# NOPWORKSHOP ON<br/>NONLINEAR OPTICS<br/>AND PLASMONICS2023September 6-8, 2023 | Lecce, Italy

# **BOOK OF ABSTRACTS**







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# About

It is our great pleasure to welcome you to NOP2023, the first international workshop on Nonlinear Optics and Plasmonics, held in Lecce, Italy, on September 6-8, 2023. This conference gathers an excellent group of keynote speakers from different topics and covers both fundamental aspects and applications, featuring several internationally renowned invited speakers, as well as poster contributions. This conference is supported by the Italian Ministry of Foreign Affairs and International Cooperation (KR23GR01), the South Korean Ministry of Science and ICT, the National Research Foundation of Korea, Istituto Italiano di Tecnologia, and Pohang University of Science and Technology in South Korea.

### Organizers



Dr. Cristian Ciracì, Istituto Italiano di Tecnologia, Italy.



Prof. Junsuk Rho, POSTECH, South Korea

### Institutional supporters









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# Venue



**Lecce** is a historic city in southern Italy, one of the most important cities of Apulia, featuring a rich collection of baroque architectural monuments and buildings. Lecce lies at the heel of Italy's boot between the Adriatic and Ionian Sea in the "Salento" area. The region offers kilometers of coastal landscape, blue sea-fringed bays, dunes and white sand beaches; it holds a rich and delicious gastronomic tradition and offers a lively and dynamic cultural scenario.

While in Lecce, enjoy a walk through the city's unique architecture. Some must-see places are: Piazza Duomo, Basilica di Stanta Croce, Anfiteatro Romano, Ex Convitto Palmieri, Porta Napoli, and Porta San Biagio

Lecce offers many food and drinks options within walking distance from the conference location. **Osteria da Angiulino** is a great and affordable option for a taste of Salento's traditional cuisine. Two of the best pizza places are **400 Gradi** and **La Gigante** where you can find "Naples" or "Salento" style pizza, respectively. **La Barca di Mario** is one of the best seafood option in the city center. Meat lovers cannot miss **La Locanda del Macellaio**, one of the best traditional "braceria" In Lecce, or **II Carrettino** for the best burgers in town. If you are looking for a fancy dinner, **3 Rane Restaurant** is a little hidden gem. For late night drinks, check-out **Quanto Basta** and **Prohibition** for cocktails, or **Cantiere Hambirreria** for beers. This is not by all means an exhaustive list. Lecce is continuously evolving and new places to try out pop up every week. Do not be afraid to explore.

The conference will be held at **Officine Cantelmo**, a modern and vibrant conference center in the heart of Lecce's historic center, in **V.Ie M. de Pietro, 8a**.

The **poster sessions** will be held at **Ex-Convento degli Agostiniani**, situated at 350 m from the main conference location.

# **Conference dinner**



The **conference dinner** will be held on Thursday night, September 7th, at **Palazzo Tamborrino Cezzi** situated **Via Guglielmo Paladini, 50**, within 15 minutes walk (through the city center) from the main conference location.

Palazzo Tamborrino Cezzi was erected in the mid-XVI century by Giacomo Mele, who came from an old family in the town. The house still has the renaissance rooms on the ground floor from this period. At the beginning of the seventeenth century it passed into the hands of the de' Giudici family, who had lived in the town for a long time and were of Genoan origin: Cola Maria de' Giudici embellished the house to such an extent that her contemporaries defined it as a "sumptuous palace". Following this the owners were briefly the Jesuits, the Staybano family, then the Capece family and from the beginning of the eighteenth century to the Unification of Italy, the Paladini family, a family of French origin, aristocrats, intellectuals and politicians. From the second half of the nineteenth century the mansion has belonged to the Tamborino family, who have been responsible for the structural and artistic changes that characterize its present appearance: the Cezzi family, who now own it, are descendants on the maternal side. The nineteenth century rebuilding of the house expressed the neoclassical taste of the period, which in the South of Italy was linked to the eclectic elegance of the Art Nouveau movement, and also to the Moorish and Oriental details which are still alive in this area.

# Maps

### From Officine Cantelmo to Ex-Convento degli Agostiniani



### From Officine Cantelmo to Palazzo Tamborrino Cezzi



# Awards



We are delighted to announce the support to young researchers of the De Gruyter, peerreviewed, open access journal **Nanophotonics** (IF: 7.923). The journal covers recent international research results, specific developments in the field and novel applications. Every issue contains a balanced combination of invited review articles, regular articles, letters and opinionated reports. Nanophotonics focuses on the interaction of photons with nano-structures, such as carbon nano-tubes, nano metal particles, nano crystals, semiconductor nano dots, photonic crystals, tissue and DNA. The journal covers the latest developments for physicists, engineers and material scientists, working in fields related to plasmonics, metamaterials, and nanophotonics.

Nanophotonics is pleased to offer prizes to the **best posters** of NOP2023.

# Instructions



The following information is provided to assist authors with the preparation of their presentation. You shall comply with these instructions and guidelines in order to make sure that your presentation will go smoothly. If you need any further information please write to cristian.ciraci@iit.it.

### For oral presenters

You shall arrive at the room where your presentation is scheduled 15 minutes prior to the first presentation in that session and make yourself known to the session Chair. You shall stay for the whole session – for your paper and the papers of other speakers in that session – in order to enable the delegates who wish to speak with you at the end of the session to be able to find you easily. Your presentation shall last 20 minutes (40 minutes for the Keynotes) including questions. The Questions & Answers sessions should not exceed 5 minutes (10 minutes for the Keynotes). The Chair will time your presentation and provide you with a 2-minute warning. Please keep a strict eye on the time during your presentation.

### For poster presenters

Your poster shall have a vertical or "portrait" orientation not exceeding the following dimensions:  $841 \times 1189$  mm (A0 format). The poster shall be readable from a distance of 2-3 meters. Avoid fuzzy images; make sure all graphics are high-resolution and easily visible. Lists with your name and assigned poster ID will be available and displayed in the venue. The **poster sessions** will be held at **Ex-Convento degli Agostiniani**, situated at 350 m from the main conference location.

Program at a Glance

Wed, Sep 6		Thu, Sep 7		Fri, Sep 8	
10:00-10:50	Welcome & registration	09:00-10:40	Session 4	09:00-10:40	Session 8
10:50-11:00	Opening	10:40-11:10	Coffee	10:40-11:10	Coffee
11:00-12:40	Session 1	11:10-12:30	Session 5	11:10-12:10	Session 9
				12:10-12:30	Awards & Closing
12:40-14:00	Lunch	12:30-14:00	Lunch		
14:00-15:20	Session 2	14:00-15:20	Session 6		
15:20-15:50	Coffee	15:20-15:50	Coffee		
15:50-17:10	Session 3	15:50-17:10	Session 7		
17:20–18:40	<b>Posters 1</b> Agostiniani	17:20–18:40	<b>Posters 2</b> Agostiniani		
		20:00–22:30	<b>Dinner</b> Palazzo Tamborino Cezzi		

# Wednesday, September 6<sup>th</sup>

10:00 - 10:50	Welcome reception & Registration		
10:50-11:00	Opening statement		
	Session 1 – Chair:	Cristian Ciracì	
11:00-11:40	Philippe Lalanne CNRS	Non-Hermitian resonators: the divergence, the Qs and Vs of their quasinormal modes	
11:40-12:00	<b>Stéphane Kena-Cohen</b> Polytechnique Montreal	Nonlocal effects in ENZ antennas and new materials for second-order nonlinear optics	
12:00-12:20	<b>Prineha Narang</b> University of California, Los Angeles	Nonlinear optics in topological quantum materials and axion electrodynamics	
12:20-12:40	<b>Stefano Palomba</b> University of Sydney	On-chip SOI hybrid plasmonic nanofocusser	
12:40-14:00		Lunch	
Session 2 – Chair: Michele Celebrano			
14:00-14:40	<b>Daniele Brida</b> University of Luxembourg	Ultrafast electron transport at the nanoscale	
14:40-15:00	Alexandre Bouhelier CNRS	(On-line) Photonic memristors	
15:00-15:20	<b>Yonatan Sivan</b> Ben-Gurion University	The nonlinear optical response and electron dynamics in ITO	
15:20-15:50	Coffee break		
	Session 3 – Chair:	Prineha Narang	
15:50-16:10	<b>Martin, Schultze</b> Graz University of Technology	Strong light waves controlling charge and spin dynamics	
16:10-16:30	Giovanni Manfredi CNRS	Driving orbital magnetism in metallic nanoparticles through plasmonic effects	
16:30–16:50	<b>Luca Sortino</b> Ludwig-Maximilians- Universität München	Monolithic van der Waals metasurfaces	
16:50–17:10	<b>Alexandre Baron</b> University of Bordeaux	Optical properties of self-assembled dense spherical clusters of plasmonic nanoparticles	
17:20-18:40	Poster Session 1 with Drink	ks & Food (Ex Convento degli Agostiniani)	

# Thursday, September 7<sup>th</sup>

Session 4 – Chair: Junsuk Rho			
09:00-09:40	Bumki Min KAIST	Photonic Temporal and Time Crystals	
09:40-10:00	<b>Andreas Tittl</b> Ludwig-Maximilians- Universität München	Photonic bound states in the continuum for spectrally selective nanophotonics	
10:00-10:20	<b>Carlo Forestiere</b> University of Naples	Plasmonic and dielectric resonances of homogeneous objects: from quasistatic to the full-wave regime	
10:20-10:40	<b>Marcus Ossiander</b> Graz University of Technology	Holes in silicon enable extreme ultraviolet metaoptics	
10:40-11:10		Coffee break	
	Session 5 – Chair:	Yonatan Sivan	
11:10-11:30	<b>Angela Demetriadou</b> University of Birmingham	Persistent sub-radiant states with plasmonic nanocavities	
11:30–11:50	<b>Antonio I.</b> <b>Fernandez-Dominguez</b> Universidad Autónoma de Madrid	Inverse design for quantum nanophotonics: qubit entanglement and Bell state preparation	
11:50-12:10	<b>Martijn Wubs</b> Technical University of Denmark	Modeling collective light emission by a few solid-state quantum emitters	
12:10-12:30	Kyoung-Duck Park POSTECH	Tip-enhanced cavity-spectroscopy to control excitonic behaviors at the nanoscale	
12:30-14:00		Lunch	
	Session 6 – Chair: Mari	a Cristina Larciprete	
14:00-14:40	<b>N. Asger Mortensen</b> University of Southern Denmark	Surface-response formalism for mesoscopic electrodynamics in plasmonic nanostructures	
14:40-15:00	<b>Sang-Hyun Oh</b> University of Minnesota	(On-line) CMOS-based terahertz camera based on quantum-dot-enhanced upconversion	
15:00-15:20	<b>Domenico de Ceglia</b> University of Brescia	Optical image differentiation with nonlinear flat optics	
15:20-15:50		Coffee break	
	Session 7 – Chair:	Martin Schultze	
15:50-16:10	<b>Crina Cojocaru</b> Universitat Politècnica de Catalunya	Large nonlinear efficiency enhancement in the visible and UV ranges from plasmonic gold nanogratings	

16:10-16:30	Michael Scalora Second and third harmonic genera		
	US Army	from aluminum nanostructures	
16:30–16:50	Fan Yang	A transformation optics approach to	
	Sichuan University	nonlinear and nonclassical plasmonics	
16:50–17:10	<b>Michele Celebrano</b> Politecnico di Milano	Free-space interferometric routing of	
		upconverted light by dielectric	
		metasurfaces	
17:20–18:40	Poster Session 2 with Drink	ks & Food (Ex Convento degli Agostiniani)	
20:00-22:30	<b>Conference dinner</b> (Palazzo Tamborino Cezzi)		

# Friday, September 8<sup>th</sup>

Session 8 – Chair: Michael Scalora			
09:00-09:40	Natalia Litchinitser	Beam shaping and frequency conversion	
	Duke University	in nonlinear all-dielectric metasurfaces	
09:40-10:00	Mihail Petrov	Optical torque induced by resonant	
	ITMO University	harmonic generation in dielectric	
		nanostructures	
10:00-10:20	Su-Hyun Gong	2D semiconductor multilayers for	
	Korea University	ultra-thin nanophotonic platform	
	Antoine Moreau		
10:20-10:40	Université Clermont	Making spatial dispersion useful	
	Auvergne		
10:40-11:10	Coffee break		
Session 9 – Chair: Francesco Todisco			
	Joel Cox	Nonlinear papenlacmonics with atomically	
11:10-11:30	University of Southern	this sectorial	
	Denmark	thin materials	
11:30-11:50	Tommaso Venanzi	Free-electron infrared nonlinearities in	
	Istituto Italiano di Tecnologia	heavily doped InGaAs nanoantennas	
11:50-12:10	Arseniy Kuznetsov	Passive and tunable flat optics with	
	A*STAR	dielectric nanoantennas	
12:10-12:30	Awards and Closing Remarks		

# **Detailed program**

### Wednesday, September 6<sup>th</sup>, 2023

### Session 1 [11:00 – 12:40] Chair: Cristian Ciracì, Istituto Italiano di Tecnologia, Italy

# 11:00 Non-Hermitian resonators: the divergence, the Qs and Vs of their quasinormal modes (Keynote)

### Tong Wu and Philippe Lalanne

*Laboratoire Photonique, Numérique et Nanosciences, CNRS-IOGS-Université de Bordeaux, Bordeaux, France* 

Micro and nanoresonators enhance many light-matter-interaction processes and are used in various modern applications in photonics. They are open systems and their modes (often called quasinormal modes) are always leaky, have a finite lifetime and thus, are defined as source-free solutions of Maxwell equations at a complex frequency. The analysis of the non-Hermitian behavior of such resonator in their modal basis has posed great difficulty in the past, but the situation has changed drastically recently thanks to recent progresses in the complex analysis of open electromagnetic systems.

### $11{:}40$ Nonlocal effects in ENZ antennas and new materials for second-order nonlinear optics

### Stéphane Kena-Cohen

### Polytechnique Montréal, Montreal, Canada

In the first part of the talk, we will discuss the experimental realization of epsilon near-zero photonic gap antennas. We will show how the inclusion of nonlocal effects in the electronic response of the ENZ material is essential to improve the quantitative agreement between full wave simulations and experiment. In particular, nonlocal effects lead to sharp resonances in the field enhancement that are completely absent in the local model. These resonances, corresponding to points of vanishing group velocity, lead to nonlocal field enhancements 4-6x greater than in the local simulations. In the second part of the talk, we will talk about our efforts to develop a flexible platform for second-order optical nonlinearities based on thermally evaporated thin films of small molecules. Using the interplay between permanent dipole and hyperpolarizabilities, we obtain values of d31 and d33 ranging from 5-10 pm/V on arbitrary substrates.

### 12:00 Nonlinear optics in topological quantum materials and axion electrodynamics

### Prineha Narang

### University of California, Los Angeles, U.S.A

Parametric optical nonlinearities are critical to a wide spectrum of photonic technologies, from optical parametric oscillators to frequency combs to quantum information processing. Optical nonlinearities also serve as a powerful method for mapping material properties including the symmetries of electronic structure. Optical nonlinearities are generally very small in conventional materials as they depend on higher order effects. Parallel to these technical needs, the field of topological materials has seen the prediction and discovery of a large number of massless, three-dimensional linear dispersion systems known as Dirac and Weyl semimetals. It was soon realized that these materials may offer a rich new material phase space for extending the nonlinear effects of graphene including the role of topology and Berry connection. In this context, I will present our recent work on predicting the optoelectronic

and nonlinear properties of Dirac and Weyl semimetals with an emphasis on figures of merit (FoMs) that we will evaluate for these new Weyl and Dirac semimetals that captures the confinement and nonlinearity, to describe the second and third order susceptibilities and electro-optic coefficients of the materials. Next, I will discuss our recent results on the multiphoton spectroscopy of a dynamical axion insulator. We demonstrate a two-step protocol for the unambiguous optical identification of the collective axion mode in such a system. Looking ahead, I will discuss how collective responses in topological quantum materials can be unambiguously identified in nonlinear electrodynamical probes.

### 12:20 On-Chip SOI Hybrid Plasmonic Nanofocusser

<u>Stefano Palomba</u><sup>1,2</sup>, Oliver Bickerton<sup>1,2</sup>, Fernando Diaz<sup>1</sup>, Thomas Kasebier<sup>3</sup>, Stefanie Kroker<sup>4</sup>, Ernst B. Kley<sup>3</sup>, C. Martijn de Sterke<sup>1,2</sup>, Alessandro Tuniz<sup>1</sup>

<sup>1</sup>Institute of Photonics and Optical Science, School of Physics, The University of Sydney, Australia; <sup>2</sup>The University of Sydney Nano Institute, The University of Sydney, Australia; <sup>3</sup>Institute of Applied Physics, Friedrich Schiller Universiat Jena, Germany; <sup>4</sup>Physikalisch-Technische Bundesanstalt, Germany

Silicon-on-insulator (SOI) chip-based hybrid-plasmonics combines advantages of all-integrated optoelectronic functionality and deep sub-wavelength optical confinement. However, the large difference in modal areas limits the coupling efficiency from conventional waveguides, in turn reducing the efficiency of light-matter interactions like nonlinear optical functions. Here we design, fabricate, and experimentally characterize an efficient on-chip SOI hybrid plasmonic nanofocussing waveguide, with a tip as small as 10 nm. The device operates by rotating the fundamental TE mode of a standard SOI waveguide to a TM mode of a hybrid plasmonic waveguide, and subsequently focussing it to the nanoscale. Since plasmonic nanofocussing cannot be demonstrated by far-field, linear experiments, we measure the second harmonic generation (SHG) at the tip. The wavelength-scale propagation distances make phase matching unnecessary - we can thus correlate the measured SHG to the degree of nanofocussing of the pump at the tip. The dramatic increase in SHG intensity for the sharpest tip indicates strong focusing; by comparing the slopes of the three curves, we can experimentally quantify the degree of maximum intensity enhancement – which is a factor of  $\times$ 7.5 with respect to the strip case. Thanks to the excellent agreement between experiment and simulations we conclude through simulations that the pump light is focused down to a mode area of approximately 40 nm2, resulting in a 1200 intensity enhancement with respect to the silicon waveguide input. This represents the first TM plasmonic nano-focusser to monolithically interface with an industry-standard TE-input SOI waveguide. This work lays the foundations for efficient and compact on-chip, deep sub-wavelength sources and sensors, bridging integrated photonic circuits and metallic nanostructures - potentially down to single-atoms -enabling operation in nonlinear plasmonic and quantum regimes.

### Session 2 [14:00 – 15:20] Chair: Michele Celebrano, Politecnico di Milano, Italy

### 14:00 Ultrafast electron transport at the nanoscale (Keynote)

Daniele Brida

Department of Physics and Materials Science, University of Luxembourg, Luxembourg

We demonstrated that single-cycle pulses of minute energy content may result in extremely nonlinear optical phenomena at the nanoscale by exploiting an electronic circuit with a few-nanometre gap between the tips of an optical antenna. The strong electrical bias provided by the field contained in ultrashort optical pulses was harnessed to drive tunnelling and ballistic acceleration of electrons to generate a current through the free-space gap with PHz bandwidth

### 14:40 Photonic memristors

### Alexandre Bouhelier

Laboratoire Interdisciplinaire Carnot de Bourgogne CNRS UMR 6303, Université de Bourgogne, France

Electronic components integrating nanometer scale gap in their design were also crucial to the advent of novel form of computing. Memristors for instance are programmable voltage-dependant resistive devices deployed nowadays in cognitive hardware systems such as artificial neural networks, neuromorphic and reservoir computing. Memristive operation relies on resistance switching triggered by the electroformation and disruption of conductive path- ways within a nanometer-scale dielectric gap. Charge transport occurs by an electro-chemical reduction of metal ions aggregating to conductive filaments, or by migration of mobile defects, such as oxygen vacancies and nanoclusters. In this presentation, we introduce an atomic scale memristive device capable of emitting photons during resistive switching, superseding thus the need for an external optical source. Our device features the compact footprint of transistors and compatibility with the emerging memristive technology. We identified three mechanisms producing photons with vastly different properties. The crossover between emission regimes depends on the history of the memristor and its operating conductance. Our results suggests that this new generation of memristor pave the way for multidimensional neural networks using both electrons and photons as information carrier.

### 15:00 The nonlinear optical response and electron dynamics in ITO

Subhajit Sarkar, leng-Wai Un, Yonatan Sivan

### Ben-Gurion University, Israel

Low electron density Drude (LEDD) materials such as transparent conducting oxides, plasmonic nitrides, became popular candidates for high-efficiency nonlinear optical applications, due to their unique near-infrared "epsilon near zero" point. Their nonlinearity is extremely large, reaching 100's of percent of the refractive index/permittivity. Peculiarly, despite the large body of related impressive experimental demonstrations, their theoretical modeling was mostly coarse, and has not yet conclusively elucidated the origins of the giant optical response. Here, we close this knowledge gap and provide a ""first principles"" model of the response of LEDD materials to ultrafast illumination. For concreteness, we focus on Indium Tin Oxide (ITO). Our model includes the Boltzmann equation (BE) complemented by a phonon dynamics equation, and an easy-to-use coarse-grained extended two temperature model (eTTM). We find the electron heat capacity of ITO to be smaller, but the electron-phonon energy transfer rate to be comparable to that in noble metals. This leads to stronger heating of the electrons, and to a faster cooling compared to noble metals [Fig. left(a)]. Surprisingly, the intense illumination and associated high electron temperature, may cause the effective chemical potential to become negative, thus, transiently converting the ITO into a semiconductor [Fig. left(b)]. We also find that the drastic increase of the real part of the permittivity shifts the resonance from the pump such that the absorptivity drops rapidly with increased illumination intensity [Fig. right(a)]. Consequently, the phonon temperature increases sub-linearly with the pump peak-intensity, reaching the melting point of at 500 GW/cm2 [Fig. right(b)]. This explains, for the first time to our knowledge, the experimental observation of the high damage threshold of ITO and shows that the ITO nonlinearity is not saturable, but rather thermal as for noble metals.

### Session 3 [15:50 – 17:10] Chair: Prineha Narang, University of California Los Angeles, USA

### 15:50 Strong light waves controlling charge and spin dynamics

### Martin Schultze

### Graz University of Technology, Austria

I will discuss a set of experiments demonstrating the capability of single cycle optical fields to deliberately drive currents and alter the magnetization state of solid bulk materials and nano-heterostructures.

### 16:10 Driving orbital magnetism in metallic nanoparticles through plasmonic effects

<u>Giovanni Manfredi<sup>1</sup></u>, J. Hurst<sup>1</sup>, P.-A. Hervieux<sup>1</sup>, R. Sinha-Roy<sup>2</sup>, P. Oppeneer<sup>3</sup>

<sup>1</sup>Université de Strasbourg, CNRS, Franc; <sup>2</sup>Aix-Marseille Université, France; <sup>3</sup>Uppsala University, Sweden

The topic of this contribution is the generation of large magnetic fields in non-magnetic materials through polarized laser fields. Transfer of angular momentum from helicity-controlled laser fields to a nonmagnetic electronic system can lead to the creation of magnetization. The underlying mechanism in metallic nanoparticles has been identified as the inverse Faraday effect (IFE), whereby a quasi-static magnetic field is generated by an external oscillating lase field and is proportional to the laser intensity. Here, we show that the IFE can be strongly amplified in small gold nanoparticles thanks to plasmonic effects. If the laser frequency matches the plasma frequency of the conduction electrons in the metal (surface plasmon resonance), a strong oscillating electric field is excited in the nanoparticle. Through the IFE, this internal self-consistent field generates a sizeable magnetization, of the order of tens of Bohr magnetons. The primary contribution to the magnetization comes from surface currents generated by the self-consistent field. The effect is maximum for circularly polarized laser fields and disappears for linearly polarized fields. This plasmonic IFE is studied here using both a simplified quantum hydrodynamic model and fully quantum simulations based on the time-dependent density functional theory. This is an important step in the ultrafast manipulation of magnetic effects in nano-objects via electromagnetic waves, which may find applications for the storage, writing, and reading of information based on optical means.

### 16:30 Monolithic van der Waals metasurfaces

### Luca Sortino<sup>1</sup>, Stefan A. Maier<sup>1,2,3</sup>, Andreas Tittl<sup>1</sup>

<sup>1</sup>Chair in Hybrid Nanosystems, Faculty of Physics, Ludwig-Maximilians-Universität München, Munich, Germany; <sup>2</sup>School of Physics and Astronomy, Monash University, Clayton, Australia; <sup>3</sup>Department of Physics, Imperial College London, London, United Kingdom

Van der Waals (vdW) materials, such as hexagonal boron nitride (hBN) and Transition Metal Dichalcogenides (TMDCs) semiconductors, are layered crystals with exceptional properties to investigate light-matter interaction at the nanoscale. In their atomically thin form they exhibit appealing features, such as tightly bound excitons and optically addressable defects, while in their bulk form they exhibit giant optical anisotropy and large refractive index values (n>4), larger than common semiconductor materials, making them a favorable candidate for the realization of low-loss optical resonances in all-dielectric nanophotonic structures. In our work, we leverage the physics of quasi bound states in the continuum (qBIC) to achieve high quality (Q) factors optical resonances in symmetry-broken dielectric metasurfaces. Notably, our approach is monolithic, meaning that it is exclusively composed of vdW materials, and allows to realize optical resonances with Q factors above 102 through a two-step fabrication process. We demonstrate spectral tuning over the whole visible spectrum in hBN gBIC metasurfaces and enhanced light-matter coupling with intrinsic spin defects in hBN. In the latter, we observe a remarkable 25-fold enhancement of the photoluminescence intensity and spectral narrowing of the defect emission, with linewidth below 4 nm full width at half-maximum. Moreover, our platform opens exciting opportunities for strong light-matter coupling, demonstrated in the clear anti-crossing behavior between qBIC resonances and intrinsic excitons in monolithic TMDC WS2 metasurfaces, exhibiting Rabi splitting up to 116 meV under ambient conditions and independent on the material's intrinsic losses. Our results demonstrate how merging gBIC photonic metasurfaces with vdW materials paves the way to the realization of novel hybrid nanophotonic platforms and room temperature polaritonic devices.

16:50 **Optical properties of self-assembled dense spherical clusters of plasmonic nanoparticles** <u>Alexandre Baron</u><sup>1,2</sup>, Ranjeet Dwivedi<sup>3</sup>, Ashod Aradian<sup>1</sup>, Virginie Ponsinet<sup>1</sup>, and Kevin Vynck<sup>4</sup> <sup>1</sup>Univ. Bordeaux, CNRS, CRPP, UMR 5031, France; <sup>2</sup>Institut Universitaire de France, France;

### <sup>3</sup>ENSEMBLE3, Centre of Escellence, Wolczynska Poland; <sup>4</sup>Univ. Claude Bernard Lyon 1, CNRS, iLM, France

Densely-packed spherical colloidal clusters of metallic inclusions, also known as plasmonic balls, have garnered a lot of interest recently, owing to their remarkable scattering behaviors and potential applications. Using an emulsion route, we have been able to produce dense spherical balls and show that they act as resonant Huygens scatterers, where the intereferences of mullipoles of even and odd parity lead to coherent forward scattering. We shall review recent designs and realizations of such Huygens scatterers using self-assembly. The interaction of light with inhomogenous spheres such as these clusters is notoriously difficult to describe theoretically. We have studied numerically the electromagnetic behavior of plasmonic balls composed of many particles using high-precision T-matrix calculations. We have shown that it is empirically possible to find an equivalent effective medium description for the clusters, taking into account spatial dispersion. We find that the average scattered field as well as the average inner field of a spherical cluster as computed from the T-matrix approach can be equivalently obtained by an extended Mie theory where three effective parameters are used to describe the inner effective medium, namely an electric permittivity  $\varepsilon_{eff}$ , a magnetic permeability  $\mu_{\rm eff}$ , and a longitudinal wavevector  $k_{\rm L}$ . The latter two account for strong interparticle couplings entailing spatial dispersion effects , which cannot be neglected in dense systems near the plasmonic resonance. Our study therefore shows that (within the range of studied sizes), it is possible to treat a cluster of plasmonic particles as a sphere made of a spatially-dispersive effective equivalent medium, even for high concentration in particles. This work broadens the range of effective parameters that can be obtained and exploited in the design of meta-atoms and metamaterials.

### Poster Session 1 [17:20 - 18:40]

1. Second-harmonic generation in monocrystalline gold nanostructures: implications of anisotropic second-order susceptibility

Sergejs Boroviks, Olivier J.F. Martin

- 2. **Plasmonic-like hot-electron nonlinear photoluminescence from patterned ITO thin films** F. Dell'Ova, G. Colas-des-Francs, E. Dujardin, <u>Alexandre Bouhelier</u>
- 3. Quasi-periodic snap-buckling mechanisms in polymeric nano-bubbles: Toward highly efficient radio-acoustic energy transducers Salvatore Buonocore, Aliaksandr Hubarevich, Francesco De Angelis
- 4. Ab initio study of a metal-molecule system for polaritonic chemistry applications Lucia Cascino, S. Corni and S. D'Agostino
- Mid-infrared Berreman modes tuning in GaN/AlGaN visible multilayer cavities on Sapphire for broadband nonlinear frequency conversion <u>Marco Centini</u>, Alessandro Bile, Alessandro Belardini, Daniele Ceneda, Adriana Passaseo, David Maria Tobaldi, Concita Sibilia, and Maria Cristina Larciprete
- 6. Electro-optic imaging of electric fields in irradiated CdTe detectors Adriano Cola, Lorenzo Dominici, Antonio Valletta
- 7. **Quantum dynamics and entanglement with multiple plasmonic modes** Angus Crookes, Ben Yuen, Angela Demetriadou
- 8. K-space hyperspectral imaging of microcavities and metasurfaces by an ultrastable commonpath interferometer

Armando Genco, <u>Cristina Cruciano</u>, Benedetto Ardini, Matteo Corti, Kirsty McGhee, Luca Sortino, Ludwig Hüttenhofer, Tersilla Virgili, David G. Lidzey, Stefan A. Maier, Andrea Bassi, Gianluca Valentini, Giulio Cerullo, Cristian Manzoni

9. Anomalous thermally activated delayed fluorescence (TADF) response for a phenothiazine

### derivative: a TD-DFT study

L. Cascino, A. Maggiore, I. Rivalta, G. P. Suranna, R. Grisorio, D. Conelli , V. Maiorano, Stefania D'Agostino

- Nonlinear and linear spatiotemporal reshaping of polariton fluids <u>Lorenzo Dominici</u>, Nina Voronova, Amir Rahmani, Antonio Gianfrate, Daniele Sanvitto, Michał Matuszewski, Marzena Szymańska, Ricardo Carretero, Fabrice Laussy
- 11. Advanced electromagnetism using FEniCSx Stefano Greco, Michele Castriotta, Cristian Ciracì
- 12. **Microscopic theory for active plasmonics in THz-pumped metal nanoparticles** Jonas Grumm, Robert Salzwedel and Andreas Knorr
- 13. Ultrafast Thermo-Optical Response of Drude Metals <u>Na'ama Harcavi</u>, Peleg Fishgrund, Yonatan Sivan
- 14. **Theory of free-electron third order nonlinearities in heavily doped InGaAs nanoantennas** <u>Huatian Hu</u>, F. De Luca, T. Venanzi, M. Ortolani, V. Giliberti, A. Rossetti, T. Deckert, D. Brida, M. Pea, A. Bousseksou, L. Lucia, R. Colombelli, and C. Ciracì
- 15. Squeezing free space with nonlocal metasurfaces towards ultrathin imaging systems Imon Kalyan, Nir Shitrit

### Thursday, September 7<sup>th</sup>, 2023

### Session 4 [09:00 - 10:40] Chair: Junsuk Rho, POSTECH, South Korea

### 09:00 Photonic temporal and time crystals (Keynote)

### Bumki Min

Department of Physics, Korea Advanced Institute of Science and Technology (KAIST), Republic of Korea

A photonic temporal crystal is characterized by its optical properties being periodically modulated over time, rendering it a temporal counterpart to spatially periodic photonic crystals. As early as 1966, the electromagnetic wave dynamics in a space-time periodic medium were theoretically explored, with the aim to describe time-growing instabilities in distributed parametric media. However, only a handful of theoretical studies followed this initial investigation, until an experiment utilizing a dynamic transmission line confirmed the existence of a shallow, yet genuine, momentum gap. Following this pioneering work, the popularity of time-varying photonics surged recently, with a primary focus on the conceptual extension of photonic spatial crystals and metamaterials into the space-time domain. More specifically, the enhanced dispersion and band structure engineering capabilities, facilitated by the additional temporal degree of freedom, have been the subject of extensive research. A multitude of intriguing phenomena such as colossal broadband nonreciprocity, efficient one-way amplification, parametric oscillation, pulse compression, and harmonic generation, have been theoretically or numerically considered for potential applications. It was only recently, however, that Floquet systems analysis was employed to shed more light on photonic temporal crystals. The momentum gap was confirmed to be the broken phase of parity-time (PT) symmetry along the wavenumber axis, while its edges were identified as non-Hermitian degeneracies, or exceptional points. In this talk, I will discuss non-Hermitian band structures, local density of states, and light-matter interactions in photonic temporal crystals. Moreover, I will delve into how nonlinearity can lead to discrete time translational symmetry breaking and result in the so-called time-crystalline behaviour within photonic temporal crystals.

### 09:40 Photonic bound states in the continuum for spectrally selective nanophotonics

Andreas Tittl

### Chair in Hybrid Nanosystems and Center for NanoScience, Faculty of Physics, Ludwig-Maximilians-Universität München, Germany

Photonic bound states in the continuum (BICs) have enabled far-reaching nanophotonic applications in high-harmonic generation, biospectroscopy, and lasing. BIC-based metasurfaces with tailored structural asymmetry have empowered these advances, but still face constraints related to large metasurface footprints, the need for complex polarization states, or fabrication limits requiring constant resonator heights throughout the structure. In this talk, I will present several recent concepts for obtaining additional photonic functionalities in such systems, including the arrangement of BIC-based unit cells in semi-infinite radial configurations for polarization invariance and reduced footprints as well as height-driven BICs leveraging resonators with different thicknesses as an additional degree of freedom for resonance engineering.

# 10:00 Plasmonic and dielectric resonances of homogeneous objects: from quasistatic to the full-wave regime

### Carlo Forestiere and Giovanni Miano

### Università degli Studi di Napoli Federico II, Napoli, Italy

The electromagnetic scattering resonances of a non-magnetic object much smaller than the incident wavelength in vacuum can be either described by the electroquasistatic approximation of the Maxwell's equations if its permittivity is negative, or by the magnetoquasistatic approximation if its permittivity

is positive and sufficiently high. Nevertheless, these two approximations fail to correctly account for the frequency shift and the radiative broadening of the resonances when the size of the object becomes comparable to the wavelength of operation. Starting from the full-wave eigenvalue problem, we introduce radiation corrections to the electroquasistatic and magnetoquasistatic resonances of arbitrarily-shaped objects are derived. Closed-form expressions of the frequency-shift and the radiative Q-factor of both plasmonic and dielectric modes of small objects are introduced.

### 10:20 Holes in Silicon enable extreme ultraviolet metaoptics

<u>Marcus Ossiander<sup>1,2</sup></u>, Hana K. Hampel<sup>1</sup>, Maryna L. Meretska<sup>2</sup>, Soon Wei D. Lim<sup>2</sup>, Nico Knefz1, Thomas Jauk<sup>1</sup>, Federico Capasso<sup>2</sup>, Martin Schultze<sup>1</sup>

<sup>1</sup>Institute of Experimental Physics, Graz University of Technology, Graz, Austria; <sup>2</sup>John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, USA We realize the first metalens operating in the extreme ultraviolet spectrum (50 nm wavelength) and demonstrate its performance using high-harmonic-generation.

### Session 5 [11:10 – 12:30] Chair: Yonatan Sivan, Ben-Gurion University, Israel

### 11:10 Persistent sub-radiant states with plasmonic nanocavities

Angela Demetriadou, Kalun Bedingfield, Angus Crookes, Ben Yuen

School of Physics and Astronomy, University of Birmingham, Birmingham, United Kingdom Plasmonic nanocavities have gaps of just 1-2nm and allow for light-matter strong coupling between molecular emitters and plasmons to be realized at room temperature. The continious energy exchange between the emitters and the plasmon leads to Rabi oscillations that dissipate quickly due to the high losses of the plasmonic system. Here, we show that when two (or more) emitters are placed in a plasmonic nanocavity, one can generate persistent sub-radiant states between the emitters that live up to 100 times longer than the Rabi oscillations.

### 11:30 **Inverse design for quantum nanophotonics: qubit entanglement and Bell state preparation** Antonio I. Fernandez-Dominguez

Departamento de Física Teórica de la Materia Condensada and Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, Spain

In this talk, I will explore the generation of entanglement between two quantum emitters through the inverse-design engineering of their photonic environment. By means of a topology-optimization approach, I will show how dielectric cloaks can be generated that operate at different inter-emitter distances and incoherent pumping strengths. I will show that the structures obtained yield steadystate concurrence values much larger than those attainable in free space, approaching the limit of maximum-entangled-mixed-states. Next, I will show how the emitter pair can be prepared, with fidelities approaching unity, into the symmetric and anti-symmetric Bell states under coherent pumping, again, through the inverse-design of the dielectric medium hosting them.

### 11:50 Modeling collective light emission by a few solid-state quantum emitters

Martijn Wubs<sup>1,2</sup>, Mads A. Jørgensen<sup>1</sup>, Devashish Pandey<sup>1</sup>, Nicolas Stenger<sup>1,2</sup>, Sanshui Xiao<sup>1,2</sup> <sup>1</sup>Dept. of Electrical and Photonics Engineering, Technical University of Denmark, Denmark; <sup>2</sup>NanoPhoton – Center for Nanophotonics, Technical University of Denmark, Denmark We present recent results on collective emission (superradiance and subradiance) by quantum emitters. The first part is about the effect of making the rotating-wave approximation. This is a very common approximation and already Dicke made it in his seminal work on superradiance. While single-emitter spontaneous emission rates are the same whether the approximation is made, this does not hold for collective emission rates: for two identical emitters, the sub- and superradiance decay rate are still the same whether ones makes the approximation or not, but for two slightly detuned emitters or for three or more identical emitters, the collective rates that one finds will generally be different, and the difference can be considerable. In the second part we present results on solid-state quantum emitters in photonic nanostructures. These emitters are open quantum systems in a double sense, as they typically couple both to the electromagnetic field and to phonons. In state of the art experiments it has become possible to engineer collective light emission despite the presence of phonons. We present and compare two methods to calculate superradiant spectra that take both Markovian and non-Markovian effects of phonons into account.

### 12:10 Tip-enhanced cavity-spectroscopy to control excitonic behaviors at the nanoscale

### Kyoung-Duck Park

Department of Physics, Pohang University of Science and Technology (POSTECH), Pohang, Republic of Korea

The tunability of the bandgap, radiative emission, and energy transfer in transition metal dichalcogenide (TMD) monolayers provides a new class of functions for a wide range of ultrathin photonic devices. Additionally, understanding and controlling the nanoscale transport of excitonic quasiparticles, such as excitons and trions, in atomically thin 2D semiconductors are crucial to produce highly efficient nano-excitonic devices. In this work, we present a dynamic nano-mechanical strain-engineering of naturally-formed wrinkles in a WSe2 monolayer, with real-time investigation of nano-spectroscopic properties using tip-enhanced cavity-spectroscopy. We reveal the modified nano-excitonic properties by the induced tensile strain at the wrinkle apex, exhibiting the exciton funneling phenomenon. In addition, we demonstrate a nanogap device to selectively confine excitons or trions of 2D TMDs at the nanoscale, facilitated by the drift-dominant exciton funneling into the strain-induced local spot. Furthermore, we present a method for the all-optical control of the exciton-to-trion conversion process and its spatial distributions in a MoS2 monolayer. We exploit propagating surface plasmon polaritons (SPPs) to localize hot electrons in a 2D TMD transferred on a metal–insulator–metal (MIM) waveguide. Our work provides a new strategy for robust, tunable, and ultracompact nano-excitonic devices using atomically thin semiconductors.

### Session 6 [14:00 – 15:20] Chair: Maria Cristina Larciprete, Sapienza, University of Rome, Italy

# 14:00 Surface-response formalism for mesoscopic electrodynamics in plasmonic nanostructures (Keynote)

### N. Asger Mortensen

*Center for Polariton-driven Light–Matter Interactions, University of Southern Denmark, Denmark; Danish Institute for Advanced Study, University of Southern Denmark, Denmark* 

The electrodynamics of matter and optical phenomena are commonly explored within the framework of classical electrodynamics and semiclassical models for the interactions of light with matter. Materials are commonly assumed homogeneous, and light-matter interactions are treated in an intuitive local manner. The plasmonic response of metal nanostructures is one such example, where the understanding of mesoscopic electrodynamics at metal surfaces is, however, becoming increasingly important for both fundamental developments in quantum plasmonics and potential applications in emerging light-based quantum technologies. The addition of surface-response formalism to classical electrodynamics is a way to represent quantum aspects and microscopic details of the electrodynamics at metal surfaces. The talk will discuss recent examples of nonlocal effects that emerge in surface-plasmonic systems, including metal surfaces, 2D materials, and combinations thereof.

### 14:40 CMOS-based terahertz camera based on quantum-dot-enhanced upconversion

### Sang-Hyun Oh

Department of Electrical and Computer Engineering, University of Minnesota, Minneapolis, USA

This work focuses on the developed of new terahertz cameras based on CMOS image sensors. Detection of terahertz (THz) radiation has numerous potential applications, but currently faced with limitations in detector performance such as sensitivity, speed, bandwidth, and operating temperature. Most of THz detectors also lack the ability to determine THz polarization states. However, the recent discovery of THz-driven luminescence in quantum dots offers a viable detection mechanism through field-driven inter-quantum-dot charge transfer. We introduce a THz camera and polarimeter that functions at room temperature, utilizing a complementary metal-oxide-semiconductor and a quantum-dot-enhanced THz-to-visible upconversion mechanism. With optimized luminophore geometries and fabrication designs, this nanoslit-based sensor achieves broadband and fast responses, and is capable of detecting THz pulses with peak fields as low as 10 kV/cm. Furthermore, we present a new coaxial nanoaperture-type device that possesses a hitherto unexplored ability to record the THz polarization state and field strength simultaneously, with comparable sensitivity.

### 15:00 Optical image differentiation with nonlinear flat optics

Domenico de Ceglia<sup>1,2</sup>, Andrea Alù<sup>3,4</sup>, Dragomir N. Neshev<sup>5</sup>, Costantino De Angelis<sup>1,2</sup> <sup>1</sup>Department of Information Engineering, University of Brescia, Italy; <sup>2</sup>CNR-INO (National Institute of Optics), Italy; <sup>3</sup>Photonics Initiative, Advanced Science Research Center, City University of New York, USA; <sup>4</sup>Physics Program, Graduate Center, City University of New York, USA; <sup>5</sup>The Australian National University, Canberra, Australia

We show that flat-optics elements with nonlinear response can be used to engineer Volterra kernels capable of real-time image processing. To illustrate this concept, we present an edge detection system that exploits the nonlinear response of a simple flat optics element. This approach offers several advantages compared to linear flat-optics-based edge detection and differentiation, including broad operation across different frequencies due to its non-resonant mechanism, significantly enhanced contrast, and improved performance in the presence of noise. Our findings suggest that the implementation of Volterra kernels in nonlinear flat optics opens up new possibilities for analog processing and computing using nonlocal nonlinear metasurfaces.

### Session 7 [15:50 – 17:10] Chair: Martin Schultze, Graz University of Technology, Austria

# 15:50 Large nonlinear efficiency enhancement in the visible and UV ranges from plasmonic gold nanogratings

<u>Crina Cojocaru</u><sup>1</sup>, S. Mukhopadhyay<sup>1</sup>, M. A. Vincenti<sup>2</sup>, L. Rodriguez-Sune, K. Hallman<sup>3</sup>, M. Scalora<sup>4</sup> and J. Trull<sup>1</sup>

<sup>1</sup>Department of Physics, Universitat Politècnica de Catalunya, Terrassa (Barcelona), Spain; <sup>2</sup>Department of Information Engineering – University of Brescia, Italy; <sup>3</sup>PeopleTec, Inc. 4901-I Corporate Dr., Huntsville, AL 35805, USA; <sup>4</sup>Aviation and Missile Center, US Army CCDC, Redstone Arsenal, AL 35898-5000, USA

We report a combined experimental/theoretical investigation on second and third harmonic generation from a plasmonic gold nanograting, resonant in the near IR. The intense field localization leads to more than three orders of magnitude enhancement in nonlinear optical processes, compared to flat gold nanolayer. The qualitative and quantitative spectral and angular dependence of the harmonics were experimentally recorded and validated within the framework of our microscopic, hydrodynamic model for linear and nonlinear material dispersion.

### 16:10 Second and third harmonic generation from aluminum nanostructures

<u>Michael Scalora</u><sup>1</sup>, K. Hallman<sup>2</sup>, S. Mukhopadhyay<sup>3</sup>, R. Vilaseca<sup>3</sup>, C. Cojocaru<sup>3</sup>, J. Trull<sup>3</sup>, D. de Ceglia<sup>4</sup>, M. A. Vincenti<sup>4</sup>

<sup>1</sup>Aviation and Missile Center, US Army, Redstone Arsenal, AL 35898-5000, USA; <sup>2</sup>PeopleTec, Inc. 4901-I Corporate Dr., Huntsville, AL 35805, USA; <sup>3</sup>Department of Physics, Universitat Politècnica

# de Catalunya, Terrassa (Barcelona), Spain; <sup>4</sup>Department of Information Engineering – University of Brescia, Italy

Plasmonics is concerned with the interaction of light with free charges on conductive surfaces. Historically, gold and silver have been the preferred choices because of low losses in the visible and near-IR portions of the spectrum. Here we report theoretical predictions and experimental observations of second and third harmonic generation from aluminum nanolayer so that we extract the intrinsic linear and nonlinear optical properties and establish baseline behavior, and then apply them to different grating structures in the hope of enhancing those basic properties. The push toward the ultraviolet range and beyond calls for additional studies of Au and Ag and alternative materials to determine their viability, a search that naturally highlights AI [1]. In some studies AI has been reported to outperform silver in the visible range due to its superior surface and interface properties [2]. However, most studies have been conducted over a limited wavelength range [3] with simple effective models. Unlike most noble metals, which display Lorentz-like behavior (interband transitions) in the UV range, AI is characterized by an absorption resonance near 850nm, which uncharacteristically splits the plasmonic range, and sets its linear and nonlinear optical properties apart. We set out to study AI with the aid of a hydrodynamic-Maxwell model that accounts for linear and nonlinear material dispersions, surface and volume nonlinear sources to study harmonic generation first from a simple AI layer a few tens of nanometers in thickness, and then from gratings and nanoantenna arrays that may display a combination of plasmonic and longitudinal Fabry-Perot resonances that localize the field inside a small volume. Our preliminary results suggest that bound charges play an outsized role in SHG, suggesting that predictions solely based on the free electron model may not adequately be used for prediction purposes, and that the absorption resonance play a pivotal role in THG.

### 16:30 A transformation optics approach to nonlinear and nonclassical plasmonics

### Fan Yang

# College of Physics, Key Laboratory of High Energy Density Physics and Technology of the Ministry of Education, Sichuan University, China

The plasmonic effects in the nanostructure provide a strong light-matter interaction between the structure and light, resulting in a giant nonlinear and nonclassical optical response. However, the analytical study of nonlinear and nonclassical effects in plasmonic structures is limited to basic geometries, such as flat surfaces, cylinders, or spheres. For a more complex geometry, an analytical solution becomes unobtainable. Fortunately, transformation optics, as a powerful analytical tool, has been employed in nonlinear and nonclassical plasmonics to solve this dilemma. For nonlinear plasmonics, second and third-harmonic generations from a nanowire dimer and a singular metasurface have been analytically studied. We found that the direct and the cascaded THG possess different size-dependence that can be used for experimental characterization of a nonlinear signal. Moreover, the SHG from a singular metasurface with a hidden dimension weakly depends on the pump field's incident angle, making it a perfect candidate as an all-angle harmonic-generation device. Regarding nonclassical plasmonics, nonlocal and electron spill-out effects have been thoroughly explored in the singular plasmonic system featured with a sharp point or a sub-nanometer metallic gap. A direct analytical approach to singular structures is complicated, but an indirect transformation optics approach that converts a complex nanostructure into a flat layered geometry becomes favorable. Obtaining the mapping rule of nonlocal parameter or Feibelman d parameter between the physical space and transformed virtual space, the complex nonclassical response of a singular nanostructure is significantly simplified.

### 16:50 Free-space interferometric routing of upconverted light by dielectric metasurfaces

A. Di Francescantonio<sup>1</sup>, A. Zilli<sup>1</sup>, D. Rocco<sup>2</sup>, F. Conti<sup>1</sup>, L. Coudrat<sup>3</sup>, M. Morassi<sup>4</sup>, A. Lemaître<sup>4</sup>, P. Biagioni<sup>1</sup>, L. Duò<sup>1</sup>, C. De Angelis<sup>2</sup>, G. Leo<sup>3</sup>, M. Finazzi<sup>1</sup>, <u>Michele Celebrano<sup>1</sup></u>

<sup>1</sup>*Physics Department, Politecnico di Milano, Italy;* <sup>2</sup>*Department of Information Engineering, University* of Brescia, Italy; <sup>3</sup>Université de Paris, CNRS, Laboratoire Matériaux et Phénomènes Quantiques, France; <sup>4</sup>Centre de Nanosciences et de Nanotechnologies, CNRS, Université Paris-Saclay, France We recently investigated frequency upconversion in both plasmonic and dielectric nanoantennas. Thanks to the adopted dual-beam pump scheme, where an ultrashort pulse at telecom wavelength impinges on the sample along with its frequency-doubled replica, Third Harmonic Generation (THG) and Sum Frequency Generation (SFG) processes are degenerate in energy. Although this should yield interference between the two processes, in all-delectric AlGaAs nanocylinders the symmetry of the system fully suppresses the interference term. Here, by applying the above dual-beam pump scheme to a periodic AIGaAs metasurfaces, we attain all-optical routing of the upconverted telecom photons in the visible range. This is attained by tuning the metasurface diffraction with respect to the meta-atom nonlinear emission in the Fourier plane, hence breaking the detection symmetry. Using the relative phase between the pump pulses as a tuning knob, we routed the upconverted radiation among different metasurface diffraction orders with an efficiency up to 90%. In particular, this is enabled by the maximization of constructive/destructive interference between SFG and THG in specific k-space directions. We also demonstrate that the polarization state of both pump and emission allows to reconfigure the routing between different sets of diffraction orders. The proposed approach can be envisioned as an all-optical method to route upconverted telecom photons into various detection channels. The combination of the interferometric and nonlinear character of the emitted light could be also extremely appealing for applications to nonlinear sensing.

### Poster Session 2 [17:20 - 18:40]

16. Crossover from non-thermal to thermal photoluminescence from metals excited by ultrashort light pulses

Imon Kalyan, leng Wai Un, Kaiqiang Lin, John M. Lupton, Sebastian Bange, Yonatan Sivan

- 17. Thermoplasmonic optical fiber probe: An experimental and computational analysis of the heating characteristics for neuroscience applications <u>Muhammad Fayyaz Kashif</u>, Di Zheng, Linda Piscopo, Cristian Ciracì ,Massimo de Vittorio, Ferruccio <u>Pisanello</u>
- 18. LNOI reconfigurable optical phased arrays for on-chip wireless switches <u>Muhammad Khalid</u>, G. Bellanca, Y. Pezhman, V. Petruzzelli, G. Calò
- 19. Polycrystalline MoO3 films fabricated by pulsed laser deposition for infrared multilayer photonics

Maria Cristina Larciprete, Daniele Ceneda, Chiyu Yang, Sina A. Dereshgi, Federico Vittorio Lupo, Maria Pia Casaletto, Roberto Macaluso, Mauro Antezza, Zhuomin M. Zhang, Marco Centini, Koray Aydin

20. Design of a room-temperature topological exciton-polariton laser in a ZnO/TiO2 photonic crystal slab

Charly Leblanc, I. Septembre, L. Hermet, H. S. Nguyen, X. Letartre, D. D. Solnyshkov, and G. Malpuech

- 21. **On-chip PI-excitonic materials: Manipulating multiple quantum states in a single quantum emitter and metallic nano-cavities accurately coupled systems** Kun Liang, Li Yu
- 22. **Split Bowtie nanoantennas for electron acceleration** Giovanni Magno, Marco Grande, Béatrice Dagens
- 23. **Computational study of interaction between Channelrhodopsin and a gold nanocluster** <u>Roberto Messina</u>, Luca Bellucci, Stefano Corni, Stefania D'Agostino, Giuseppe Gigli, Laura Zanetti Polzi
- 24. Effect of electron spill-out on the surface plasmon polariton propagation at dielectricmagnetized plasma interface.

Tadele O. Otomalo, Muhammad Khalid, Cristian Ciracì

- 25. **Exciton-Plasmon Hybridization at interfaces of metal nanoparticles and 2D semiconductors** <u>Robert Salzwedel</u>, Lara Greten, Stefan Schmidt, Stephen Hughes, Andreas Knorr, Malte Selig
- 26. **Monolithic van der Waals metasurfaces** Luca Sortino, Stefan A. Maier, Andreas Tittl
- 27. **Manipulating light-matter interactions by strain modulation in two dimensional semiconductors** <u>Francesco Todisco</u>, L. Polimeno, A. Di Renzo, R. Mastria, S. Rizzato, M. Mannoccio, M. Esposito, V. Tasco, K. Kurselis, R. Kiyan, M. De Giorgi, G. Maruccio, B. Chichkov, D. Sanvitto
- 28. Metallic nanoislands-decorated tapered optical fibers for remote SERS sensing and heat generation

<u>Di Zheng</u>, Filippo Pisano1, Liam Collard, Antonio Balena, Muhammad Fayyaz Kashif, Linda Piscopo, Cristian Ciracì, Massimo De Vittorio, Ferruccio Pisanello

29. **Spontaneous parametric down-conversion beaming from a Lithium Niobate nanoresonator** <u>Attilio Zilli</u>, Vitaliy Sultanov, Michael Poloczek, Marzia Ferrera, Yigong Luan, Emmanouil Kokkinakis, Tomás Santiago-Cruz, Luca Carletti, Andrea Toma, Marco Finazzi, Maria Chekhova, Michele Celebrano

### Friday, September 8<sup>th</sup>, 2023

### Session 8 [09:00 - 10:40] Chair: Michael Scalora, US Army, USA

09:00 **Beam shaping and frequency conversion in nonlinear all-dielectric metasurfaces (Keynote)** Jiannan Gao<sup>1</sup>, Dmitrii Tsvetkov<sup>1</sup>, Danilo Gomes Pires<sup>1</sup>, Maria Antonietta Vincenti<sup>2</sup>, Yun Xu<sup>3</sup>, Ivan Kravchenko<sup>4</sup>, Jesse Frantz<sup>5</sup>, Anthony Clabeau<sup>6</sup>, Xingdu Qiao<sup>7</sup>, Liang Feng<sup>8</sup>, Michael Scalora<sup>9</sup>, Natalia M. Litchinitser<sup>1</sup>

<sup>1</sup>Department of Electrical and Computer Engineering, Duke University, USA; <sup>2</sup>Department of Information Engineering, University of Brescia, Italy; 31Dassault Systemes Simulia Corp (CST), USA; <sup>4</sup>Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, USA; <sup>5</sup>US Naval Research Laboratory, USA; <sup>6</sup>University Research Foundation, USA; <sup>7</sup>Department of Electrical and Systems Engineering, University of Pennsylvania, USA; <sup>8</sup>Department of Materials Science and Engineering, University of Pennsylvania, USA; <sup>9</sup>Aviation and Missile Center, US Army CCDC, Redstone Arsenal, USA

The emergence of nonlinear flat-optics nanostructures, optical metasurfaces, enabling unprecedented enhancement of light-matter interactions without phase-matching requirements may revolutionize future applications of integrated nonlinear optics. We demonstrate ultrafast tunable, near-infrared to ultraviolet frequency conversion in a chalcogenide glass metasurface based on Mie resonances and quasi-bound states in the continuum (qBIC) resonances, enabled by a phase-locking mechanism between the pump and the inhomogeneous portion of the TH signal. Through phase locking, the pump pulse and the inhomogeneous harmonic component can co-propagate, resulting in the acquisition of the same refractive index and absorption coefficient as the pump. If this process occurs within a cavity, efficient frequency conversion can take place, even in the presence of strong material absorption at the wavelengths of the harmonics. As for all nonlinear processes, a resonant condition at the pump field boosts the nonlinear interactions. We also experimentally show the simultaneous generation of phase-locked structured light beams, including optical vortices and Hopf-links at fundamental and tripled frequencies in all-dielectric nonlinear optical metasurfaces despite the fact that the tripled frequency is corresponding to the region of high absorption of the dielectric material. This work may have useful prospects in optical manipulation, optical communication, and quantum information transmission.

### 09:40 Optical torque induced by resonant harmonic generation in dielectric nanostructures

Mihail Petrov<sup>1</sup>, Ivan Toftul<sup>1,2</sup>, Gleb Fedorovich<sup>1</sup>, Denis Kislov<sup>1</sup>, Kristina Frizyuk<sup>1</sup>, Kirill Koshelev<sup>2</sup>, Yuri Kivshar<sup>2</sup>

### <sup>1</sup>Department of Physics and Engineering, ITMO University, Russia; <sup>2</sup>Australian National University, Australia

Optically induced mechanical torque driving rotation of small objects requires the presence of absorption or breaking cylindrical symmetry of a scatterer. A spherical nonabsorbing particle cannot rotate due to the conservation of the angular momentum of light upon scattering. Here, we suggest a novel physical mechanism for the angular momentum transfer to nonabsorbing particles via nonlinear light scattering. The breaking of symmetry occurs at the microscopic level manifested in nonlinear negative optical torque due to the excitation of resonant states at the harmonic frequency with higher projection of angular momentum. The proposed physical mechanism can be verified with resonant dielectric nanostructures, and we suggest some specific realizations.

### 10:00 2D semiconductor multilayers for ultra-thin nanophotonic platform

Su-Hyun Gong

Department of Physics, Korea University, South Korea

The emergence of 2D materials stimulated intensive research on both electronic and photonic

applications. Especially, transition metal dichalcogenides (TMDs) provided an excellent platform for photonic applications due to their strong light-exciton interaction. Various photonic devices such as a light-emitting device, laser, and exciton-polariton device have been successfully demonstrated experimentally using TMD monolayers. However, multilayered TMDs have attracted far less attention than TMD monolayers because they become indirect bandgap materials. Here I will present that multilayered TMD itself is a good platform for controlling light-matter interaction without integrating an external photonic structure. A TMD multilayer can be utilized for a passive optical structure because it possesses a high dielectric constant. For example, light guiding is possible along a multilayered TMD, which is very thin compared to the wavelength of light. Because a high dielectric constant is owing to the exciton resonances, guided light along a TMD layer is referred to as exciton-polariton. I will also show that light can be further controlled using a patterned TMD multilayer. A patterned WS2 disk structure has a very high confinement factor for lasing action because the TMD disk offers both optical modes and optical gains. As a result, we observed the lasing operation under continuous-wave excitation at room temperature. We believe our results show potential for the TMD-based nanophotonics offering a small mode volume but with a lower loss compared to the surface plasmon polaritons.

### 11:00 Making spatial dispersion useful

Antoine Moreau<sup>1</sup>, Émilie Sakat<sup>2</sup>, Jean-Paul Hugonin<sup>3</sup> and Thierry Taliercio<sup>4</sup>

<sup>1</sup>Université Clermont Auvergne, CNRS, Institut Pascal, Clermont-Ferrand, France; <sup>2</sup>Université Paris Saclay, Center for Nanoscience and Nanotechnology, CNRS, Palaiseau, France; <sup>3</sup>Université Paris-Saclay, Institut d'Optique Graduate School, CNRS, Palaiseau, France; <sup>4</sup>IES, Université de Montpellier, UMR CNRS 5214, Montpellier, France

For more than a century, the accuracy of the Drude model for describing the response of media containing an electron gas has been such that there was no use of more advanced models. This has changed ten years ago when the first signs of spatial dispersion in metals have been observed – however the importance of this effect is relatively modest. This may not be the case for doped semi-conductors, in which the effective mass is small enough for spatial dispersion to have a distinctive impact that can not be neglected or explained by any other phenomenon. This can even be leveraged to retrieve, thanks to a single optical measurement, all the characteristics of the material (doping and effective mass) making spatial dispersion truly useful in that case.

### Session 9 [11:10 – 12:10] Chair: Francesco Todisco, CNR NANOTEC, Italy

### 11:10 Nonlinear nanoplasmonics with atomically thin materials

### Joel Cox

POLIMA–Center for Polariton-driven Light-Matter Interactions, University of Southern Denmark, DK-5230 Odense M, Denmark; Danish Institute for Advanced Study, University of Southern Denmark, DK-5230 Odense M, Denmark

Plasmons–collective oscillations in the free electron plasma–constitute nanoscale optical resonators that are imbued with a nonlinear response by their supporting conductive media. In the 2D limit represented by atomically thin materials, plasmon resonances provide unprecedented levels of optical field confinement, while exhibiting relatively lower losses in pristine samples. The appealing properties of 2D plasmons are ideal for nonlinear plasmonics, which seeks to overcome the weak nonlinear response of available materials by exploiting the large near field enhancement supplied by plasmon resonances. Here we theoretically explore nonlinear light-matter interactions of 2D plasmons hosted in atomically thin materials and their heterostructures. Our investigations are based on nonclassical methods to describe graphene plasmons, characterized by high confinement and electrical tunability, plasmons supported by ultrathin crystalline noble metal films, with thickness-dependent properties and lower losses than their amorphous counterparts, and nanostructured phosphorene, an anisotropic two-dimensional semiconductor that hosts plasmons in highly-doped samples. We further explore

possibilities to trigger nonlinear interactions on the few-plasmon level and to enhance harmonic generation through synergetic interactions between plasmons in atomically-thin heterostructures.

### 11:30 Free-electron infrared nonlinearities in heavily doped InGaAs nanoantennas

Tommaso Venanzi<sup>1</sup>, M. Ortolani<sup>2</sup>, V. Giliberti<sup>1</sup>, A. Rossetti<sup>3</sup>, T. Deckert<sup>3</sup>, D. Brida<sup>3</sup>, M. Pea<sup>4</sup>, A. Bousseksou<sup>5</sup>, L. Lucia<sup>5</sup>, I. Sagnes<sup>5</sup>, G. Beaudoin<sup>5</sup>, R. Colombelli<sup>5</sup>, H. Hu<sup>6</sup>, F. De Luca<sup>6</sup>, C. Ciracì<sup>6</sup> <sup>1</sup>Istituto Italiano di Tecnologia, Rome, Italy; <sup>2</sup>Sapienza University of Rome, Italy; <sup>3</sup>University of Luxembourg, Luxembourg; <sup>4</sup>CNR Institute for Photonics and Nanotechnologies, Italy; <sup>5</sup>CNRS Center of Nanoscience and Nanotechnology (C2N), University of Paris-Saclay, France; <sup>6</sup>Istituto Italiano di Tecnologia, Lecce, Italy;

Hydrodynamic models of free electrons in metals and in degenerately doped semiconductors can describe accurately a range of nanoscale plasmonic phenomena that occur at interfaces, nanoantennas, metasurfaces. Within the hydrodynamic model, third-order nonlinear terms arise, especially when the free electrons are driven at frequencies close to their plasma frequency. In the project NEHO, we aim to employ free-electron nonlinearities in n-doped InGaAs as non-linear process to make optical computation in mid-infrared integrated photonic circuits. As first step of the project, we quantify the third-order free-electron non-linearity by measuring third-harmonic generation of plasmonic antenna arrays. We fabricated antennas with different plasma frequencies and, therefore, with different plasmonic resonance frequencies (8.5, 9.8, 11.3 um). The third harmonic generation (THG) is strongly enhanced when the mid-infrared pump wavelength matches the plasmonic resonance of the array. No THG is observed from the undoped antenna array confirming that the TH originates from the non-linear response of free electrons.

### 11:50 Passive and tunable flat optics with dielectric nanoantennas

### Arseniy Kuznetsov

# Institute of Materials Research and Engineering, A\*STAR (Agency for Science, Technology and Research), Singapore

Dielectric nanoantennas and metasurfaces have recently emerged as a new nanophotonic platform paving way to new generation of flat optical components. They can control light at nanometer dimensions in an unprecedented fashion, not achievable by conventional bulk optics. This led to demonstrations of flat optics with extraordinary performance, e.g. very large numerical aperture or very large field of view. One of the existing challenges of metasurface-based flat optics is strong grating-like dispersion of metasurfaces, which typically limits their ability to perform high-quality imaging in a broad spectral range, e.g., with white light. In this talk, I will first show how combining a few large-field of view quadratic metalenses with image-processing it is possible to achieve white-light wide-field-of-view imaging potentially applicable to regular smartphones or laptops. On the other hand, I will also demonstrate that the metasurface dispersion can be used in a constructive way to achieve hyperspectral imaging systems for space applications. I will then demonstrate that integrating these nanoantennas on top of actively controlled 1D and 2D electrode arrays embedded in liquid crystals it is possible to achieve fully controllable and dynamically switchable metasurfaces with a pixel size down to 1 micrometer. These metasurface-based spatial light modulators can generate arbitrary wavefront patterns and can be used for beam steering in LiDAR devices or tunable holography in 3D holographic displays.

**Full length abstracts** 

Talks

### Non-Hermitian resonators: the divergence, the Qs and Vs of their quasinormal modes

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Micro and nanoresonators enhance many light-matter-interaction processes and are used in various modern applications in photonics. They are open systems and their modes (often called quasinormal modes) are always leaky, have a finite lifetime and thus, are defined as source-free solutions of Maxwell equations at a complex frequency. The analysis of the non-Hermitian behavior of such resonator in their modal basis has posed great difficulty in the past, but the situation has changed drastically recently thanks to recent progresses in the complex analysis of open electromagnetic systems [1].

For instance, the problem of the mode normalization (with its sterile and misleading debate) is fully clarified today, see the easy-to-read Section 4 in the recent review article [2]; the same can be said, hopefully, for the completeness of the mode basis and for the regularization of the exponentially diverging field [2]. We have now reached a point where non-Hermitian freeware packages, like the one developed in my group for 10 years, now exist and compute/normalize/regularize the quasinormal modes of virtually any resonator [3].

The physics of non-Hermitian systems is significantly richer than that of Hermitian ones, hopefully. We will try:

- to explain that the exponential growth of the modal field outside the resonator in the leaky region is not unphysical, contrarily to what could be read in the literature, but can even be observed in some conditions that will be defined [4].
- to present what is the complex mode volume [1,5], an important notion that comprehensively helps understanding key phenomena observed in non-Hermitian nanocavities. The real part of V has the usual sense defined for Hermitian systems. We will show that the imaginary part of V can be measured [6] and can be easily interpreted [5].
- to illustrate how the quasinormal mode formalism can be used to design resonators and interpret experimental works, see [7] for an example in nonlinear optics.

### References

- P. Lalanne et al., "Light interaction with photonic and plasmonic resonances", Laser & Photonics Reviews 12, 1700113 (2018).
   S. Sauvan et al., "Normalization, orthogonality, and completeness of quasinormal modes of open systems: the case of electromagnetism", Opt. Express, **30**, 6846-85 (2022).
- [3] T. Wu et al., "Modal analysis of electromagnetic resonators: User guide for the MAN program", Computer Physics Communications 284, 108627 (2023). <u>https://zenodo.org/record/7400937</u>
- [4] T. Wu, J. L. Jamarillo, P. Lalanne et al., submitted.

[6] K. G. Cognée et al., "Mapping complex mode volumes with cavity perturbation theory", Optica 6, 269-273 (2019).

<sup>[5]</sup> T. Wu, M. Gurioli, P. Lalanne, "Nanoscale Light Confinement: the Q's and V's", ACS Photonics 8, 1522-1538 (2021).

<sup>[7]</sup> C. Gigli et al., "Quasinormal-Mode Non-Hermitian Modeling and Design in Nonlinear Nano-Optics",

ACS Photonics 7, 1197–1205 (2020).

### Nonlocal effects in ENZ antennas and new materials for second-order nonlinear optics

Stéphane Kéna-Cohen<sup>1</sup>

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In the first part of the talk, we will describe the realization epsilon-near-zero photonic gap antennas (ENZ-PGAs) enclosing a thin layer of indium tin oxide serving as the ENZ material [1]. Such antennas can provide field enhancements of >100 and large (but lossy) Purcell factors. We will show, by comparing experimental results from single antenna dark field scattering and third harmonic generation to those of full wave simulations that nonlocal effects play an important role in these structures [2]. The inclusion of nonlocal effects within the simplified hydrodynamic model of Ref. [3] in our full wave simulations show much better agreement for both peak positions and feature shape in dark field scattering as shown in Figure 1. In particular, nonlocal simulations show that the volume-averaged field enhancement is 4–6 greater than that predicted by the local model. Moreover, sharp resonances occur near wavelengths of vanishing group velocity that are completely absent in the local model. These sharp resonances are consistent with our experimental observations.



Figure 1: Dark field scattering along major and minor axes of the elliptical ENZ-PGAs comparing experiment (black) to full wave simulation results (red).

In the second part of the talk, we will describe our attempts to grow aligned thin films of small molecules with large hyperpolarizabilities as a flexible platform for implementing  $\chi^{(2)}$  optical nonlinearities. Vacuum thermal evaporation allows for the deposition of organic materials on arbitrary substrates at room temperature. However, thermally evaporated films of small molecules have typically shown vanishing  $\chi^{(2)}$  nonlinearities due the random arrangement of molecular orientations, and thus centrosymmetry in the bulk. We will show that by harnessing the interplay between the permanent dipole moment and the hyperpolarizability, it is possible to spontaneously break centrosymmetry during thermal evaporation, without the need for poling. Using a range of materials, we will show that values of d<sub>31</sub> and d<sub>33</sub> ranging from 5-10 pm/V are readily achievable, without resorting any special alignment procedures.

### References

- [1] A. Patri, Phys. Rev. B 105, 165126 (2022).
- [2] F. Thouin et al., arXiv:2211.05214 (2022).
- [3] Y. Luo et al., Phys. Rev. Lett. 111, 093901 (2013).

### Nonlinear Optics in Topological Quantum Materials and Axion Electrodynamics Prineha Narang, UCLA

Parametric optical nonlinearities are critical to a wide spectrum of photonic technologies, from optical parametric oscillators to frequency combs to quantum information processing. Optical nonlinearities also serve as a powerful method for mapping material properties including the symmetries of electronic structure. Optical nonlinearities are generally very small in conventional materials as they depend on higher order effects. Parallel to these technical needs, the field of topological materials has seen the prediction and discovery of a large number of massless, three-dimensional linear dispersion systems known as Dirac and Weyl semimetals. It was soon realized that these materials may offer a rich new material phase space for extending the nonlinear effects of graphene including the role of topology and Berry connection. In this context, I will present our recent work on predicting the optoelectronic and nonlinear properties of Dirac and Weyl semimetals with an emphasis on figures of merit (FoMs) that we will evaluate for these new Weyl and Dirac semimetals include the  $\chi^{(2)}$ \* (Q/V) that captures the confinement and nonlinearity as well as  $\chi^{(2)}/\alpha$  and  $\chi^{(3)}/\alpha$ , where  $\alpha$  is the absorption rate, to describe the second and third order susceptibilities and electro-optic coefficients of the materials. Next, I will discuss our recent results on the multiphoton spectroscopy of a dynamical axion insulator. Here, the axion receives contributions from the collective motion of electrons, leading to a nonlinear topological magnetoelectric effect. Identifying this collective axion response faces a number of major experimental difficulties, which necessitate a theory-predicted smoking gun signature. We demonstrate a two-step protocol for the unambiguous optical identification of the collective axion mode in such a system. First, we show how collective oscillations of the axion mode can be induced by two-photon absorption or stimulated Raman spectroscopy, with the magnetoelectric nature of the excitation manifesting in the polarization dependence of the excitation beams. Second, we show how the axion oscillations can be confirmed through their manifestations in the time-resolved Kerr-rotation, which again carries signature polarization dependence due to the magnetoelectric nature of the coupling. Looking ahead, I will discuss how collective responses in topological quantum materials can be unambiguously identified in nonlinear electrodynamical probes, as well as identify potential avenues for intersections with particle physics in axion electrodynamics.

### **On-Chip SOI Hybrid Plasmonic Nanofocusser**

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Silicon-on-insulator (SOI) chip-based hydrid-plasmonics combines advantages of all-integrated optoelectronic functionality and deep sub-wavelength optical confinement [1]. However, the large difference in modal areas limits the coupling efficiency from conventional waveguides [2], in turn reducing the efficiency of lightmatter interactions like nonlinear optical functions [3]. Here we design, fabricate, and experimentally characterize an efficient on-chip SOI hybrid plasmonic nanofocussing waveguide, with a tip as small as 10 nm. The device operates by rotating [4] the fundamental TE mode of a standard SOI waveguide  $(350 \times 220 \text{ nm})$  to a TM mode of a hybrid plasmonic waveguide (gold thickness: 50 nm; SiO<sub>2</sub> spacer thickness: 20 nm), and subsequently focussing it to the nanoscale. We consider the three devices in Fig. 1(a), with increasingly sharp gold tips with measured apexes of 300 nm (blue), 100 nm (red), and 10 nm (magenta). These are deposited on three identical SOI waveguides using in-house nanolithography which result in high alignment precision.

Since plasmonic nanofocussing cannot be demonstrated by far-field, linear experiments, we measure the second harmonic generation (SHG) at the tip. The wavelength-scale propagation distances make phase matching unnecessary - we can thus correlate the measured SHG to the degree of nanofocussing of the pump at the tip. In our experiments, we couple light from an optical parametric oscillator ( $\lambda_p$ =1320 mm, pulse width: 200 fs; repetition rate: 80 MHz) onto a waveguide input grating coupler from free space using a 100×microscope objective (NA = 0.85), and observe the scattered light at the pump and second harmonic ( $\lambda_{SHG}$ =660 mm) wavelengths. Figure 1b shows simulated and measured diffraction-limited spots at the 10 nm tip at the pump and SHG wavelengths. The measured SHG spectrum is shown in the inset of Fig. 1(c). We measure the spectral yield (in counts/30 minutes) versus incident power (Fig. 1c, circles). A linear relation between the square root of the yield and the input power confirms the required quadratic input power dependence of the second harmonic light ( $I_{SHG}^{\frac{1}{2}} \propto P_{in}$ , Fig. 1c). The dramatic increase in SHG intensity for the sharpest tip indicates strong focusing; by comparing the slopes of the three curves, we can experimentally quantify the degree of maximum intensity enhancement - which is a factor of  $\times$  7.5 with respect to the strip case. The modelled increase in  $|E|^2$  (COMSOL 3D) is shown via a relative slope increase with respect to the widest gold strip (Fig. 1c, dashed blue line) as indicated by the shaded regions in Fig. 1c, with the upper- and lower- bounds stemming from uncertainty of the tip apex width. The agreement between experiment and simulations is excellent, allowing us to conclude through simulations that the pump light is focused down to a mode area of approximately 40 nm<sup>2</sup> [5], resulting in a  $\sim$ 1200 intensity enhancement with respect to the silicon waveguide input. This represents the first TM plasmonic nano-focusser to monolithically interface with an industry-standard TE-input SOI waveguide. This work lays the foundations for efficient and compact on-chip, deep sub-wavelength sources and sensors, bridging integrated photonic-circuits and metallic nanostructures potentially down to single-atoms -enabling operation in nonlinear plasmonic and quantum regimes.



Fig. 1 (a) SOI hybrid plasmonic nano-focusser SEMs, with tip width of 300 nm (blue), 138 nm (red) and 15 nm (magenta). (b) Simulated and measured intensity scattered out-of-plane tips at 1320 nm and 660 nm. (c) Circles: square root of the measured yield for each sample (colour coding as in (a)). Dashed lines: linear fits confirming quadratic dependence on incident power  $((I_{SHG})^{\frac{1}{2}} \propto P_{in})$ . Shaded regions contain bounds of the modelled maximum intensity for a tip apex width of 10 ± 5 nm (purple) and 100 ± 30 nm (red), relative to 300 nm case (blue dashed line).

### References

[1] V. Sorger, R. F. Oulton, R. M. Ma and X. Zhang, "Toward integrated plasmonic circuits," MRS Bull. 37, 728-738 (2012)

[2] Y. Luo, M. Chamanzar, A. Apuzzo, R. Salas-Montiel, K. N. Nguyen, S. Blaize, and A. Adibi, "On-chip hybrid photonic-plasmonic light concentrator for nanofocusing in an integrated silicon photonics platform," Nano Lett. 15, 849-856 (2015).

[3] S. Kim and M. Qi, "Polarization rotation and coupling between silicon waveguide and hybrid plasmonic waveguide," Opt. Express 23, 9968-9978 (2015).

[4] M. P. Nielsen, X. Shi, P. Dichtl, S. A. Maier, R. F. Oulton, "Giant nonlinear response at a plasmonic nanofocus drives efficient four-wave mixing", Science 358, 1179-1181 (2017).

[5] L. Lafone, T. P. H Sidiropoulos, and R. F. Oulton, "Silicon-based metal-loaded plasmonic waveguides for low-loss nanofocu 😚 ", Opt. Lett. 38, 4356-4359 (2014).

### Ultrafast electron transport at the nanoscale

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Recently, we demonstrated that single-cycle pulses of minute energy content may result in extremely nonlinear optical phenomena at the nanoscale by exploiting an electronic circuit with a few-nanometre gap between the tips of an optical antenna. The strong electrical bias provided by the field contained in ultrashort optical pulses was harnessed to drive tunnelling and ballistic acceleration of electrons to generate a current through the free-space gap with PHz bandwidth [1]. This non-perturbative process is fully coherent with the driving radiation and occurs within a half-cycle of the near-IR carrier wavelength. In addition, we further explored this concept by gaining direct temporal information via interferometric autocorrelation measurements with two identical replicas of truly single-cycle driving pulses.



Figure 1: Conceptual sketch for ultrafast electron transport at the nanoscale. The optical field biases the junction between two metal thus inducing tunnelling and transport on a sub-cycle timescale. Left panel: experiments in a metallic junction made by a bowtie plasmonic antenna with the electron transport occurring in its feedgap. Right panel: same concept applied to a scanning tunnelling microscope with atomic resolution.

These experiments exploit the electric currents coherently driven at the gap of a single nanodevice by optical pulse with pJ energies. The full width at half maximum of the current autocorrelation amounts to less than one femtosecond, demonstrating that we can transfer individual electrons between the two contacts on an attosecond time scale. These concepts are currently being applied, with promising results, to a scanning tunnelling microscope with the bias provided solely by the optical field. In the future, we are aiming at a regime where the Coulomb interaction between electrons becomes important at truly atomic time and length scales. In addition, by a careful selection of the tip material, it will be possible to access the spin degree of freedom in the characterization of electronic wavefunctions.

### References

[1] D. Brida, G. Krauss, A. Sell and A. Leitenstorfer, Laser Phot. Rev. 8, 409-428 (2014).

[2] T. Rybka, M. Ludwig, M. F. Schmalz, V. Knittel, D. Brida und A. Leitenstorfer, Nature Photon. 10, 667–670 (2016).

[3] M. Ludwig, G. Aguirregabiria, F. Ritzkowsky, T. Rybka, D.C. Marinica, J. Aizpurua, A. G. Borisov, A. Leitenstorfer, D. Brida, Nature Physics 16 (3), 341-345 (2020)

### **Photonic Memristors**

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Electronic components integrating nanometer scale gap in their design were also crucial to the advent of novel form of computing. Memristors for instance are programmable voltage-dependant resistive devices deployed nowadays in cognitive hardware systems such as artificial neural networks, neuromorphic and reservoir computing [1]. Memristive operation relies on resistance switching triggered by the electroformation and disruption of conductive path- ways within a nanometer-scale dielectric gap [2]. Charge transport occurs by an electro-chemical reduction of metal ions aggregating to conductive filaments [2], or by migration of mobile defects, such as oxygen vacancies [3] and nanoclusters [4]. In this presentation, we introduce an atomic scale memristive device capable of emitting photons during resistive switching, superseding thus the need for an external optical source. Our device features the compact footprint of transistors and compatibility with the emerging memristive technology. We identified three mechanisms producing photons with vastly different properties [5, 6]. The crossover between emission regimes depends on the history of the memristor and its operating conductance. Our results suggests that this new generation of memristor pave the way for multidimensional neural networks using both electrons and photons as information carrier.



Figure 1: **a**: Memristive output properties of the planer gap geometry shown in **b**. **c**: time traces showing the voltage pulse applied to the device, the current flowing and the light emission.

### References

- [1] L. Ye, Z. Gao, J. Fu, W. Ren, C. Yang, J. Wen, X. Wan, Q. Ren, S. Gu, X. Liu et al., Frontiers in Physics 10, 839243 (2022)
- [2] T. Tsuruoka, K. Terabe, T. Hasegawa, M. Aono, Nanotechnology 21, 425205 (2010)
- [3] C. Funck, S. Menzel, ACS Appl. Elec. Mat. 3, 3674 (2021)
- [4] Y. Yang, P. Gao, L. Li, X. Pan, S. Tappertzhofen, S. Choi, R. Waser, I. Valov, W.D. Lu, Nature Communications 5, 4232 (2014)
  [5] B. Cheng, T. Zellweger, K. Malchow, X. Zhang, M. Lewerenz, E. Passerini, J. Aeschlimann, U. Koch, M. Luisier, A. Emboras
- et al., Light: Science & Applications 11, 78 (2022)
  [6] S. Hamdad, K. Malchow, E. Dujardin, A. Bouhelier, B. Cheng, T. Zellweger, J. Leuthold, Overbias light emission from memristive nanojunctions (2022), ArXiv 2212.02320

### The nonlinear optical response and electron dynamics in ITO

Subhajit Sarkar, leng-Wai Un, Yonatan Sivan, Ben-Gurion University

Low electron density Drude (LEDD) materials such as transparent conducting oxides, plasmonic nitrides, became popular candidates for high-efficiency nonlinear optical applications, due to their unique near-infrared "epsilon near zero" point. Their nonlinearity is extremely large, reaching 100's of percent of the refractive index/permittivity. Peculiarly, despite the large body of related impressive experimental demonstrations, their theoretical modeling was mostly coarse, and has not yet conclusively elucidated the origins of the giant optical response. Here, we close this knowledge gap and provide a "first principles" model of the response of LEDD materials to ultrafast illumination. For concreteness, we focus on Indium Tin Oxide (ITO).

Our model includes the Boltzmann equation (BE) complemented by a phonon dynamics equation, and an easy-to-use coarse-grained extended two temperature model (eTTM). We find the electron heat capacity of ITO to be smaller, but the electron-phonon energy transfer rate to be comparable to that in noble metals. This leads to stronger heating of the electrons, and to a faster cooling compared to noble metals [Fig. left(a)]. Surprisingly, the intense illumination and associated high electron temperature, may cause the effective chemical potential to become negative, thus, transiently converting the ITO into a semiconductor [Fig. left(b)].



Fig. Left: (a) Effective electron temperatures (orange lines) and phonon temperatures (green lines) as a function of time for pulse of width 220 fs with peak intensity,  $2.5 \ GW/cm^2$ . Solid and dotted lines correspond to temperatures obtained from the BE and the eTTM, respectively. Dot-dashed lines correspond to the temperature dynamics of gold (Au) obtained from the eTTM simulation. (b) The (instantaneous) chemical potential following illumination by a pulse of width 220 fs with peak intensity is  $50 \ GW/cm^2$ , obtained from the effective electron temperature shown in the inset. Fig. Right (a) The total absorbed pump pulse energy (normalized to the pump pulse energy) as a function of the pulse peak intensity. (b) The maximum phonon temperature achieved for different pump peak intensities.

We also find that the drastic increase of the real part of the permittivity shifts the resonance from the pump such that the absorptivity drops rapidly with increased illumination intensity [Fig. right(a)]. Consequently, the phonon temperature increases sub-linearly with the pump peak-intensity, reaching the melting point of at 500 GW/cm<sup>2</sup> [Fig. right(b)]. This explains, for the first time to our knowledge, the experimental observation of the high damage threshold of ITO and shows that the ITO nonlinearity is not saturable, but rather thermal as for noble metals.
## Strong light waves controlling charge & spin dynamics

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The V/Ångstrom-class electric field strength offered by ultrafast laser waveforms allows to steer electronic motion and control electronic excitation so rapidly, that many collective processes including carrier scattering and coupling to lattice motion that in more conventional excitation conditions conspire to disrupt coherence have hard time catching up – even in condensed phase systems.

We investigate the opportunities this temporal segregation offers and strive to transfer ideas of coherent control to the manipulation of electronic and magnetic states of room temperature solids. I will discuss a set of experiments demonstrating the capability of single cycle optical fields to deliberately drive currents and alter the magnetization state of solid bulk materials and nano-heterostructures:

Sub-femtosecond photo-doping within a materials band-structure preserves the wave-packet properties of the excited carriers at early times. In this regime, via carrier injection in dielectric materials interfaced to electronic circuitry we demonstrate the feasibility of ultrafast, coherent optoelectronic applications up to 1 Petahertz frequencies[1]. In a similar fashion, ultrafast and strong optical fields permit the modification of the electronic properties of a nanometer metallic foil towards dynamical refraction. In this case of super-nonlinear optics a set of unexpected phenomena are observed, including the conversion of band-gap materials into subcycle optical limiters, saturation of saturable absorption and the quasi-instantaneous formation of a negative



refractive index,

As a corollary of this ultrafast coherent modification of the electronic system, in suitably chosen hetero-structures also the spin system can be manipulated coherently.

Optically induced spin transfer is demonstrated as a route to the direct, all-optical manipulation of macroscopic magnetic moments on previously inaccessible attosecond timescales[2] and a path to arrange magnetic domain patterns with light.

### Figure:

Light wave driven population transfer across a ferromagnet/metal interface causes a rapid spin transfer between both constituents of the interface region despite the electric field not coupling directly to the magnetic moment.

### References

[1] M. Ossiander et al., "The speed limit of optoelectronics," Nat. Commun., 2022.

[2] F. Siegrist et al., "Light-wave dynamic control of magnetism," Nature, vol. 571, 7764, 240–244, 2019.

### Driving orbital magnetism in metallic nanoparticles through plasmonic effects

<u>Giovanni Manfredi</u><sup>1</sup>, J. Hurst<sup>1</sup>, P.-A. Hervieux<sup>1</sup>, R. Sinha-Roy<sup>2</sup>, P. Oppeneer<sup>3</sup> <sup>1</sup>Université de Strasbourg, CNRS, France, manfredi@unistra.fr <sup>2</sup>Aix-Marseille Université, France <sup>3</sup>Uppsala University, Sweden

The topic of this contribution is the generation of large magnetic fields in non-magnetic materials through polarized laser fields.

Transfer of angular momentum from helicity-controlled laser fields to a nonmagnetic electronic system can lead to the creation of magnetization. The underlying mechanism in metallic nanoparticles has been identified as the inverse Faraday effect (IFE), whereby a quasi-static magnetic field is generated by an external oscillating lase field and is proportional to the laser intensity.

Here, we show that the IFE can be strongly amplified in small gold nanoparticles thanks to plasmonic effects. If the laser frequency matches the plasma frequency of the conduction electrons in the metal (surface plasmon resonance), a strong oscillating electric field is excited in the nanoparticle. Through the IFE, this internal self-consistent field generates a sizeable magnetization, of the order of tens of Bohr magnetons. The primary contribution to the magnetization comes from surface currents generated by the self-consistent field. The effect is maximum for circularly polarized laser fields and disappears for linearly polarized fields.

This plasmonic IFE is studied here using both a simplified quantum hydrodynamic model [1] and fully quantum simulations based on the time-dependent density functional theory [2].

This is an important step in the ultrafast manipulation of magnetic effects in nano-objects via electromagnetic waves, which may find applications for the storage, writing, and reading of information based on optical means.



Figure 1: Circularly polarized laser excitation of a potassium K561 cluster. From top to bottom, the panels show: the time dependence of the x (red) and y (blue) components of the laser electric field (e); the three components of the dipole moment (f); the total energy absorbed by the electronic system (g); and the three components of the magnetic moment, in units of the Bohr magneton  $\mu$ B (h). From Ref. [2].

### References

[1] J. Hurst, P. M. Oppeneer, G. Manfredi, P.-A. Hervieux, Magnetic moment generation in small gold nanoparticles via the plasmonic inverse Faraday effect, Phys. Rev. B **98**, 134439 (2018).

[2] R. Sinha-Roy, J. Hurst, G. Manfredi, P.A. Hervieux, Driving Orbital Magnetism in Metallic Nanoparticles Through Circularly Polarized Light: A Real-Time TDDFT Study, ACS Photonics 7, 2429-2439 (2020).

### Monolithic van der Waals metasurfaces

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Van der Waals (vdW) materials, such as hexagonal boron nitride (hBN) and Transition Metal Dichalcogenides (TMDCs) semiconductors, are layered crystals with exceptional properties to investigate light-matter interaction at the nanoscale. In their atomically thin form they exhibit appealing features, such as tightly bound excitons and optically addressable defects, while in their bulk form they exhibit giant optical anisotropy and large refractive index values (n>4), larger than common semiconductor materials, making them a favorable candidate for the realization of low-loss optical resonances in all-dielectric nanophotonic structures.

In our work, we leverage the physics of quasi bound states in the continuum (qBIC) to achieve high quality (Q) factors optical resonances in symmetry-broken dielectric metasurfaces. Notably, our approach is monolithic, meaning that it is exclusively composed of vdW materials, and allows to realize optical resonances with Q factors above  $10^2$  through a two-step fabrication process. We demonstrate spectral tuning over the whole visible spectrum in hBN qBIC metasurfaces [1] and enhanced light-matter coupling with intrinsic spin defects in hBN [2]. In the latter, we observe a remarkable 25-fold enhancement of the photoluminescence intensity and spectral narrowing of the defect emission, with linewidth below 4 nm full width at half-maximum. Moreover, our platform opens exciting opportunities for strong light-matter coupling, demonstrated in the clear anti-crossing behavior between qBIC resonances and intrinsic excitons in monolithic TMDC WS<sub>2</sub> metasurfaces, exhibiting Rabi splitting up to 116 meV under ambient conditions and independent on the material's intrinsic losses [3].

Our results demonstrate how merging qBIC photonic metasurfaces with vdW materials paves the way to the realization of novel hybrid nanophotonic platforms and room temperature polaritonic devices.

### References

[1] L. Kühner, L. Sortino, et al. "High-Q Nanophotonics Over the Full Visible Spectrum Enabled by Hexagonal Boron Nitride

Metasurfaces" Advanced Materials 35, 13 (2023)

[2] L. Sortino, et al. "Optically addressable spin defects coupled to bound states in the continuum metasurfaces" arXiv:2306.05735 (2023)

[3] T. Weber, L. Kühner, L. Sortino, et al. "Intrinsic strong light-matter coupling with self-hybridized bound states in the continuum in van der Waals metasurfaces" Nature Materials http://dx.doi.org/10.1038/s41563-023-01580-7 (2023)

## Optical properties of self-assembled dense spherical clusters of plasmonic nanoparticles

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Densely-packed spherical colloidal clusters of metallic inclusions, also known as plasmonic balls [1], have garnered a lot of interest recently, owing to their remarkable scattering behaviors and potential applications. Using an emulsion route, we have been able to produce dense spherical balls and show that they act as resonant Huygens scatterers [2], where the intereferences of multipoles of even and odd parity lead to coherent forward scattering [3, 4]. We shall review recent designs and realizations of such Huygens scatterers using self-assembly.

The interaction of light with inhomogenous spheres such as these clusters is notoriously difficult to describe theoretically. Even when the inclusions behave as small resonant electric dipoles, the ensemble properties of the cluster are radically different to those of the inclusion because of multiple scattering interactions. These systems are of particular interest because they involve localized resonant inclusions that are assembled into a Mie resonator that provide a lot of leverage experimentally in the engineering of the spectral scattering characteristics of the clusters. We have studied numerically the electromagnetic behavior of plasmonic balls composed of many particles using high-precision T-matrix calculations. We have shown that it is empirically possible to find an equivalent effective medium description for the clusters, taking into account spatial dispersion. We find that the average scattered field as well as the average inner field of a spherical cluster as computed from the T-matrix approach can be equivalently obtained by an extended Mie theory where three effective parameters are used to describe the inner effective medium, namely an electric permittivity  $\mathcal{E}_{eff}$ , a magnetic permeability  $\mu_{eff}$ , and a longitudinal wavevector  $k_{L}$ . The latter two account for strong interparticle couplings entailing spatial dispersion effects, which cannot be neglected in dense systems near the plasmonic resonance. Our study therefore shows that (within the range of studied sizes), it is possible to treat a cluster of plasmonic particles as a sphere made of a spatially-dispersive effective equivalent medium, even for high concentration in particles. This work broadens the range of effective parameters that can be obtained and exploited in the design of meta-atoms and metamaterials.



Figure 1: Effective medium description of plasmonic balls. (a) The cluster of radius R is composed of a dense ensemble of gold inclusions of radius r embedded in a medium of refractive index 1.4. The entire cluster itself is embedded in a medium of refractive index 1.42. (b) Equivalent homogeneous sphere with effective parameters  $\varepsilon_{\rm eff}$ ,  $\mu_{\rm eff}$  and  $k_{\rm L}$  that enable the computation of the field scattered by the cluster using Mie theory. Comparison between the total averaged electric field of the plasmonic cluster (c) and equivalent homogeneous sphere at  $\lambda = 800$  nm (d).

- [1] P. Yazhgur, N. Muller, and F. Scheffold, ACS Photonics 9, 2809-2816 (2022).
- [2] R. Elancheliyan, R. Dezert, S. Catano, A. Bentaleb, E. Nativ-Roth, O. Regev, P. Barois, A. Baron, O. Mondain-Monval, and V. Ponsinet, Nanoscale 12, 24177-24187 (2020).
- [3] R. Dezert, P. Richetti, and A. Baron, Phys. Rev. B 96, 180201 (2017).
  [4] R. Dezert, P. Richetti, and A. Baron, Optics Express 27, 26317-26330 (2019).

### **Photonic Temporal and Time Crystals**

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A photonic temporal crystal is characterized by its optical properties being periodically modulated over time, rendering it a temporal counterpart to spatially periodic photonic crystals. As early as 1966, the electromagnetic wave dynamics in a space-time periodic medium were theoretically explored, with the aim to describe time-growing instabilities in distributed parametric media [1]. However, only a handful of theoretical studies followed this initial investigation, until an experiment utilizing a dynamic transmission line confirmed the existence of a shallow, yet genuine, momentum gap [2].

Following this pioneering work, the popularity of time-varying photonics surged recently, with a primary focus on the conceptual extension of photonic spatial crystals and metamaterials into the space-time domain. More specifically, the enhanced dispersion and band structure engineering capabilities, facilitated by the additional temporal degree of freedom, have been the subject of extensive research [3]. A multitude of intriguing phenomena such as colossal broadband nonreciprocity, efficient one-way amplification, parametric oscillation, pulse compression, and harmonic generation, have been theoretically or numerically considered for potential applications.

It was only recently, however, that Floquet systems analysis was employed to shed more light on photonic temporal crystals [4]. The momentum gap was confirmed to be the broken phase of parity-time (PT) symmetry along the wavenumber axis, while its edges were identified as non-Hermitian degeneracies, or exceptional points [5]. In this talk, I will discuss non-Hermitian band structures, local density of states, and light-matter interactions in photonic temporal crystals. Moreover, I will delve into how nonlinearity can lead to discrete time translational symmetry breaking and result in the so-called time-crystalline behaviour within photonic temporal crystals.

- [1] E. S. Cassedy, Proceedings of the IEEE 55, 1154 (1967).
- [2] J. R. Reyes-Ayona and P. Halevi, App. Phys. Lett. 107, 074101 (2015).
- [3] C. Caloz and Z-L. Deck-Léger, IEEE Trans. Antennas Propag. 68, 1583 (2018).
- [4] N. Wang, Z.-Q. Zhang, and C. T. Chan, Phys. Rev. B 98, 085142 (2018).
- [5] J. Park et al., Sci. Adv. 8, eabo6220 (2022).

### Photonic bound states in the continuum for spectrally selective nanophotonics

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Photonic bound states in the continuum (BICs) have become a prominent and influential concept in nanophotonics [1,2], with applications spanning the gamut from high-harmonic generation and biospectroscopy to lasing and generalized phase front control. BIC-based metasurfaces where the asymmetry of the constituent unit cells is carefully controlled have emerged as a particularly flexible design, providing versatile control over the resonance properties, including the spectral location and the linewidth. To further develop these advances, new BIC-based configurations are needed, which overcome some of the current constraints related to generally large metasurface footprints, the need for complex polarization states, or fabrication limits requiring constant resonator heights throughout the structures. In my talk, I will present several recent concepts for obtaining additional nanophotonic functionalities in BIC-based systems. First, I will describe how radial BICs can be realized by arranging asymmetric double rod unit cells in a semi-infinite ring structure to avoid edge effects and provide a polarization-invariant optical response [3]. Notably, radial BIC modes retain the geometry-induced resonance tuning capabilities of traditional 2D metasurface arrays, but at much more compact footprints as low as 2 square microns, enabling applications for biomolecular sensing and enhanced second harmonic generation from transition metal dichalcogenides. Second, I will introduce the experimental realization of height-driven BICs, where an additional degree of freedom for metasurface design is created by tailoring the height difference of multiple resonators within an individual unit cell, which provides an ideal platform for the efficient interaction of the structures with circularly polarized light [4]. Such advanced metasurface concepts are important building blocks for future applications, especially in ultra-compact and chip-integrated settings.

- S. I. Azzam and A. V. Kildishev, Photonic Bound States in the Continuum: From Basics to Applications, Advanced Optical Materials 9, 2001469 (2021).
- [2] K. Koshelev, G. Favraud, A. Bogdanov, Y. Kivshar, and A. Fratalocchi, Nonradiating photonics with resonant dielectric nanostructures, Nanophotonics 8, 725-745 (2019).
- [3] L. Kühner, L. Sortino, R. Berté, J. Wang, H. Ren, S. A. Maier, Y. S. Kivshar, and A. Tittl, Radial bound states in the continuum for polarization-insensitive nanophotonics, Nature Communications 13, 4992 (2022).
- [4] L. Kühner, F. J. Wendisch, A. A. Antonov, J. Bürger, L. Hüttenhofer, L. S. Menezes, S. A. Maier, M. V. Gorkunov, Y. Kivshar, and A. Tittl, Unlocking the out-of-plane dimension for photonic bound states in the continuum to achieve maximum optical chirality, arXiv:2210.05339 (2022).

## Plasmonic and dielectric resonances of homogeneous objects: from quasistatic to the full-wave regime

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In the linear regime, resonant electromagnetic scattering from non-magnetic and small objects, occurs according to different mechanisms. If the real part of the permittivity is negative (e.g., as in metals), resonances arise from the interplay between the electric field energy of the electro-quasistatic current modes and the polarization energy. It is well established that when the size of the object is much smaller than the incident wavelength in vacuum, plasmon resonances can be predicted by the electroquasistatic approximation of the Maxwell-equations, and they are associated with the values of permittivity for which source-free electrostatic fields exist [1].

A small dielectric object with positive permittivity may also resonate when the free-space wavelength is large in comparison with the object dimensions if the permittivity is sufficiently high. In the first part of this presentation, we show that these resonances can be described by the magnetoquasistatic approximation of Maxwell's equations in which the normal component of the displacement current density field vanishes on the surface of the particle. These resonances are associated with the values of permittivity and frequency for which source-free quasistatic magnetic fields exist, which are connected to the eigenvalues of a magnetostatic integral operator [2].



Figure 1: Magnetoquasistatic current modes of a cylindrical nanoprism.

Unfortunately, both the electroquasistatic and magnetoquasistatic descriptions fail to correctly account for the frequency shift and the radiative broadening of the plasmon and dielectric resonances when the size of the object becomes comparable to the wavelength of operation. Thus, in the second part of this presentation, starting from the full-wave eigenvalue problem [3, 4] we introduce radiation corrections to the electroquasistatic and magnetoquasistatic resonances and modes of arbitrarily shaped objects, which only depend on the quasistatic current density modes. These radiation corrections enable to derive closed-form expressions of the frequency shift and the radiative quality factor (Q-factor) of both plasmonic and dielectric modes of small objects, where the dependencies on the material and the size of the object are factorized. In particular, it is shown that the radiative Q-factor explicitly depends on the multipolar components of the quasistatic mode and its corrections [5].

- [1] I. D. Mayergoyz, D. R. Fredkin, and Z. Zhang, Phys. Rev. B 72 155412 (2005).
- [2] C. Forestiere, G. Miano, G. Rubinacci, M. Pascale, A. Tamburrino, R. Tricarico, and S. Ventre, Phys. Rev. Research, 2 013158 (2020).
- [3] D. J. Bergman and D. Stroud, Phys. Rev. B 22, 1980.
- [4] C. Forestiere and G. Miano, Phys. Rev. B 94 201406 (2016)
- [5] C. Forestiere, G. Miano, and G. Rubinacci, Phys. Rev. Research 2 043176 (2020)

### Holes in Silicon enable Extreme Ultraviolet Metaoptics

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Extreme ultraviolet (EUV) radiation is crucial for attosecond science and semiconductor lithography. Yet, due to the physics of the refractive index and absorption in solids, optics for this range are scarce. In our study [1], we push the boundaries of dielectric metasurfaces [2–5], creating affordable EUV metalenses that operate at 50 nm wavelength.

To achieve EUV metasurfaces, we exploit that the real part of the refractive index of crystalline Silicon is smaller than one between 35 nm and 190 nm wavelength. Accordingly, we invert the design paradigm of most dielectric metasurfaces: by etching holes in a Silicon membrane (n < 1), we create light-guiding high refractive index perforations (n = 1) (Fig. 1a). Varying the hole diameter allows phase-shifting transmitted 50-nm wavelength light by up to 1.5  $\pi$  (Fig. 1a). Absorption is limited because most light travels within the holes.

To prove the concept, we matched the radial phase profile of an aspheric lens with 10 mm focal length and 1 mm diameter at each position with the correct hole diameter. We then fabricated a metalens using electron beam lithography and reactive ion etching on a Silicon-on-insulator wafer (Fig. 1d). We generated broadband EUV radiation through high-harmonic upconversion in Argon gas, driven by a near-infrared fiber laser (1030 nm wavelength). By focusing the 21st harmonic (49 nm wavelength) with the metalens and using a knife-edge measurement, we determine that the metalens achieves a minimum beam waist of  $w_0 = 0.7 \pm 0.3$  um (Fig. 1b, c). This is only 1.6 times larger than the diffraction limit set by the incoming beam profile and the nominal lens parameters.

This breakthrough represents the first universal EUV optics platform. As it can realize arbitrary phase profiles, we are now exploring how it can enable holography, orbital-angular-momentum generation, and diffraction-limited focusing in the EUV spectrum.



Figure 1: Extreme ultraviolet metalens. a) metaatom library: hole diameter-dependent transmission phase of a perforated 220 nm membrane. Inset: metaatom setup. b) Knife edge characterization of the metalens focusing extreme ultraviolet light at different positions z along the propagation direction. c) Waist size of the metalens-focused light beam around the focus. d) Scanning electron micrograph of one zone of the fabricated metalens compared to the design (red).

- [1] M. Ossiander et al., Science 380, pp. 59-63 (2023).
- [2] M. L. Tseng et al., Sci. Adv. 8, eabn5644 (2022).
- [3] Y. Deng et al., Adv. Mater. **30**, 1802632 (2018).
- [4] K. Huang et al., Laser Photonics Rev. 13, 1800289 (2019).
- [5] C. Zhang et al., Light Sci. Appl. 9, 55 (2020).

## Persistent sub-radiant states with plasmonic nanocavities

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Light-matter strong coupling at room temperature can be reached with molecular emitters placed in plasmonic gaps of just 1-2nm [1, 2]. This has paved the way for controlling quantum matter with light, and ultimately generating quantum states at room temperature, without the complex and cumbersome experimental methods required at cryogenic temperatures.

Since the first experimetal demonstration of strong coupling at room temperature, we have gained a lot of understanding on how light-matter interactions occur at such small plasmonic gaps [3, 4]. Here [5], we demonstrate persistent sub-radiant states formed between 2 or more quantum emitters residing within such plasmonic systems. We develop a quantum electrodynamics description for an open cavity to obtain the quantum dynamics of the system. Figure 1 shows the quantum dynamics of 2 emitters in a plasmonic nanocavity and reveal persistent sub-radiant states formed between them. Although the Rabi oscillations between the plasmon and emitters decay very fast (within few tens of fsec), the sub-radiant states persist for up to 100 fsec, depending on the inherent non-radiative losses of the molecular emitters chosen.



Figure 1: The nanoparticle on mirror (NPoM) with two emitters within the cavity (left) and the quantum dynamics of the system with the symmetric and anti-symmetric states shown in black (right)

Using the same model, we also obtain the quantum extinction cross section:

$$\langle \sigma_{ext} \rangle = -\sqrt{\kappa_{in}} \frac{\alpha \langle a^{\dagger} \rangle + \alpha^* \langle a \rangle}{c_0 |\alpha|^2} \tag{1}$$

where  $\sqrt{\kappa_{in}}$  is the rate that energy couples into the system by a monochromatic source,  $\alpha$  is the amplitude of the coherent state defined by the incident source's photon flux  $c_0|\alpha|^2$  and  $a, a^{\dagger}$  are the bosonic creation and annihilation operators. This quantum extinction cross-section allows us to theoretically predict experimental observables, and in fact in the absence of quantum emitters reduces to the classical behavior of the plasmonic nanocavity.

- Chikkaraddy, R., de Nijs, B., Benz, F., Barrow, S.J., Scherman, OA, Rosta, E., Demetriadou, A., Fox, P., Hess, O., Baumberg, JJ., Nature, 535, 127-130 (2016).
- [2] Zengin, G., WersĤll, M., Nilsson, S., Antosiewicz, T.J., KĤll, M., Shegai T., Physical Review Letters, 114, 157401 (2015).
- [3] Kongsuwan, N., Demetriadou, A., Chikkaraddy, R., Benz, F., Turek, VA, Keyser, UF, Baumberg, JJ., Hess O., ACS Photonics, 5, 186 (2018).
- [4] Demetriadou, A., Hamm, JM., Luo, Y., Pendry, JB., Baumberg, JJ., Hess O., ACS Photonics, 4, 2410 (2017).
- [5] Bedingfield, K, Yuen, B., Demetriadou, A., submitted (2023).

# Inverse Design for quantum nanophotonics: qubit entanglement and Bell state preparation

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In this talk, I will explore the generation of entanglement between two quantum emitters through the inverse-design engineering of their photonic environment. By means of a topology-optimization approach, I will show how dielectric cloaks can be generated that operate at different inter-emitter distances and incoherent pumping strengths. I will show that the structures obtained yield steady-state concurrence values much larger than those attainable in free space, approaching the limit of maximum-entangled-mixed-states. Next, I will show how the emitter pair can be prepared, with fidelities approaching unity, into the symmetric and anti-symmetric Bell states under coherent pumping, again, through the inverse-design of the dielectric medium hosting them.

[1] A. Miguel-Torcal et al., Nanophotonics 11, 4387 (2022).

[2] A. Miguel-Torcal et al., in preparation (2023).

## Modeling Collective Light Emission by a few Solid-State Quantum Emitters

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We present recent results on collective emission (superradiance and subradiance) by quantum emitters. The first part is about the effect of making the rotating-wave approximation. This is a very common approximation and already Dicke made it in his seminal work on superradiance [1]. While single-emitter spontaneous emission rates are the same whether the approximation is made, this does not hold for collective emission rates: for two identical emitters, the sub- and superradiance decay rate are still the same whether ones makes the approximation or not, but for two slightly detuned emitters or for three or more identical emitters, the collective rates that one finds will generally be different, and the difference can be considerable [2], as illustrated for the case of two detuned emitters in the Figure.



Figure 1: The collective decay rates (in units of free-space decay rate  $\gamma$ ) of two atoms in the RWA (dashed lines) as compared to full-interaction (solid lines) formalisms. The red curve is the difference of these scaled decay rates in both formalisms. (a) Detuning dependence of two-atom collective decay rates at a fixed distance with  $k_0R = 1/2$ . (b) Super- and subradiant decay rates at a finite frequency detuning  $\delta = \gamma$ , as a function of interatomic separation. In both panels, the maximal error due to making the RWA is indicated by a vertical dashed line. (Figure reproduced from Ref. [2]).

In the second part we present results on solid-state quantum emitters in photonic nanostructures. These emitters are open quantum systems in a double sense, as they typically couple both to the electromagnetic field and to phonons. In state of the art experiments it has become possible to engineer collective light emission despite the presence of phonons [3]. We present and compare two methods to calculate superradiant spectra that take both Markovian and non-Markovian effects of phonons into account [4].

- [1] R. H. Dicke, Phys. Rev. 93, 99-110 (1954).
- [2] M. A. Jørgensen and M. Wubs, J. Phys. B: At. Mol. Opt. Phys. 55, 195401 (2022).
- [3] A. Tiranov et al., Science **379**, 389–393 (2023).
- [4] D. Pandey et al., in preparation (2023).

### Tip-enhanced cavity-spectroscopy to control excitonic behaviors at the nanoscale

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The tunability of the bandgap, radiative emission, and energy transfer in transition metal dichalcogenide (TMD) monolayers provides a new class of functions for a wide range of ultrathin photonic devices. Additionally, understanding and controlling the nanoscale transport of excitonic quasiparticles, such as excitons and trions, in atomically thin 2D semiconductors are crucial to produce highly efficient nano-excitonic devices. In this work, we present a dynamic nano-mechanical strain-engineering of naturally-formed wrinkles in a WSe<sub>2</sub> monolayer, with real-time investigation of nano-spectroscopic properties using tip-enhanced cavity-spectroscopy [1-4]. We reveal the modified nano-excitonic properties by the induced tensile strain at the wrinkle apex, exhibiting the exciton funneling phenomenon. In addition, we demonstrate a nanogap device to selectively confine excitons or trions of 2D TMDs at the nanoscale, facilitated by the drift-dominant exciton funneling into the strain-induced local spot. Furthermore, we present a method for the all-optical control of the exciton-to-trion conversion process and its spatial distributions in a MoS<sub>2</sub> monolayer. We exploit propagating surface plasmon polaritons (SPPs) to localize hot electrons in a 2D TMD transferred on a metal–insulator–metal (MIM) waveguide. Our work provides a new strategy for robust, tunable, and ultracompact nano-excitonic devices using atomically thin semiconductors.



Figure 1: **a.** Schematic diagram of all-optical trion control platform operating with adaptive optical excitation. Green dashed line indicates transferred  $MoS_2$  ML. **b**. Illustration of all-optical trion control platform facilitated by nanoscale strain gradient, plasmon-induced hot electrons, and resultant exciton-to-trion conversion.

- [1] Yeonjeong Koo et al., Advanced Materials 33, 2008234 (2021).
- [2] Hyeongwoo Lee et al., Advanced Functional Materials **31**, 2102893 (2021).
- [3] Hyeongwoo Lee et al., Science Advances 8, eabm5236 (2022).
- [4] Hyeongwoo Lee et al., Nature Communications 14, 1891 (2023).

### Beam Shaping and Frequency Conversion in Nonlinear All-Dielectric Metasurfaces

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The emergence of nonlinear flat-optics nanostructures, optical metasurfaces, enabling unprecedented enhancement of light-matter interactions without phase-matching requirements may revolutionize future applications of integrated nonlinear optics. Nonlinear wavelength conversion, including second harmonic generation (SHG) and third harmonic generation (THG), plays a key role in extending the spectral coverage of laser sources to wavelengths that are more difficult to access by standard laser gain media. Moreover, the demand for compact, high-efficiency, and multifunctional devices that can replace traditional bulky, free-space technologies, is continuously rising especially in the ultraviolet (UV) region. Indeed, coherent UV sources are essential for many applications. Currently, efficient wavelength conversion in an integrated, compact, tunable UV source, is possible only using nonlinear optical materials that are simultaneously transparent in the UV, possess high and fast nonlinear susceptibility, and are, at the same time, birefringent. Chalcogenide glasses have emerged as a promising platform for many on-chip applications at mid- and near-infrared wavelengths due to their high linear refractive indices and strong Kerr nonlinearity [1-3].

We demonstrate ultrafast tunable, near-infrared to ultraviolet frequency conversion in a chalcogenide glass metasurface based on Mie resonances and quasi-bound states in the continuum (qBIC) resonances, enabled by a phase-locking mechanism between the pump and the inhomogeneous portion of the TH signal. Through phase locking, the pump pulse and the inhomogeneous harmonic component can co-propagate, resulting in the acquisition of the same refractive index and absorption coefficient as the pump [4-8]. If this process occurs within a cavity, efficient frequency conversion can take place, even in the presence of strong material absorption at the wavelengths of the harmonics. As for all nonlinear processes, a resonant condition at the pump field boosts the nonlinear interactions.

Finally, we experimentally show the simultaneous generation of phase-locked structured light beams, including optical vortices and Hopf-links at fundamental and tripled frequencies in all-dielectric nonlinear optical metasurfaces despite the fact that the tripled frequency is corresponding to the region of high absorption of the dielectric material. This work may have useful prospects in optical manipulation, optical communication, and quantum information transmission.

### References

[1] J. Gao et al., Near-infrared to ultra-violet frequency conversion in chalcogenide metasurfaces. Nature Communications 12, 5833 (2021).

[2] J. Gao et al., All-optical tunable wavelength conversion in opaque nonlinear nanostructures. Nanophotonics 11, 4027-4035 (2022).
 [3] M. Vincenti et al., Stacked chalcogenide metasurfaces for third harmonic generation in the UV range. New Journal of Physics 24, 035005 (2022).

[4] N. Bloembergen, P. S. Pershan, Light Waves at the Boundary of Nonlinear Media. Phys. Rev. 128, 606-622 (1962).

[5] V. Roppo et al., Role of phase matching in pulsed second-harmonic generation: Walk-off and phase-locked twin pulses in negative-index media. Phys. Rev. A 76.3: 033829. (2007).

[6] M. Mlejnek, E. Wright, J. Moloney, N. Bloembergen, Second harmonic generation of femtosecond pulses at the boundary of a nonlinear dielectric. Phys. Rev. Lett. 83, 2934 (1999).

[7] V. Roppo et al., Anomalous momentum states, non-specular reflections, and negative refraction of phase-locked, second-harmonic pulses. Metamaterials 2, 135-144 (2008).

[8] M. Centini et al., Inhibition of linear absorption in opaque materials using phase-locked harmonic generation. Phys. Rev. Lett. 101, 113905 (2008).

### CMOS-based terahertz camera based on quantum-dot-enhanced upconversion

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This work focuses on the developed of new terahertz cameras based on CMOS image sensors. Detection of terahertz (THz) radiation has numerous potential applications, but currently faced with limitations in detector performance such as sensitivity, speed, bandwidth, and operating temperature. Most of THz detectors also lack the ability to determine THz polarization states. However, the recent discovery of THz-driven luminescence in quantum dots offers a viable detection mechanism through field-driven inter-quantum-dot charge transfer. We introduce a THz camera and polarimeter that functions at room temperature, utilizing a complementary metal-oxide-semiconductor and a quantum-dot-enhanced THz-to-visible upconversion mechanism. With optimized luminophore geometries and fabrication designs, this nanoslit-based sensor achieves broadband and fast responses, and is capable of detecting THz pulses with peak fields as low as 10 kV/cm. Furthermore, we present a new coaxial nanoaperture-type device that possesses a hitherto unexplored ability to record the THz polarization state and field strength simultaneously, with comparable sensitivity.

### References

[1] J. Shi, D. Yoo, F. Vidal-Codina, C.-W. Baik, K.-S. Cho, N.-C. Nguyen, H. Utzat, J. Han, A. M. Lindenberg, V. Bulovic, M. G. Bawendi, J. Peraire, S.-H. Oh, K. A. Nelson, Nature Nanotechnology (2022) 17, 1288

## Optical image differentiation with nonlinear flat optics

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We show that flat-optics elements with nonlinear response can be used to engineer Volterra kernels capable of real-time image processing [1]. To illustrate this concept, we present an edge detection system that exploits the nonlinear response of a simple flat optics element. This approach offers several advantages compared to linear flat-optics-based edge detection and differentiation [2-4], including broad operation across different frequencies due to its non-resonant mechanism, significantly enhanced contrast, and improved performance in the presence of noise. Our findings suggest that the implementation of Volterra kernels in nonlinear flat optics opens up new possibilities for analog processing and computing using nonlocal nonlinear metasurfaces. In the linear regime (Fig. 1a), a flat-optics structure has an optical transfer function  $\widehat{\chi}_{\ell m}^{(1)}(\mathbf{f})$ , i.e., a linear susceptibility that links an input function (the electric field component  $\widehat{E}_{m}^{\omega_{0}}(\mathbf{f})$ , where **f** is the spatial frequency), to an output function (the polarization component  $\widehat{P}_{\ell}(\mathbf{f})$ ). In the nonlinear regime, in addition to the linear term, the flat-optics structure supports a nonlinear contribution that can be described by a Volterra filter (Fig. 1b) that mixes spatial frequencies originating from two distinct input functions, for example two orthogonal electric-field components. For second-order nonlinear effects, the Volterra filter is then described by the kernel  $\chi^{(2)}_{\ell m n}(\mathbf{f}', \mathbf{f} - \mathbf{f}')$ , i.e., the quadratic nonlinear susceptibility. Although the nonlinear response of Volterra filters can be synthesized for a variety of functionalities, here we focus on image differentiation, specifically on edge detection. For edge detection, we need to suppress low spatial frequency components, and this condition is easily accomplished having only  $\chi_{lmn}^{(2)}$  that forbids second harmonic propagation in the far field for fundamental-frequency light impinging at normal incidence, i.e., we need a flat-optics system with  $\chi_{lmn}^{(2)}$  tensor elements having m = z and/or n = z. If Fig. 1c we show edge-detection capabilities from a  $\chi^{(2)}_{zzz}$  flat-optics element. This scenario may occur (i) at a metal surface, (ii) in multi-quantum well slabs grown in the z-direction, (iii) in a properly designed metasurface.



Figure 1: a. Image processing with a linear flat-optics system; b. image processing with a nonlinear flatoptics system; c. In red, the input image formed by an aperture with the "UNIBS" shape illuminated at fundamental frequency  $\omega_0$  and with circular polarization. In green, the output image from at the second harmonic frequency  $2\omega_0$  filtered by the Volterra kernel of a  $\chi_{zzz}^{(2)}$  sheet.

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- [1] A. Silva, F. Monticone, G. Castaldi, V. Galdi, A. Alù, N. Engheta, Science 343, 160 (2014).
- [2] A. Komar, R. A. Aoni, L. Xu, M. Rahmani, A. E. Miroshnichenko, D. N. Neshev, ACS Photonics 8, 864 (2021).
- [3] A. Overvig, A. Alù, Laser and Photonics Reviews 16, 1863 (2022)
- [4] T. Zhu, Y. Zhou, Y. Lou, H. Ye, M. Qiu, Z. Ruan, S. Fan, Nature Communications 8, 15391 (2017).

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## Large nonlinear efficiency enhancement in the visible and UV ranges from plasmonic gold nanogratings

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Nonlinear frequency conversion in metals has been a subject of theoretical and experimental research since the emergence of nonlinear optics, partially motivated by their high third order nonlinearities and despite its high absorption in the visible and near infrared spectral ranges. Enhancement of harmonic generation from gold surface has been demonstrated in different nanostructures, especially when excited by plasmonic resonances, and represents nowadays an active field of research with applications to functional nano-photonic devices. However, most of experimental results presented in literature often show only qualitative aspects of this enhancement, do not quantify the efficiency (presenting only arbitrary units) and are not in good agreement with the theoretical models set forward. Understanding the properties of light propagation and localization at the nanoscale, when light-matter interaction displays completely new and interesting physical phenomena, which are not well explained by conventional approximations, is a crucial step to design structures capable of enhancing the naturally low efficiency of harmonic generation in metals in the visible and UV ranges.

We recently carried out a combined theoretical and experimental study of second (SH) and third-harmonic (TH) generation in thin-layered gold surfaces, to extract the basic physical intrinsic properties of the material [1]. We use an expanded version of the hydrodynamic model [2] to account for the linear and nonlinear material dispersions associated with both free and bound electrons. Using those parameters, we can accurately predict the enhancement of SH and TH signals in a nanostructure showing a plasmonic resonance.

We report experimental results from a gold nano-grating having a resonance in reflection tunable in the IR from 700 nm to 1100 nm. We measure the second and third harmonic radiation as a function of the input wavelength tuned around the resonance at 800nm for a fixed incident angle. The SH (400 nm) and TH (266 nm) are emitted in diffraction orders that can be measured individually in reflection. When summed up, the SH efficiency given by the grating is about 1400 times the total SH efficiency from the planar gold nanolayer. Similarly, the total TH efficiency shows an enhancement factor of nearly 4000.



Fig. 1 (a) Schematic representation of the nanograting. (b) Grating reflectance map for TM incident polarization, as a function of incident angle and wavelength. Experimental measurements are shown as black dots on top of the simulations. (c) Reflectance as a function of incident angle for carrier wavelength tuned at 800 nm, normalized to the value obtained from the unpatterned gold layer: simulation (solid blue curve) and experimental measurements (dashed curve with red dots). The resonance is centered at  $16^{\circ}$  (d) Electric and magnetic field distributions at 800 nm and  $16^{\circ}$  angle of incidence around a single groove, normalized with respect to the incident electric and magnetic field amplitudes. (e) Simulated and measured enhancement 52 effected SHG and THG, while pump wavelength is tuned at 800 nm and incident on the sample at  $16^{\circ}$ .

The reflected SH efficiencies (at the zero-diffraction order) were also measured around 1000 nm and around 1100 nm, finding similar enhancement factors. A large set of simulations were carried out with our model, obtaining SH and TH

### Second and third harmonic generation from aluminum nanostructures

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Plasmonics is concerned mostly with the interaction of light with free charges on conductive surfaces. Historically, noble metals like gold (Au) and silver (Ag) have been the preferred choices because of their relatively low losses in the visible and near-IR portions of the spectrum. Here we report a combination of theoretical predictions and experimental observations of second and third harmonic generation (SHG and THG) from aluminum (Al) nanolayer to extract the intrinsic linear and nonlinear optical properties and establish baseline behavior, and then apply them to different grating structures in the hope of enhancing those basic properties. The push toward the ultraviolet range and beyond calls for additional studies of Au and Ag and alternative materials to determine their viability, a search that naturally highlights Al [1]. Al is relatively inexpensive and is naturally abundant on Earth. It has material stability, unique spectral capability in the ultraviolet range, and is compatible with metal-oxide-semiconductor technology. Additionally, in some studies Al has been reported to outperform silver in the visible range due to its superior surface and interface properties [2]. However, most studies have been conducted over a limited wavelength range [3] with simple effective models. Unlike most noble metals, which display Lorentz-like behavior (interband transitions) in the UV range, Al is characterized by an absorption resonance near 850nm, which uncharacteristically splits the plasmonic range, and sets its linear and nonlinear optical properties apart. Indeed, these basic properties, along with surface and volume characteristics, have yet to be explored in detail and so they may not be wellunderstood. Therefore, we set out to study Al with the aid of a hydrodynamic-Maxwell model that accounts for linear and nonlinear material dispersions, second and third order surface and volume nonlinear sources to study harmonic generation first from a simple Al layer a few tens of nanometers in thickness, and then from gratings and nanoantenna arrays that may display a combination of plasmonic and longitudinal Fabry-Perot resonances that localize the field inside a small volume. Our preliminary results suggest that bound charges play an outsized role in SHG, pointing to the fact that predictions based solely on the free electron model may not adequately be used for prediction purposes, and that the absorption resonance plays a pivotal role in THG. The intrinsic properties of Al may be more pronounced compared to either Au or Ag, and nanostructured geometries can enhance the nonlinear response while unraveling some important, fundamental aspects of the optical properties of Al at the nanoscale. In Fig. 1 we plot (a) Complex dielectric permittivity of Al fitted with a 1-Drude plus 1-Lorentz oscillator system peaked between 800nm and 850nm; (b) Transmittance, Reflectance, and Absorption of a 100fs pump incident at 30° on a 20nm-thick Al layer; (c) Transmittance, Reflectance, and Absorption of 100fs pump pulses normally incident on a resonant aluminum grating.



- [1] Gérard, Davy, and Stephen K. Gray. "Aluminium plasmonics." Journal of Physics D: Applied Physics 48.18 (2014): 184001
- [2] Zhu, Xiangchao, et al. "Beyond noble metals: high Q-factor aluminum nanoplasmonics." ACS Photonics 7.2 (2020): 416-424.
  [3] Krause, Debra, et al., "Optical surface second harmonic measurements of isotropic thin-film metals: Gold, silver, copper, aluminum, and tantalum" J. Appl. Phys. 96, (2004).

### A Transformation Optics Approach to Nonlinear and Nonclassical Plasmonics

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The success of classical plasmonics endows us with a wide range of applications, such as strong field confinement and enhancement, ultra-compact optical wave-guiding, super-resolution imaging, surfaceenhanced Raman scattering, fluorescence emission, bio-sensing, nanolasers, etc. The development of highpower laser and nano-fabrication technology further prospers the field of plasmonics, where novel nonlinear and nonclassical optical effects appear. The plasmonic effects in the nanostructure provide a strong lightmatter interaction between the structure and light, resulting in a giant nonlinear and nonclassical optical response. However, the analytical study of nonlinear and nonclassical effects in plasmonic structures is limited to basic geometries, such as flat surfaces, cylinders, or spheres. For a more complex geometry, an analytical solution becomes unobtainable. Fortunately, transformation optics, as a powerful analytical tool, has been employed in nonlinear and nonclassical plasmonics to solve this dilemma.

For nonlinear plasmonics, second and third-harmonic generations from a nanowire dimer and a singular metasurface have been analytically studied. We found that the direct and the cascaded THG possess different size-dependence that can be used for experimental characterization of a nonlinear signal. Moreover, the SHG from a singular metasurface with a hidden dimension weakly depends on the pump field's incident angle, making it a perfect candidate as an all-angle harmonic-generation device.

Regarding nonclassical plasmonics, nonlocal and electron spill-out effects have been thoroughly explored in the singular plasmonic system featured with a sharp point or a sub-nanometer metallic gap. A direct analytical approach to singular structures is complicated, but an indirect transformation optics approach that converts a complex nanostructure into a flat layered geometry becomes favorable. Obtaining the mapping rule of nonlocal parameter or Feibelman d parameter between the physical space and transformed virtual space, the complex nonclassical response of a singular nanostructure is significantly simplified.

- F. Yang and C. Ciracì, A Transformation Optics Description of Direct and Cascaded Third-harmonic Generation, ACS Photonics, ACS Photonics 10.1021/acsphotonics.3c00343 (2023).
- [2] F. Yang and C. Ciracì, Second-harmonic generation from singular metasurfaces, Phys. Rev. B, 105, 235432 (2022).
- [3] F. Yang and K. Ding, Transformation optics approach to mesoscopic plasmonics, Phys. Rev. B, 105, L121410 (2022). (EDITORS' SUGGESTION)
- [4] F. Yang and K. Ding, Electron Spill-Out Effect in Singular Metasurfaces, Photonics, 8, 154 (2021). (Editor's Choice) (Invited paper)
- [5] F. Yang, E. Galiffi, P. A. Huidobro, and J. B. Pendry, Nonlocal effects in plasmonic metasurfaces with almost touching surfaces, Phys. Rev. B, 101, 075434 (2020).
- [6] F. Yang, Y. T. Wang, P. A. Huidobro, and J. B. Pendry, Nonlocal effects in singular plasmonic metasurfaces, Phys. Rev. B, 99, 165423 (2019). (EDITORS' SUGGESTION)

### Free-space interferometric routing of upconverted light by dielectric metasurfaces

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Frequency upconversion of near-infrared photons to the visible range is strategical for information technology, as it can provide an alternative for the read out of telecom signals using efficient silicon-based detectors. Light upconversion is a nonlinear process mediated by matter that consists in the interaction of either energy-degenerate photons, such as in second-harmonic and third-harmonic generation (THG), or photons with different energies, such as in sum-frequency generation (SFG). We recently investigated frequency upconversion in both plasmonic and dielectric nanoantennas [1,2]. Thanks to the adopted dual-beam pump scheme, where an ultrashort pulse ( $\omega$ ) at telecom wavelength ( $\lambda = 1551$  nm) impinges on the sample along with its frequency-doubled replica ( $2\omega$ ), THG and SFG are degenerate in energy. This, along with coherence, enables the interference between the processes. Yet, we found that in individual nanoantennas symmetry plays a major role in enhancing/suppressing the interference between SFG and THG. By tuning the relative phase between the two impinging pulses, we performed all-optical switching of upconverted light with efficiency > 50% in asymmetric plasmonic antennas [2].

Optical metasurfaces are rapidly emerging as flexible, ultrathin and multi-functional platforms to manipulate light [3]. Recently, they were also applied to efficient nonlinear light conversion and steering [4]. Here, by applying the above dual-beam pump scheme to a periodic AlGaAs metasurfaces, we attain all-optical routing of the upconverted telecom photons in the visible range as in [1,2]. This is attained by tuning the metasurface diffraction with respect to the meta-atom nonlinear emission in the Fourier plane, hence breaking the detection symmetry. Using the relative phase between the pump pulses as a tuning knob, we routed the upconverted radiation among different metasurface diffraction orders with an efficiency up to 90%. In particular, this is enabled by the maximization of constructive/destructive interference between SFG and THG in specific k-space directions. We also demonstrate that the polarization state of both pump and emission allows to reconfigure the routing between different sets of diffraction orders. The proposed approach can be envisioned as an all-optical method to route upconverted telecom photons into various detection channels. The combination of the interferometric and nonlinear character of the emitted light could be also extremely appealing for applications to nonlinear sensing.

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### References

[1] A. Zilli, D. Rocco, M. Finazzi, A. Di Francescantonio, L. Duò, C. Gigli, G. Marino, G. Leo, C. De Angelis, M. Celebrano "Frequency Tripling via Sum-Frequency Generation at the Nanoscale" ACS Photonics 8, 1175-1182 (2021).

[2] A. Di Francescantonio, A. Locatelli, X. Wu, A. Zilli, T. Feichtner, P. Biagioni, L. Duò, D. Rocco, C. De Angelis, M. Celebrano, B. Hecht, M. Finazzi "Coherent Control of the Nonlinear Emission of Single Plasmonic Nanoantennas by Dual-Beam Pumping"

Adv. Opt. Mat. 2200757, 1-8 (2022).

[3] W. T. Chen, A. Y. Zhu, F. Capasso, "Flat Optics with Dispersion-Engineered Metasurfaces". Nat. Rev. Mater. 5, 604–620 (2020).

[4] L. Carletti, A. Zilli, F. Moia, A. Toma, M. Finazzi, C. De Angelis, D. N. Neshev, M. Celebrano "Steering and Encoding the Polarization of the Second Harmonic in the Visible with a Monolithic LiNbO3 Metasurface" ACS Photonics 8, 731-737 (2021).

## Surface-response formalism for mesoscopic electrodynamics in plasmonic nanostructures

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The electrodynamics of matter and optical phenomena are commonly explored within the framework of classical electrodynamics and semiclassical models for the interactions of light with matter. Materials are commonly assumed homogeneous, and light-matter interactions are treated in an intuitive local manner. The plasmonic response of metal nanostructures is one such example, where the understanding of mesoscopic electrodynamics at metal surfaces is, however, becoming increasingly important for both fundamental developments in quantum plasmonics and potential applications in emerging light-based quantum technologies. The addition of surface-response formalism to classical electrodynamics is a way to represent quantum aspects and microscopic details of the electrodynamics at metal surfaces. The talk will discuss recent examples of nonlocal effects that emerge in surface-plasmonic systems, including metal surfaces, 2D materials, and combinations thereof.

- [1] N. A. Mortensen, Nanophotonics 10, 2563-2616 (2021).
- [2] T. Christensen, W. Yan, A.-P. Jauho, Soljačić, and N. A. Mortensen, Phys. Rev. Lett. 118, 157402 (2017).
- [3] P. A. D. Gonçalves, T. Christensen, N. Rivera, A.-P. Jauho, N. A. Mortensen, and M. Soljačić, Nat. Commun. 11, 366 (2020).
- [4] P. A. D. Gonçalves, T. Christensen, N. M. R. Peres, A.-P. Jauho, I. Epstein, F. H. L. Koppens, M. Soljačić, and N. A. Mortensen, Nat. Commun. 12, 3271 (2021).
- [5] Y. Yang, D. Zhu, W. Yan, A. Agarwal, M. Zheng, J. D. Joannopoulos, P. Lalanne, T. Christensen, K. K. Berggren, and M. Soljačić, Nature 576, 248 (2019).
- [6] S. Boroviks, Z.-H. Lin, V. A. Zenin, M. Ziegler, A. Dellith, P. A. D. Gonçalves, C. Wolff, S. I. Bozhevolnyi, J.-S. Huang, and N. A. Mortensen, Nat. Commun. 13, 3105 (2022).
- [7] L. Zurak, C. Wolff, J. Meier, R. Kullock, N. A. Mortensen, B. Hecht, and T. Feichtner, arXiv:2307.01423.

### Optical torque induced by resonant harmonic generation in dielectric nanostructures

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Rotation and spinning of micro- and nanoscale objects is one of the central goals of optical manipulation since the discovery of optical tweezers. Optically-induced mechanical torque leading to rotation of small objects requires breaking cylindrical symmetry or absorption of a scatterer. A spherical non-absorbing particle can not rotate due to the conservation of angular momenta of light upon scattering, thus limiting the tools of optical manipulation. Here, we propose a novel physical mechanism for the angular momentum transfer to azimuthally symmetric non-absorbing particles via nonlinear light scattering. The breaking of symmetry occurs at the microscopic level manifested in the parametric generation of higher harmonics. By performing theoretical analyses and full-numerical calculations, we predict nonlinear negative optical torque due to the radiative state at the double frequency with higher projection of angular momentum.



Fig. 1 Circularly polarized light at the frequency  $\omega$  is launched onto a cylindrical dielectric particle and generates second-harmonic fields at the frequency  $2\omega$  that might have different angular momentum due to a crystalline lattice structure, producing a nonlinearity-induced optical torque enhanced by the Mie resonances.

We present the general theory of nonlinearity-induced optical torque, originating from the angular momentum transfer from the photonic field to a nanostructure via harmonics generation (see Fig. 1). We demonstrate [1] that a nonzero optical torque can appear for the case of a non-absorptive dielectric structure with a rotational symmetry and can be greatly enhanced by the Mie resonances. Notably, the resulting angular frequency can be as high as 100 kHz. Additionally, the stable rotation of circular TMDC flakes of single-layer WS2 under the circularly polarized light excitation is also possible. We believe that our work paves a way towards novel intriguing phenomena driven by nonlinearity-induced optomechanical manipulation.

### References

[1] I. Toftul, G. Fedorovich, D. Kislov, K. Frizyuk, K. Koshelev, Y. Kivshar, M. Petrov, Nonlinearity-Induced Optical Torque. Phys. Rev. Lett. 130, 243802 (2023).

### 2D semiconductor multilayers for ultra-thin nanophotonic platform

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The emergence of 2D materials stimulated intensive research on both electronic and photonic applications. Especially, transition metal dichalcogenides (TMDs) provided an excellent platform for photonic applications due to their strong light-exciton interaction. Various photonic devices such as a light-emitting device, laser, and exciton-polariton device have been successfully demonstrated experimentally using TMD monolayers. However, multilayered TMDs have attracted far less attention than TMD monolayers because they become indirect bandgap materials. Here I will present that multilayered TMD itself is a good platform for controlling light-matter interaction without integrating an external photonic structure. A TMD multilayer can be utilized for a passive optical structure because it possesses a high dielectric constant. For example, light guiding is possible along a multilayered TMD, which is very thin compared to the wavelength of light [1-2]. Because a high dielectric constant is owing to the exciton resonances, guided light along a TMD layer is referred to as exciton-polariton. I will also show that light can be further controlled using a patterned TMD multilayer [3-4]. A patterned WS2 disk structure has a very high confinement factor for lasing action because the TMD disk offers both optical modes and optical gains. As a result, we observed the lasing operation under continuouswave excitation at room temperature. We believe our results show potential for the TMD-based nanophotonics offering a small mode volume but with a lower loss compared to the surface plasmon polaritons.

### References

[1] D. -J. Shin, H. Cho, J. Sung, and S.-H. Gong\*, "Direct observation of self-hybridized exciton-polaritons and their valley polarizations in a bare WS<sub>2</sub> layer", Advanced Materials, 34, 2207735 (2022)

[2] S. W. Lee, J. S. Lee, W. H. Choi, and S.-H. Gong\*, Ultra-thin WS<sub>2</sub> polariton waveguide for efficient light guiding, Advanced Optical Materials, DOI: 10.1002/adom.202300069 (2023)

[3] J. Sung, D. Shin, H. Cho, S. Lee, S. Park, Y. D. Kim, J. S. Moon, J.-H. Kim, S. H. Gong\*, "Room-temperature continuous-wave indirect bandgap transition lasing in an ultrathin WS<sub>2</sub> disk", Nature Photonics 16, 792 (2022)

[4] H. Cho, D.-J. Shin, J. Sung and S.-H. Gong\*, "Ultra-thin grating coupler for guided exciton-polaritons in WS<sub>2</sub> multilayers", Nanophotonics, 12, 2563–2571(2023)

## Making spatial dispersion useful

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Spatial dispersion, due to repulsive interactions between electrons, have been expected to be strong enough to be experimentally observed since the 70s. Drude's model has however proved to be so accurate to describe even the response of the smallest nanoparticles than the first experimental proof that spatial dispersion (aka nonlocality) has an impact on the response of plasmonic nanostructures has been published ten years ago[1]. This has triggered a lot of excitement and subsequent publications. We are now more equipped than ever with theoretical and numerical methods to describe this phenomenon[2, 3]. This has, also, allowed to assess its real significance – and for metals in the visible the correction brought by taking nonlocality into account is just that: a correction. As a consequence, it is often difficult to distinguish its impact from the influence of other phenomena like the spill-out[4].

It seems however that doped semiconductors, whose plasma frequency is in the mid infra-red and for which the effective mass of electrons is very low, are extremely sensitive to spatial dispersion. Recent experimental data decisively shows that the longitudinal wave (actual plasmons, often called bulk plasmons) is responsible for very characteristic resonances[5]. We have experimental data with doped InAsSb pointing in this direction too. Interestingly, optical methods are routinely used to determine the doping level of such materials. We will show how, using a nonlocal model instead of a simple Drude model for such materials coupled to an optimization method[6], could allow to retrieve in a single measurement both the doping level and the effective mass of electrons – making, for once, spatial dispersion truly useful.

Considering spatial dispersion has further merits. It allows to better understand the nature of the "Berreman mode"[7], excited when the Drude permittivity is supposed to vanish, and whose characteristics are actually completely dependent on the nonlocal response of the medium. Other phenomenon, like the interferometric control of the absorption, quite unexpectedly occur in a single slab of nonlocal material illuminated from one side only. This shows that, conversely to metals in the visible, spatial dispersion can not be neglected for semiconductors in the mid-infrared range.

- [1] C. Ciracì, R. T. Hill, J.J. Mock et al, "Probing the ultimate limits of plasmonic enhancement", Science, 337 1072-1074 (2012).
- [2] E. Sakat, A. Moreau and J. P. Hugonin, "Generalized electromagnetic theorems for nonlocal plasmonics", Physical Review B, 103 235422 (2021).
- [3] J. Benedicto, R. Polles, C. Ciracì et al, "Numerical tool to take nonlocal effects into account in metallo-dielectric multilayers", JOSA A, 32 1581-1588 (2015)
- [4] M. Khalid, O. Morandi, E. Mallet et al. "Influence of the electron spill-out and nonlocality on gap plasmons in the limit of vanishing gaps", Physical Review B,104, 155435 (2021).
- [5] D. De Ceglia, M. Scalora, M. A. Vincenti et al. "Viscoelastic optical nonlocality of low-loss epsilon-near-zero nanofilms", Scientific reports, 8 9335 (2018).
- [6] M. A. Barry, V. Berthier, B. D. Wilts et al. "Evolutionary algorithms converge towards evolved biological photonic structures", Scientific reports, 10 12024 (2020).
- [7] S. Vassant, J.P. Hugonin, F. Marquier et al., "Berreman mode and epsilon near zero mode", Optics express, 20, 23971-23977 (2012).

### Nonlinear nanoplasmonics with atomically thin materials

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Plasmons—collective oscillations in the free electron plasma—constitute nanoscale optical resonators that are imbued with a nonlinear response by their supporting conductive media. In the 2D limit represented by atomically thin materials, plasmon resonances provide unprecedented levels of optical field confinement, while exhibiting relatively lower losses in pristine samples. The appealing properties of 2D plasmons are ideal for nonlinear plasmonics, which seeks to overcome the weak nonlinear response of available materials by exploiting the large near field enhancement supplied by plasmon resonances. Here we theoretically explore nonlinear light-matter interactions of 2D plasmons hosted in atomically thin materials and their heterostructures. Our investigations are based on nonclassical methods to describe graphene plasmons [1], characterized by high confinement and electrical tunability, plasmons supported by ultrathin crystalline noble metal films [2], with thickness-dependent properties and lower losses than their amorphous counterparts, and nanostructured phosphorene [3], an anisotropic two-dimensional semiconductor that hosts plasmons in highly-doped samples. We further explore possibilities to trigger nonlinear interactions on the few-plasmon level [4] and to enhance harmonic generation through synergetic interactions between plasmons in atomically-thin heterostructures [5].



**Figure 1: Second-harmonic generation in co-planar asymmetric graphene nanoribbon pairs.** (a) Schematic illustration of second-harmonic generation (SHG) in two co-planar graphene nanoribbons separated by a distance *d* and characterized by widths  $W_j$  and Fermi energies  $E_{Fj}$  for  $j \in \{1,2\}$ . (b) For a dimer formed by arranging ribbons of width  $W_1$ =160 nm and  $W_2$ =40 nm at the same doping  $E_{Fj}$ =0.4 eV, such that the frequency of the lowest-order dipolar plasmon resonance in ribbon *j*=1 is half of that in ribbon *j*=2, the effect of inter-ribbon interaction is explored in both the linear absorption cross-section (upper panel) and the SHG susceptibility (lower panel) by varying the separation distance *d*. (c) Similar to (d), but for ribbons of equal width  $W_j$ =100 nm and different doping  $E_{Fi}$ =0.2 eV and  $E_{F2}$ =0.8 eV. (d-f) SHG from co-planar ribbons spaced d=25 nm apart when: (d) the ribbons have the same doping  $E_{Fi}$ = $E_{F2}$ =0.4 eV and varying widths  $W_j$ ; (e) equal widths  $W_i$ = $W_2$ =100 nm and varying doping  $E_{Fj}$ ; (f) both the width and doping of ribbon *j*=2 are changed. (g) Similar to (f), but for a small ribbon separation d=2.5 nm. In panels (d-g) the impinging light frequency is maintained at the lowest-order dipolar plasmon resonance frequency.

- [1] J. D. Cox and F. J. García de Abajo, Acc. Chem. Res. 52, 2536 (2019).
- [2] Á. Rodríguez Echarri, J. D. Cox, F. Iyikanat, and F. J. García de Abajo, Nanophotonics 10, 4149 (2021).
- [3] L. Jelver and J. D. Cox, arXiv:2305.09532 (2023).
- [4] G. Calajò, P. K. Jenke, L. A. Rozema, P. Walther, D. E. Chang, and J. D. Cox, Phys. Rev. Research 5, 013188 (2023).
- [5] T. P. Rasmussen, Á. Rodríguez Echarri, F. J. García de Abajo, and J. D. Cox, Nanoscale 15, 3150 (2023).

### Free-electron infrared nonlinearities in heavily doped InGaAs nanoantennas

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Hydrodynamic models of free electrons in metals and in degenerately doped semiconductors can describe accurately a range of nanoscale plasmonic phenomena that occur at interfaces, nanoantennas, metasurfaces [1]. Within the hydrodynamic model, third-order nonlinear terms arise, especially when the free electrons are driven at frequencies close to their plasma frequency [2].

In the project NEHO, we aim to employ free-electron nonlinearities in n-doped InGaAs as non-linear process to make optical computation in mid-infrared integrated photonic circuits. As first step of the project, we quantify the third-order free-electron non-linearity by measuring third-harmonic generation of plasmonic antenna arrays. Figure 1a shows a sketch of the antenna and a SEM picture. Figure 1b shows the FTIR spectra of three different antenna arrays where two resonances are observed for each array. The antennas of different arrays have different doping and, therefore, different plasma frequencies that are  $\lambda_{\text{plasma}} = 7.3$ , 8.0, 9.6  $\mu$ m. The antennas are so short that their plasmonic resonance frequencies ( $\lambda_r = 8.5$ , 9.8,11.3  $\mu$ m) are pinned to the plasma frequency of the material and are not influenced much by the geometry the antennas (2.2  $\mu$ m x 0.8  $\mu$ m).

Figure 1c shows the emission spectra emitted by pumping the antenna array that have  $\lambda_r = 9.8 \ \mu m$  (medium doping) with 400fs pulses at 10.3  $\mu m$ . The third harmonic generation (THG) is observed when the mid-infrared spot is on top of the antenna array and it is strongly enhanced when the mid-infrared pump wavelength matches the plasmonic resonance of the array. No THG is observed from the undoped antenna array confirming that the TH originates from the non-linear response of free electrons.



Figure 1: (a) Sketch and SEM picture of the plasmonic antennas. (b) FTIR spectra of the antenna arrays. The vertical arrows correspond to the plasmonic resonances of the different arrays, i.e. different doping densities. (c) Emission spectra by pumping with 400fs pulses at 10.3 mm. TH emissions is observed.

### References

[1] F. De Luca and C. Ciracì, Phys. Rev. Lett. 129, 123902 (2022)

[2] F. De Luca, M. Ortolani and C. Ciracì, Phys. Rev. B 103, 115305 (2021)

### Passive and tunable flat optics with dielectric nanoantennas

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Dielectric nanoantennas and metasurfaces have recently emerged as a new nanophotonic platform paving way to new generation of flat optical components. They can control light at nanometer dimensions in an unprecedented fashion, not achievable by conventional bulk optics. This led to demonstrations of flat optics with extraordinary performance, e.g. very large numerical aperture or very large field of view. One of the existing challenges of metasurface-based flat optics is strong grating-like dispersion of metasurfaces, which typically limits their ability to perform high-quality imaging in a broad spectral range, e.g., with white light. In this talk, I will first show how combining a few large-field of view quadratic metalenses with image-processing it is possible to achieve white-light wide-field-of-view imaging potentially applicable to regular smartphones or laptops. On the other hand, I will also demonstrate that the metasurface dispersion can be used in a constructive way to achieve hyperspectral imaging systems for space applications. I will then demonstrate that integrating these nanoantennas on top of actively controlled 1D and 2D electrode arrays embedded in liquid crystals it is possible to achieve fully controllable and dynamically switchable metasurfaces with a pixel size down to 1 micrometer. These metasurface-based spatial light modulators can generate arbitrary wavefront patterns and can be used for beam steering in LiDAR devices or tunable holography in 3D holographic displays.

**Full length abstracts** 

**Posters** 

## Second-harmonic generation in monocrystalline gold nanostructures: implications of anisotropic $\chi^{(2)}$

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Second-order nonlinear processes, for example second-harmonic generation (SHG), normally do not occur in bulk gold and other noble metals, since they naturally render as centrosymmetric face-centered cubic (FCC) crystals. The centrosymmetry, however, is broken at the metal surface, giving rise to non-zero surface-normal components of the second-order susceptibility tensor  $\overline{\chi}^{(2)}$ . Exploiting these surface nonlinearities and enhancing them using plasmonic resonances is one of the central themes in the field of nonlinear plasmonics [1, 2].

Apart from the surface-normal component  $\chi_{\perp\perp\perp}^{(2)}$  of the second-order susceptibility tensor, which was shown to be dominant at the surface of polycrystalline gold [3], the tangential components  $\chi_{\parallel\parallel\parallel}^{(2)}$ ,  $\chi_{\perp\parallel\parallel}^{(2)}$  and  $\chi_{\parallel\perp\parallel}^{(2)}$  are observed only at monocrystalline surfaces [4]. These components are associated with the trigonal symmetry of the FCC crystal and lead to the anisotropic second-order nonlinear response. This manifests in a six-fold pattern in the dependence of SHG on the sample orientation angle at 111 monocrystalline gold surfaces (see Fig. 1a).

In this work we investigate the possibility to enhance the nonlinear response of plasmonic nanostructures fabricated from monocrystalline gold flakes, which emerged recently as an alternative high-quality material platform for plasmonics [5]. Having grown especially large gold flakes [6], we fabricated plasmonic meta-gratings using focused ion-beam (FIB) milling (Fig. 1b–d). These meta-gratings exhibit resonance at the excitation wavelength of 800 nm and have different orientations with respect to the crystalline axis of the flake. Thus, we aim to exploit both plasmonic resonance and strong  $\chi^{(2)}_{\parallel\parallel\parallel\parallel}$  at 111 gold surface. Preliminary nonlinear optical characterization results, shown in Fig. 1e–h, indicate that SHG has a stronger dependence on the orientation of plasmonic resonances with respect to the crystal axis, as compared with nonlinear photoluminescence (NPL), a third-order nonlinear incoherent process that is often used for the characterization of plasmonic field-enhancement.



Figure 1: (a) Co- and cross-polarized SHG signal dependence as a function of crystal orientation with respect to the excitation polarization. (b) Optical and (c) SEM micrographs of FIB-patterned monocrystalline gold flake (scale bars:  $20\mu$ m); (d) close-up tilted-view of the milled grating (scale bar: 200 nm). (e-f) SHG and (g-h) NPL confocal scan images (logarithmic scale) of the sample at (e and f) 0° and (f and h) 30° angle orientations with respect to the excitation polarization.

- [1] Kauranen, M. and Zayats, A.V. 2012. Nat Photon., 6, 737
- [2] Butet, J.; Brevet, P.F.; Martin, O.J.F. 2015. ACS Nano, 9, 10545
- [3] Wang, F.X.; Rodríguez F.J.; Albers W.M.; et al. 2009 Phys. Rev. B 80, 233402
- [4] Boroviks, S.; Yezekyan T.; et al. 2021. Opt.Lett., 46, 833
- [5] Huang, J-S.; Callegari, V.; et al. 2010. Nat. Comm., 1, 150
- [6] Kiani, F. and Tagliabue, G. 2022 Chemistry of Materials 34, 1278

## Plasmonic-like hot-electron nonlinear photoluminescence from patterned ITO thin films

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Nonlinear photoluminescence (NPL) is known as a property of noble metals to generate a broad upconverted signal under near-infrared excitation [1]. This broad emission is spanning the whole visible region decaying towards the short wavelength range, see Figure 1. For low laser irradiance, NPL emission is attributed to a nonlinear absorption process, while at large power density, it is the radiative signature of an electron bath brought at temperatures exceeding thousands of degrees [2]. In the realm of nonlinear transparent conductive oxides, indium tin oxide (ITO) is a widely used material [3] sustaining a freeelectron system with dispersive properties enabling controllable nonlinear properties notably in the context of epsilon-near-zero material [5]. Here we demonstrate the emergence of NPL emission from ITO- coated glass substrate [?]. The signal shares strong similarities with that commonly detected from noble metals. The emission is activated by the action of a focused Ga ion beam (Ga-FIB) on the surface, which modifies the electronic band structure of the film. An in-depth analysis of the NPL dependency on laser intensity unambiguously unveils the role of a heated electron gas in the emission process.



Figure 1: NPL emission emerging from Ga-FIB-patterned thin ITO layer under NIR pulsed laser excitation.

- [1] K. Malchow, and A. Bouhelier, J. Opt. Soc. Am. B 38, 576-583 (2021)
- [2] L. Roloff, P. Klemm, I. Gronwald, R. Huber, J. M. Lupton, and S. Bange, Nano Lett. 17, 7914-7919 (2017)
- [3] A. Capretti, Y. Wang, N. Engheta, and L. Dal Negro, ACS Photonics 2, 1584-1591 (2015)
- [4] G. K. Dalapati, et al., J. Mater. Chem. A 9, 1662-16684 (2021)
- [5] F. Dell'Ova, et al., ACS Applied Optical Materials 1, 244-25 (2023)

## Quasi-periodic snap-buckling mechanisms in polymeric nano-bubbles: Toward highly efficient radio-acoustic energy transducers

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This work reports the results of a theoretical and numerical study showing the occurrence of stochastically resonating bistable dynamic in nanometric bubbles made of polymer with stiffer core and softer shell. The bubble is submerged in water, excited with a pulsed laser working in the Mega-Hertz band and tuned to match both an optical and acoustic resonance of the bubble [1,2,5]. The laser interacts with the carbon nanotubes embedded in the bubble shell thus generating heat. The concurrent action of the generated heat with the standing acoustic oscillations, gives rise to a stochastically resonating bistable system. The system in fact is forced to switch between two states (identifiable with the creation and organized disruption of a quasi-hexagonal tessellation [3]) via a snap-through-buckling mechanism [4]. This phenomenon generates pressure oscillations of unprecedented intensity. These results open the way to develop a new type of nano-transducers for radio-acoustic imaging applications able to work in the Mega-Hertz band [5]. From a general thermodynamic perspective, the reported mechanism shows remarkable characteristics of periodicity and energy conversion efficiency.

### References

[1] Ali Hatef, Behafarid Darvish, Adrien Dagallier, Yevgeniy R Davletshin, William Johnston, J Carl Kumaradas, David Rioux, and Michel Meunier. *Analysis of photoacoustic response from gold–silver alloy nanoparticles irradiated by short pulsed laser in water*. The Journal of Physical Chemistry C, 119(42):24075–24080, 2015.

[2] Peitao Xie, Zhongyang Wang, Zidong Zhang, Runhua Fan, Chuanbing Cheng, Hu Liu, Yao Liu, Tingxi Li, Chao Yan, Ning Wang, et al. *Silica microsphere templated self-assembly of a three-dimensional carbon network with stable radio-frequency negative permittivity and low dielectric loss.* Journal of Materials Chemistry C, 6(19):5239–5249, 2018.

[3] Fan Xu, Shichen Zhao, Conghua Lu, and Michel Potier-Ferry. *Pattern selection in core-shell spheres*. Journal of the Mechanics and Physics of Solids, 137:103892, 2020.

[4] Alessandro Speciale, Raffaele Ardito, Marco Baù, Marco Ferrari, Vittorio Ferrari, and Attilio A Frangi. *Snap-through buckling mechanism for frequency-up conversion in piezoelectric energy harvesting*. Applied Sciences, 10(10):3614, 2020.

[5] Ricardo Martin Abraham-Ekeroth and Francesco De Angelis. *Radioplasmonics: Plasmonic transducers in the radiofrequency regime for resonant thermo-acoustic imaging in deep tissues.* ACS Photonics.

## Ab initio Study of a Metal-Molecule System for Polaritonic Chemistry Applications

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In this work a fully-QM scheme, based on an atomistic description in the linear response framework of the TD-DFT of both the matter and light components, is proposed to analyze a chosen isomerization pathway for an azobenzene molecule interacting with a plasmonic nanocluster put in its proximities (Fig. 1). Azobenzene represents one of the simplest photochromes able to interconvert between trans- and cis- forms. Despite its isomerization process has not yet been fully understood, numerous applications have been developed: in biomedicine, in bioimaging, in photopharmacology, etc. The metallic counterpart is a tetrahedral cluster of  $Ag_{20}$ . This choice is due to the fact that this cluster has a simple spectrum dominated by only one narrow excitation in the same spectra range of Azobenzene which can be associated to a localized plasmon [1]. Recent works have shown that the strong coupling between surface plasmon-polariton mode and excitons could lead to the formation of hybrid states, namely polaritons or plexcitons [2]. Here we show the onset of these new states for particular conditions and how these can modify the potential energy landscape of the molecule. Moreover, we introduce a new figure of merit defined *Polaritonic Index* (PI) which allow us to identify all the hybrid excitations and among them to distinguish polaritonic from charge-transfer states [3]. Beyond polaritonic states, responsible for the perturbation of the S<sub>2</sub> and S<sub>3</sub> PoPESs, through a PI-based analysis, a second category of hybrid states appeared: the charge-transfer (CT) states. They represent a cardinal phenomenon in many areas of science and a challenging theoretical issue.



Fig. 1: (a) Sketches of the 4 hybrid systems corresponding to Azobenzene put 5Å away from the cluster. (b) Reconstruction of the first PESs of the hybrid system. Dots represent the TD-DFT collective excitations with a polaritonic index larger than 1.99. Colored ones refer to the configurations shown on the left side. The PESs of the first 5 electronic states of the Azobenzene alone are reported (in black continuous line) for a comparison.

### References

[1] S. D'Agostino, R. Rinaldi, G. Cuniberti and F. Della Sala, "Density functional tight binding for quantum plasmonics", J. Phys. Chem. C., Vol. 122, No. 34, 19756–19766, 2018.

[2] G. Giannone, F. Della Sala and S. D'Agostino, "Atomistic investigation of hybrid plasmonic systems", Nanomater. Nanotechnol., Vol. 9, 1847980419856534, 2019.

[3] L. Cascino, S. Corni and S. D'Agostino, "Plexcitonic Chemistry: Revealing the Interplay between Polaritonic and Charge Transfer Excitations", submitted to ACS NANO (2023).

## Mid-infrared Berreman modes tuning in GaN/AlGaN visible multilayer cavities on Sapphire for broadband nonlinear frequency conversion.

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The interest in the infrared optical response of polar materials is mainly motivated by the existence of the Reststrahlen bands, high reflectivity bands situated between longitudinal and transverse optical phonon frequencies. In this region the electric permittivity becomes negative, allowing for the excitation of surface waves named surface phonon polaritons. By combining different polar materials, it becomes feasible to induce leaky coupling modes known as Berreman modes [1], without requiring grating or prism coupling mechanisms.

In our work, we explore the potential of tuning and controlling a Berreman mode through AlGaN/GaNtype heterostructures on a Sapphire substrate. The interest for GaN and its Al- alloys ranges from the visible to the THz. Because of the large energy gap, it is possible to design and tailor optical and electromagnetic properties of this kind of structure to be used in nonlinear frequency conversion, for example [2-3]. Here we show that it is possible to take advantage of two effects of field localization, in different frequency regimes. Starting with a multilayer microcavity, properly designed for applications in the visible range we analyze the mid-IR reflection spectrum varying both polarization angles and incidence angle.

We show that in this way it is possible to optimize both fields' localization in the visible, by tuning the multiple interference effects at each interface, and the field's confinement related to the excitation of the Berreman mode in the mid-infrared. This scheme is particularly promising for enhancing the efficiency of nonlinear second-order effects like different frequency generation in the mid-IR triggered by visible pumps.

IR reflectivity measurements were performed using FT-IR interferometer (Invenio-R, Bruker) in the spectral range of 4-25  $\mu$ m with a spectral resolution of 2 cm<sup>-1</sup>. The FT-IR platform is equipped with a reflectance unit allowing to set the angles of incidence and reflectance, from almost normal incidence (about 13°) to grazing angles (85°). Polarization state of incident light can be selected using a wide range holographic polarizing filter with motorized mounter. Corresponding spectra were obtained using Fourier transform infrared (FTIR) system in the frequency of 400-1000 cm<sup>-1</sup>. Visible transmittance spectra were performed using Hamamatsu optical spectrometer in the 350-1050 nm wavelength range, fed by a quartz tungsten halogen (QTH) lamp.

Our preliminary results show that it is possible to properly design GaN/AlGaN multilayer cavities to tune both visible and IR resonances and take advantage of the two different effects of field localizations to enhance nonlinear frequency conversion effects on a wide band.

### References

[1] Khan, I., Fang, Z., Palei, M., Lu, J. Nordin, L., Simmons, E. L., Dominguez, O., Islam, S.M., Xing, H.G., Jena, D., Podolskiy, V.A., Wasserman, D., Hoffman, A.J., Engineering the Berreman mode in mid-infrared polar materials, Opt. Express 28, 28590-285599 (2020).

[2] Nikolai Christian Passler, I. Razdolski, D. Scott Katzer, D. F. Storm, Joshua D. Caldwell, Martin Wolf, and Alexander Paarmann, "Second Harmonic Generation from Phononic Epsilon-Near-Zero Berreman Modes in Ultrathin Polar Crystal Films", ACS Photonics, 6 (6), 1365-1371, (2019)

[3] M. C. Larciprete; M. Centini; A. Belardini; L. Sciscione; M. Bertolotti; C. Sibilia; M. Scalora; A. Passaseo; B. Potì, "Second harmonic generation in films deposited by metal-organic chemical vapor deposition", Appl. Phys. Lett. 89, 131105 (2006)

### Electro-Optic Imaging of Electric fields in irradiated CdTe detectors

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In radiation detectors, the spatial distribution of the electric field plays a fundamental role in their operation. Access to this field distribution is of strategic importance, especially when investigating the perturbing effects induced by incident radiation. Here, we probe the two-dimensional electric field in a planar Schottky CdTe detector using the nonlinear Pockels effect and report on its local perturbation after exposure to an optical beam [1,2]. Our electro-optical imaging setup, together with a custom processing routine, allows the extraction of the electric-field vector maps and their dynamics during a voltage bias-optical exposure sequence. For optical irradiation on the cathode side of the CdTe detectors, a sort of optical doping effect is observed: a highly stable, flux-dependent, reversible and spatially localized space charge is induced in correspondence of the opposite anode electrode.



Figure 1. Normalized Pockels images (left), corresponding electric-field maps (right). Images correspond to situation under light, after 5 min of optical irradiation (top row), and later on under dark, 15 min after the light switch-off (bottom row). Isopotential lines (white lines) in 50 V steps are also reported in right panels. Panels taken from the open access publication Sensors, Ref. [2].

The results are in agreement with numerical simulations, allowing us to confirm a two-level model based on a dominant deep level. Such a simple model is indeed able to fully account for both the temporal and spatial dynamics of the perturbed electric field. This approach thus allows a deeper understanding of the main mechanisms affecting the non-equilibrium electric-field distribution in CdTe Schottky detectors, such as those leading to polarization.

### References

Cola, Dominici, Valletta, Sensors 22, 1579 (2022)
 Cola, Dominici, Valletta, Sensors 23, 4795 (2023)

### **Quantum Dynamics and Entanglement with Multiple Plasmonic Modes**

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In the past decade, there has been tremendous progress towards plasmonic cavities that facilitate rapid energy exchange between molecules and a single plasmonic resonance [1]. The paradigmatic example is the nanoparticle on mirror (NPoM) geometry – where sub nanometer gap control has yielded extreme field enhancements and enabled single molecule strong coupling even at room temperature [2]. Formalisms employed to describe these systems commonly assume the interaction is entirely with a single plasmonic resonance, ignoring a large collection of spectrally overlapping higher order modes [3,4].

In this work, we show that the multiple modes in plasmonic cavities dominate the quantum dynamics of the system and the emerging entangled states. We focus on the truncated sphere on mirror (TSoM) morphology, which is treated as an open system supporting a large collection of quasi-normal modes (QNM's) with complex eigenfrequencies. We show that the simultaneous strong coupling of a molecule to the near field of multiple QNMs can lead to (1) additional high frequency oscillations controlled by the mode detuning and coupling strength (2) a shift in the fundamental Rabi interaction and cavity decay rates and (3) qubit mediated mode-mode interactions. We further show that two molecules symmetrically coupled to a large collection of high-order modes can exhibit persistent sub-radiant entanglement and it can be selectively generated with multiple atoms in the gap. Understanding and controlling the interaction between molecules and a large set of plasmonic modes is crucial for harnessing the quantum properties of the realistic plasmonic system – for example in the generation of ultra-fast and non-linear operations and room temperature entanglement.



Figure 1: (A-C) Schematic of the TSoM cavity and the first 15 quasi-normal modes. (D) Single molecule excited state population as a function of time and cumulative numbers of higher order spherical QNMs. (E) Fourier transform of the excited state population as a function of dipole moment for ten spherical QNMs.

#### References

[1] M. S. Tame, K.R. McEnery, S.K. Özdemir, J. Lee, S. A. Maier & M. S. Kim, Nature Physics, 2013, Vol 9, p-329.

- [2] R. Chikkaraddy et. al, Nature Physics 2016, Vol 535, p-127
- [3] N. Kongsuwan et al, ACS Photonics 2018, 5, 1, 186–191
- [4] P. Törmä and W. L. Barnes, Rep. Prog. Phys, 2015, 78 013901

## Anomalous *Thermally Activated Delayed Fluorescence* (TADF) Response for a Phenothiazine Derivative: a TD-DFT Study

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In classical fluorescent systems, especially at room temperature, the triplet electronic states can decay only through a nonradiative way by vibration or phonon interactions, while radiative decay results forbidden. On the other hand, in phosphorescence metalorganic complex presenting heavy metals, the strong spin orbit coupling enables a radiative emission from triplet excited states. In order to overcome the drawbacks of the latter (toxicity and high costs), in the last few years, new full organic systems that recovering triplet excitation through a mechanism known as delayed fluorescence have been strongly investigated. Such process also noted as Thermally Activated Delayed Fluorescence (TADF) is possible thanks to a fast reverse intersystem crossing (rISC) which up convert triplet to singlet thanks to thermal energy. In 2012 Adachi and co-workers [1] introduced the term Thermally Activated Delayed Fluorescence to indicate the phenomenon for which the singlet state can be populated-back by electrons coming from the nearest triplet state according to what is called a reverse intersystem crossing (rISC) process. In this work we take in exam the TADF performance of a phenothiazine derivative consisting of a strong donor (phenothiazine) and an acceptor (fluorenone) in the quasi-equatorial and axial conformations [2], see Fig. 1. All simulations were performed within Density Functional Theory (DFT) framework. Electronical and optical properties were analyzed for the monomers in the non-polar cyclohexane solvent this allowing to identify the first singlet  $(S_1)$  and triplet  $(T_3)$  charge transfer states. We performed several DFT simulations to select the most stable dimer conformations and once identified the most probable systems, we compared their behavior to the monomer ones. What we found was that, with respect to the equatorial conformation, where no significant changes were noted in the aggregated form, for the axial one we obtained a reduction of 88% of the energy gap. For what concerns the spin-orbit couplings among the states, no significant changes were found out for the equatorial geometry, while for the axial geometry, by moving from the monomer to the dimer, it was registered an increase of more than one order of magnitude of the spin-orbit coupling. These results seem to be in agreement with the experimental measurements showing that, while in the monomer only a strong prompt fluorescence (PF) emission is present, the TADF emission is activated only in the aggregated state, which notably emits in the deep red (c.a. 630-650 nm, depending on the medium polarity) [3].



Figure 1. Geometry of the four analyzed systems relaxed in their ground-state S0.

- Uoyama, H., K. Goushi, K. Shizu, H. Nomura and C. Adachi, "Highly efficient organic light-emitting diodes from delayed fluorescence", Nature, Vol. 492, No. 7428, 234-238, 2012.
- Zhang, J., M. Zhu, Y. Lu, X. Zhang, S. Xiao, H. Lan and T. Yi, "Design of Stimuli-Responsive Phenothiazine Derivatives with Triplet-Related Dual Emission and High-Contrast Mechanochromism Guided by Polymorph Prediction", Eur. J. Chem., Vol. 28, No. 29, 2022. 71
- 3. Maggiore A. et al., "Activation of RED delayed fluorescence emission through aggregates formation in new fluorenone based D-A materials", in preparation (2023).

### Nonlinear and linear spatiotemporal reshaping of polariton fluids

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Polaritons are very interesting quasiparticles produced in semiconductors as a hybrid mixture of light and optical excitation of the material. They inherit a strong nonlinearity from the exciton component while retaining high coherence and a nonparabolic dispersion from the photon counterpart. These features can activate, among other effects, Bose-Einstein condensation, nonlinear quantum fluid dynamics and even quantum correlations [1].

Here, we will show diverse nonlinear spatio-temporal deformation phenomena in microcavity polaritons, where the whole fluid can be described by a collective wavefunction characterised by bistability regions, solitons and quantum vortices.

We will also discuss the fundamental repulsive nonlinearity of exciton polaritons, which can trigger the formation of two-dimensional X-waves [2] or ignite expanding shock waves and sustain stable dark soliton rings [3]. In particular, we describe a novel effect of delayed nonlinearity inversion leading to the dynamical formation of a bright soliton. The simultaneous presence of the central density singularity and the radially expanding cloud is reminiscent of the exotic structures seen in condensed matter bosonic supernovae.

We will also show how we can seed and track quantum vortices in the polariton fluid on the picosecond timescale. These vortices are characterised by a central phase singularity surrounded by an azimuthally winding cloud. The observations reveal a rich nonlinear phenomenology, such as vortex spiralling, splitting and ordered branching into newly generated secondary couples [4]. These events are reminiscent of the particle pair generation effect. Remarkably, we also observe that vortices placed in close proximity experience attractive-repulsive scenarios. Such nonlinear vortex pair interactions can be described by a tunable effective potential [5], reminiscent of the Lennard-Jones potential that exists between molecules.

Finally, we show novel forms of complex light structuring using Rabi oscillations and coherent control of quantum vortices. This results in ultrafast swirling vortices characterised by one or more internal phase singularity tubes spiralling around their propagation axis [6,7]. The emitted photonic beams are endowed with the recently described property of time-varying orbital angular momentum and self-torque [8], and their topologies subtend conformal mappings including a Bloch sphere to a real-plane stereographic projection. Such a homeomorphism provides grounds for an original perspective on the meaning and role of Berry curvature [9].

- [1] Lerario et al., Nat. Phys. 13, 837 (2017)
- [2] Gianfrate et al., Light Sci. Appl. 7, e17119 (2018)
- [3] Dominici et al., Nat. Commun. 6, 8993 (2015)
- [4] Dominici et al., Sci. Adv. 1, e1500807 (2015)
- [5] Dominici et al., Nat. Commun. 9, 1467 (2018)
- [6] Dominici et al., Phys. Rev. Research 3, 013007 (2021)
- [7] Dominici et al., Opt. Express **29**, 37262 (2021)
- [8] Rego et al., Science **364**, 1253 (2019)
- [9] Dominici et al., Commun. Phys., in press (2023)
## K-space Hyperspectral Imaging of Microcavities and Metasurfaces by an Ultrastable Common-path Interferometer

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Fourier-plane optical microscopy is a powerful technique for studying the angularly-resolved optical properties of a plethora of materials and devices [1]. The information about the direction of the emission of light by a sample is extracted by imaging the objective back focal plane on a camera, via a suitable optical system [2]. This imaging technique is able to resolve the angular spectrum of the light over a wide angular field of view, but typically it doesn't provide any spectral information, since it integrates the light intensity over a broad wavelength range. On the other hand, advanced hyperspectral imaging techniques are able to record the spectrum of the transmitted/reflected/emitted light at each pixel of the detector.



Figure 1: Sketch of the k-space hyperspectral microscope employing the common-path interferometer, which enables FT spectroscopy. Varying the delay between the image replicas in the interferometer, each pixel records an interferogram, whose FT yields a spectrum for each element of the angular field of view. The full 3D energy-angular dispersion of a microcavity can be reconstructed from the resulting hypercube.

In this work, we combine an innovative hyperspectral imaging system with Fourier-space microscopy (see Fig.1), and we apply the novel device to the characterization of planar organic microcavities and optical metasurfaces. In our system, hyperspectral imaging is performed by Fourier-transform spectroscopy thanks to an innovative common-path birefringent interferometer [3]: it generates two delayed replicas of the light field, whose interference pattern is recorded as a function of their delay. The Fourier Transform of the resulting interferogram yields the intensity spectrum for each element of the microscope angular field-of-view. This system provides an angle-resolved hyperspectral view of the fluorescence from the microcavities, after excitation by a laser at 365 nm. The hyperspectral k-space image clearly evidences the cavity modes in photoluminescence, from which we reconstruct a 3D view of the parabolic cavity dispersion across the whole Fourier space. Furthermore, we apply our technique for the characterization of a dielectric nanodisks metasurface, evidencing the non-trivial angular and spectral behaviour of its anapole mode in reflection.

#### References

[1] Zhang, Y., et al., Momentum-Space Imaging Spectroscopy for the Study of Nanophotonic Materials. Sci. Bull. 2021, 66 (8), 824–838

[2] Kurvits, J. A. et al., *Comparative Analysis of Imaging Configurations and Objectives for Fourier Microscopy*. JOSA A 2015, 32 (11), 2082–2092.

[3] Candeo, A. et al., *A Hyperspectral Microscope Based on an Ultrastable Common-Path Interferometer*. APL photonics 2019, 4 (12), 120802.

### Advanced electromagnetism using FEniCSx

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Computational physics is mostly based on the Finite Element Method (FEM): it can be used to solve Maxwell's equations, enabling the analysis and design of electromagnetic devices and systems. The FEM can handle a heterogeneous mesh refinement, allowing for higher accuracy in regions of interest, while keeping the overall computational cost contained: this feature is especially advantageous in nanophotonics, where a multi-scale approach is needed.

FEniCSx (<u>https://fenicsproject.org/</u>), an open-source computing platform for solving partial differential equations, is a very powerful tool for the application of the FEM in electromagnetism and nanophotonics. In FEniCSx, while setting-up a variational problem, is straightforward to define a suitable finite element function space, while the computational domain can be generated using a free mesh-generator tool. In addition, FEniCSx is ready for parallel computation. We exploit its high-level Python interface to provide an open-source computational package for solving electromagnetic problems, starting from the mesh building up to the computation of the output relevant physical quantities.

In detail, we use of FEniCSx to solve a series of common problems in electromagnetism, each requiring a specific set of boundary conditions (BCs), such as perfect electric and magnetic conductor BCs, perfectly matched layers, active ports, etc. Some examples are the computation of S-parameters of 3D periodic grating, scattering efficiencies for 3D and axis-symmetric. Moreover, our goals include the development of some examples of multi-physics coupling in which Maxwell's equations are coupled to a hydrodynamic-like equation for the description of advanced plasmonic effects in nanoparticles, in the limit of the Thomas-Fermi approximation and the quantum hydrodynamic theory.



Figure 1: Comparison between some analytical and numerical results for the Thomas-Fermi model, where the longitudinal modes are visible in the bulk region above the plasma frequency

## Microscopic theory for active plasmonics in THz-pumped metal nanoparticles

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The optical response of metal nanoparticles (MNPs) is dominated by the formation of collective electronic resonances, forming localized plasmons. To gain insight into their dynamics and for switching applications, it would be beneficial to influence the plasmonic resonances actively.

Here, we present a theoretical model for an active tuning of the plasmonic resonance in optical spectra by using strong THz fields and analyze a THz pump-optical probe scheme, as illustrated in Fig. 1 (left). The combined THz and optical dynamics of MNP plasmons are described microscopically in the Heisenberg equation of motion framework, and a Boltzmann transport equation is established in the Wigner formalism.

Additionally in a coarse-grained approach, we apply a non-perturbative extension of the hydrodynamic model of the electron gas to achieve a polarization of the MNP. The formalism allows us to merge the electron dynamics and Maxwell equations analytically to obtain self-consistent solutions.



Figure 1: Left: A spherical gold nanoparticle is polarized by a strong THz field in the pump-probe experiment. The influence of the THz field on the optical probe signal is studied. Right: We have evaluated the spectrum of the imaginary part of the polarizability and observe a shift and broadening of the plasmon resonance with increasing strength of the THz pump field up to  $2 \frac{MV}{cm}$ .

We obtain a renormalization of the optical MNP plasmon resonance by the THz field, as depicted in Fig. 1 (right). The magnitude of the renormalization depends strongly on the field strength and frequency of the THz pump field and allows an active tuning of the plasmon resonance. Physically, this process should be thought of as a renormalization of the pressure contribution addressed in the hydrodynamic model.

# Theory of Free-electron third order nonlinearities in heavily doped InGaAs nanoantennas

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Free electrons in plasmonic structures can provide extra degrees of freedom in modulating nonlinearities in the system when the nonlocal effect is considered (eqs. 1-3). The hydrodynamic model reveals that the third harmonic nonlinearity due to the free electrons could even be orders of magnitude higher than that of the conventional bulk  $\chi^{(3)}$  when the free-electron density is low.[1] By employing this theory to heavily doped semiconductors, in the project NEHO, we aim to achieve advanced nonlinear integrated photonic circuits working in the mid-infrared by taking advantages of the high nonlinearity from the plasmonic aspect and low loss from the semiconductor aspect.

While the hydrodynamic theory has been used to successfully reproduce second order processes in noble metals [2,3], third order nonlinearities have remained unexplored experimentally, since in noble metals these are negligible compared to the crystalline lattice nonlinearities (i. e.  $\chi^{(3)}$ ). Here, we use a proof-of-principle n-doped InGaAs antenna design (Fig. 1a) to demonstrate and achieve high hydrodynamic nonlinearity. The experiment shows that the third-harmonic nonlinearity has an evident doping dependence, which is predicted by hydrodynamic model. Numerical simulation reproduces the qualitative behavior observed experimentally. Moreover, from the simulation, we show both linear (Fig. 1b) and nonlinear (Fig. 1c) efficiencies, where the third-harmonic efficiency due to the free electrons is expected to be 3 orders of magnitude higher than that of the conventional bulk  $\chi^{(3)}$  (Fig. 1c).

$$\ddot{\mathbf{P}} + \gamma \dot{\mathbf{P}} = \frac{n_0 e^2}{m} \mathbf{E} + \beta^2 \nabla (\nabla \cdot \mathbf{P}) + \mathbf{S}_{\mathrm{NL}}^{(2)} + \mathbf{S}_{\mathrm{NL}}^{(3)}$$
(1)

$$\mathbf{S}_{\mathrm{NL}}^{(2)} = \frac{e}{m} \mathbf{E} \nabla \cdot \mathbf{P} - \frac{e\mu_0}{m} \dot{\mathbf{P}} \times \mathbf{H} + \frac{1}{en_0} (\dot{\mathbf{P}} \nabla \cdot \dot{\mathbf{P}} + \dot{\mathbf{P}} \cdot \nabla \dot{\mathbf{P}}) + \frac{1}{3} \frac{\beta^2}{en_0} \nabla (\nabla \cdot \mathbf{P})^2 \tag{2}$$

$$\mathbf{S}_{\mathrm{NL}}^{(3)} = -\frac{1}{e^2 n_0^2} (\nabla \cdot \mathbf{P} (\dot{\mathbf{P}} \nabla \cdot \dot{\mathbf{P}} + \dot{\mathbf{P}} \cdot \nabla \dot{\mathbf{P}}) + \dot{\mathbf{P}} \cdot \dot{\mathbf{P}} \nabla \nabla \cdot \mathbf{P}) - \frac{1}{27} \frac{\beta^2}{e^2 n_0^2} \nabla (\nabla \cdot \mathbf{P})^3$$
(3)



Figure 1: (a) Sketch and SEM picture of the plasmonic antennas. Linear (b) and nonlinear (c) efficiencies of the antenna. The comparison shows that the third-harmonic efficiency due to the free electrons is over 3 orders of magnitude larger than that due to the bulk  $\chi^{(3)}$ .

- [1] F. De Luca, M. Ortolani and C. Ciracì, Phys. Rev. B 103, 115305 (2021)
- [2] M. A. Vincenti, D. De Ceglia, A. Ciattoni and M. Scalora. Phys. Rev. A, 84, 063826 (2011)
- [3] C. Ciracì, E. Poutrina, M. Scalora and D. R. Smith, Phys. Rev. B 85, 201403 (2012)

## Crossover from Non-thermal to Thermal Photoluminescence from Metals excited by Ultrashort light pulses

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We provide a complete quantitative theory for the important, decades-old, yet unresolved problem of nonlinear photoluminescence (PL) from metals [1]. Remarkably, the countless previous experimental works shored radically different behaviours, and were not accompanied by a complete theory because of the lack of knowledge of the hard-to-calculate electron non-equilibrium distribution under illumination. This resulted in disagreements on a long series of fundamental aspects associated with this emission.

Based on our recently derived *steady-state* non-equilibrium quantum electron distribution in metals [2], we first compute numerically and analytically the emission, electron and lattice temperatures from metals illuminated by *CW* light [3]. This solution reveals what is to our knowledge, the first ever explanation of the dependence of the metal emission on the electric field, and its dependence on the electron temperature; we show that the emission is a primarily *non-thermal* phenomenon, and identify the unique signatures of the deviation from thermal equilibrium on the emission spectra and electric field dependence.

We then analyze *transient* nonlinear PL following illumination by intense *ultrashort pulses*. Our detailed calculations and heuristic model enable us to elucidate the crossover from the non-thermal emission characteristic of weak (CW-like) illumination to thermal emission occurring after illumination by strong shorter pulses [4]. Specifically, we identify the characteristics of nonthermal and thermal emission in the transient emission spectra and its dependence on the illumination frequency and intensity. Our work puts to rest the many decades-long arguments and motivates improved thermometry protocols based on the anti-Stokes emission [1].

Relying on the extension of our point-emission theory [3,4] to macroscopic bodies using the local Kirchhoff's law [5,6], we study the dependence of the PL from nanospheres and films under continuous and pulsed illumination. Specifically, we show that the non-thermal emission grows linearly with the nanoparticle volume for small sizes, and that once retardation kicks in, the PL growth with size is slower. The crossover from nonthermal emission to thermal emission is also intensity and size-dependent, see Fig. 1(e).



Figure 1: (a) Calculated emission spectra from an Au rod normalized by the photonic density of states. The early stages of the emission (black & blue curves) exhibit a step-like structure characteristic of non-thermal emission while the emission at the later stages is smooth, as appropriate for thermal emission. (b) The exponent extracted from a fit of the emission intensity to a power-law in the incident intensity. The black (blue) curves correspond to non-thermal (thermal) emission. (c) Measured spectra at various illumination intensities and (d) the corresponding power-law exponent extracted from the measured data; an excellent match to the theoretical prediction (black line) is observed. (e) The emission power at wavelength 950 nm for intensities 40 kW/cm<sup>2</sup> (continuous line) and 10 kW/cm<sup>2</sup> (dash-dot line) due to a pulsed illumination of 520 nm. Contributions from thermal emission, one photon absorption and two photon absorption are indicated by black, blue and red, respectively.

#### References

[1] G. Baffou, ACS Nano 15, 5785-5792 (2021).

[2] Y. Dubi, Y. Sivan, Light: Science & Applications 8, 89 (2019). Y. Sivan et al., Faraday Discussion 214, 215-233 (2019).

- [3] Y. Sivan, Y. Dubi, ACS Nano 15, 8724 (2021).
- [4] Y. Sivan, I.W. Un, I. Kalyan, K. Lin, J. Lupton, S. Bange, ACS Nano 17, 11439-11453 (2023).

[5] J.-J. Greffet et al.. Light emission by nonequilibrium bodies: Local Kirchhoff law. Phys. Rev. X, 8:021008, 2018.

[6] A. Pelous-Loirette et al, in preparation.

## Crossover from Non-thermal to Thermal Photoluminescence from Metals excited by Ultrashort light pulses

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#### References

[1] G. Baffou, ACS Nano 15, 5785-5792 (2021).

- [2] Y. Dubi, Y. Sivan, Light: Science & Applications 8, 89 (2019). Y. Sivan et al., Faraday Discussion 214, 215-233 (2019).
- [3] Y. Sivan, Y. Dubi, ACS Nano 15, 8724 (2021).
- [4] Y. Sivan, I.W. Un, I. Kalyan, K. Lin, J. Lupton, S. Bange, ACS Nano 17, 11439–11453 (2023).
- [5] J.-J. Greffet et al.. Light emission by nonequilibrium bodies: Local Kirchhoff law. Phys. Rev. X, 8:021008, 2018.
- [6] A. Pelous-Loirette et al, in preparation.

## Thermoplasmonic optical fiber probe: An experimental and computational analysis of the heating characteristics for neuroscience applications

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Thermoplasmonics is a nanoscale optical heating phenomenon where plasmonic nanoparticles are used as nano heat generators. Plasmonic heat generation is the basis of the many applications in biomedicine, particularly in cancer therapies and drug delivery [1]. Recently, optical fiber probes with thermoplasmonic effects have been demonstrated for the modulation of neural activity [2] and lab-on-fiber applications [3]. Precise and accurate numerical investigations play a vital role in predicting and understanding the thermal effects of neural implants. Here an experimental and computational analysis of the heating characteristics of a thermoplasmonic optical fiber probe is presented.

The thermoplasmonic optical fiber probe consists of a tapered optical fiber decorated with densely packed bimetallic nanoislands (Ti and Au with 5 nm initial film thickness respectively) covering the entire taper surface. The nanoislands are fabricated by a non-planar solid-state dewetting process [4] which allows the nucleation of nanoislands with an average diameter around 60 nm, and plasmonic nanogaps around 40 nm. A multiphysics computational model of the optical fiber probe has been developed employing the finite-element (FEM) method. The optical power absorbed by the nanoislands under laser excitation is first calculated by the electromagnetic simulation. Then the heat module is used to compute the resulting spatial and temporal temperature profile around the optical fiber probe surface in various background environments such as air, water, and brain tissue. A thermographic camera was used to measure the temperature of the probe in the air, and the results are compared with numerical simulations. Based on simulations and experimental results in this work, we foresee that a thermoplasmonic optical fiber probe can be used for localized heating *in vivo* as an additional tool for neuroscience research.



Figure 1: (a) A schematic of the thermoplasmonic optical fiber probe (b) An SEM of the fabricated device (c) Simulated absorption cross-section of the probe (d) Measured and simulated steady state temperature change at the fiber surface as a function of the input laser power (e) Measured and simulated transient response of the temperature change at the fiber surface.

- [1] G. Baffou, F. Cichos, and R. Quidant, Nature Materials, 19-9, pp. 946–958, (2020)
- [2] H. Kang, W. Hong, H. J. Kwon, and Y. Nam, ACS Nano, 14-9, pp. 11406–11419, (2020)
- [3] S. Principe et al., Opt Laser Technol, vol. 132, (2020)
- [4] D. Zheng et al., Advanced Materials, p. 2200902, (2022)

#### LNOI Reconfigurable Optical Phased Arrays for On-chip Wireless Switches

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In this work, we investigate reconfigurable optical phased arrays (OPA) proposed for the realization of optical wireless switches, which can serve as basic building blocks for on-chip wireless interconnections [1]. We present the design and radiation characteristics of lithium niobate (LN) taper antenna element comprising the OPA by considering the LNOI technology. LN is considered as a highly suitable material for integrated photonic circuits as it exhibits interesting properties including high optical nonlinearities, strong electro-optic coefficient and wide optical transparency window [2]. These characteristics of LN make it significant for the implementation of OPA.

The antenna element used as a radiator in the OPA is shown in Fig. 1(a), which is composed of a LN straight waveguide of length L and with top width  $w_t$ , an inversely tapered waveguide of length TL, and a straight waveguide of length L and top width  $w_t$  forming the taper tip. The LN waveguide with height *h* and sidewall angle  $\gamma$  is immersed in silica and the operating wavelength is 1.55 µm. In our analysis, we consider an x-cut LN thin film with its optical axis along the y-direction.



Figure 1: (a) LN taper antenna element with trapezoidal cross-section. (b) Effective refractive index  $n_{\text{eff}}$  for different values of waveguide height *h* and sidewall angle  $\gamma$ . (c) Radiation diagram of OPA made of three taper antennas. The electric field |E| maps (d-f) in the xy-plane showing the beam steering to address a particular receiver when a suitable phase difference  $\alpha$  is applied between the radiators of OPA.

Figure 1(b) shows the effective refractive index  $n_{\text{eff}}$  for different values of waveguide height *h* and sidewall angle  $\gamma$ . The radiation diagram as a function of azimuthal angle  $\phi$  for the OPA made up of N<sub>a</sub>=3 identical taper antennas is shown in Fig. 1(c). The beam steering can be achieved by applying a suitable phase difference  $\alpha$ =360/N<sub>a</sub> between the antenna elements of the OPA. Figures 1(d-f) show the electric field patterns in the xy-plane obtained by 3D FDTD simulations, demonstrating that depending upon the applied phase difference  $\alpha$  (+120°, 0°, -120°), the transmitter  $T_x$  can address a particular receiver ( $R_x^{+1}$ ,  $R_x^{0}$ ,  $R_x^{-1}$ ) and thus can serve as a 1x3 optical wireless switch for on-chip communication [3].

The proposed reconfigurable OPAs are capable of communicating with different receivers and can be assembled in 1xN and NxN switching matrix configurations. These optical wireless switches can be a promising alternative to microring resonator based networks.

#### References

[2] J. Lin, F. Bo, Y. Cheng and J. Xu, Photonics Research., 8, 1910-1936 (2020).

<sup>[1]</sup> J. He, T. Dong and Y. Xu, IEEE Access, 8, 188284-188298 (2020).

<sup>[3]</sup> G. Calò, et al., Optics Express, 29, 31212-31228 (2021).

## Polycrystalline MoO<sub>3</sub> films fabricated by pulsed laser deposition for infrared multilayer photonics.

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The development of a photonic platform in the mid-infrared range is motivated by applications, such as detection of traces of biomolecules and harmful/explosive substances, passive radiative cooling as well as devices for thermal imaging and for medical diagnostics. Recent promising solutions are based on the exploitation of polar materials [1] including ultra-thin van der Waals (vdW) materials [2-3]. In this framework, Molybdenum trioxide ( $\alpha$ -MoO3) is attracting great attention [4] as it supports surface phonon polaritons (SPhPs) in three different wavelength bands for the three orthogonal directions (range 10-20 µm) being a biaxial natural hyperbolic material. However, the development of a new highly versatile and compact MoO<sub>3</sub> based platform for IR photonics is hampered by the lack of availability of good quality scalable films and/or multilayer stacks. According to literature, MoO3 for IR photonics and polaritonics is mostly used in the form of crystalline flakes, coming from bulk MoO3 crystals through a complex mechanical exfoliation process followed by a transfer to an appropriate substrate.

Here we show that pulsed laser deposition (PLD) can be employed to obtain  $\alpha$ -MoO3 films on SiO2 substrates at lower temperatures (approx. 500 °C), without using harmful precursor gases normally employed by atomic layer deposition (ALD) and without the need for any post-growth annealing. Although deposition parameter optimization is still required for the definition of a process to synthesize MoO<sub>3</sub> films with a high degree of crystallinity, our experimental findings show remarkable spectral features of the obtained polycrystalline films. All the samples have been analyzed by X-ray diffraction (XRD) and atomic force microscopy (AFM) to confirm the orthorhombic  $\alpha$ -phase of MoO<sub>3</sub> films, the polycrystalline nature, and assess film morphology. Moreover, we reported optical characterization in the IR, carried out by Fourier-transform infrared spectroscopy (FT-IR) in reflection mode with an incidence angle of 45° and a rotating linear polarizer. Reflectance spectra exhibit interesting features, i.e. a polarization-independent perfect absorption behavior at 962 cm<sup>-1</sup> (well preserved for a broad angular incidence range), and an enhanced tunability vs. light polarization angle of a narrow band reflection peak at 1006 cm<sup>-1</sup> with a Q factor of 55. More relevantly, the use of PLD technology enables the fabrication of PLD-grown MoO3 with the phase-changing properties of VO2 can lead to narrow-band, tailorable, polarization-sensitive resonances for IR sensing and photonics.

- [1] Y.Chen et al., ACS Photonics 1, 718724 (2014).
- [2] M. Tamagnone et al., Science Advances 4, eaat7189 (2018).
- [3] D. N. Basov et al., Science 354, 6309 (2016).
- [4] G.Álvarez-Pérez et al., Adv.Materials 32, 1908176 (2020).
- [5] S. Abedini Dereshgi et al, ACS Appl. Mater. Interfaces, 13, 41, 48981-48987 (2021)

## Design of a room-temperature topological exciton-polariton laser in a ZnO/TiO<sub>2</sub> photonic crystal slab

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In this article [1], we propose theoretically a scheme to get a room-temperature two-dimensional topological exciton-polariton laser with propagating topological lasing modes. The structure uses guided modes in a photonic crystal slab. A ZnO layer provides strong excitonic resonances stable at room temperature. It is capped by a  $TiO_2$  layer pierced by a triangular lattice of air holes. The exciton-polariton modes of the three-dimensional structure are computed by solving numerically Maxwell's equations including the excitonic response. The designed triangular lattice of circular air holes shows a transverse electric band gap. The triangular lattice of air holes is shown to be well described by a staggered honeycomb tight-binding lattice, associated with valley Chern numbers defining the interface states [2-3]. The interface between two shifted triangular lattices of air holes supports two counterpropagating modes lying in the gap of the bulk modes, analogous to quantum pseudospin Hall interface states. These modes show orthogonal polarizations. They can be selectively excited using polarized excitation and are well protected from backscattering. These modes can benefit from the exciton-polariton gain at room temperature because of their sufficiently large exciton fraction and favorable position in energy. The strong localization of these propagating modes makes them suitable to host topological lasing triggered by a nonresonant pump localized on the interface.



Figure 1: (a) Sketch of the 3D PCS structure hosting topological interface states. (b) Polaritonic band structure calculated for a finite ribbon in the y direction and infinite structure in the x direction, including the interface. Note the interface states that can propagate in the gap formed by the bulk modes. An excitation with circular right (left) polarization leads to propagation in the right (left) direction (red and blue lines [(e), (f)] Profiles of the interface states calculated by FDTD under (e) a circular right-polarized excitation and (f) a circular left-polarized excitation located below the interface. Note that the propagation occurs mainly in the right or left direction (dashed white line).

#### References

[1] Ismael Septembre et al. Design of a room-temperature topological exciton-polariton laser in a ZnO/TiO2 photonic crystal slab. Phys. Rev. B 107, 155304 (2023)

<sup>[2]</sup> Yun Zhou et al. On-Chip Unidirectional Waveguiding for Surface Acoustic Waves along a Defect Line in a Triangular Lattice. Phys. Rev. Applied 19, 024053 (2023)

<sup>[3]</sup> Robert J. Davis et al. Topologically protected edge states in triangular lattices. Phys. Rev. B 106, 165403 (2022).

#### Split Bowtie nanoantennas for electron acceleration

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Electron accelerators based on integrated optical and plasmonic structures represent an emerging field of research with the potential to revolutionize those applications that require hot electrons, such as catalysis, generation of x-ray, cancer therapies, and so on. In such structures, the Lorentz force acting on charged particles can be concentrated by exploiting nanoantennas and resonators, enabling electron kinetic energy enhancement. Plasmonic nanostructures stand out as powerful energy concentrators, capable of tightly confining electromagnetic energy down to subwavelength volumes, enabling the design of compact metasurfaces [1], integrated nanotweezers [2], isolators [3], sensors etc. Here we propose a novel plasmonic nanoantenna geometry, the split bowtie (SB), which is composed of a gold twofold bowtie antenna placed on top of a SOI waveguide. As shown in Fig. 1(a,b), the SB has an additional slit that can be used as a channel for an electron beam interacting with its localized nearfield. The coupling between the waveguide and the plasmonic resonator in this new geometry enables the excitation of a highly concentrated nearfield with components that promote the acceleration of an electron beam passing through the nanoantenna channel (see Fig. 1(d)). The neat energy gain of a single electron interacting with the structure excited at 1408 nm is shown in Fig. 1(c). An SB array can be leveraged to increase the acceleration gradient while maintaining integrability and compact size.



Figure 1: (a,b) Sketch of a SB resonator integrated on top of a SOI waveguide ( $h_{wg}=220 \text{ nm}$ ) depicting the different geometrical parameters. (c) Velocity and energy of an electron passing through the SB slit along the y direction as a function of the time when W = 350 nm, H = 240 nm,  $h_{Au}=80 \text{ nm } g_x=g_y=40 \text{ nm}, g_{x2}=100 \text{ nm},$  and when  $P_{in}=1 W$  and  $v_{y0}=2.488 \times 10^4 \text{ km/s}$ . (d) Electron trajectory through the slit of the SB nanoantenna.

- B. Dagens, M. Février, P. Gogol, S. Blaize, A. Apuzzo, G. Magno, R. Mégy, G. Lerondel, "Direct observation of optical field phase carving in the vicinity of plasmonic metasurfaces," Nano Lett., 16, 4014–4018 (2016).
- [2] C. Pin, G. Magno, A. Ecarnot, E. Picard, E. Hadji, V. Yam, F. de Fornel, B. Dagens, B. Cluzel, "Seven at One Blow: Particle Cluster Stability in a Single Plasmonic Trap on a Silicon Waveguide," ACS Photonics, 7, 1942–1949 (2020).
- [3] S. Abadian, G. Magno, V. Yam, B. Dagens, "Broad-band plasmonic isolator compatible with low-gyrotropy magneto-optical material," Opt. Express, 29, 4091 (2021).

## Computational study of interaction between Channelrhodopsin and a gold nanocluster

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Channelrhodopsin represents a biological system of extreme interest for optogenetics and nanomedicine being it able to control electrical excitability, the acidity and other cellular properties by means of appropriate light stimuli [1].

The work presents a study performed in the framework of classical molecular dynamics [2], of an atomistic system given by a functionalized  $Au_{25}$  gold cluster [3] and a *Channelrhodopsin* protein inserted in a cellular membrane.

The analysis consists in the simulation of 26 diffusion trajectories aimed at verifying whether the nanoparticle interacts and/or links with the membrane protein, identifying any anchoring sites and quantifying the structural modifications induced on the protein. The cluster was initially put on 9 different points on two planes located above and below the membrane (about 3 nm far from the protein). What was found is that in 40% of cases, the gold hydrophobic cluster diffuses towards the protein reaching it after less than 50 ns (an example is given in Fig. 1a). These cases together with other more favorite cases (the cluster was almost attached to the protein to promote interaction with it) were, then, analyzed in detail: preferential interaction sites between the nanoparticle and the protein were identified, as well as the main interaction mechanisms. In addition, the effect of the interaction with the nanoparticle on the protein structure (Fig. 1b) and dynamics was analyzed, also investigating possible effects on the retinal binding cavity and on the protein interior hydration. The basic idea is to find out configurations to be characterized, then, with the quantum mechanical approaches: we are confident that the use of metallic clusters or nanoparticles could affect the photochemistry of the protein and possibly represent a further tool for the control of this photoreceptor.



Fig 1: (a) ChR interacting with the Au NP; (b) RMSD of C- $\alpha$  atoms for the monomer M1 and M2 with and without NP, for NP interacting with M1 (i,ii) and M2 (iii,iv).

1. K. Deisseroth and P. Hegemann, "The form and function of channelrhodopsin", Science 15, 357-6356 (2017). 2. H.J.C. Berendsen, D. van der Spoel, R. van Drunen, "GROMACS: A message-passing parallel molecular dynamics implementation", Comp. Phys. Comm. 91, 43 (1995).

3. G. Brancolini, D. Toroz and S. Corni, "Can small hydrophobic gold nanoparticles inhibit β<sub>2</sub>-microglobulin fibrillation?" Nanoscale 6., 7903 (2014).

## Effect of electron spill-out on the surface plasmon-polariton propagation at dielectricmagnetized plasma interface

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Surface plasmon-polaritons (SPPs) propagating at dielectric-metal interfaces, due to the coupling of light with the collective oscillation of free electrons, satisfy the Lorentz reciprocity principle which is exhibited by the symmetrical propagation of the SPPs in opposite directions. This reciprocity can be broken when external magnetic field is applied to the system and may result in the creation of unidirectional SPP propagation within a certain frequency gap [1]. This phenomenon has attracted a substantial interest in the community since it carried a significant application prospect in the fields of sensing and communication. Recent works, have however, shown that this unidirectionality does not hold if one accounts for nonlocal effects in the optical response of the plasmonic system [2,3]. Contrarily to the local response approximation (LRA) model, nonlocality prevents the existence of unidirectional band gaps, since nonlocal dispersion would in principle always provide a back propagating mode. This underlines the importance of consideration of more realistic material models in the theoretical study of SPPs. Here we investigate SPP propagation in the presence of externally applied magnetic field, within the framework of quantum hydrodynamic theory (QHT), which accounts for both nonlocality (beyond the Thomas-Fermi approximation) and electron spill-out. Contrarily to Thomas-Fermi nonlocal dispersion predictions, the QHT appears to restore the existence of the unidirectional gaps. The results given by the QHT are compared with previous results, Fig. 1(**a**).



Figure 1: (a) Dispersion diagrams calculated for the SPP propagation at the air-magnetized Na interface based on the local response approximation, LRA (black curve), the Thomas-Fermi hydrodynamic theory, TFHT, nonlocal model (red curve), and the Quantum hydrodynamic theory, QHT, model (light-blue curve). (b) Real part of the magnetic field distribution at  $\omega = 0.77\omega_p$  [green dashed line in (a)] calculated with QHT framework. (c) The one-dimensional profile of the field extracted from (b) at 0.5 nm below the interface. Indicates field interference pattern on the right of the dipole source due to the contributions from the two crossing points [green dashed line in (a)] in the positive k of the QHT dispersion curve.

- [1] K. L. Tsakmakidis, L. Shen, S. A., Schulz, X. Zheng, J. Upham, A. F. Vakakis and R. W. Boyd, Science 356, 1260-1264(2017).
- [2] S. Buddhiraju, Y. Shi, A. Song, C. Wojcik, M. Minkov, Ian A. D. Williamson, A. Dutt and S. Fan, *Nat. commun.* 11(1), 674 (2020).
   [3] S. Ali Hassani Gangaraj and F. Monticone, *Optica* 6(9), 1158-1165 (2019).

# Exciton-Plasmon Hybridization at interfaces of metal nanoparticles and 2D semiconductors

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In recent years, monolayers of transition metal dichalcogenides (TMDCs) have attracted considerable attention due to their strong Coulomb and light-matter interactions, leading to tightly bound excitons with large optical oscillator strength. Due to the finite thickness of the monolayers, these excitons are very sensitive to the environment, which allows their properties to be tailored, e.g., by functionalization with molecules or metal nanoparticles (MNPs) [1, 2].



Figure 1: (left) Macroscopic TMDC polarization in real space shows localized excitons within the TMDC layer near the metal nanoparticle. (right) Avoided-crossing behavior can be observed in electric near-field, indicating the system to operate in the strong coupling regime.

We present a self-consistent Maxwell-Bloch theory to analytically study the interaction between a nanostructure consisting of a metal nanoparticle and a monolayer of transition metal dichalcogenide. For the combined system, we identify an effective eigenvalue equation that governs the center-of-mass motion of the dressed excitons in a plasmon-induced potential. Examination of the dynamical equation of the exciton-plasmon hybrid reveals the existence of bound states with negative eigenenergies, which we interpret as excitons localized in the plasmon-induced potential. The appearance of these bound states in the potential indicates strong coupling between excitons and plasmons. We quantify this coupling regime by computing the scattered light in the near-field explicitly and identify signatures of strong exciton-plasmon coupling with an avoided crossing behavior and an effective Rabi splitting of tens of meV [3].

- Carlson, C., Salzwedel, R., Selig, M., Knorr, A., and Hughes, S. (2021). Strong coupling regime and hybrid quasinormal modes from a single plasmonic resonator coupled to a transition metal dichalcogenide monolayer. Phys. Rev. B, 104, 125424.
- [2] Denning, E. V., Wubs, M., Stenger, N., Mørk, J., and Kristensen, P. T. (2022). Quantum theory of two-dimensional materials coupled to electromagnetic resonators. Phys. Rev. B, 105, 085306.
- [3] Salzwedel, R., Greten, L., Schmidt, S., Hughes, S., Knorr, A., and Selig, M. (2023). Spatial Exciton Localization at Interfaces of Metal Nanoparticles and Atomically Thin Semiconductors. arXiv preprint arXiv:2305.11099.

## Manipulating Light-Matter Interactions by Strain Modulation in Two Dimensional Semiconductors

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In the last years, two dimensional (2D) materials emerged as extremely interesting platforms for optoelectronic and quantum devices. Among all the layered van der Waals materials, transition metal dichalcogenides (TMDs) constitute one of the most promising systems. Interestingly, their optical properties are dominated by excitons, providing very large oscillator strength. Due to large quantum confinement and reduced dielectric screening, excitons in TMDs have very large binding energy, and are thus visible at room temperature. As a result, TMDs have been exploited in several nanophotonic architectures for studying light-matter interactions, including microcavities, individual plasmonic and dielectric nanoparticles, Bloch surface waves and plasmonic and dielectric lattices[1-3].

TMDs have recently shown interesting properties enabling active manipulation of carriers and excitons, with applications in solid state physics and nano-optoelectronic devices, including high sensitivity to the dielectric environment and to external electromagnetic fields, valley polarization, in-plane polarized dipole and extreme sensitivity to local strain fields.

Here we show our latest results on different approaches for manipulating the optical properties of TMDs via strain modulation. In particular, we show the emergence of quantum and high-energy anomalous emission by nano-structuring localized strain fields at the monolayer level by laser micromachining[4], as well as the possibility of manipulating the monolayer morphology by using surface acoustic waves in a piezoelectric cavity configuration[5].



#### References

[1] Maggiolini, E., Polimeno, L., Todisco, F. et al. Strongly enhanced light-matter coupling of monolayer WS2 from a bound state in the continuum. Nat. Mater. (2023).

[2] Mathias Geisler, Ximin Cui, Jianfang Wang, Tomas Rindzevicius, Lene Gammelgaard, Bjarke S. Jessen, P. A. D. Gonçalves, Francesco Todisco, Peter Bøggild, Anja Boisen, Martijn Wubs, N. Asger Mortensen, Sanshui Xiao, and Nicolas Stenger, ACS Photonics 2019 6 (4), 994-1001.

[3] Barachati, F., Fieramosca, A., Hafezian, S. et al. Interacting polariton fluids in a monolayer of tungsten disulfide. Nature Nanotech 13, 906–909 (2018).

[4] Zywietz, U., Evlyukhin, A., Reinhardt, C. et al. Laser printing of silicon nanoparticles with resonant optical electric and magnetic responses. Nat Commun 5, 3402 (2014).

[5] D. Scolfaro, M. Finamor, L. O. Trinchao, et al. ACS Nano 2021, 15, 15371-15380.

## Metallic Nanoislands-Decorated Tapered Optical Fibers for Remote SERS Sensing and Heat Generation

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Tapered optical fibers (TF) have been widely used as a versatile tool in the optogenetic field to deliver and collect light from the brain with minimal invasiveness<sup>[1]</sup>. Integrating TFs with metallic nanoparticles can create a new generation of multifunctional neural probes which can bring highly sensitive surface-enhanced spectroscopy, such as surface-enhanced Raman scattering (SERS)<sup>[2]</sup>, or surface-enhanced fluorescence (SERF) in deep brain regions. Meanwhile, the highly absorbing nature of metallic nanoparticles can provide efficient local heat generation, which is highly desirable for applications such as photo-thermal cancer therapy<sup>[3]</sup> and neural activity stimulation<sup>[4]</sup>.

Here, a universal and high-throughput non-planar solid-state dewetting approach has been developed to fabricate metallic nanoislands (NIs) with tunable distribution patterns around the whole taper surface. The fabrication approach mainly includes two steps, thin metal film deposition and high-temperature dewetting. To maximize the SERS performance of the probe, densely packed pure Au NIs patterns with an average diameter of 50 nm, and effective gaps <10 nm can be obtained by repeating twice the 5 nm Au film evaporation and 600 °C dewetting procedure. The rapid remote SERS sensing experiments show the NIs-TFs probes have a limit of detection (LOD) of  $10^{-7}$  M for aqueous solutions of rhodamine 6G (R6G), and  $10^{-5}$  M for serotonin and dopamine in the near-infrared region. The obtained LOD for neurotransmitters surpasses the current report for SERS-active TF detecting dopamine employing the same configuration (excitation and collection field guided in the same waveguide) by two orders of magnitude<sup>[5]</sup>, and is compatible with the upper bound level of physiological concentrations for neurotransmitters, measured to be in the tens of  $\mu M$ range for both dopamine and serotonin<sup>[6,7]</sup>. To improve the stability of the device, bi-metallic (Ti and Au) alloy NIs with an average diameter of 62 nm, and effective gap of 40 nm can be obtained by evaporating 5 nm Ti film and 5 nm gold film, then go through the 600 °C dewetting procedure only one time. The fabricated devices show improved stability with a SERS LOD of 10<sup>-6</sup> M for R6G, and a stable temperature increase slope of 2.5 °C/mW in air with 473 nm laser injection. Based on those results, we believe that the metallic NIs-TFs fabricated by the non-planar dewetting approach show promises as a multifunctional neural probe to explore neural activity in-vitro or in-vivo.



Figure1 (a) Schematic illustration for NIs-TFs fabrication. (b) SEM inspections on the NI-TF. (c) The configuration of LOD experiment for R6G, serotonin and dopamine. (d-f) Spectra response for NIs-TF detecting R6G, serotonin and silica background subtracted dopamine. Figures modified from ref.2.

- [1] F. Pisanello, G. Mandelbaum, M. Pisanello, et al, *Nature Neuroscience* **2017**, *20*, 1180.
- [2] D. Zheng, F. Pisano, L. Collard, et al, *Advanced Materials* **2023**, *35*, 2200902.
- [3] X. Huang, P. K. Jain, I. H. El-Sayed, M. A. El-Sayed, Nanomedicine 2007, 2, 681.
- [4] H. Kang, W. Hong, Y. An, S. Yoo, H.-J. Kwon, Y. Nam, ACS Nano 2020, 14, 11406.
- [5] M. Volkan, D. L. Stokes, T. Vo-Dinh, *Appl Spectrosc* 2000, *54*, 1842.
- [6] T. Patriarchi, J. R. Cho, K. Merten, et al, Science 2018, 360, eaat4422.
- [7] J. Wan, W. Peng, X. Li, et al, *Nat Neurosci* **2021**, *24*, 746.

#### Spontaneous Parametric Down-Conversion Beaming from a LiNbO<sub>3</sub> Nanoresonator

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Lithium niobate (LiNbO<sub>3</sub>) is a prime material for photonics thanks to its unique mix of properties, including a strong electro-optical response, a large second-order nonlinearity, and a broad transparency window (0.35 to 4.5  $\mu$ m). Albeit its hardness and resistance to chemical etching make for a challenging nanofabrication, recent advances enabled the realization of deeply sub- $\mu$ m features in LiNbO<sub>3</sub>, underpinning a series of breakthroughs in the field of nano and meta-photonics [1]. Specifically, several LiNbO<sub>3</sub> metasurfaces have been designed to enhance and steer second-harmonic emission [1] up to the visible range [2]. Similar designs were also exploited for generating photon pairs via spontaneous parametric down-conversion (SPDC) [3], whereby a single photon is converted into two photons of the same total energy (Fig. 1c, right).



Figure 1 a) Spatial distribution of the electric field norm for the targeted eigenmode (wavelength  $\lambda$ =1450 nm, quality factor Q = 14) simulated with the finite-element method. b) Same as panel a for a spectrally adjacent non-radiative eigenmode ( $\lambda$ =1430 nm, Q = 40). c) Left: Scanning electron micrograph of the resonator. Right: Energy-level diagram of degenerate SPDC pumped at frequency  $\omega$ . d) Photon correlation measurement on a single resonator (excitation 5 mW power at  $\lambda$ =785 nm, 10 minutes exposure time). A peak of "true" coincidence counts,  $N_{coi}$ , emerges at zero delay against the background of accidental coincidences,  $N_{acc}$ .

We report here the numerical design, fabrication, and optical characterization of a nanostructured LiNbO<sub>3</sub> photon-pair source in the telecom range. The relaxation of the phase-matching constraint at the nanoscale allows us ample freedom for engineering the resonant behavior, Specifically, we aimed at: (i) enhance the inherently low SPDC yield in such a small interaction volume; and (ii) shape the two-photon emission pattern for efficient light harvesting in the far-field. We optimized the geometry starting from the systematic numerical study of the eigenmodes of a LiNbO<sub>3</sub> cylinder (height 500 nm). Inspired by annular Bragg resonators, we found that the directionality of the selected mode (Fig. 1a) is improved by the insertion of an annular groove, yielding a tightly beamed emission (numerical aperture NA~0.2) perpendicular to the substrate. Additional lateral cuts are introduced to suppress competing non-radiative modes (Fig. 1b). The resonators are directly milled into a 500 nm-thick commercial LiNbO3 film on a silica substrate by a Ga<sup>+</sup> focused ion beam, see Fig. 1c (left). An exemplary photon

correlation histogram in Fig. 1d displays a clear coincidence peak corresponding to the detection of photon pairs, with rates exceeding 300 Hz/W. For context, this preliminary characterization can be compared to the only other report—to the best of our knowledge—of SPDC from LiNbO<sub>3</sub> particles, namely 4  $\mu$ m-sized colloidal microcubes [4], where 15 Hz/W were achieved despite a much larger volume.

In perspective, designs like the one we propose can find their application as miniaturized room-temperature sources of non-classical light in integrated devices for quantum communication and computing.

- [1] A. Fedotova et al., ACS Photonics 9, 3745-3763 (2022).
- [2] L. Carletti et al., ACS Photonics 8, 731-737 (2021).
- [3] T. Santiago-Cruz et al., Nano Lett. 21, 4423-4429 (2021).

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<sup>[4]</sup> N. M. H. Duong et al., Opt. Mater. Express 12, 3696-3704 (2022).

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