface plasmon resonance, fluorescence emission or Rayleigh scatter. Additionally, Raman measurements from the identical field of view can be captured, which provides quantitative molecular fingerprint confirmation of the nanoparticles or other sample elements. Finally Raman mapping of unique sample elements can be performed when required. This presentation will provide a detailed overview of this new multi-modal imaging and spectral measurement capability. Specific illustrations of plasmonic, SERS active, metal oxide and lipid based nanoparticles interacting with cells and tissue will be presented.

**Wednesday, December 11<sup>th</sup>**

**Session : NANO-OPTICS & NANOPHOTONICS 1**

*Santenay-Chablis room*

**10h45 - 13h15**

**Keynote speaker: Natalia DEL FATTI**

*Linear and ultrafast plasmonics with a single nano-object*

**14h30 - 16h30**

**Keynote speaker: Fabrice RAINERI**

*III-V semiconductors on Silicon nanophotonics*

# Abstracts



**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** Thermoplasmonics, Pump-probe, 2Tmodel, heat process

**Generalized Two-Temperature Fitting Algorithm  
for Ultrashort Laser Heating of Metal Film and Nanoparticles  
to spatially and temporally study heat propagation**

**J-F Bryche<sup>1,2</sup>, P. Bresson<sup>1,2,3</sup>, J. Moreau<sup>3</sup>, M. Besbes<sup>3</sup>, P-L. Karsenti<sup>1,2</sup>, D. Morris<sup>1,2</sup>, P.G. Charette<sup>1,2</sup>, M. Canva<sup>1,2</sup>**

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2. *Institut Interdisciplinaire d'Innovations Technologiques 3IT – Université de Sherbrooke - Sherbrooke, Canada.*
3. *Laboratoire Charles Fabry; Institut d'Optique Graduate School; CNRS UMR-8501; Université Paris-Sud, Université Paris-Saclay, Palaiseau, France.*

**Abstract:**

The dynamics of photothermal phenomena are governed by a three-step mechanism involving electronic heating through photon absorption ( $\sim 100$  fs), thermalization via electron-phonon interaction ( $\sim$  ps), and thermal diffusion outside the focal point ( $\sim$  ns). These mechanisms can be studied and resolved separately using pump-probe measurements with short ( $< 100$  fs) laser pulses. A pump-probe based measurement setup was developed with the capability to acquire the spectro-temporal optical response of ultrashort laser heating of metallic thin films or nanostructures, with high temporal and spatial resolution. In order to quantitatively explain these optical signals, we have developed a complete thermal and electromagnetic numerical model which can be used to fit the experimental data with a minimum of parameters. This model combines the 2-temperatures model, well-known in the field of ultrafast heating processes, with a FEM electromagnetic code to take into account the 3D geometry of the sample. Electronic and lattice thermal conduction through the metal and substrate are also included. To validate our model, we have realized multiple experiments on thin gold films and periodic array of gold nanorods, showing an excellent agreement with our model on a large range of pump fluence and film thicknesses. Interestingly, the thermal losses can be a significant source of energy for photosynthesis, chemical reactions or bio-surgery and need to be study.

**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** gold, nanorods, inelastic light scattering, acoustic vibrations

### Inelastic light scattering by vibrations of long narrow gold nanocrystals

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2. MONARIS, UMR 8233 CNRS-Sorbonne Université, Paris, France
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4. ILM, UMR 5306 CNRS-Université Claude Bernard, Villeurbanne, France

Gold nanorods and nanobipyrisms have attracted a wide interest in the last decade due to their potential applications in many fields. This results from their shape-dependent optical and electronic properties and from their crystallinity that is likely to play a key role for tailoring properties and functionalities of metal nanoobjects [1]. The goal of this work is to investigate the vibrational dynamics of nanorods and nanobipyrisms through inelastic light scattering experiments and numerical models in order to assess its sensitivity to the shape, size and crystallinity of the nanocrystals. To this end, we synthesized single crystalline gold nanorods and twinned pentagonal nanobipyrisms using a method based on ref [2]. Their length is typically of several tens of nanometers and the aspect ratios vary between about two and five.

Very low frequency Raman spectra measured with a tandem Fabry-Pérot interferometer exhibit several narrow bands in the 20-200 GHz frequency range. These Raman peaks originate from acoustic vibrations confined in the nanocrystal. Their frequencies vary mainly as either the reciprocal length or the reciprocal diameter of the nanoobjects depending on the nature of the mode as shown by calculations performed in the framework of continuum elasticity and taking into account the elastic anisotropy of gold [3] and the shape of the nanocrystals. The comparison of the measured and calculated frequencies help assign the different bands observed in the Raman spectra and highlight the sensitivity of low-frequency Raman spectra to the dimensions (length and diameter) and the inner crystalline structure of the nanocrystals.

- 1- Y. Tang and M. Ouyang, Nature Mater. 6, 754 (2007).
- 2- L. Vigderman and E. R. Zubarev, Chem. Mater. 25, 1450 (2013).
- 3- L. Saviot, Phys. Rev. B 97, 155420 (2018).



**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** plasmonic, Rayleigh scattering, remote Raman, EELS, nanowire

## Optical properties of advanced gold-based nanowires and exploitation as plasmon mediated remote Raman sensor

**Funes-Hernando Daniel<sup>1</sup>, Peláez-Fernández Mario<sup>2</sup>, Winterauer Dominik<sup>3</sup>, Mevellec Jean-Yves<sup>1</sup>, Arenal Raúl<sup>2,4</sup>, Batten Tim<sup>3</sup>, Humbert Bernard<sup>1</sup>, Bayle Maxime<sup>1</sup>, Duvail Jean-Luc<sup>1</sup>**

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### Abstract

Plasmonic nanowires make possible to exploit altogether the propagative nature of surface plasmon polaritons (SPP) in a guided way, and the strong field enhancement at the nanowire tips. They are thus of great interest to be exploited as building blocks for plasmonic based devices, such as nano-sensors.

Here, we report on the proof-of-concept of a SPP-mediated remote Raman effect for specially designed coaxial nanowires [1]. Remote Raman spectroscopy is based on the separation by many micrometres of the excitation laser spot on one tip of the nanowire, and the Raman detection at the opposite tip. Cylindrical coaxial nanowires consisting of a gold core to propagate SPP and a Raman-emitting shell of poly(3,4-ethylene-dioxythiophene) (PEDOT) were synthesized and exploited for the proof-of-concept.

However, the in-coupling of an excitation optical signal with the nanowire and the out-coupling of the plasmon-mediated signal at a remote location are challenging aspects to obtain a strong enough signal to be exploited. This challenge has been tackled by transforming the gold nanowire tips with dry laser heating treatments to obtain dog bones like nanowires. The plasmonic properties of these original nanowires have been determined by an EELS-STEM study. As shown by a Rayleigh scattering study at the nanoscale, the excrescence located at one or at the two tips of the nanowires provides enhanced in and out coupling.

[1] D. Funes-Hernando, M. Peláez-Fernández, D. Winterauer, J.-Y. Mevellec, R. Arenal, T. Batten, B. Humbert and J.L. Duvail *Nanoscale* (2018) 10, 6437 – 6444



**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** DNA Nanotechnology, Plasmonics, Strong-coupling, Spontaneous emission

## Reaching a strong coupling regime between fluorescent emitters and a plasmonic resonator using DNA

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DNA self-assembly is a flexible and robust technique to controllably introduce a controlled number of quantum emitters in the gap of plasmonic resonators. In particular, DNA-templated gold particle dimers enhance the spontaneous emission rates of single molecules by more than two orders of magnitude (Nat. Commun. **3** (2012) 962), providing bright single-photon sources with lifetimes below 10 ps and quantum yields above 50% (ACS Nano **10** (2016) 4806).

It is important to note that increasing the coupling between fluorescent molecules and plasmonic resonators, or maximizing the emission yield, require different structural parameters. Using electrodynamic simulations, we will show how reaching a strong coupling regime between a single dipolar emitter and a dimer of gold spheres is only feasible for interparticle spacings below 4 nm and with 40 nm gold particles, conditions in which the emission quantum yield is negligible. Furthermore, we will demonstrate how optimizing the antenna properties, in particular using plasmonic nanocubes, should allow strong coupling conditions to be observed in conjunction with high luminescence quantum yields at visible frequencies.

Taking into account these geometrical constraints, we will discuss recent experimental results on the optimization of the interaction between multiple identical fluorescent molecules and a single gold resonator in order to reach a strong coupling regime. This allows us to observe reproducibly, in scattering spectroscopy, plasmon mode splitting in the longitudinal resonance of single 40 nm gold particle dimers coupled to 5 ATTO647N molecules.

**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** quantum plasmonics, Purcell factor, Fano profile, pseudo-modes

### Fano profile and plasmonics Purcell factor

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Light-matter interaction is generally quantified thanks to the Purcell factor  $Q/V$  where  $Q$  and  $V$  refer to the quality factor and mode volume of the cavity, respectively. Cavity quantum electrodynamics (cQED) relies on the extremely high quality factor but at the price of diffraction limited sizes. That is why strong efforts have been done since a decade to transpose cQED concepts to nanophotonics and plasmonics, taking benefit from the deeply subwavelength confinement of localized surface plasmon polaritons (LSP) [1].

Therefore, an exact definition of the plasmonic Purcell factor is of strong interest to engineer quantum plasmonics devices but also for a better understanding of the light matter interaction at the nanoscale. When defining the mode volume of LSP, a great difficulty originates from the radiation leakages that prevents to extrapolate standard cQED definition [2]. Recently, Sauvan and Lalanne consider a fully classical approach based on quasi normal modes to define a complex mode volume that exactly describes the dipolar-plasmon coupling process [3]. Here, we will discuss a fully cQED approach and will show that the imaginary part of the mode volume can be ascribed as a Fano parameter measuring the contribution of the radiation leakages into the quantum emitter/LSPs coupling process [4].

[1] Tame *et al*, *Quantum plasmonics*, Nat. Phys. **9**, 329 (2013)

[2] Colas des Francs *et al*, *Plasmonic Purcell factor and coupling efficiency to surface plasmons. Implications for addressing and controlling optical nanosources*, J. Opt. **18**, 094005 (2016).

[3] Sauvan *et al*, *Theory of the spontaneous optical emission of nanosize photonic and plasmon resonators*, Phys. Rev. Lett. **110**, 237491 (2013).

[4] Varguet *et al*, *Non-hermitian Hamiltonian description for quantum plasmonics : from dissipative dressed atom picture to Fano states*, J. Phys. B : At., Mol. Opt. Phys. **52**, 055404 (2019).



**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** Surface plasmon, Semiconductor, Nanocrystals, Tungsten oxide

## Structure Property Relationship in Plasmonic Semiconductor Nanocrystals

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Doped semiconductor nanocrystals are an emerging class of materials hosting localized surface plasmon resonance (LSPR). Their wide spectral range (from visible to the entire IR regions) and post-synthetic tunability through doping promise new plasmon-assisted active optical materials and devices. Recent studies discovered different semiconductor species that perform efficient LSPR. However, the nanocrystals' structural impact on their LSPR remain poorly explored.

In this presentation, we illustrate how the structural factors collaborate to exhibit novel LSPR properties that are unseen from metal hosts. For instance, in hexagonal cesium-doped tungsten oxide ( $Cs_xWO_{3-y}$ ) nanocrystals, the crystalline anisotropy causes a strong LSPR band-splitting into two distinct and intense peaks. This finding highlights that multiple structural factors can be controlled to create and exquisitely tune the multimodal LSPR bands from semiconductors, while the shape anisotropy has been the unique means to tune metal LSPR. We also show the rapid and reversible modulation of LSPR by chemical and electrochemical charging/discharging of carriers in the plasmonic semiconductor nanocrystals, which is applicable to sensors and electrochromic smart windows.



**Thematic Session:** Nano for imaging, diagnosis & theranostics

**Keywords:** gold nanoparticles, photothermal therapy, NIR laser, magnetic hyperthermia

## Visible/NIR photothermia with gold bipyramids and nanostars

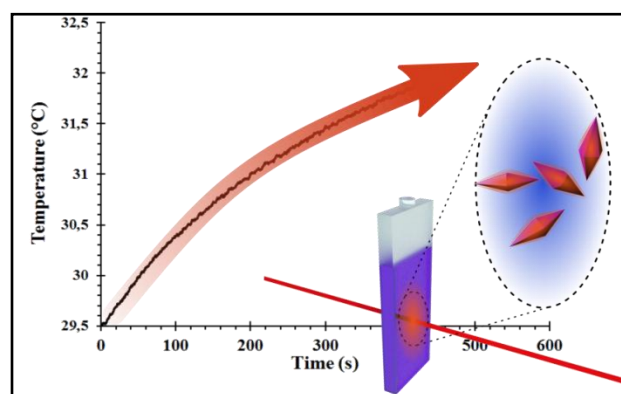
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### Abstract

Photothermal efficiency of anisotropic gold nanoparticles (AuNPs) is studied as a potential way of cancerous cells photothermal treatment (PTT). Specific AuNPs with bipyramidal and star-like shapes were synthesized following a control seeded-growth method[1, 2] in order to shift the plasmonic absorption peak in the near-infrared (NIR) range of transparency for living tissues. Their photothermal properties were investigated by irradiating aqueous samples in the visible (658 nm) and the NIR (785 nm) ranges, showing a neat increase in local temperature, up to 3°C, at low gold concentration (20-60 µg/mL). The photothermal phenomenon was found to vary linearly with the laser power and to depend strongly on the position of plasmon resonance peak, with respect to the laser irradiation wavelength. Corresponding specific absorption rate (SAR) values were calculated, in order to quantify the photothermal efficiency independently from the nanoparticle concentration, and resulted in high values, up to 3300 W/g. In addition, experiments were performed with a medical NIR LED FluoBeam800™ apparatus (750 nm illumination at a 13mW/cm<sup>2</sup> flux) to replicate biomedical treatment conditions. The SAR evaluation showed a relevant temperature increase, with the particular case of gold bipyramids that exhibited synergistic heating when applying simultaneously an alternating magnetic field at 146 kHz (22.5 kA/m) and the NIR irradiation, as presumably ascribed to Eddy currents produced in the suspension of electrically conducting nanoparticles. Such photothermal behavior underlines the promising use of gold nanoparticles in mild hyperthermia cancer treatment with NIR local irradiation.



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**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** gold nanoparticles, pulsed-lasers, ophthalmology, eye floaters

### Photoablation of human vitreous opacities by light-induced vapor nanobubbles

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Floater are collagen aggregates that can form in the vitreous of the eye due to structural changes and which cast shadows on the retina. Current therapies are based on laser treatment with an yttrium garnet laser (YAG) or vitrectomy (i.e. the replacement of the vitreous by a saline solution) but most of the time, floaters are left untreated. Besides, the efficacy of YAG laser treatment is still controversial and vitrectomy can be associated with important side effects (cataract formation, endophthalmitis).

In this work we propose a nanotechnology-based approach for the treatment of floaters using the plasmon properties of gold nanoparticles (AuNPs) which we named 'photo-cleaning'. When exposed to pulsed-laser light (typically a nanosecond laser), AuNPs able to bind floaters, heat up and generate vapour nanobubbles (VNBs) due to the evaporation of the surrounding water. These VNBs will then burst providing sufficient mechanical energy to fragment floaters.



To maximize the probability of AuNPs to reach and bind floaters after intravitreal injection, we coated them with hyaluronic acid (HA-AuNPs) and observed they could bind and fragment collagen I fibers and human floaters obtained from patients.

Since smaller AuNPs have a higher threshold to generate VNBs we hypothesized smaller HA-AuNPs (10 nm), would be able to clusterize on the floaters to locally decrease the threshold to generate VNBs preserving the surrounding vitreous and ocular tissues from being damaged. Besides, using smaller particles implies less scattering for the patient and higher mobility in the vitreous to further maximize binding to floaters.

Our strategy can effectively and rapidly destroy floaters by using lower energy levels compared to YAG therapy and paves the way for the use of pulsed-lasers and nanotechnologies in the posterior segment of the eye.



**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** All-dielectric resonators, Nonlinear metasurfaces, Second harmonic generation, Mie-resonances, Polarization control

## Polarization- and diffraction-controlled second harmonic generation from semiconductor metasurfaces

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### Abstract

We demonstrate the possibility of controlling the polarization state of second harmonic (SH) generation by periodic nanostructures. The knowledge of Mie-resonances in all-dielectric nanoantennas enables to exploit the peculiar scattering features of isolated resonators to design the polarization state of emitted beam.

The building block of the proposed nonlinear metasurfaces are (100)  $\text{Al}_{0.18}\text{Ga}_{0.82}\text{As}$  nanocylinders laying on a monolithic aluminum oxide optical substrate. These resonators are designed to operate at 1550 nm and the SH generated signal is collected in back-reflection. We propose two control mechanisms for SH polarization. In the former we set the fundamental frequency (FF) linear polarization and we exploit the orientation of elliptical base nanocylinders to break the symmetry of the system. In the latter we fix the geometry of circular base nanocylinders and rotate FF polarization. In the assumption of independent resonators, the total SH radiated intensity from a periodic array can be expressed as the product between the far-field response of single emitters and the array factor of the metasurface.

Experimental characterization confirms that our 2D arrays enhance collection efficiency of more than one order of magnitude in a numerical aperture  $\text{NA} = 0.1$ , and we detect either horizontally or vertically polarized SH with 80% polarization degree. Therefore, with our metasurface we are able to decouple polarization and directivity properties of SH signal and redirect the emission close to the normal direction, enabling the integration of these devices for on-axis applications.



**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** Optomechanics, Nonlinear dynamics, Coupled systems, Chaos

## Noise-enhanced detection and chaos with a driven electromechanical resonator

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In ordinary consciousness, noise is associated with the term “hindrance”; it is considered as a nuisance that can lead to communication or signal transmission failure or prevent the detection of a weak signal to be measured. However, under certain conditions the impact of noise can be counter-intuitively a resource. In nonlinear systems, noise can induce novel regimes leading to the formation of synchronized structures, coherence between the output and the input of resonators or even to the amplification of weak signals. In other words, noise can play a constructive role, with potential benefits for signal processing or measurement for instance. Single or coupled Nano-Opto-ElectroMechanical resonators whose design is based on suspended photonic crystal membrane could be used as a toy-system to pursue two main goals: stochastic amplification and chaos. As such noise-aided processes might have a strong impact in noise-assisted applications including signal processing or sensing.



**Thematic Session:** Nanophotonics&nano-optics

**Keywords:** Brillouin scattering, acoustic phonons, nanomechanics, optomechanics, nanophononics

## Brillouin/Raman spectroscopy in optophononic resonators working in the 20-300 GHz frequency range

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Inelastic scattering of light by acoustic phonons has potential for the tailored generation of frequency combs, laser-line narrowing, and all-optical data storage. To be efficient, these applications require strong optical fields and a large overlap between the optical and acoustic modes. Control over the shape of the acoustic spectrum is highly desirable. In this presentation, we introduce a monolithic Brillouin generator based on high-frequency nanoacoustic resonators operating in the 20-300 GHz range. These devices allow independent design of the Brillouin spectrum and the optical mode profile while presenting an optimal spatial overlap between the acoustic and optical fields. We developed a free-space filtering technique enabling the measurement of relatively low-frequency signals with a simple experimental setup. The presented one-dimensional optophononic structures can be extended into tridimensional micropillars, which can, at the same time, be readily integrated into fibered and on-chip architectures.

**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** photonic crystal, stress-engineering, cylindrical cavity resonator, slow Bloch modes

## 3D confinement of light inside rolled-up 2D photonic crystal membranes

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Since pioneer works of Prinz [1], nanofabrication techniques based on self-rolling of pre-stressed membranes have developed in a thriving way and led to the creation of 3D nanostructures with complex geometries (tubes, coils, Origamis [2]). In particular, opto-fluidic detectors based on rolled-up semi-conducting membranes have been developed using whispering gallery modes in the membrane wall [3]. Meanwhile, remarkable dispersion features of photonic crystal membrane (PCM) were exploited to build efficient and compact mirrors as an alternative to traditional Bragg stacks in vertical emission laser applications [4]. Combining the two previous approaches allows the production of a new family of versatile 3D photonic microstructures.

We report here the design, the fabrication and the characterization of “photon cages” under the form of rolled-up 2D PCM mirror which can confine light in 3D and most importantly in low index media (air). Parameters of the PCM were adjusted to obtain an efficient reflector (reflectivity  $R > 95\%$ ) over a large spectral range ( $> 100$  nm) in near infrared. The cylindrical cavity resonator model and FDTD simulations were used to predict the optical response of the rolled-up membrane. Tubular cavities were then fabricated using stress engineering technique. Finally, near-field (SNOM) measurements were carried out to investigate the modes in the hollow of the cavity.

Near-field experimental characterization has revealed the presence of cavity modes as predicted by numerical and analytical computations. It brought the proof of concept for photon cages and also attested the quality of self-rolling fabrication technique.

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**Thematic Session:** Nanophotonics & nano-optics, nanomaterials

**Keywords:** Symmetry protected modes, photonic crystal, Lithium Niobate, sensing applications, Fano resonance

## Excitation of symmetry-protected modes in photonic crystals for electric field detection applications

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Electric field sensors based on light-matter interaction are highly desirable in many applications such as medical (EEG, ECG) or military applications. To allow confinement of the electromagnetic field in a non-linear material (lithium niobate LN) and thus obtain an exaltation of the electro-optical effect [1], a resonance with a high quality factor (Q) is necessary. A Fano resonance, well known in quantum physics resulting from the interference between a discrete mode and a continuum, would be an ideal solution. Based on this type of resonance, a sensor based on photonic crystals in LN has already been proposed by our team and manufactured on a membrane in a configuration called Suzuki [2]. In trying to improve performance, we have designed a structure based on a square-array photonic crystal of cylindrical patterns etched in a LN membrane with resonance having a better quality factor. After some investigations, it was found that this is a protected symmetry mode that was revealed by a numerical simulation based on a centered finite difference algorithm. This type of spatial discretization inevitably introduces a break in the symmetry of the object and leads to the excitation of this type of protected mode [3,4].

We have demonstrated by FDTD numerical simulation that with resonance having a high quality factor Q (up to 105) and a suitable extinction factor (up to 50%) we can develop an electric field sensor with a sensitivity of 70 mV / m resulting from a local confinement of the electromagnetic field reaching a factor of 200. Comparing with reference [5], a temperature sensor based on our configuration will have a sensitivity lower than  $\Delta T = 10^{-4}$  °C instead of 17 °C obtained in the reference. A technological mastery at the nanometer scale would be essential for the exploitation of this type of resonance.

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**Thematic Session:** Nanophotonics & nano-optics

**Keywords:** symmetry breaking, nanocavity, driving, dissipative, coherent

## Symmetry breaking in coherently driven-dissipative coupled nanocavities

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Spontaneous symmetry breaking (SSB) is a fundamentally important process that underlies many outstanding physical phenomena, from phase transition in metamaterials to the emergence of the Nambu-Goldstone and Higgs modes in particle physics, and can be exploited for a wide variety of applications. A highly promising platform is nanodevices that could be harnessed for targeting diminution of the photon number or to improve integration capacity. SSB has recently been observed in evanescently coupled nanolasers, driven above the laser threshold, as the breaking of the mirror ( $Z_2$ ) symmetry. However, most theoretical developments on few photon SSB in nonlinear coupled cavities have been carried out in a coherently driven-dissipative framework, because of the relative simplicity of quantum models implementing coherent driving. In this respect, these driven-dissipative nonlinear coupled-cavity models constitute a paradigm of open quantum systems. Importantly, SSB in a coherently driven-dissipative system has not been demonstrated to date.

Here, we report on the first experimental realization of symmetry breaking in coherently-driven dissipative nanocavities. Our experiments are based on two evanescently coupled Indium Phosphide-based photonic crystal nanocavities, that are free-space and coherently driven at wavelengths that ensure low absorption Kerr-like nonlinearity. Upon symmetric driving conditions, in which one expects the measured cavity intensities to be equal, we show the coexistence of two different mirror states, where light is localized in either of the two cavities, over a broad range of driving power. Furthermore, we demonstrate the possibility to induce a switch between the two symmetry-broken states by perturbing with a single incoherent short-pulse.