NanoPlasm 2022

4th International Conference

13 - 17 June 2022
Grand Hotel San Michele - Cetraro (CS), Italy

Frontiers in Plasmonics and Nano-Photonics

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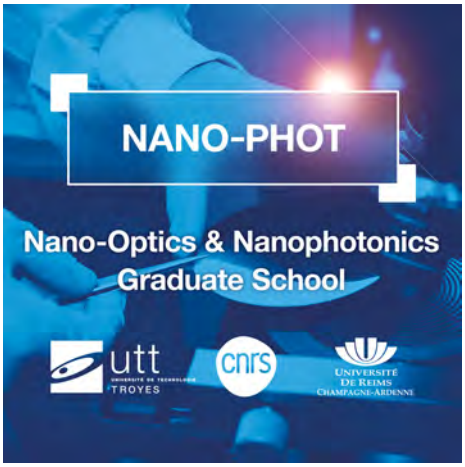

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Detailed Program

	SUN - JUNE 12	MON - JUNE 13		TUE - JUNE 14		WED - JUNE 15		THU - JUNE 16	FRI - JUNE 17	
08:30		**OPENING CEREMONY**								
		Session Chair G. Strangi		Session Chair G. Cerullo		Session Chair R. Quidant		Session Chair F. De Angelis		**LEGEND**
8:45-9:25		**F. Capasso**		**E. Hu**		**N. Engheta**		**H. Altug**		**6 Plenary Talks** 40'(35+5)
9.25-9:50		**M. Wegener**		**R. Advincula**		**G. Cerullo**		**G. Baffou**		
9:50-10:10		**F. Pineider**		**J. Huang**		**G. Della Valle**	9:50-10:15	**O. Marago'**		
10:10-10:40		*coffee break*		*coffee break*		*coffee break*		*coffee break*		**4 Keynote Talks** 35'(30+5)
		Session Chair J. Khurgin		Session Chair A. Boltasseva		Session Chair R. Grange		Session Chair O. Marago'		
10:40-11:15		**V. Shalaev**		**R. Grange**		**B. Palpant**		**J. Garcia De Abajo**		
11:15-11:40		**F. de Angelis**		**M. Celebrano**		**F. Capolino**		**V. Giannini**		**16 Invited Talks** 25'(20+5)
11:40-12:00		**C. Ciracì**		**B. Zappone**		**S. De Liberato**		**S. Bernatova**		
12:00-12:20	**ARRIVALS**	**R. Hasan**		**A. Magazzu**						**12 Oral Talks** 20'(15+5)
12:30-14:30		*lunch*	12:30-14:30	*lunch*	12:30-14:30	*lunch*	12:30-14:30	*lunch*		
		Session Chair A. De Luca		Session Chair M. Wegener				Session Chair F. Capolino	**DEPARTURES**	
16:00-16:40		**A. Boltasseva**	16:00-16:40	**M. Soljacic**			16:00-16:35	**N. Zheludev**		
16:40-17:00		**A. Foti**	16:40-17:05	**R. Quidant**			16:35-17:00	**H. Caglayan**		
17:05-17:30		*coffee break*	17:05-19:30	*Elevator Pitch SESSION*			17:00-17:30	*coffee break + POSTERS*		
17:30-17:55		**J. Khurgin**		*coffee break + posters*		***Social Event*** with dinner	17:30-17:55	**C. De Angelis**		
17:55-18:15		**M. G. Donato**					17:55-18:20	**G. Tagliabue**		
18:15-18:40		**R. Bachelot**								
18:40-19:00	**WELCOME RECEPTION**						18.30-19:30	Closing Remarks and Awards		
20:15-21:30	**Dinner**	**Dinner**	20:15-21:30	**Dinner**			20:15-21:30	**Gala Dinner**		

4th International Conference

NanoPlasm 2022

Frontiers in Plasmonics and Nano-Photonics

13-17 June, 2022- Grand Hotel San Michele (Italy)

Detailed Program

For this 4th edition, NanoPlasm2022 has chosen to be green and eco-sustainable. Please consider in this same spirit to download it only via the QR code without printing it

NanoPlasm2022 Abstract Book

MONDAY, June 13th 2022

08:30	*OPENING CEREMONY*	
Chair: G. Strangi		
8:45-9:25	F. Capasso	***Plenary:*** *Structured Light and Dark by Metaoptics*
9:25-9:50	M. Wegener	***Invited:*** *The rise of two-step absorption 3D laser nanoprinting*
9:50-10:10	F. Pineider	***Oral:*** *Detection of local magnetic fields with plasmonic nanoantennas*
10:10-10:40	*Coffee Break*	
Chair: J. Khurgin		
10:40-11:15	V. Shalaev	***Keynote:*** *Hybrid Quantum Photonics*
11:15-11:40	F. de Angelis	***Invited:*** *Plasmonic Solid-state Nanopores: Toward Single-molecule Protein Identification*
11:40-12:00	C. Ciracì	***Oral:*** *Quantum Hydrodynamic Theory for Plasmonics: from Molecule-Coupling to Nonlinear Optics*
12:00-12:20	R. Hasan	***Oral:*** *Trapping of biological nanoparticles by quasi-bound state in the continuum*
12:30-14:30	*Lunch*	
Chair: A. De Luca		
16:00-16:40	A. Boltasseva	***Plenary:*** *Advancing Photonics with Machine Learning*
16:40-17:05	A. Foti	***Oral:*** *Controlled Aggregation of Gold Nanorods on Graphene by Radiation Pressure for SERS Detection of Biomolecules*
17:05-17:30	*Coffee Break*	
17:30-17:55	J. Khurgin	***Invited:*** *Epsilon (and mu) Near Zero Materials – Photonics on Steroids?*
17:55-18:15	M. G. Donato	***Oral:*** *Light-induced forces in front of epsilon-near-zero metamaterials.*
18:15-18:40	R. Bachelot	***Invited:*** *hybrid plasmonic nano-sources of light: on the importance to control the spatial distribution of the active medium*
20:15-21:30	*Dinner*	

TUESDAY, June14th 2022		
	Chair: G. Cerullo	
8:45-9:25	E. Hu	***Plenary:*** *Nano-Photonic Emitters for the "Quantum Age": where imperfections lead to opportunities*
9:25-9:50	R. Advincula	***Invited:*** *Plasmonics and Molecular Imprinted Polymer (MIP) Sensing*
9:50-10:10	J. Huang	***Oral:*** *Plasmonic Nanogap-enhanced Single-molecule Raman Spectroscopy: Towards Single-protein Raman Sequencing*
10:10-10:40	*Coffee Break*	
	Chair: A. Boltasseva	
10:40-11:15	R. Grange	***Keynote:*** *Bottom-Up Assembly of Random Polycrystals for Nonlinear, Electro-Optic, and Quantum Devices*
11:15-11:40	M. Celebrano	***Invited:*** *Controlling and steering the nonlinear emission in nanoantennas and metasurfaces*
11:40-12:00	B. Zappone	***Oral:*** *Understanding and controlling mode hybridization in multi-cavity optical resonators using quantum theory and the surface forces apparatus*
12:00-12:20	A. Magazzu	***Oral:*** *Investigation of individual dust particle grains by optical tweezers for space applications*
12:30-14:30	*Lunch*	
	Chair: M. Wegener	
16:00-16:40	M. Soljacic	***Plenary:*** *Nanophotonics for tailoring of electron-light interactions*
16:40-17:05	R. Quidant	***Invited:*** *Dielectric meta-optics for reconfigurable planar optics and biosensing*
17:05-19:30	*Elevator Pitch SESSION* *Coffee Break* + *Posters*	
20:15-21:30	*Dinner*	

WEDNESDAY, June 15th 2022

	Chair: R. Quidant	
8:45-9:25	N. Engheta	***Plenary:*** *Extreme Metamaterials in Four Dimensions*
9:25-9:50	G. Cerullo	***Invited:*** *Nonlinear optical response of two-dimensional materials*
9:50-10:10	G. Della Valle	***Oral:*** *Ultrafast All-optical Light Management with Plasmonic Metasurfaces*
10:10-10:40	*Coffee Break*	
	Chair: R. Grange	
10:40-11:15	B. Palpant	***Invited:*** *Ultrafast plasmonics in complex nano-objects*
11:15-11:40	F. Capolino	***Invited:*** *What's exceptional at exceptional degeneracy points?*
11:40-12:00	S. De Liberato	***Oral:*** *Mid-infrared nonlocality and electrical generation*
12:30-14:30	*Lunch*	
16.00 - Night	***Social Event:***	

Excursion to the Tyrrhenian Coast: Colavolpe Company (Belmonte) and Fiumefreddo Bruzio: one of the most beautiful Hamlet in Calabria

THURSDAY, June16th 2022		
Chair: F. De Angelis		
8:45-9:25	H. Altug	***Plenary:*** Frontiers in Nanophotonics: Enabling Technology for Next-Generation Optical Biosensors
9:25-9:50	G. Baffou	***Invited:*** *Quantitative phase microscopy for nanophotonics*
9:50-10:10	O. Maragò	***Invited:*** *Optical tweezers: from space to the nanoscale... and back*
10:10-10:40	*Coffee Break*	
Chair: O. Marago'		
10:40-11:15	J. Garcia De Abajo	***Keynote:*** *Atomically thin polaritonics: Challenges and opportunities*
11:15-11:40	V. Giannini	***Invited:*** *Exploring "Unconventional" Topological Nanoparticle Photonics Situations*
11:40-12:00	S. Bernatova	***Oral:*** *Optical force aggregation of gold nanorods as a tool to fabrication a multifunctional sensor*
12:30-14:30	*Lunch*	
Chair: F. Capolino		
16:00-16:35	N. Zheludev	***Keynote:*** *Optical Parametric Processes, Nonlinearity and Bistability in Nano-Opto-Mechanical Metamaterials*
16:35-17:00	H. Caglayan	***Invited****: Linear all-optical switching via plasmonic analogue of Enhancement of Index of Refraction*
17:00-17:30	*Coffee Break*	
17:30-17:55	C. De Angelis	***Invited:*** *Reconfigurable nonlinear photonic metasurfaces*
17:55-18:20	G. Tagliabue	***Invited:*** *Nanoengineering of Light Absorption: from Hot Carriers to Photothermal Effects*
18:30-19:30	Closing Remarks and Awards	
20:15-21:30	*Gala Dinner*	

Plenary Talks

Frontiers in Nanophotonics: Enabling Technology for Next-Generation Optical Biosensors

Hatice Altug

Ecole Polytechnique Federale de Lausanne (EPFL), Lausanne, Switzerland

Emerging healthcare needs and initiatives, including global health care, personalized medicine, and point-of-care applications are demanding breakthrough advancements in diagnostic tools. Biosensors play an essential role in bioanalytics, but traditional methods are limited in precision, affordability, integration or portability. Furthermore, they require long detection times, sophisticated infrastructure, and trained personnel. Our research group addresses these challenges by developing next-generation nanophotonic biosensors, spectroscopy and bioimaging technologies [1-5]. Our lab's expertise covers a variety of techniques, including nanophotonics, nanofabrication, microfluidics, surface chemistry, and data science. In particular, we exploit nanoplasmonic and dielectric metasurfaces because of their ability to confine light below the fundamental diffraction limit and generate strong electromagnetic fields at nanometric volumes. The increased light-matter interaction provided by nanophotonics is ideal to realize highly sensitive biosensors performing measurements from small sample volumes. We develop nanofabrication methods for high-throughput and low-cost manufacturing of nanophotonic structures and integrate them with microfluidics for automated sample handling. We also leverage smart data science tools with hyperspectral imaging and spectroscopy to achieve high device performance. In this talk, I will present some of our recent effort in these directions [6-13]. For example, I will introduce ultra-sensitive Mid-IR biosensors based on surface-enhanced infrared spectroscopy for chemical-specific detection of molecules in aqueous environment and large-area chemical imaging. I will describe ultra-compact, portable, rapid, and low-cost nanophotonic microarrays and their use for early disease diagnostics in real-world settings. I will also highlight label-free optofluidic biosensors that can perform one-of-a-kind measurements on live cells down at the single-cell level, and provide their overall prospects in biomedical and clinical applications.

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Advancing Photonics with Machine Learning

Alexandra Boltasseva[1,2*]

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Discovering unconventional optical designs via machine-learning promises to advance on-chip circuitry, imaging, sensing, energy, and quantum information technology. In this talk, we discuss photonic design approaches and emerging material platforms for showcasing machine-learning-assisted topology optimization for optical metasurface designs with applications in thermophotovoltaics, reflective optics, and lightsail technology. We demonstrate the effectiveness of autoencoders for compressing the vast design space of metasurfaces into a smaller search space. By employing global optimization via adjoint methods or quantum annealing, one can find the optimal metasurface designs within the smaller space constructed by the autoencoder. The quantum-assisted machine learning framework, named bVAE-QUBO, presented in this work is the first demonstration of a generic machine learning framework that compresses an arbitrary continuous optimization problem into an Ising-model formalism for quantum sampling. When compared to other global optimization techniques, bVAE-QUBO has the potential for quantum speedups and achieving higher quality designs than traditional adjoint optimization methods. The techniques employed in this work extend well beyond the metasurface optimization space and into many inverse design problems for engineering and physics.

Acknowledgements: This work is supported by the U.S. Department of Energy (DOE), Office of Science through the Quantum Science Center (QSC), a National Quantum Information Science Research Center, and Purdue's Elmore ECE Emerging Frontiers Center 'The Crossroads of Quantum and AI'. The authors also acknowledge support from the National Science Foundation award 2029553-ECCS.

Structured Light and Dark by Metaoptics

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Abstract

Metaoptics offer fresh opportunities for structuring light as well as dark. I will discuss metasurfaces that enable light's spin and OAM to evolve, simultaneously, from one state to another along the propagation direction[1,2], along with nonlocal supercell designs that demonstrate multiple independent optical functions at arbitrary large deflection angles with high efficiency.[3] In one implementation the incident laser is simultaneously diffracted into Gaussian, helical and Bessel beams over a large angular range and in another one a compact wavelength-tunable external cavity laser with arbitrary beam control capabilities including hologram lasing is demonstrated.[3] We also propose a new class of computer-generated holograms whose far-fields have designer-specified polarization response, dubbed Jones matrix holograms.[4] We provide a simple procedure for their implementation using form-birefringent metasurfaces. Jones matrix holography generalizes a wide body of past work with a consistent mathematical framework, particularly in the field of metasurfaces, and suggests previously unrealized devices, examples of which are demonstrated here. In particular, we demonstrate holograms whose far-fields implement parallel polarization analysis and custom waveplate-like behavior. Finally, the realization of 2D phase and polarization singularities and the unique applications that they will open will be discussed.[5]

1. Ahmed H. Dorrah, Noah A. Rubin, Aun Zaidi, Michele Tamagnone & Federico Capasso *Nature Photonics* **15**, 287 (2021)
2. Ahmed H Dorrah, Noah A Rubin, Michele Tamagnone, Aun Zaidi, & Federico Capasso *Nature Communications* **12,** 6249 (2021)
3. Ahmed H Dorrah, Noah A Rubin, Michele Tamagnone, Aun Zaidi, & Federico Capasso *Nature Communications*, **12**, 6249 (2021)
4. Noah A. Rubin, Aun Zaidi, Ahmed H. Dorrah, Zhujun Shi, & Federico Capasso *Science Advances*, **7**, eabg7488 (2021)
5. Soon Wei Daniel Lim, Joon-Suh Park, Maryna L. Meretska, Ahmed H. Dorrah, & Federico Capasso *Nature Communications*, **12**, 4190 (2021)

Extreme Metamaterials in Four Dimensions

N. Engheta

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Four-dimensional (4D) metamaterials, in which the material parameters can change with time in addition to (or instead of) changing in three dimensions of space, can offer extra degrees of freedom in manipulating and tailoring light-matter interaction [1]. This provides novel possibilities in exploring ideas for wave-based devices and components. In recent years there have been significant interests and growing activities in studying waves in temporal and spatiotemporal media with exciting photonic and electromagnetic features [2, 3]. In my group we have been investigating some of the fundamental aspects of waves in such 4D material platforms. We have introduced and developed the concepts of temporal aiming [4], temporal deflecting [5], anti-reflection temporal coatings [6], temporal equivalent of the Brewster angle [7], effective medium in temporal media [8], and asymmetric diffusion [9], to name a few. Moreover, we have also explored the generalization of the Kramers-Kronig relation for the time-varying media [10], and have studied in details how such media behaves in the presence of Lorentzian dispersion [11]. While there are interesting analogies between the wave interaction in temporal inhomogeneities and spatial inhomogeneities, there are also certain important differences, which can lead to interesting unconventional possibilities.

In this talk, I will present an overview of some of our ongoing work in this area, will discuss the physical insights into our findings, and will forecast future directions and possibilities in the field of 4D metamaterials.

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Nano-Photonic Emitters for the "Quantum Age": where imperfections lead to opportunities

Evelyn L. Hu

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Harvard University

Although we usually assume that a "perfect" material is required to produce the best emitters for nano-optical devices, *defect states* in wide bandgap semiconductors are defining a new frontier for nano-optics. In the 2018 meeting, I spoke of *silicon vacancies* in 4H-SiC, integrated within nanobeam photonic crystal cavities, which achieved an 80-fold optical enhancement of a Si-vacancy transition with emission at about 860 nanometers. That example showed the use of high-quality cavities as exquisitely sensitive optical amplifiers [1].

More recently, we have used the photonic crystal cavities as "nanoscopes" that allow us to better understand the local environment of the silicon vacancies, the other defects that they encounter, the possible interactions with those defects and pathways to better processing and control of the defects [2], [3]. The accompanying spin signatures [4], and the correlations between photon emission and spin modulation provide insights into further applications of these *quantum defects.*

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Nanophotonics for tailoring of electron-light interactions

Marin Soljačić

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Maxwell's equations are a very accurate representation of optical phenomena in most settings, but their accuracy often fails at nanoscale. In particular, for nanoplasmonic phenomena, whenever the relevant scales are below 10nm or so, there can be substantial discrepancies between the experimental observations and the predictions of Maxwell's theory. We present our theoretical and experimental framework [1] that extends their accuracy to this nano-scale regime.

We also present our work on Smith-Purcell radiation which occurs when fast electrons interact with nano-structured materials to emit light. We present our novel theoretical framework [2] to understand and tailor such phenomena, as well as how to enhance Smith-Purcell radiation.

Finally, we also present our theoretical framework for modelling and tailoring scintillation phenomena using nanophotonic techniques [3], as well as our experimental results in this field.

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Keynote Talks

Atomically thin polaritonics: Challenges and opportunities

J. Garcia De Abajo

ICFO- The Institute of Photonic Sciences, Barcelona, Spain

Atomically thin materials have emerged as a robust platform for manipulating and exploiting light at the nanoscale thanks to a wide variety of polaritonic modes, ranging from plasmons in highly doped graphene to excitons in transition metal dichalcogenides and photons in ionic insulators. The electromagnetic behavior of these modes can be well understood in terms of effective surface conductivities, which in addition can capture their strong dependence on temperature and external static electric and magnetic fields. Recent advances have also been produced in the synthesis of thin noble-metal films, which open new possibilities for exploring entirely new regimes of nanometallic plasmonics. In this talk, I will overview the general characteristics of the optical response of these materials, which can be understood in terms of simple theoretical descriptions. We will also cover more sophisticated models, aiming at exploring genuinely quantum-mechanical effects. We will further overview recent advances in the control of the ultrafast optical response and nonlinear optics, as well as the potential application of these materials in light modulation, quantum-optics, and optical sensing. The in/out coupling problem between external light and polaritons of short wavelength remains as a major challenge, for which we will propose innovative solutions. The presentation will conclude with some emerging directions in the design of polaritonic materials, including the intriguing possibility of exploiting quantum phase effects.

Bottom-Up Assembly of Random Polycrystals for Nonlinear, Electro-Optic, and Quantum Devices

Viola V. Vogler-Neuling, Artemios Karvounis, Andrea Morandi, Helena Weigand, and Rachel Grange

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Nonlinear and electro-optic devices are present in our daily life with many applications: light sources for microsurgery, green laser pointers, or modulators for telecommunication [1]. Most of them use bulk materials such as glass fibres or high-quality crystals, hardly integrable or scalable. Even the fast developments of thin film lithium niobate face the challenging etching of metal-oxides [2]. Therefore, the quest for a non-centrosymmetric material system, easy to fabricate and to scale up while maintaining its functionality is still ongoing. Here we will present our recent advances on bottom-up photonic assemblies of randomly oriented nanocrystals and how we can produce electro-optic, nonlinear and parametric down conversion signals. First, barium titanate metalenses synthesized by a sol-gel technique will be demonstrated (Fig. 1a) [3]. Then, we will show how the electro-optic response in assembled nanostructures can be as strong as certain other perfect crystalline structure (Fig. 1b) [4]. Finally, we will describe the more fundamental aspect of random quasi phase matching at the nanoscale. The field of metal-oxides at the sub-wavelength scale is developing fast with potential applications in nanophotonics, integrated optics and telecommunication [5-6].

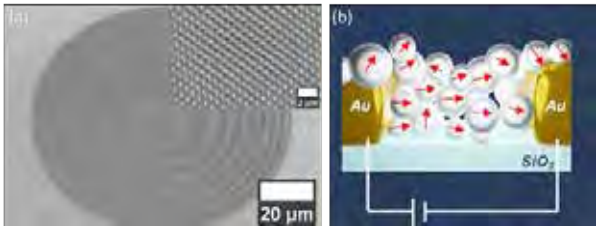

Fig. 1: Examples of barium titanate polycrystalline assemblies. (a) Nonlinear sol-gel metalens by nanoimprinting with a close-up view in the inset. (b) Schematic of an electro-optic device with MHz frequency.

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Hybrid Quantum Photonics

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Integrated photonic technologies are essential for efficient generation, manipulation, and detection of quantum states of light and can potentially enable a high density of on-chip photonic qubits and the level of performance required for the practical realization of various applications in the quantum domain. Our group recently discovered bright, stable, linearly polarized, and high-purity sources of single-photon emission at room temperature in scalable platform based on SiN. We also show how the enhancement of the light-matter interaction with plasmonic materials can shorten the spontaneous emission time to beat the dephasing time and achieve coherence even at non-cryogenic temperatures. Our findings spark further studies of quantum emitters toward deeper understanding of their nature, deterministic formation, and scalable integration with on-chip quantum photonic circuitry. We also demonstrate how hybrid quantum sensors with record-high sensitivity can be developed, by employing spin qubits controlled by light and coupled via magnons.

Optical Parametric Processes, Nonlinearity and Bistability in Nano-Opto-Mechanical Metamaterials

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The changing balance of forces at the nanoscale allows nanomachines that can alter optical properties of metamaterials with light, electromagnetic and acoustic forces and heat. We report new plasmonic and dielectric nano-opto-mechanical metamaterials that explore optical parametric phenomena for controlling light with light and achieving optical memory in such media.

Invited Talks

Plasmonics and Molecular Imprinted Polymer (MIP) Sensing

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Plasmonics and metamaterials have a common thread of enhanced phenomena and shape/composition independent dielectric and light behavior. A common application of plasmonic materials in sensing where the "enhanced light" can be used to further the use in detecting low levels of analyte and combined with a waveguide or guided layer detection. This is especially useful when combined with spectroscopic and imaging methods for characterizing the potential for artificial intelligence and machine learning (AI/ML) paradigm. This work will report on the use of plasmon, localized surface plasmon resonance SPR and SPR waveguiding methods to demonstrate high sensitivity and selectivity for the detection of chemical and biological analytes at a very low level of concentrations. Using electrochemically molecularly imprinted polymers enabling tuning of the dielectric parameters based on the oxidation state of the polymer to facilitate the MIP process and evaluate the figures of merit of a sensor device. The main sensitivity is achieved by the plasmonic enhancement and relationship with multilayer films and dielectric behavior closer to a guided wave platform. Thus, there is a high possibility of combining in-situ and in-operando measurements methods that can be relevant for future AI/ML applications.

hybrid plasmonic nano-sources of light: on the importance to control the spatial distribution of the active medium

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In microscale optoelectronics, the possibility to precisely control the spatial distribution of the active medium allows for the optimization of systems and devices. At the nanoscale, this issue still constitutes a challenge, especially within the frame of hybrid plasmonic nano-emitters [1].

Here, we report on the study of the spatial overlap between the exciting optical near-field and the nanoscale active medium whose position in space is controlled via plasmon-assisted two-photon polymerization [2] of a photosensitive formulation containing nano-emitters [3]. By using different geometries of gold nanoparticles and different modes of plasmon excitation, nano-emitter-containing active medium can be structured selectively with different degrees of symmetry in the close vicinity of the metal nanoparticles. The resulting hybrid plasmonic nano-emitters (e.g. Figures 1(a), 1(b)) are shown to be highly sensitive to the direction of polarization used for exciting the system (Figures 1(c), 1(e)). This sensitivity is quantified through the photoluminescence contrast between different states of emission, and related polarization-sensitive nanoscale spatial overlap integral between the local exciting optical field and the active medium [4].

By decreasing the concentration of the nano-emitters within the formulation, single quantum emitters can be trapped in the vicinity of gold nanocubes. As a result, we report preliminary observation of a nano-switch in the single photon regime that is driven by polarization [4].

Finally, we report a very recent new approach. The photopolymer has been functionalized to grab the emitters to its surface by electrostatic interaction after polymerization [5]. In this way, we can control both the position of emitters attached to the polymer surface and the average emitter-metal surface distance by adjusting the thickness of the polymer on the plasmonic structure. As a result, the emitter fluorescence life time can be controlled.

This approach has resulted in the promising concept of “smart photopolymer” for hybrid nanoplasmonics. The cunning use of the polymer is actually twofold. First, it

"records" both the selected site and the future emitters-GNC distance through plasmon-assisted photopolymerization. Second, because the polymer is chemically functionalized, it makes it possible to attach the nano-emitters right at the preselected polymerized sites which subsequently "recognize" the nano-emitters to get attached [6].

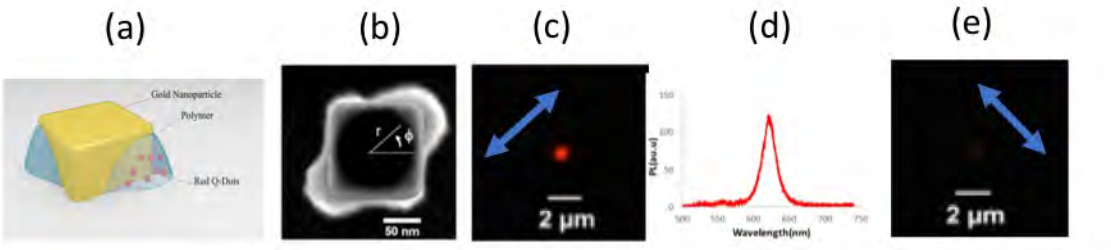

Fig. 1 (a) Schematic representation of a nanocube-based hybrid plasmonic nanoemitter. (b) SEM image of the hybrid nanosystem. (c) Far field image of the photoluminescence (PL) from a single hybrid nanosuurce excited with an incident polarization (**λ**=405 nm) parallel to the polymer lobes shown in (b). (d) PL spectrum from a single hybrid nanosource. (c) Far field image of the photoluminescence (PL) from a single hybrid nanosource excited with an incident polarization (**λ**=405 nm) perpendicular to the polymer lobes shown in (b).

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Quantitative phase microscopy for nanophotonics

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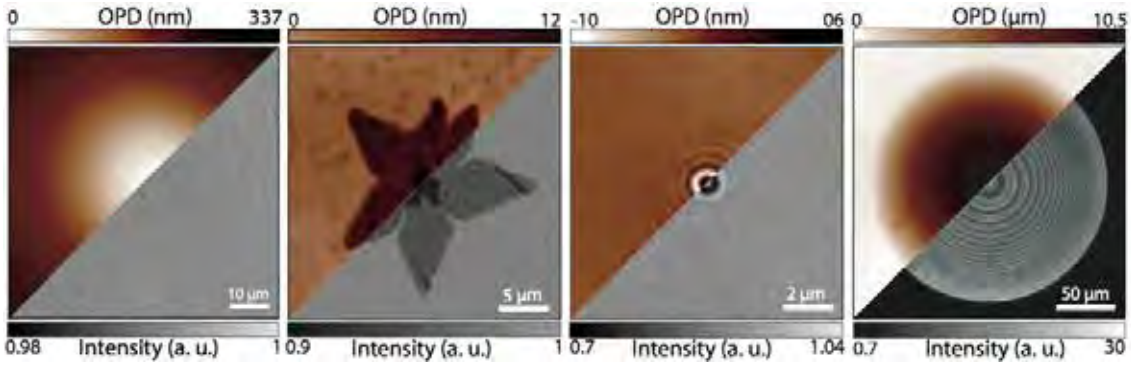
Quantitative phase microscopy (QPM) designs a family of optical microscopy techniques capable of mapping the phase of a light beam. Oddly enough, mapping the phase of a light beam seems to be only of interest for biologists, who have been extensively using QPMs technique for two decades, not really for physicists.

In this presentation, I show how we have been using a particularly sensitive and simple QPM technique called **cross-grating phase microscopy (CGM)** [1] for applications nanophotonics, opening a new route of investigation for QPM, besides biology. This contribution is aimed to explain (i) how CGM can be used as a **temperature microscopy technique** to map the temperature of gold nanoparticles under illumination [2], enabling applications in physics [3], chemistry [4] and biology [5] at the nano/microscale; (ii) how CGM can map the complex optical conductivity and complex refractive index of **2D materials** [6] ; (iii) how CGM can retrieve the complex optical polarizability of **nanoparticles**, along with the extinction, scattering and absorption cross section from a single interferogram image [7,8], and finally (iv) how CGM can fully characterize **metasurfaces** [9].



Examples of intensity and optical-path-difference (OPD) images acquired using CGM. From left to right: an assembly of gold nanoparticles under illumination, creating a localized, radial temperature gradient ; a MoS2 flake ; a single 100-nm gold nanosphere and a metalens.

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Linear all-optical switching via plasmonic analogue of Enhancement of Index of Refraction

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All-optical nano-switches and modulators operating at minimal energy consumption with extreme confinement, are highly desirable in nanophotonics for development of efficient all-optical computing and processing devices [1]. While, plasmonic equivalents of quantum optical phenomena in atomic physics such as Fano interference and electromagnetically induced transparency (EIT) lead to several remarkable applications for ultra-sensitive sensors and slow light [2, 3]. In this work, for the first time, we experimentally realize the plasmonic analogue of the Enhancement of Index of Refraction (EIR) effect in quantum optics and demonstrate linear all-optical switching mechanism through a particular plasmonic metasurface consisting of square array of L-shaped metamolecule. This is attributed to the coherent control of polarizability of nanoantennas by properly tailoring the phase, amplitude and polarization of the control beam to achieve the unprecedented modulation strength of signal beam at ultra low power. Our approach further shows how the transparency in the nearly whole plasmon band of metasurface is created without implementing gain materials or nonlinear processes and can be controlled as per demand using the properties of control beam as a tool.

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What's exceptional at exceptional degeneracy points?

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There are various kinds of degeneracies that can occur in mechanical, electromagnetic, photonic, quantum systems, and here we discuss an important class of them: the case when two or more eigenstates of a system coalesce, in both eigenvalues and eigenvectors. Such degeneracies can happen in circuit and optical resonators, plasmonic systems, electron beam devices, multimode waveguides, etc, either in fully passive systems or also with active components. In photonic systems, this kind of degeneracy also involves the polarization states, and can happen in a surprisingly large number of systems. We will discuss the common aspects of these degeneracies. When exceptional points of degeneracy (EPDs) occur in waveguides, we will also discuss the role of symmetries in their occurrence. Despite at the beginning EPDs were associated to PT symmetry in a system [1], now it is clear that PT symmetry is only a special case that leads to a real eigenvalue. Also, it has been said that EPDs belong to non-Hermitian systems, but they are found also in linear systems that are lossless and gainless [2]. We will show that, as a particular case, linear time variation is an enabling condition to find EPDs in single resonators [3].

What is so special about these degeneracies? Is it just an academic curiosity or they lead to interesting new physics that can be exploited to improve devices? In answering these questions, we will show that there are a variety of applications that can benefit from the special degeneracies of these points. We will focus on two possible applications: lasers/oscillators and sensing, showing the dynamics of these systems when operating at an EPD and possible advantages.

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Controlling and steering the nonlinear emission in nanoantennas and metasurfaces

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The last decades witnessed major efforts in nanophotonics devoted to secure the efficient generation of nonlinear effects in nanoscale volumes. This endeavor is driven by the utmost prospect to perform light conversion in integrated devices for optical information processing, sensing and nonlinear digital holography. The main hindrance to the technological deployment of nonlinear nanophotonics is the perturbative character of nonlinear interactions, whose intrinsic weakness is compounded by the small volume of matter involved at the nanoscale. To overcome such fundamental limit the key blueprint consists in exploiting the plasmonic and photonic (i.e. Mie-type) resonances – and the associated localized field enhancements – in engineered nanoantennas and metasurfaces to enhance light–matter interaction. Specific care has also to be paid to considerations about symmetry, in view of optimizing well-defined nonlinear processes [1].

In the first part of my talk, I will report on the sum frequency generation (SFG) process in both AlGaAs nanodiscs [2] and asymmetric gold nanoantennas [3]. In particular, we studied SFG using a semi-degenerate configuration, were one of the two pump beams is tuned at ω, in the telecom frequency domain (λ = 1550 nm), and at the second is its frequency-doubled at 2ω. In this way, SFG (at 3ω) is inherently degenerate with the third harmonic generation (THG) of the pump at ω. For symmetry reasons that I will describe, while in the AlGaAs nanoantennas SFG and THG do not show any interplay mechanism [4], in the plasmonic nanoantennas a sizeable interference between the two effects can be retrieved. The interference term can be coherently controlled by the phase delay between the pump beams, inducing an intensity modulation of the signal

up to 50%. Concurrently, a sizeable tilt in the polarization of the emission at 3ω can be detected, which is extremely sensitive to the nanoantenna geometrical parameters [5].

In the second part, I will describe our attempts to selectively direct the second harmonic generation (SHG) from all-dielectric metasurfaces. Firstly, we devised a lithium niobate monolithic metasurface for enhanced SHG. Thanks to the employment of a z-cut substrate and the exploitation of the Mie resonances in the individual meta-atoms, we were able to engineer the polarization and the intensity in the first diffraction orders by selecting the pump polarization [6]. Secondly, we demonstrated the possibility to modulate the SHG by nonlinear AlGaAs metasurfaces embedded in a E7 liquid crystal (LC) matrix. An almost 1 order of magnitude modulation is attained by changing the relative in-plane orientation between the LC director and the linear pump polarization [7]. A key role is also played by the orientation of the LC director and the lattice periodicity.

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Nonlinear optical response of two-dimensional materials

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Two-dimensional materials such as graphene and transition metal dichalcogenides (TMDs) show extraordinarily strong second- and third-order nonlinear optical responses which, thanks to their unique electronic properties and despite their atomic thickness, allow a wide range of fundamental studies and technological applications. In this presentation we will discuss some of our recent results on the study of the nonlinear optical properties of two-dimensional materials.

We show that the third-harmonic generation efficiency in graphene can be increased by almost two orders of magnitude by controlling the Fermi energy and the incident photon energy [1]. This enhancement is due to logarithmic resonances in the imaginary part of the nonlinear conductivity arising from resonant multiphoton transitions. Thanks to the linear dispersion of the massless Dirac fermions, gate controllable third-harmonic enhancement can be achieved over an ultrabroad bandwidth, paving the way for electrically tunable broadband frequency converters.

Using semiconducting TMDs, we demonstrate single-pass optical parametric amplification at the ultimate thickness limit, down to a single atomic layer [2]. Second-order nonlinear interaction at the two-dimensional limit bypasses phase-matching requirements and achieves ultrabroad amplification bandwidths. In agreement with first-principle calculations, we observe that the amplification process is independent of the in-plane polarization of signal and pump fields.

We investigate 3R-stacked TMD crystals which combine broken inversion symmetry and aligned layering. By measuring second harmonic generation (SHG) of 3R-MoS_2 with various thickness, from monolayer (0.65 nm) to bulk (≈1 μm), we present the first measurement of the SHG coherence length (≈ 530 nm) at 1520nm and achieve record nonlinear optical enhancement from a van der Waals material, > 10^4 stronger than a monolayer. We find that 3R-MoS_2 exhibits similar conversion efficiency as lithium niobate, but with more than 100-fold shorter propagation lengths.

Finally, we demonstrate a novel approach for the all-optical control of SHG polarization in MoS_2 and show that this can be used for all-optical modulation of the SHG efficiency with modulation depth close to 100% and speed limited only by the fundamental frequency pulse duration [3].

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Reconfigurable nonlinear photonic metasurfaces

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Optically resonant metasurfaces are an emerging and promising area of nanophotonics. Recently, active tuning of the linear response and nonlinear effects of these components has received an increasing amount of interest. However, so far, these research directions have remained separated with only few sporadic works that study their combination beginning to appear in the literature. The evolution of nonlinear metasurfaces toward reconfigurable and dynamic components could potentially answer the demand of integrated on-chip components that realize essential functionalities such as frequency conversion, active switching, optical isolation, and all-optical routing. I will present here our recent investigations in this field [1].

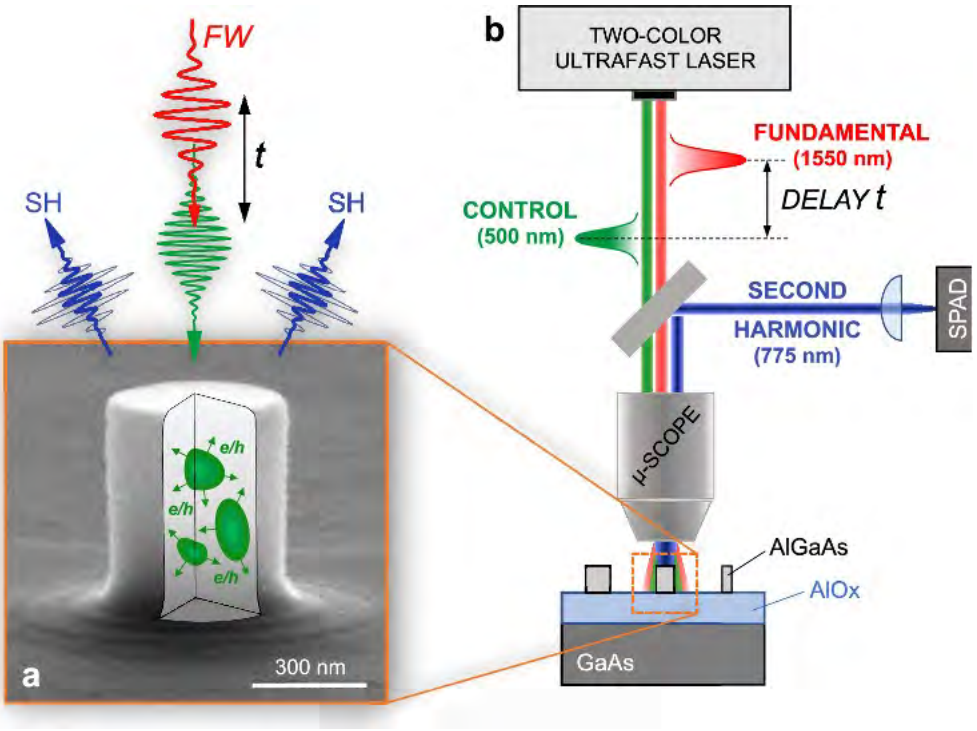

Figure 1: Concept of the ultrafast all-optical control of nanoscale second-harmonic generation. (a) Scanning electron microscopy image of the nonlinear nanoantenna. (b) Sketch of the experimental setup combining a two-color ultrafast laser source.

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Plasmonic Solid-state Nanopores: Toward Single-molecule Protein Identification

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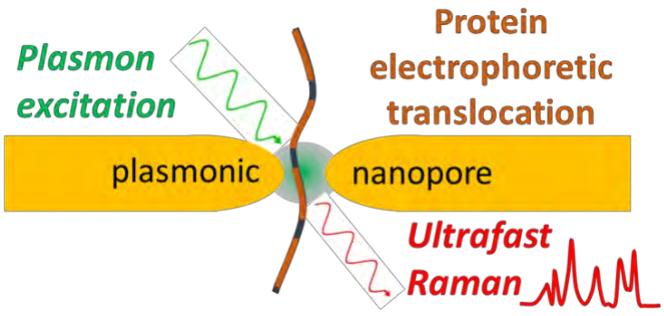
Sequence identification of peptides and proteins is central to proteomics. Protein sequencing is mainly conducted by insensitive mass spectroscopy because proteins cannot be amplified, which hampers applications such as single-cell proteomics and precision medicine. The commercial success of portable nanopore sequencers for single DNA molecules has inspired extensive research and development of single-molecule techniques for protein sequencing. Among them, three challenges remain: (1) discrimination of the twenty amino acids as building blocks of proteins; (2) spatial and temporal resolution (sensitivity) to detect single amino acids within the same molecule; and (3) controlling the motion of proteins into the nanopores. In this context, the emergence of label-free optical analysis techniques for single amino acids and peptides by solid state nanopores shows promises to address the first two challenges. Here we show our latest results on label-free optical methods to show how they address the single-amino-acid identification within single peptides. They include localized surface plasmon resonance detection and surface-enhanced Raman spectroscopy on plasmonic nanopores. Notably, we report new data to show the ability of plasmon-enhanced Raman scattering to record and discriminate the twenty amino acids at a single-molecule level. In addition, we discuss briefly the manipulation of molecule translocation and liquid flow in plasmonic nanopores for controlling molecule movement to allow high-resolution reading of protein sequences. We envision that a combination of Raman spectroscopy with plasmonic nanopores can succeed in single-molecule protein sequencing in a label-free way.



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Exploring “Unconventional” Topological Nanoparticle Photonics Situations

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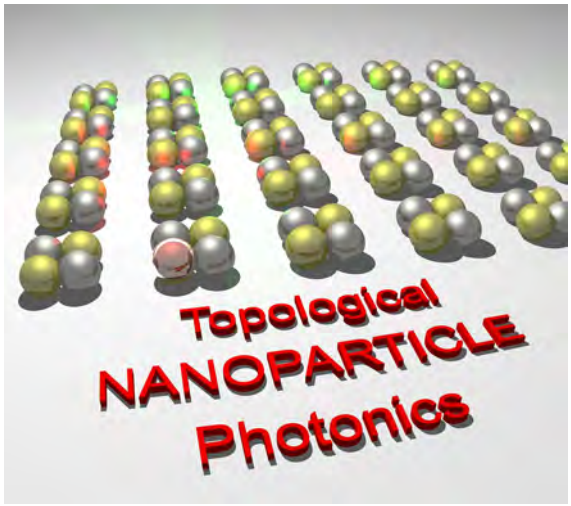
Topological nanophotonics is a new avenue for exploring nanoscale systems from visible to THz frequencies with unprecedented control. A driving force of such a new field has been condensed matter physics. This talk will explore some phenomena that cannot be obtained in condensed matter but only in photonics. In particular, we will see that simple anisotropic nanoparticle systems allow us to project the dipolar moment in a particular direction obtaining a simple system that presents hidden symmetries in a first analysis[1].



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Epsilon (and mu) Near Zero Materials – Photonics on Steroids?

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Abstract:

We examine the characteristics of optical materials with near zero real part of permittivity (ENZ), and compare them with other materials relying on resonance where Re(ε)~0 (e.g. plasmonics) or Re(1/ε)~0 (e.g. slow light, microresonators, e.t.c.). Despite being seemingly very diverse phenomena all of the resonant effects share a key common characteristics – slow group velocity. Consequently, whether one operates near a zero or a pole in optical response, one is bound to gain the same very useful enhancement of some properties such as nonlinearity, and, regrettably, the commensurate increase of loss and reduction in bandwidth. We show that all the purported enhancement of nonlinear properties of ENZ material comes from (a) the fact that the group velocity is very low ("slow light effect") (b) the response due to hot carriers is sufficiently fast but not instant – that is definitely the most attractive feature. If in addition to ε~0 the magnetic permeability μ~0 as well (EMNZ materials), group velocity is high, and consequently, no enhancement of nonlinear optical properties ensues. As nonlinear or electro-optic materials ENZ's do have another advantage – slow group velocity can be achieved in them without fabricating nanostructures such as resonators, photonic crystals etc. But this advantage needs to be weighed against high insertion loss. If time remains, we shall discuss "zero-index-materials" based on photonic crystals, which unlike ENZ offer no advantage whatsoever when it comes to switching and modulation

Optical tweezers: from space to the nanoscale… and back

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Optical tweezers are powerful tools based on focused laser beams [**1**]. They are able to trap, manipulate and investigate a wide range of micro and nanoscopic particles in different media, such as liquids, air, and vacuum [**2**]. After an introduction to optical forces, I will give an overview of results on optical trapping, optical binding, and characterization of particles at the nanoscale [**3-6**]. Furthermore, we will describe how optical tweezers can be used to trap and characterize extraterrestrial particulate matter. On one side, we exploit light scattering theory in the T-matrix formalism to calculate radiation pressure and optical trapping properties of a variety of complex particles of astrophysical interest [**7**]. On the other side, we show results on microparticles in controlled laboratory experiments. Our results open perspectives to non-destructive, non-contact and non-contaminating investigation of extraterrestrial particles, aiming for space tweezers applications.

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Ultrafast plasmonics in complex nano-objects

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The irradiation of plasmonic nanoparticles by ultrashort laser pulses produces a series of transient phenomena that can be exploited in various fields, from targeted therapies to nanoscale broadband light radiation sources [1]. These phenomena can be ascribed to the ultrafast dynamics of the metal hot electron gas induced by multiphoton absorption, which can be monitored by recording the transient evolution of the nanoparticle optical response. The plasmon modes of noble metal nanoparticles cover the visible and near-infrared spectral ranges. Although the nanoparticle size, shape, and composition influence the characteristics of these modes, it is impossible to access strong resonances in the near-ultraviolet, while this would yet be potentially beneficial for many applications.

In this communication, we study both the stationary and transient optical properties of different complex plasmonic nano-objects: (*i*) silica nanowires decorated with gold nanodisks [2]; (*ii*) bimetallic AuNR@Ag nanocuboids consisting of a gold nanorod core (AuNR) coated with a silver shell with variable thickness (Fig. 1) [3]. Due to the dynamics of the hot electrons generated by ultrashort pulses, the efficacy of plasmonic nanoparticles shined at their resonance for near-field enhancement, electron photoemission, and photothermal conversion is sublinear with the incoming intensity [4]. Despite this nonlinearity, we show that the optical properties of complex nano-objects with inner interfaces exhibit robust, sharp, and ultrafast variations at their high-order plasmon modes in the near-ultraviolet range (Fig. 1) [5].

Furthermore, analyzing the transient absorption signal by fast Fourier transform reveals vibration modes from 15 to 150 GHz frequency in the AuNR@Ag bimetallic nano-objects. Simulations allow us to address this vibrational landscape. While bare AuNRs exhibit extensional and breathing modes [6], the AuNRs@Ag undergo complex motions involving facets, edges, and corners. Their amplitude and frequency depend on the Ag-shell thickness, as the silver load modifies the nanoparticle aspect ratio and mass. Moreover, the acousto-plasmonic coupling results in a wavelength-dependent vibrational spectrum that we elucidate in simulations combining the nanoparticle elastic and optical properties.

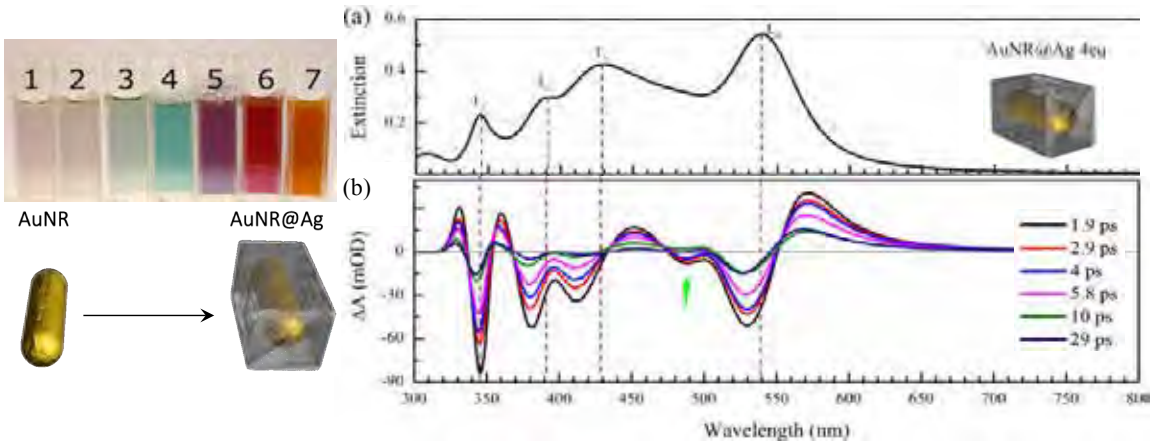

Fig. 1. Left: Photograph of AuNR@Ag suspensions with increasing Ag thickness from 1 to 7. Right: (a) Experimental stationary extinction spectrum of AuNRs@Ag nanoparticles with an equivalent Ag:Au molar ratio 4 (labeled as 4eq). One longitudinal dipolar mode (L_0) and three multipolar transverse modes (T_i) can be identified. (b) Ultrafast transient variation of the absorbance spectrum induced by interaction with a pump laser pulse, probed at different time delays. Strong and narrow features can be associated with the plasmon modes. The green arrow points a bleaching zone which reveals the existence of an additional resonance mode, hidden in the stationary case (a).

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Dielectric meta-optics for reconfigurable planar optics and biosensing

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In this talk, we present our latest advances in the development of novel nanophotonic platforms for both imaging and biosensing. In the first part, we introduce our most recent advances in the development of reconfigurable planar optical elements, focusing on two original technologies. The first technology relies on dynamically controlling the distribution of refractive index by means of an engineered micro-resistor embedded in a thermo-optical material. We first show how our approach can be used to create varifocal lenses with 100ms response time. When combined with machine learning, this approach additionally enables to go beyond a simple lens and create complex phase fronts. Our second approach to reconfigurability relies on an optomechanical control. Upon illumination with a control beam, the meta-atoms forming the lens mechanically rearrange, inducing a fast change of focus. In the second part of the talk, we discuss the use of dielectric nanoresonators for biosensing and lab-on-a-chip technology. In our approach, Si nanoresonators are integrated into a state-of-the-art PDMS microfluidic environment. We first demonstrate that arrays of Si nanocylinders can be used for the specific detection of cancer markers in human serum with sensitivity levels comparable to the one obtained with gold nanoantennas. We also show how dielectric nanoresonators can benefit chiral molecular sensing, demonstrating enantio-selective differentiation with improved performance over plasmonics. Finally, we discuss different novel directions toward improved sensing performance and detection of emerging biomarkers.

Nanoengineering of Light Absorption: from Hot Carriers to Photothermal Effects

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In the last decade, optical nanoantennas have revolutionized light manipulation and control at the nanoscale. Light absorption was initially considered a purely detrimental process, reducing the efficiency of optoelectronic devices. Recently, however, it has attracted growing interest, enabling novel light-energy conversion pathways, such as hot carrier harnessing, but also interesting application opportunities based on nanoscale photothermal effects.

In this talk, I will first present fundamental aspects of plasmonic hot carrier generation and collection towards photoelectrochemical devices for light energy storage. In particular, I will report the construction, optoelectronic and photoelectrochemical characterization of plasmon-driven devices that operate via hot-hole injection, and I will compare them to their hot electron counterparts [1-4]. I will also show emerging opportunities for plasmonic hot carriers in redox-couple based photoelectrochemical devices and advanced optical design strategies for improving the efficiency [5]. Overall, these results demonstrate the critical role of the metal properties onto hot carrier generation and transport. Thus, I will next report on our recent effort towards the synthesis of record-high aspect ratio gold monocrystalline flakes [6]. In particular, I will discuss how we can leverage them for understanding fundamental electronic processes in metals as well as plasmonic photo-electrochemistry. Finally, I will focus on collective photothermal effects in plasmonic networks and discuss a fast computational method we developed to estimate local temperature maxima and minima. I will conclude with an outlook on applications of hot carriers and photothermal effects in nanoengineered optical antennas.

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The rise of two-step absorption 3D laser nanoprinting

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Two-photon absorption based 3D laser printing on the micro- and nanometer scale has become a commercially available workhorse for the making of complex 2D and 3D architectures for optics and other areas [1]. Two-photon absorption provides the nonlinearity that is required to spatially confine the excitation. In order to make two-photon absorption reasonably efficient, femto- or picosecond lasers are used. These lasers alone cost several tens of thousands of Euro and take up a major part of the volume of commercial instruments.

Here, we review our recent efforts on replacing two-photon absorption by two-step absorption. In essence, we replace the "virtual" or dressed state that the light field induces in between the ground state and an excited state of a photoinitiator molecule by a "real" electronic state. As a result, still two photons need to be absorbed to lead to a population of the excited state, but these two photons need not be absorbed simultaneously, they can be absorbed sequentially. This process can be mediated by a compact continuous-wave (cw) low-power (< 1 mW) semiconductor laser diode [2]. Due to the used wavelength of 405 nm, the spatial resolution is better than for two-photon absorption based 3D laser nanoprinting using wavelengths at around 800 nm. We work on building a 3D laser nanoprinter that fits into a shoe box and that is orders of magnitude less expensive than current commercially available instruments.

In addition to such one-color two-step absorption, two-color two-step light-sheet 3D laser printing using cw lasers effectively works with 33,000 independent laser foci in parallel und approaches printing rates of 10^7 voxels/s [3].

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Orals

Optical force aggregation of gold nanorods as a tool to fabrication a multifunctional sensor

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Optical nano-printing provides a versatile platform to print various nanometer size particles into arbitrary configurations with very high precision. Optical printing, the use of light to direct the formation of a desired structure, has been of significant interest in the last two decades [1, 2, 3, 4, 5].

For particles much smaller than the laser wavelength, optical forces can be well described in the dipole approximation[6, 7, 8]. For a focused laser beam, two main optical force components are identified[9, 10]: the gradient force, which attracts particles toward the high-intensity focal spot, and the scattering force, which tends to push particles along the beam propagation direction. When the light is nearly resonant with the particle localised surface plasmons resonance, optical forces are dominated by radiation pressure[11] and can be used to efficiently push nanoparticles along the beam optical axis onto a substrate [12, 1, 13, 2]. In this context, optical forces can be applied to optically print nanoparticles into patterns aggregated elements [12, 1, 3, 5] on surfaces such as glass.

Here, we summarize recent progress in our experiments that use optical nanoprinting of plasmonic nanoparticles to create an active aggregate in a solution containing biomolecules or nanoplastics. The active aggregate, produced on the base of optical forces, serves as a very sensitive sensor which is used to detect biomolecules in concentration below the limit of detection for common Raman spectroscopy and/or to detection of nano meter size plastic particles.

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Quantum Hydrodynamic Theory for Plasmonics: from Molecule-Coupling to Nonlinear Optics

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Metals support surface plasmons at optical wavelengths and have the ability to localize light to sub-wavelength regions. Nano-gap plasmonic systems – in which two or more metallic nanoparticles are separated only few nanometers from each other by an insulating spacer – have been predicted to produce enormous field enhancements (as much as thousands of times that of the incident radiation) [1]. For the narrowest (< 1 nm) gaps, light can be so tightly confined that the nonlocality associated with the dielectric response of the metal and quantum effects can have a strong impact on the scattering properties of the system, placing strict bounds on the ultimate field enhancement [2].

A reliable way to theoretically describe and numerically model optical properties of plasmonic nanostructures with different length scales requires methods beyond classical electromagnetism. In this context, it becomes very important to develop simulation techniques to take into account quantum microscopic features at the scale of billions of atoms. A promising solution is given by the hydrodynamic theory, which takes into account the nonlocal behavior of the electron response by including the electron pressure and it can be generalized so that it can describe electron spill-out and tunneling effects [3, 4]. This method allows to explore light-matter interactions in extreme scenarios in which microscopic features can strongly affect the macroscopic optical response.

In this talk, we will present the quantum hydrodynamic theory for plasmonics and will discuss some applications including, photon emission [6], strong-coupling [7] and nonlinear optics [8].

Finally, we point out the limitations of conventional functionals in QHT and propose possible corrections [5, 9].

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Mid-infrared nonlocality and electrical generation

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The development of phonon polariton technology has allowed some concepts developed in plasmonics to be translated to the mid-infrared spectral range. Due to their longer lifetime, phonon polaritons easily enter the strong coupling regime with other sample-hosted excitations, creating highly tunable hybrid quasiparticles with on-demand characteristics. Examples of matter resonances that have been demonstrated to have strong coupling with phonon polaritons include epsilon-near-zero modes [1], monopolar resonances [2], and weak phonons [3].

The maturity of nanoscale fabrication of polar dielectrics has also made possible the realization of complex designs on truly nanometric scales, entering a regime in which standard dielectric modelling breaks down. As a result, density-functional numerical approaches have had to be used to interpret the resulting spectra (see Fig. 1a) [4], creating a serious limit to the quantitative electromagnetic simulation of full-sized devices. In a series of papers, we have developed a semi-analytical theory of the nonlocal optical response of polar dielectrics [5,6,6,7], demonstrating how the novel features observed in nanoscale phonon-polariton samples can be understood and described quantitatively using such a theory, which can be easily integrated in commercial numerical solvers.

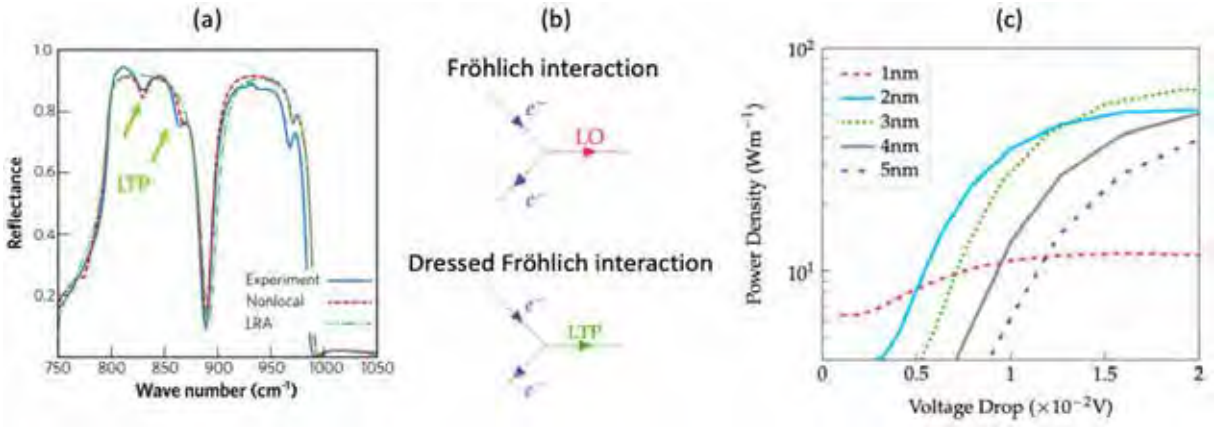

Fig. 1 (a) Reflectance of a GaN-AlN superlattice. Local response approximation (LRA) doesn't allow to model the experimentally visible LTP modes. (b) Schematic demonstration of Fröhlich electron-phonon interaction dressed by strong coupling. (c) Emission of LTP by electrical current flowing through SiC nanolayers of different thickness due to the dressed Fröhlich interaction.

Crucially, and in contrast to what happens in plasmonic nonlocality, optical phonons disperse toward the red. This implies that longitudinal and transverse charge oscillations can become resonant and hybridise, generating what we named Longitudinal-Transverse polaritons (LTP). In addition to providing a tool to model the optical response of nanoscopic polar dielectrics without requiring cumbersome numerical methods, our theory has thus demonstrated how hybridization between longitudinal and transverse phonons is a generic feature of nanoscale polar dielectrics. The practical relevance of this discovery to mid-infrared nanphotonics is due to the fact that while transverse charge oscillations couple with far-field radiation, electrical currents couple strongly to longitudinal oscillations through the Fröhlich interaction.

In the last part of this talk, we will show how the hybrid longitudinal-transverse nature of LTPs can solve one of the major problems in mid-infrared photonics by allowing resonant emission of mid-infrared photons via electric currents, without the need for complex quantum cascade structures (see Fig. 1b-c).

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Ultrafast All-optical Light Management with Plasmonic Metasurfaces

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Resonant plasmonic effects in gold metaatoms are exploited to achieve all-optical control of light with unprecedented speed. Photoinduced broadband dichroism, fully reversible and transiently vanishing in less than 1 picosecond, has been experimentally demonstrated in plasmonic metasurfaces with nanocross metaatoms. Also, we designed a nonlinear plasmonic metagrating where the photoinduced hot-electron symmetry breaking enables ultrafast reconfiguration of diffraction orders via control laser pulses.

Hot electrons in plasmonic nanostructures have been the subject of intensive research, with particular interest to the temporal dynamics following excitation with femtosecond laser pulses [1]. However, the effects linked to spatial local inhomogeneities of hot electrons have been overlooked until very recently [2-4].

We designed a plasmonic metasurface (square lattice arrangement with ~270-nm periodicity) of C4-symmetric gold nanocrosses with 45-nm thickness, 60-nm width and 165-nm length of the nanocross arms. The metasurface was fabricated by electron beam lithography on a transparent substrate (CaF2). Due to its high symmetry, this nanomaterial provides a polarization-independent static transmittance at normal incidence, characterized by a broad extinction peak at 800 nm, arising from the degenerate longitudinal plasmonic resonances of the two arms. Such a degeneracy can be broken by the resonant absorption of an ultrashort control pulse with linear polarization parallel to the direction of one of the arms (Fig. 1a). Indeed, photoexcitation creates a highly inhomogeneous near field, mostly because of the retardation-based nature of plasmonic modes in relatively large nanostructures. The inhomogeneous absorption pattern in each meta-atom locally affects the electronic energy distribution of gold, inducing a non-uniform out-of-equilibrium hot-carrier distribution that anisotropically modifies the metal permittivity on a time scale of 1 ps (Fig. 1b). The ultrafast photoinduced symmetry breaking is revealed as a transient transmission anisotropy from polarization-resolved pump-probe measurements (Fig. 1c). The degeneracy between the two polarizations is restored in ~1 ps, i.e. much earlier than the onset of electron-phonon relaxation processes (taking place on the time scale of 10 ps) [3].

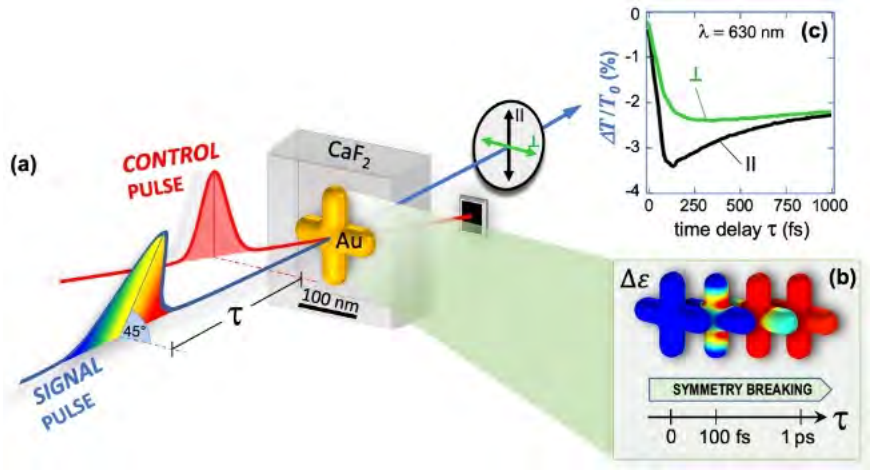

Figure 1. (a) Sketch of the Au-nanocross metaatom supported on a dielectric substrate. The meta-atom is excited with ~30-fs control laser pulses (~400 μJ/cm^2 fluence) at 860 nm (red), generating hot electrons at the nanoscale. (b) The C4 symmetry of the nanocross is broken by the highly inhomogeneous spatial pattern of the photoinduced permittivity modulation on the sub-picosecond time scale. (c) Polarization-resolved analysis of the transmitted signal at a time delay τ with respect to the control pulse [3].

By exploiting the same approach, we designed a plasmonic metagrating with 800 nm periodicity and unit cell made of a bent gold nanostrip supported on a sawtooth CaF2 substrate. A p-polarized ultrashort laser pulse at 600 nm shined at 45° angle of incidence is capable of inducing a highly inhomogeneous spatial pattern of the photogenerated hot electrons. The subsequent nonlinear permittivity change thus breaks the left-right symmetry of the cell and causes a transient imbalance between symmetric diffraction orders, for a broadband signal pulse impinging at normal incidence. Thanks to the high thermal conductivity of the metal, causing high speed homogenization of electronic temperature, the degeneracy between +1 and -1 orders is restored in about 2 ps [4].

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Light-induced forces in front of epsilon-near-zero metamaterials.

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The extraordinary properties of Epsilon-near-Zero (ENZ) materials allow many interesting applications [1, 2, 3]. Among these, the levitation of an electric dipole in front of an ENZ metasurface, resembling the Meissner effect for magnets in front of semiconductors, has been shown [4]. Optical tweezers [5, 6] can be used as extremely sensitive probes of force fields in front of surfaces. This approach allows force sensing in the femtoNewton range, with a spatial resolution controlled by the size of the trapped probe [7]. In this work, we aim to extend the study of light-induced forces on nano- and micro-sized particles in front of an ENZ surface. We use both a theoretic and an experimental approach. Dipole approximation, finite elements calculations, and T-matrix modeling are used to calculate forces expected on dielectric particles, core-shell SiO_2-Ag spheres and Ag ellipsoids. The experimental investigation of layered materials, behaving as ENZ at a fixed wavelength (560 nm), allow the observation of a thermophoretic force in front of the metasurface, suggesting a high non-linear light-to-heat conversion efficiency of this structure. Our results may be used as a guide for the design of new metasurfaces enabling the tailoring of light-induced forces in optomechanics.

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Controlled Aggregation of Gold Nanorods on Graphene by Radiation Pressure for SERS Detection of Biomolecules

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Radiation pressure can be used to control the aggregation of gold nanorods (AuNRs) on substrates in order to create plasmonic clusters suitable for Surface-Enhanced Raman Spectroscopy (SERS) of biomolecules in liquid at low traces [1,2]. In this work the effects of the substrate where the aggregation occurs is studied. Namely the technique is used to push AuNRs on multilayered graphene and create hybrid active surfaces for SERS in liquid [3]. As a proof of concept, ultrasensitive detection of bovine serum albumin (BSA) is shown, and the aggregation kinetics is studied as a function of the irradiation time. We observe that optical aggregation on graphene is 3.5 times slower compared to glass, while no stable aggregation is obtained on gold. We attribute the differences to the destabilization effect of the standing wave produced on the metallic substrates, due to their higher reflectivity, and to the reduced thermophoretic effects, related to the higher heat dissipation. Despite the slowdown of the aggregation kinetics, the usage of graphene as substrate offers manifold benefits: an almost negligible fluorescence background when using near-infrared light (785 nm), the absence of thermal absorption as well as the possibility to easily functionalize the surface to enhance the affinity with the analytes. Our results enlarge the spectrum of materials that can be used for optical aggregation and SERS detection of biomolecules, highlighting the importance of controlling the physical properties of the surfaces [3].

Our results open perspectives non-destructive, non-contact and non-contaminating investigation of extraterrestrial particles, aiming for space tweezers applications.

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Trapping of biological nanoparticles by quasi-bound state in the continuum

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Bound states in the continuum (BICs) are nonradiating states of light showing an infinite Q-factor. Dielectric metasurfaces can support quasi-BIC modes with a finite Q-factor and large electric field enhancement [1, 2]. In this contribution, we investigate quasi-BIC modes for near-visible light and for trapping of biological nanoparticles. A near-visible wavelength, i.e. 785 nm, is studied for possible combination of trapping and Raman-spectroscopy in the future. Silicon nitrite (Si_3N_4) is chosen for the metasurface, as it is transparent for this wavelength range.

A vast number and variety of biological nanoparticles circulate in the body and are involved in diverse physiological and pathological activities. They can be biomarkers of diseases and contribute to intercellular communications. Thus, trapping and analysis of biological nanoparticles is important to investigate their biochemical properties. Using the large field enhancement of a silicon quasi-BIC metasurface, trapping of small nanoparticles has recently been reported at 1550 nm [2]. Optical nanoantennas based tweezers have also been investigated in several works to trap and manipulate nanoscale objects [3-5]. However, they have limited field enhancement, hence requires a high laser power for stable trapping. In plasmonic tweezers, Joule heating due to absorption losses generates a thermally induced flow that moves the trapped particle from the hotspots, thus decreasing the trapping stability. Therefore, a system that allows stable trapping of nanoparticles with a negligible heating is desirable. Here, we numerically investigate a Si_3N_4 quasi-BIC metasurface for trapping of biological nanoparticles at near-visible wavelength. At this wavelength, Si_3N_4 has no absorption, and heating can thus be ignored.

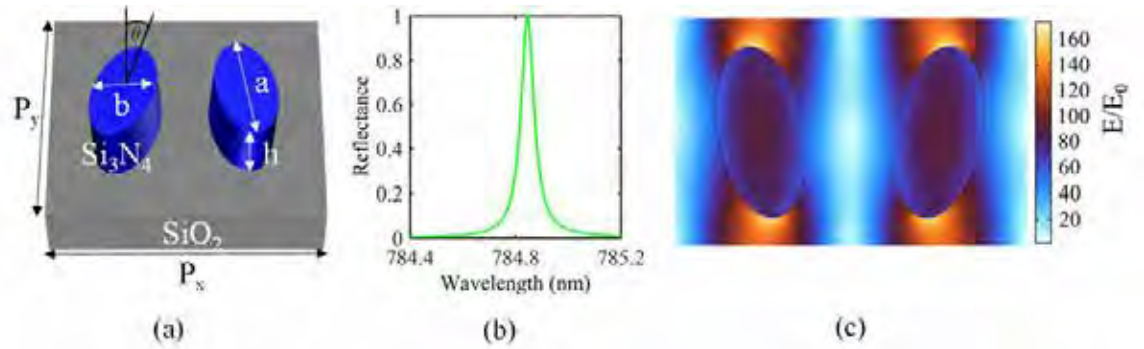


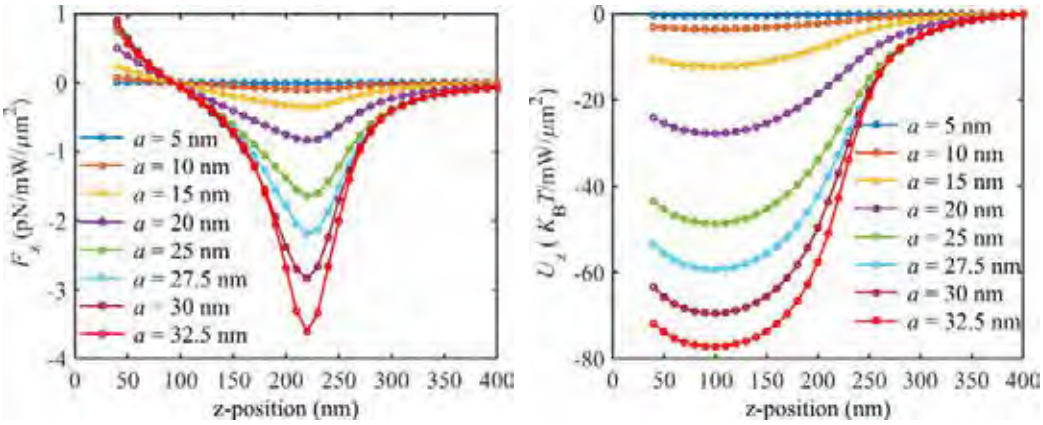
Fig. 1 (a) Unit cell with dimensions P_x = 522 nm, P_y = 336 nm, a = 255 nm, b = 125 nm, θ = 10°, and h = 230 nm. (b) Reflectance spectrum showing a peak at λ = 784.84 nm. (c) Electric field enhancement at z = 115 nm (mid-height of bar) and λ = 784.84 nm.

In this work, we design and simulate a dielectric metasurface consisting of an infinite array of a unit cell. The unit cell contains a pair of Si_3N_4 elliptical bars on a SiO_2 substrate (Fig. 1a). We use COMSOL Multiphysics 5.6 to simulate the unit cell. By varying the design parameters of the unit cell, the resonance wavelength of the quasi-BIC mode is adjusted to 784.84 nm (Fig. 1b). For these parameters, the tip-to-tip gap between two unit-cells (where the particle is trapped) is 81 nm, and the structure is thus not suitable for trapping nanoparticles with a larger

diameter than this gap. At this wavelength and for bars with 10º angle, a high electric field enhancement of ~ 178 is achieved on the tips of the bars (Fig. 1c). The metasurface contains a periodic array of unit cells, many 'hotspots' can be obtained, allowing simultaneous trapping of a single nanoparticle at each gap.

Next, we use the Maxwell stress tensor (MST) to determine the optical forces exerted on a trapped nanosphere. A refractive index of $n = 1.39$ is used for the nanosphere, to resemble that of a biological nanoparticle. Due to the high field enhancement, the nanosphere experiences strong optical forces, which increase with nanosphere radius (Fig. 2a). The vertical position showing zero force F_z indicates the equilibrium position (at approx. $z = 100$ nm), where the nanosphere is trapped. We obtain the trapping potential by path integration of the optical forces along the vertical direction. The potential depth increases strongly with particle radius a (Fig. 2b), as expected. A potential depth of 10 k_BT is considered sufficient for stable trapping, and a relatively modest power of 1 mW/μm^2 is thus sufficient to trap particles with a diameter of 30 nm.

This work shows that quasi-BIC can be used to trap biological nanoparticles with diameters of 30–65 nm using near-visible light. However, the fabrication tolerances will be tight and making the structure will be challenging. Regarding Raman-spectroscopy of trapped nanoparticles, the feasibility depends mainly on the Raman-background of the metasurface, which has not been considered in this work.



(a) (b)

Fig. 2 (a) Vertical force F_z and (b) trapping potentials U_z as a function of z-positions for different radii of trapping nanoparticles.

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Plasmonic Nanogap-enhanced Single-molecule Raman Spectroscopy: Towards Single-protein Raman Sequencing

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Proteins are the workhorses in biological systems that can be used to characterize cellular functions and progress of diseases as biomarkers. The fact that proteins cannot be amplified results in a serious lag of proteomics behind genomics and transcriptomics, hampering not only mechanistic studies but also clinical applications. The state-of-the-art single-molecule sequencing technologies towards protein sequencing include label-based fluorescence methods and label-free nanopore resistant pulse sensors. Compared to DNA, proteins are folded 3-dimentional structures and consist of 20 proteinogenic amino acids with different charges. These features limit these single-molecule sequencing methods to sequencing only single peptides, because they can detect 4 at maximum of the 20 proteinogenic amino acid residues comprising the primary sequence of a protein.

Surface-enhanced Raman spectroscopy (SERS) as a powerful label-free biomedical detection method for with single-molecule sensitivity. It could pave a way to new generation single-protein sequencing platforms by the single-molecule SERS spectra of the 20 types of amino acids as building blocks of proteins. In fact, each amino acid molecules have their own fingerprint Raman scattering signals that can be used for sequencing. But the Raman signals of single amino acids were too weak to be detected even by surface-enhanced Raman spectroscopy (SERS). In the tremendous amount of literature of SERS-based protein detections in past decades, the SERS spectra of proteins are mostly occupied by SERS signals of aromatic amino acids and backbones, while those of non-aromatic amino acids are invisible.

In this talk, we will present our recent work on a plasmonic nanogap biosensor that has demonstrated single-molecule Raman detection of all 20 proteinogenic amino acids.[1] By trapping single gold nanoparticle in a gold nanopore to generate a single plasmonic hot spot in the nanogap,[2] the sensor achieved detecting single aromatic and non-aromatic amino acid residues within single peptides, as shown in Figure 1.[3] Our results suggest that the signal dominance due to large spatial occupancy of aromatic rings of the peptide sidechains can be overcome by the high localization of the single hot spot. With reference to a protein database, our works have paved a ground-breaking way to nanogap-enhanced Raman identification and sequencing of single proteins.

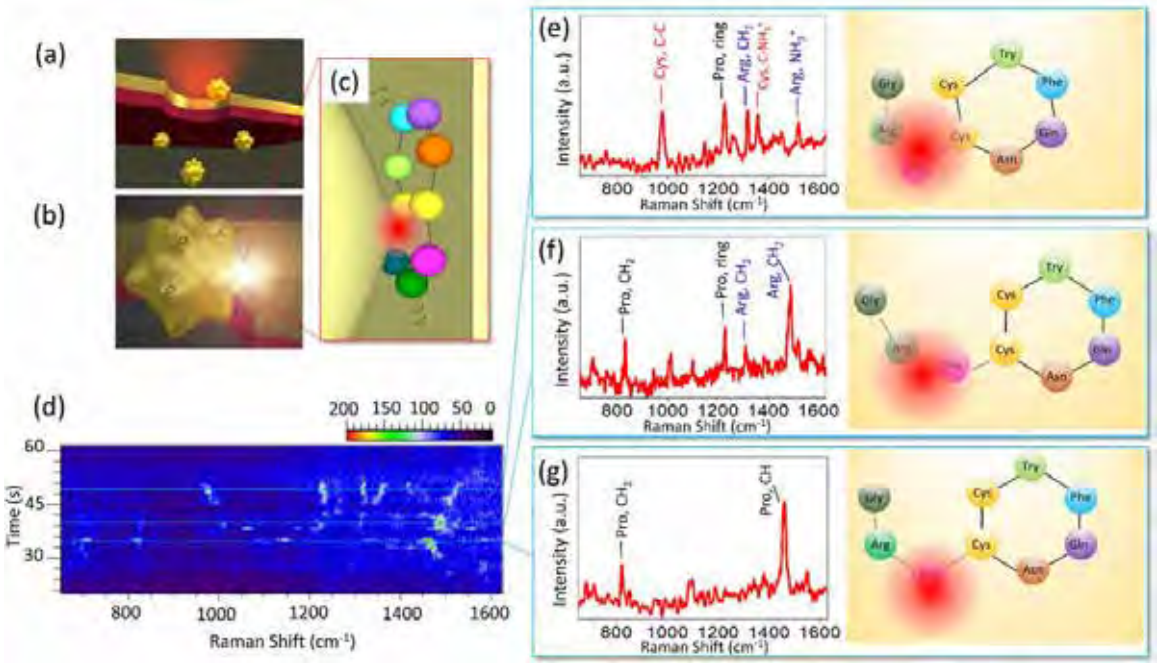

Figure 1. The plasmonic nanogap biosensor that detected single residues in single vassopresin peptide.

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Investigation of individual dust particle grains by optical tweezers for space applications

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Cosmic dust consists of small solid particles of silicon, irons, oxygen and magnesium, in the sizes range of microns to millimetres. Dust plays a dominant role in the Universe leading to the formation of galaxies, stars, planets and minor bodies and it is also the key to life's origins [1]. Extra-terrestrial dust characterisation by means of advanced analytical techniques can provide important information to understand processes occurring in different astrophysical environments, including the origins of life. Cosmic dust can be collected in space by spacecraft [2] and in the Earth's stratosphere by balloon-borne collectors [3]. Dust samples are mainly deposited on a substrate (apart few exceptions [4]) to analyse their physicochemical properties at terrestrial facilities by different techniques [5]. Conversely, the contactless manipulation of single dust particles prevents the shielding effects of surfaces or other particles. Standard and Raman Tweezers represent a powerful and an ideal tool to isolate single dust particle, to characterize their response to optical forces and to identify their mineral and organic compositions, particularly to search for biogenic signatures. [7–9].

We investigate cosmic dust samples from different meteorites by Optical Tweezers (OT) and Raman Tweezers (RT) [8, 9]. The scattered light by the trapped dust particle is analysed by a photo-detector providing information about optical forces on the trapped particle [8, 9]. This provides more accurate information to theoretical models for the calculation of radiation pressure for a variety of complex particles of astrophysical interest. Furthermore, by RT we identify minerals and organic composition of micron-sized individual meteorites fragments, having a better sight on the bench work where H_2 molecules and simple organic compounds, carbon monoxide and ammonia are produced [10]. We compare the Raman peaks of our samples obtained by RT with the Raman spectra of the components of DEW 12007 found in literature [11]. The agreements of our result with the literature documents the high potential and applicability of our high-resolution spectroscopic technique for the non-destructive and contactless analyses of planetary materials. Our results open medium term new perspectives for the investigation, in controlled laboratory conditions, of extra-terrestrial particles collected in

space and brought back to Earth by space probes (e.g. from Mars, Moon, asteroids, comets) and by balloon born instruments. On a longer-term frame, our efforts are addressed to space tweezers development for planetary space mission in situ applications.

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Detection of local magnetic fields with plasmonic nanoantennas

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The key concept of magnetoplasmonics is the possibility to alter the resonance conditions of a plasmonic system with the proper combination of light polarization and applied magnetic field [1]. Here we show that, using this principle, we can probe the local magnetic field generated in the vicinity of a plasmonic nanostructure by an assembled layer of cobalt ferrite nanoparticles (Figure 1).

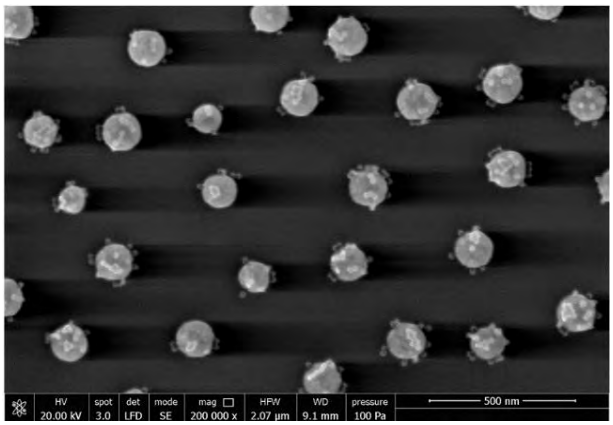

Figure 1. SEM image of magnetic NPs assembled on gold nanodisks.

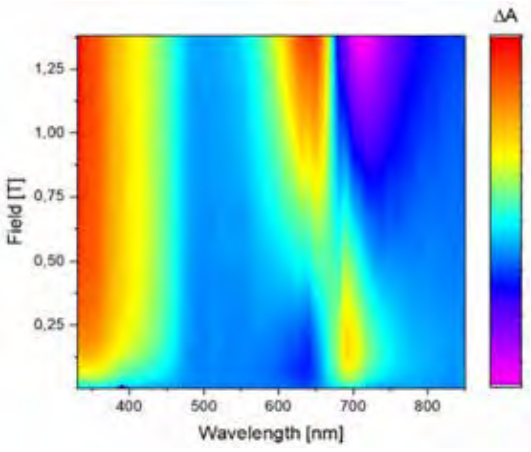

Figure 2. Field- and wavelength- dependent magneto-optical signal of the sample.

As in classical magnetometry, in a magneto-optical experiment cobalt ferrite nanoparticles exhibit a saturating behaviour with respect to an external magnetic field. Gold nanoantennas show a linear field dependence of their magneto-optical signal due to the field-induced splitting of circular magnetoplasmonic modes [4,5]. Differently from magnetometry, however, an additional degree of freedom is available in magneto-optics, i.e. the energy of the exciting photon. We can then take advantage of this distinction to separate spectroscopically the magneto-optical footprints of the two components (Figure 2).

With this procedure we were able to recover a plasmonic component of the signal (at 600-700 nm) with opposite sign to that of the applied magnetic field. This arises from the magnetized cobalt ferrite particles at the sides of the disk, whose dipolar field is opposite to the applied magnetic field (Figure 3).

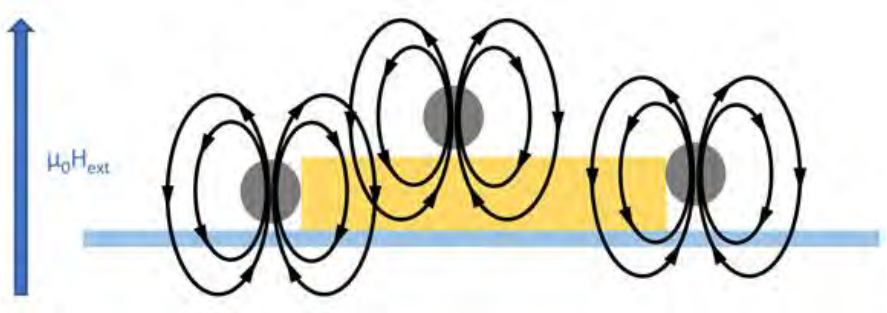

Figure 3. Sketch of the dipolar fields generated by magnetized particles around the nanoantenna.

We assume that the contribution of the particles standing on top of the disks is negligible because they are far from the hot spots of the plasmonic system. Such opposite field is directly detected and quantified as a magnetoplasmonic effect through the nanoantennas, which effectively act as a sensor of the local magnetic field around them. We believe that, with proper optimization, this method can be developed into an outstanding and powerful optical readout mechanism for magnetic field at the nanoscale.

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Understanding and controlling mode hybridization in multi-cavity optical resonators using quantum theory and the surface forces apparatus

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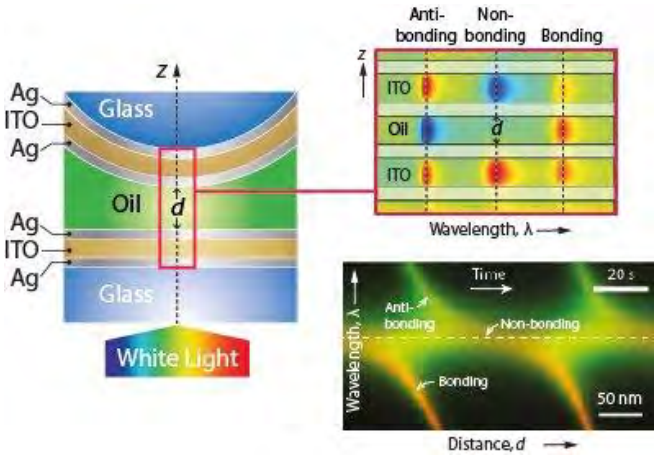

Optical waves in a metal-dielectric multilayers display quantum-like features, such as energy level quantization and avoided crossing, underpinned by an isomorphism between the Helmholtz and Schrodinger wave equations. This article builds on the fundamental concepts and methods of quantum theory to facilitate the understanding and design of multi-cavity resonators. It also introduces the surface forces apparatus (SFA) as a powerful tool for rapid, continuous, and extensive characterization of mode dispersion and hybridization. Instead of fabricating many different resonators, two equal metal-dielectric-metal microcavities were created on glass lenses and displaced relative to each other in a transparent silicone oil using the SFA. The fluid thickness was controlled in real time with nanometer accuracy from more than 50 μm to less than 20 nm, reaching mechanical contact between the outer cavities in a few minutes. The fluid gap acted as a third microcavity providing optical coupling and producing a complex pattern of resonance splitting as a function of the variable thickness. An electromagnetic wave in this symmetric three-cavity resonator emulated a quantum particle with non-zero mass in a potential comprising three square wells. Interference between the wells produced a three-fold splitting of the degenerate energy levels due to hybridization. The experimental results can be explained using the standard methods and formalism of quantum mechanics, including symmetry operators, perturbation theory, and variational method. Notably, the interaction between square wells produced bonding, anti-bonding, and non-bonding states, that are analogue to hybridized molecular orbitals and are relevant to the design of "epsilon-near-zero" devices with vanishing dielectric permittivity.

Reference: B. Zappone, V. Caligiuri, A. Patra, R. Krahne, A. De Luca, *ACS Photonics* 8 (2021) 3517–3525.

Elevator Pitch Talks

Nanostructured lipid membranes for enantio-discrimination of amino acids

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Amino acids are fundamental biological molecules that exist in L- or D-enantiomeric forms and play a key role in living systems. Chirality is a driving force in biomolecular recognition, and there is an increasing interest in exploring chiral preferences in the interaction and binding of biomolecules. In this study, we investigated the enantioselective recognition of D- and L-Tryptophan (Trp) via nano-structured lipid membranes. We performed a multi-scale analysis of the binding preference of Trp enantiomers with neutral zwitterionic dipalmitoylphosphatidylcholine (DPPC) and negatively charged dipalmitoylphosphatidylglycerol (DPPG) model membranes. The effects of the chiral Trp amino acid on the structure and dynamics of DPPC and DPPG multilamellar vesicles were examined by differential scanning calorimetry, electron spin resonance, attenuated total reflectance Fourier-transform infrared spectroscopy, and molecular docking.

The overall results revealed an association of the amino acid at the polar/apolar interface for both lipid bilayers. The interaction is mainly modulated by the formation of hydrogen bonds between the carbonyl group (C=O) and the amino acid side chain, as well as by hydrophobic contributions. However, the enantiomeric Trp specificity is driven by the nature of the lipid polar head. Such specificity is absent for neutral DPPC bilayers. In contrast, for the negatively charged DPPG membrane, the interaction with Trp is chirality-dependent.

The results indicate that lipid membranes can be used to perform chiral recognition of active biomolecules, such as amino acids. To make this enantio-discrimination more selective and specific, membrane composition can be suitably designed to maximize the interaction with one enantiomeric form. Such studies can pave the way for biomedical and pharmaceutical applications where the separation, recognition, and detection of chiral biomarkers/drugs is a necessity.

Machine learning enhanced calculations of optical forces in the geometrical optics approximation

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Optical tweezers [1,2], which trap and manipulate particles by highly focusing a laser beam, are a key tool in fields like physics, biology, or nanotechnology. Calculating the optical forces applied on a particle is essential to plan experiments and interpret their results. When the studied particles are larger than the incident wavelength, computing these forces by considering the geometrical optics approximation for the light propagation and the light matter interaction is often the most sensible approach [3]. However, the geometrical optics approximation comes with a compromise between computation speed and accuracy of the calculation. Considering more rays to discretize the light makes the calculation more accurate, however, it also takes longer to compute the exchange of momentum of all rays and therefore the total force applied by the rays on the particle. This lack of speed for accurate calculations limits the number of systems that can be numerically explored.

In this work we use machine learning to compute optical forces. We prove that it can be used to sidestep this compromise and improve not only the speed but also the accuracy of the calculation. This is first demonstrated for the simple case of a spherical particle in an optical tweezers and it is later expanded to include all the relevant parameters involved in basic optical tweezers experiments. Furthermore, this enhancement in the calculation allows us to carefully explore the dynamics of ellipsoidal particles in a double beam configuration, where we study the equilibrium points as a function of the aspect ratio (Fig 1).

We conclude that machine learning allows to improve the optical forces calculation in the geometrical optics approximation. Moreover, it opens the door to future applications of optical tweezers by exemplifying how it can allow to study systems that, due to computation limitations, were out of the scope of the conventional ray optics approach.

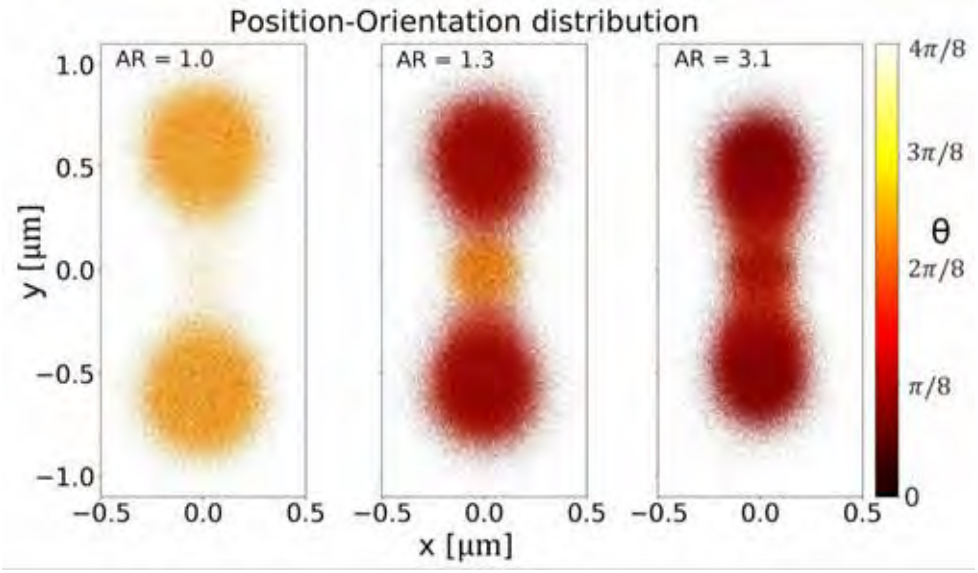

Fig 1: Position-Orientation distribution of an ellipsoidal particle in a double trap configuration at three different aspect ratios (AR). The location of the point in the plot accounts for the position of the center of mass in the x-y plane and the color indicates the particle angle with the z-axis (0 when perfectly aligned with the beam).

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Coexisting and Cooperating Light-Matter Interaction Regimes in Meta-Voltaic Systems

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Light-matter interaction is usually observed in a quantum framework where an atom or a molecule interacts with a resonant cavity. Their interaction is quantified through the amplitude of the perturbation induced in the optical response of the pristine systems (the atom and the cavity). In such systems, however, light-matter interaction occurs through a single feedback channel: an exciton relaxes by emitting a photon that is stored in the cavity for several roundtrips before being re-absorbed to create another exciton, and so on. [1] Very few has been said on the possibility for the excited system to relax through two different channels belonging to two different regimes. In this poster, we present the case in which the strong coupling and the photovoltaic regime cooperate to enhance the External Quantum Efficiency (EQE) of a photovoltaic cell specifically engineered to behave as an optical cavity tuned with the excitonic transition of the embedded active material ($CH_3NH_3PbI_3$ perovskite). We exploit the angular dispersion of the so-engineered photovoltaic cell to show that while the cavity mode approaches the energy of the exciton, the strong coupling regime is achieved and the EQE is significantly enhanced with respect to a classic configuration serving as a benchmark. Our findings demonstrate that polariton-based photovoltaics can overcome the performances of classic photovoltaics especially in critical cases like grazing-angle incidence. The proposed system constitutes also an effective toolbox to prepare and study a still vastly unexplored phenomenology associated to the coexistence of multiple light-matter interaction regimes in the same quantum system.

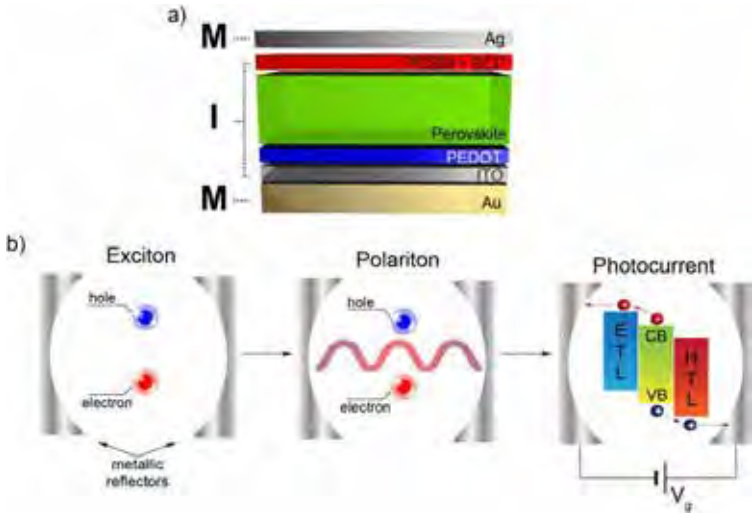

Figure 1: (a) Sketch of the Meta-Voltaic cell consisting in a perovskite-based photovoltaic cell embedded between two external metallic mirrors that constitute, at the same time, the two reflectors of a Metal/Insulator/Metal resonator and

the electrodes of the photovoltaic cell. (b) Sketch of the polariton-based photocurrent from the formation of the exciton to that of the polariton *via* strong light-matter coupling that sustain the polariton photo-current.

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Hierarchical fabrication of hybrid nanoparticles: a performing nano-matryoshka array

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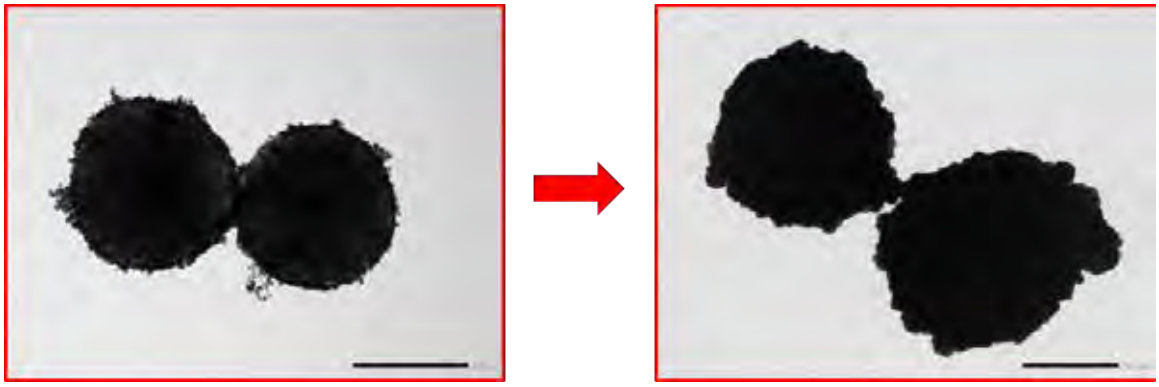
A metal-insulator-metal (MIM) nanostructure, stable in aqueous solution, was designed and synthesized using a seed-mediated growth approach from differently shaped gold cores.

In detail, the gold nanoparticles acted as a metal core on which an insulating layer (specifically silica) was grown. On the latter, in turn, a layer of gold was formed and, finally, another silica layer was added. Finally, a hybrid nano-matryoshka composed of gold/silica/gold/silica concentric shells was obtained and characterized by Uv-Vis spectroscopy and TEM.

Further studies were conducted on these nanomaterials made up of nanoparticles dispersed in solution to explore the interaction of the plasmonic properties with the photophysical features of emitting molecules. From this perspective, new application scenarios would open up for MIM materials.



Hyperbolic Metamaterials via Hierarchical Block Copolymer Nanostructures

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Hyperbolic metamaterials (HMMs) show extremely exotic optical properties that are unattainable in natural materials. HMMs are composed of metal and dielectric, indeed they show very appealing characteristics for what concerns their photonic properties due to their strong electrical permittivity anisotropy. Because of the anisotropy in the optical behavior a hyperbolic isofrequency surface is generated and it theoretically leads to two interesting features: a directional emission and an infinity local density of optical states (LDOS). This manifest itself in an enhancement of the spontaneous emission for a photon source coupled with the metamaterial [1-3]. Since most room-temperature photon sources are limited by their emission rate and broadened emission spectrum, e.g. NV centers [4], the enhancement of spontaneous emission opens up different opportunities to overcome photon source limitations. In this work we present a fabrication method to obtain HMMs with an in-plane optical axis, desirable for different applications related to nanophotonics. An Au/air HMM with a lamellar (40 nm period) structure is obtained through the exploitation of hierarchical self-assembly of block copolymers induced by a dewetting process. By exploiting the blend film instability over topographically defined substrates, see figures 1.a−1.b, droplets composed of highly ordered lamellar nanostructures in hierarchical disposition can be obtained [5]. The pattern transfer process onto a flexible substrate creates an Au/air HMM with hyperbolic dispersion in a broad wavelength range in the visible spectrum.

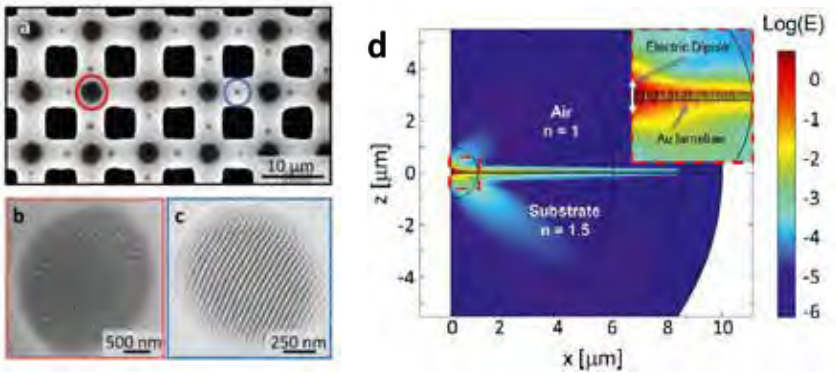

Fig. 1. **a)** SEM image of BCP blend dewetting over a large-area substrate. (**b,c**) SEM images of lamellar nanostructured droplets in a single grain configuration related to highlighted areas in figures 1.a. **d**) Magnitude of the electric field in the near field for $\lambda = 594$ nm with the actual metamaterial. The inset shows a zoom of the region marked by the dashed red line. The white arrow in the inset represents the orientation of the dipole

To estimate the optical behavior of the proposed structure we simulated with the software Comsol Multiphysics 5.3 the optical behaviour of an HMM coupled to a photon

source (an electric dipole in the model). We have computed a frequency sweep to estimate the electric field distribution in the near field and the Purcell factor (see Fig 1.a), estimated as the ratio between the total emitted power by the dipole when is placed 5 nm above the HMM and in an air domain. Moreover, we analyzed the behavior of the Purcell factor varying the height of the HMM. As shown in Fig. 2.b the peak position and value change drastically with a few nanometers of variation. Moreover, we obtained a strong reduction in the measured fluorescence lifetime of NV centers in nanodiamonds placed on top of the HMM. This measurement is compatible with the computed Purcell factor estimated for structures with 70 nm height, which is equal to 32 at $\lambda = 580$ nm (see figures 2.a and 2.b).

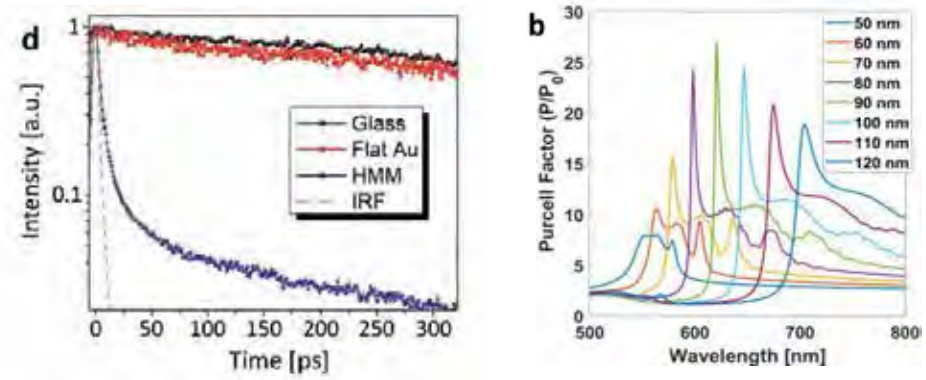

Fig. 1. **a)** Lifetime fluorescence measurements for NV centers in nanodiamonds in different conditions: above the glass (black), flat Au (red), and HMM (blue) **b)** Simulated Purcell factor for different HMM heights with the actual metamaterial.

To conclude, we have presented a fast and low-cost method to obtain hybrid metal/dielectric nanostructured metasurfaces with an in-plane optical axis whose spectral response can be further tuned.

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Non-local effects of nanoscopic field confinement in polaritonic systems

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Nanophotonic structures confine electromagnetic radiation below the Abbe diffraction limit by storing part of the electromagnetic energy into kinetic energy of moving charges [1]. The employment of semiconductors and two-dimensional materials with a plasma frequency tunable via carrier concentration has allowed to observe plasmonic excitations in the Mid-IR frequencies range and to achieve an extreme confinement of the electromagnetic field in nanostructures [2]. The propagative nature of charge excitations in nanophotonic devices cannot be neglected below certain length scale, leading to the emergence of non-local effects. In this regime the nanoscopic features confining the charges act as a diffraction grating allowing the electromagnetic field to couple to high-momenta plasmonic resonances, which act as loss channel reducing the field confinement.

In Ref. [4] we theoretically and experimentally investigated the emergence of non-local effects in Landau polaritons platforms, where the photonic field of a sub-wavelength metamaterial LC resonator, fabricated on top of a two-dimensional electron gas (2DEG), couples to collective electronic excitations dressed by the magnetic field (magneto-plasmons). In such a system, the reduction of the photonic mode volume can lead to a higher coupling strength [3]. Our finding is that as we force the subwavelength confinement of the electromagnetic field of a nanogap resonator below a certain gap size threshold, we observe the generation of a continuum of propagating high-momenta magneto-plasmons to which the photonic field couples. This determines not only a reduction of the effective polariton splitting due to a limitation of the field enhancement, but also a progressive disappearance of the upper polariton (UP) branch as a result of the plasmonic loss channel.

Our theory shows that discrete-to-discrete models underlying the polaritonic framework, such as the standard Hopfield model, are no longer valid below the threshold gap size, and the development of a novel discrete-to-continuum bosonic model was necessary to predict the main non-local effects on the polariton features. We thus developed a multimode theoretical model able to provide analytical formula for the Gauge-invariant observables, taking into account both the light-matter coupling of the photonic mode to the continuum of the propagative plasmonic modes and any possible reservoir where the photonic and matter excitations can get lost [5].

As showed in Fig.1, while above the resonator gap threshold the standard Landau polariton model displays the two polaritonic branches for any value of the magnetic field, the extreme photonic confinement due to the narrower gap dramatically changes the nature of the light-matter system and the narrow polaritonic resonances broaden and vanish into the continuum region. Experiments and finite-element simulations support this picture, demonstrating multiple novel nonlocal polaritonic features.

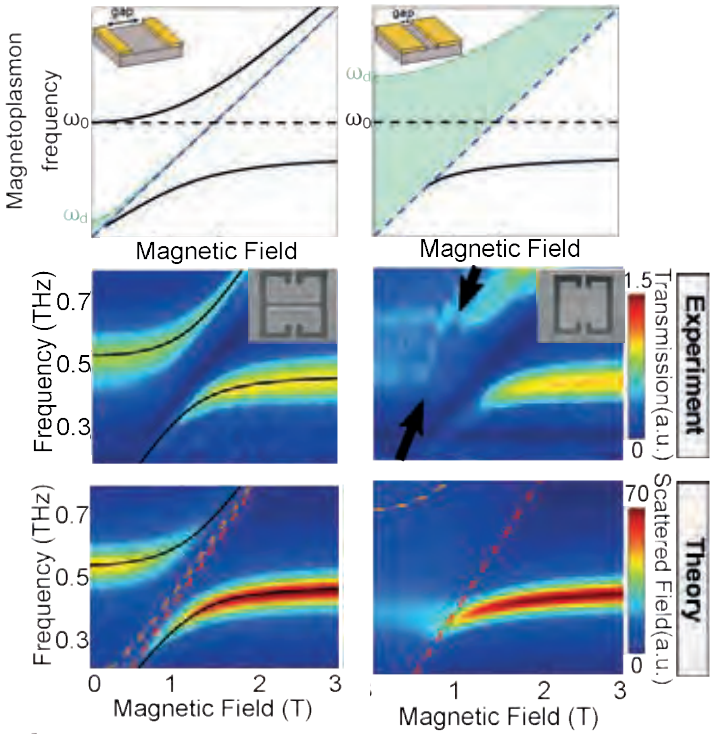

Figure 1: Top row: the continuum of magneto-plasmonic energies (shaded green region) as a function of the magnetic field for a wide (left) and narrow (right) gap size. Medium row: THz time domain spectroscopy transmission measurement for metallic complementary split-ring resonators with 4 μm (left) and 250 nm (right) gap sizes coupled to the electronic modes in a GaAs/AlGaAs QW. Resonance frequency is at 500 GHz and black lines are fitted polariton branches using Hopfield model. The insets show SEM pictures of the resonators with wide and narrow gaps. Bottom row: calculated scattered field for different gap feature sizes, d (same as the gap sizes of the experiment results).

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Modulating plasmonic nonlinearities through surface charge depletion in heavily doped semiconductors

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Within the background of near- and mid-infrared plasmonics based on heavily doped semiconductors [1], we have recently shown that free-electron (FE) third-harmonic generation (THG), when coupled to plasmonic enhancement, could be up to two order of magnitude bigger than conventional semiconductor nonlinearities [2]. Here, we apply a hydrodynamic perturbative approach within the Thomas-Fermi approximation [2, 3], with the purpose of investigating the impact of surface charge depletion on the FE nonlinear response of heavily doped semiconductor. In this context, the optical response of the FE fluid under the influence of external electric- and magnetic- fields $\mathbf{E}(\mathbf{r},t)$ and $\mathbf{H}(\mathbf{r},t)$, can be modeled through the following constitutive relation:

$$\ddot{\mathbf{P}}+\gamma\dot{\mathbf{P}}=\frac{n_0e^2}{m}\mathbf{E}+\beta^2\nabla(\nabla\cdot\mathbf{P})-\frac{1}{3}\frac{\beta^2}{n_0}(\nabla\cdot\mathbf{P})\nabla n_0+\mathbf{S}_{\mathrm{NL}}. \tag{1}$$

where $\mathbf{P}(\mathbf{r},t)$ is the polarization field, n_0 is the equilibrium charge density, and time derivatives are expressed in dot notation. Here, $\beta^2=\frac{10}{9}\frac{c_{\mathrm{TF}}}{m}n_0^{2/3}$, with $c_{TF}=\frac{\hbar^2}{m}\frac{3}{10}(3\pi^2)^{\frac{2}{3}}$, while $\mathbf{S}_{\mathrm{NL}}^{(2)}$ and $\mathbf{S}_{\mathrm{NL}}^{(3)}$ are the second- and third-order nonlinear sources, whose expressions are:

$$\begin{aligned}\mathbf{S}_{NL}^{(2)}=&\frac{e}{m}\mathbf{E}\nabla\cdot\mathbf{P}-\frac{e\mu_0}{m}\dot{\mathbf{P}}\times\mathbf{H}+\frac{1}{en_0}(\dot{\mathbf{P}}\nabla\cdot\dot{\mathbf{P}}+\dot{\mathbf{P}}\cdot\nabla\dot{\mathbf{P}})-\frac{1}{en_0^2}\dot{\mathbf{P}}(\dot{\mathbf{P}}\cdot\nabla n_0)\\&+\frac{1}{3}\frac{\beta^2}{en_0}\nabla(\nabla\cdot\mathbf{P})^2-\frac{1}{9}\frac{\beta^2}{en_0^2}(\nabla\cdot\mathbf{P})^2\nabla n_0,\end{aligned} \tag{2a}$$

$$\begin{aligned}\mathbf{S}_{NL}^{(3)}=&-\frac{1}{e^2n_0^2}\Big[\nabla\cdot\mathbf{P}(\dot{\mathbf{P}}\nabla\cdot\dot{\mathbf{P}}+\dot{\mathbf{P}}\cdot\nabla\dot{\mathbf{P}})+\dot{\mathbf{P}}\cdot\dot{\mathbf{P}}\nabla\nabla\cdot\mathbf{P}\Big]\\&+\frac{2}{e^2n_0^3}(\nabla\cdot\mathbf{P})\dot{\mathbf{P}}(\dot{\mathbf{P}}\cdot\nabla n_0)-\frac{1}{27}\frac{\beta^2}{e^2n_0^2}\nabla(\nabla\cdot\mathbf{P})^3+\frac{4}{81}\frac{\beta^2}{e^2n_0^3}(\nabla\cdot\mathbf{P})^3\nabla n_0.\end{aligned} \tag{2b}$$

where m is the electron effective mass, e the elementary charge (in absolute value), μ_0 is the magnetic permeability of vacuum and γ is the damping rate. Here, contributions proportional to ∇n_0 tackle the non-zero gradient of the equilibrium charge density.

Eqs.(1-2) can be solved numerically, assuming a time-harmonic dependence of the fields [2]. Results in the case of a TM plane wave (at a fundamental field wavelength $\lambda_{FF}=12\ \mu m$) impinging on a semi-infinite semiconductor slab are summarized in Fig.1. We take into account both cascaded (i.e. due to second-harmonic signals) and direct FE THG. A measure, ζ, of the

enhancement caused by the surface depleted charge density has been calculated normalizing all the efficiencies to that obtained for a constant $n_0 = n_b$. We predict a one order of magnitude increase of the efficiency of FE THG for an equilibrium charge density 25 smaller than that in the bulk. This enhancement factor can grow up to two orders in correspondence with the plasmonic resonance of a nanopatterned structure.

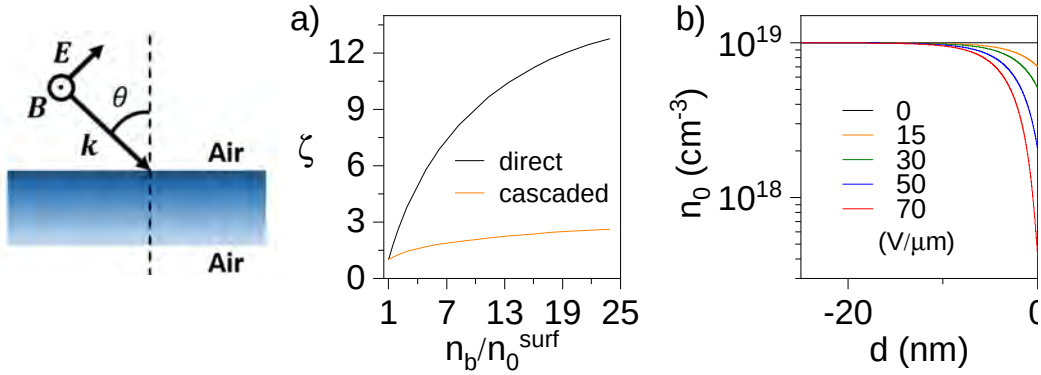

Figure 1: (a) THG efficiency enhancement factor ζ, for $\theta = 60°$, as function of the depletion factor n_b/n_0^{surf}, in the case of FE cascaded and FE direct THG from a semiconductor slab (shown in the schematics on the left), where n_b and n_0^{surf} are the values of n_0 in the bulk and for $d = 0$, respectively; (b) equilibrium charge density n_0 as a function of the distance d from the surface of the slab for different boundary conditions E_0^{surf}, this being a static bias applied on the surface. We calculated the surface depleted n_0 curves taking into account bands bending in doped semiconductors within the parabolic band approximation [4, 5]. The material considered is indium phosphide, which, assuming $n_b = 10^{25}\ m^{-3}$, has a screened plasma wavelength in the mid-infrared ($\tilde{\lambda}_{\text{p}} = 9\ \mu m$)

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Preparation and characterization of macroscopic, two-dimensional gold nanocubes plasmonic films

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Self-assembly at the water-hexane interface is one of the simplest and cheapest methodologies to obtain ordered closed-packed gold nanoparticles monolayers that show improved optical and electrical properties than stand-alone nanoparticles induced by strong plasmonic coupling. [1] Plasmon coupling consist in the combination of plasmonic modes of two or more nanoparticles when these are in close proximity each other. The combination of electric fields results in a redshift of the plasmonic band and an increase of the near field in the interparticles gap. Among the myriad of gold nanoparticles possible shapes, the cubic-one with their six planar facets and a strong plasmonic field localized on their sharp edges, are ideal candidate as building blocks for the preparation of ordered 2D monolayers. [2]

In this work, gold nanocubes (AuNC) were synthesized by a seed mediated growth technique [3], assembled in close-packed 2D array at water hexane interface, using ethanol to destabilize the colloidal aqueous dispersion and 1-dodecanethiol to promote in-situ nanoparticles ordering [4], and can be deposited both on ITO and glass substrates using a drain-to-deposit approach after the hexane evaporation. [5] The obtained films were characterized by UV-Vis spectroscopy, electronic microscopy (SEM), and X-Ray diffraction. Low magnification SEM image (Figure 1a) shows a film formed by islands of AuNC with size between 5 and 20 μm. Into these islands gold nanocubes form a dense monolayer with very few void and overlaps (Figure 1b). Uv-Vis spectrum is high representative of the film morphology, showing a plasmon band strongly enlarged and red-shifted than that of starting AuNC aqueous dispersion, due to coupling between nanoparticles (Figure 1c). These optical properties, united to the macroscopic size of the obtained deposition (Figure 1d), make these films good candidate as substrates suitable in metal-enhanced fluorescence and photothermal devices.

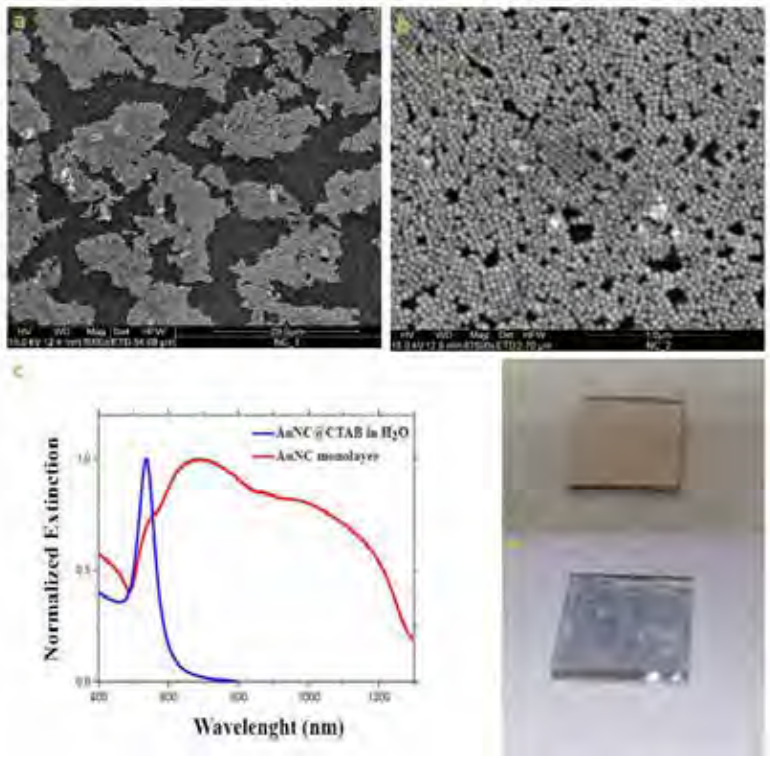

Figure 1: Representative SEM image at low (**a**) and high (**b**) magnification of a typical gold nanocubes self-assembled monolayer. Extinction spectra (**c**) of AuNCs monolayer on ITO glass (red line) and in aqueous dispersion (blue line). Optical image of a self-assembled monolayer of gold nanocubes deposited on ITO glass showing a gold coloration if observed in reflectance mode (**d**), and a dark blue coloration if observed in transmittance mode (**e**).

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Magnetoplasmonics beyond metals: Transparent Conductive Oxide Nanocrystals for High Performance Sensing

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Active modulation of the plasmonic response of nanostructures with an external stimulus enables the dynamic control of light propagation at the nanoscale, improving the performance of plasmonic sensors or optical modulators. With respect to other external tools employed in active plasmonics, magnetic field presents several advantages: it is a fast stimulus, easy to implement in devices, and its action on free charge carriers is fully reversible as it does not damage or modify the plasmonic material. However, achieving large magnetic modulation of the plasmonic resonance while maintaining a sharp optical response represents a great challenge in material choice for magnetoplasmonics. Indeed, noble metal NPs have sharp optical resonances, but weak magneto-optical signal, proportional to the cyclotron frequency (ω_C) [1]; on the other hand, nickel ferromagnetic nanodisks [2] or hybrid bimetallic nanostructures [3] have large magnetic modulation, but suffer from the high optical losses due to the magnetic metal, thus broadening the plasmonic resonance.

To overcome such limitations, we propose a paradigm shift in material choice by employing transparent conductive oxides (TCO) NPs, which are able to support a plasmonic resonance in the infrared. The carrier density in these NPs can be modulated in the range 10^{18}-10^{21} cm^{-3} by controlling the amount of aliovalent dopant introduced in the host semiconductor. Among TCOs we synthesized Sn-doped In_2O_3 (ITO) and F- and In-co-doped CdO (FICO) colloidal NPs, revealing a 20-fold and 40-fold enhanced magnetoplasmonic response compared to Au NPs. The enhancement is ascribed to the reduced effective mass (m^*) of free carriers in such TCOs with respect to most metals, which in turn boosts the cyclotron frequency. FICO NPs are characterized by a sharper resonance, which increases the magnetoplasmonic response with respect to ITO, while having a comparable m^* [4].

Finally, by using FICO NPs in a proof of concept magnetoplasmonic refractometric sensing experiment (Figure 1) we achieved a superior refractive index sensitivity with respect to metal-based magnetoplasmonic systems reported in the literature [1-3]. Remarkably, our approach is competitive with the current state of the art of plasmonic refractometric sensing employing extinction spectroscopy [5], with the advantage of not requiring complicate curve fitting.

Considering that non-magnetic TCOs have been used in this work, further enhancement of the magnetoplasmonic response is potentially achievable by introducing magnetic dopants in plasmonic TCO NPs, exploiting the interaction between free charges and the introduced dopants. Such carrier-mediated magnetic interactions have been reported in

the literature for several magnetically doped TCO films at low carrier concentration regimes [6]. If successful, the use of magnetic TCOs can represent a dramatic innovation in magnetic field modulated active plasmonics.

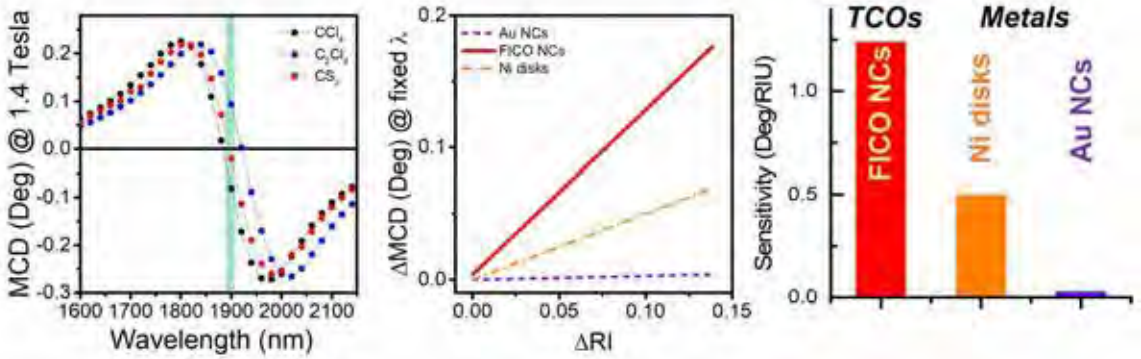

Figure 1: a) Magnetic Circular Dichroism of F^- and In^{3+} doped CdO NCs in three different solvents; b) magneto-optical signal at fixed wavelength as a function of the solvent refractive index (RI); c) derived refractive index magneto-optical sensitivity for FICO and ITO NCs compared with state of the art magnetoplasmonic nanostructures.

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Non-linear Terahertz Driving of Plasma Waves in Layered Cuprates

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Plasmon are among the fundamental collective excitations of quasi-free electron systems. A benchmark example is the one of isotropic metals, where they emerge as quantized oscillations of the conduction electrons. Plasmons can also arise, as oscillations of the superfluid electrons, when a metal undergoes a superconducting (SC) transition. In such case they are related to phase fluctuations of the SC order parameter: indeed, in a superconductor the quantum phase of electrons is the variable conjugate to the density, and the sound-like propagating phase mode typical of a neutral superfluid is converted into gapped plasma oscillations due to the Coulomb repulsion between electrons. For this reason plasmons appear in the spectrum of the phase mode, which can be determined by deriving either the equation of motion or the quantum action for phase fluctuations.

From an experimental point of view, plasmons are invisible, due to their longitudinal character, to most of the conventional electromagnetic probes used in linear spectroscopy, which have, instead, a purely transverse nature. Nevertheless, their excitation has been recently achieved, by means of non-linear Terahertz (THz) spectroscopy, in layered cuprates superconductors. These systems host arrays of stacked 2D SC layers which are coupled by a weak Josephson interaction, which pushes the energetic cost of an inter-layer Josephson-plasma mode (JPM) down to the THz range; on the other hand, the high intra-layer mobility allows the existence of an high-energy in-plane plasmon. Therefore, cuprates show a strong anisotropy along the direction orthogonal to the SC planes, thus providing a wide spectrum of excitations, ranging from few THz to the eV. The low-energy JPM can be then investigated by means of THz radiation: in particular, the use of intense THz out-of-plane polarized electric fields in cuprates has led to the observation, in the SC state, of non-linear optical phenomena, such as third-harmonic generation (THG) [1] and pump-probe oscillations [2], which undergo a strong amplification at the frequency of the JPM, and are therefore interpreted as consequences of the non-linear excitations of the latter.

From a theoretical point of view, the spectral properties of the JPM at low temperature, both in the linear and in the non-linear regime, can be well understood within the context of the classical equation of motion for the phase degrees of freedom [3]. The same approach, however, cannot provide reliable predictions on the full thermal evolution of the out-of-plane THG/pump-probe signal, from zero temperature to the critical one: at higher temperature, indeed, quantum effects become relevant and such classical approach breaks down. A full quantum treatment is

therefore needed. To this purpose we provided, in a recent work, a complete quantum description, within the effective-action formalism, of the JPM contribution to the non-linear response of cuprates below the critical temperature, focusing on both THG and pump-probe protocols [4]. We showed that the main mechanism behind the non-linear driving of the JPM consists in a zero-momentum photon exciting simultaneously two JPMs with opposite momenta. We then repeated the same analysis for the in-plane plasmon, since recent experiments, performed in the last two years, suggest that also strong in-plane-polarized THz pulses can induce important non-linear effects in cuprates [5][6]. The latter effects have been discussed so far only within the context of the SC Higgs mode, which is linked to amplitude fluctuations of the SC order parameter and whose energy lies in the THz range in cuprates, thus appearing as a better candidate than the high-energy in-plane plasmon; nevertheless, the observed monotonic temperature dependence of the non-linear response does not match the expectations for the Higgs mode [5][6]. In the final part of the talk, I will show how our theoretical predictions well reproduce the experimental findings in the case of both out-of-plane and the in-plane polarized external fields: in the out-of-plane case, where the plasmon energy lies in the THz range, there is an high thermal probability of exciting two plasmons, which causes the non-linear response to vanish only in the close proximity of the critical temperature. On the other hand, in the in-plane case, the plasmon energy lies far beyond the THz range, thus the resonant excitation of two plasmons is only attained very close to the critical temperature, where the signal is already suppressed by the vanishing of the superfluid stiffness: this makes the non-linear response monotonically decreasing towards the critical temperature, in perfect agreement with the experiments [5][6].

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Photonic and Plasmonic Multilayer Metamaterials with Tunable Properties Based on Alternative Plasmonic Materials

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The raising need to extend the excitation range and to tailor the optical characteristics of plasmonic metals has pushed the research towards alternatives (doped semiconductors, e.g. Transparent Conductive Oxides TCOs, or transition metal nitrides, e.g. TiN) which lay the foundations of a new class of artificial metamaterials exhibiting novel functionalities. Multi-phase nanocomposites are fascinating because their building blocks assembled in multilayer configuration can accomplish "effective" properties unattainable in independent components [1]. For instance, hyperbolic metamaterials present unique electromagnetic phenomena (high-k modes) activated by the anisotropic permittivity resulting from the periodic alternation of conductors and dielectrics of subwavelength thicknesses [2]. When increasing the characteristic dimension of layers, Bragg's reflections start to appear and one-dimensional (1D) photonic crystals can be achieved straightforward [3]. Playing with material selection and device geometry is a key point for engineering efficiently plasmonic characteristics as well as light propagation within the nanostructure. Besides, control over nanoscale morphology, structure and electrical performance is fundamental for designing next-generation devices, with ad-hoc behavior for targeted applications in optoelectronics, photonics or bio-sensing [4]. At present, multilayers consisting of noble metals (Au, Ag) and oxide-based dielectrics (In_2O_3, Al_2O_3), known to display optical features in the visible spectrum [5], have been widely investigated.

In this work, multilayer meta-structures have been developed with a less-explored approach, involving alternative materials (nitrides, TCOs) and original design routes (via pulsed laser deposition). The final goal is to explore electrical/optical responses resulting from unusual combinations of materials and properties (transparency, conductivity, tunable VIS-IR plasmonics, active modulation).

For instance, novel TiN-TiO_2 plasmonic multilayers have been realized as hyperbolic metamaterial in the visible (**Figure 1a**), with added advantages determined by the good material affinity and the versatility of TiO_2 as a wide-band gap semiconductor. In addition, the resulting features can benefit from the refractory nature and CMOS compatibility of TiN, along with the possibility to modify the plasmonic response through stoichiometry. Then, original transparent conducting multilayers based on the less-explored Tantalum-doped TiO_2 (Ta:TiO_2) TCO [6] (**Figure 1b**) have been obtained directly in a simple one-step synthesis, by alternating conductive (compact) and dielectric (nanoporous) layers of the same TCO achieved by varying the deposition pressure (1-6 Pa O_2). With a material science perspective, structural and electrical properties have been optimized to control optical/plasmonic outputs as a function of deposition conditions, doping content and geometrical parameters. The overall crystalline structure and electrical performances are ruled by the interplay of single features of metal-like and dielectric units, showing good TCO performances (electrical resistivity $<10^{-2}$-10^{-3} Ωcm). Future applications are foreseen as hyperbolic platforms in the IR, while 1D photonic crystals can be accomplished by customizing compact/porous fraction and dimension of the layers. The proper

optimization of material properties leads to an intense photonic band gap, spanning from green to red wavelengths in the visible, which in turn can be actively modulated with an external bias (**Figure 1c**).

Paramount importance should be given to the inherent tunability of optical/plasmonic properties achievable in the aforementioned systems, where the possibility to adjust the carrier concentration in alternative conductors (directly at synthesis or with an "active" approach) is unattainable in metals due to the fixed carrier density (i.e. fixed plasma frequency). Moreover, these multifunctional meta-devices are application-oriented and possess a cross-disciplinary attitude, thus are appealing as TCO-based electrodes, nano-architectures for sensing/thermo-photovoltaics or optoelectronic elements for active manipulation of colors.

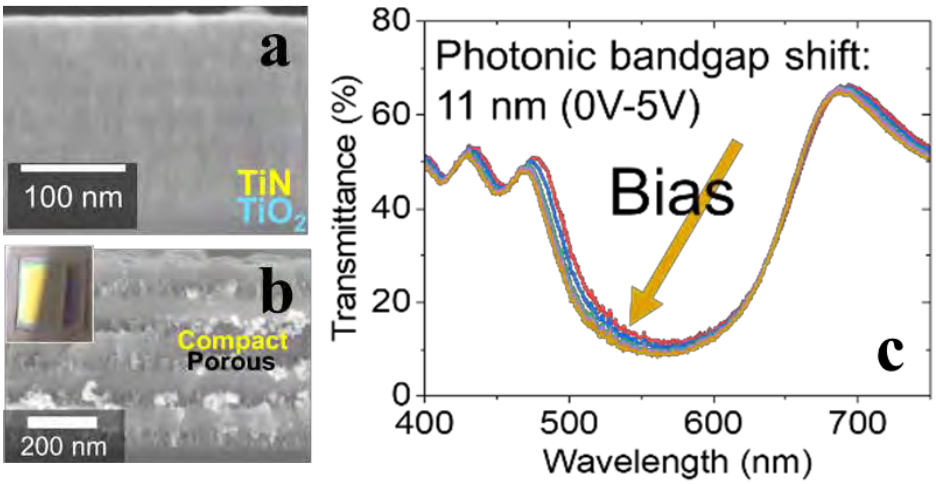

Figure 1: SEM images of TiN-TiO_2 multilayer (a), Ta:TiO_2 porous/compact multilayer (b); (c) transmittance spectrum of a Ta(5%):TiO_2-based photonic crystal with external bias applied.

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Open cavities for molecular strong coupling

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Abstract We report on the modification of photoluminescence from within a dielectric slab cavity supported by a metallic/dielectric substrate. Dielectric slab cavities have been studied in the past in the context of 'self' strong coupling. However, the modification of photoemission has not yet been reported. We show that a dielectric slab placed on silicon substrate does not show any significant modification of the photoemission, whilst a cavity on a gold substrate does.

Introduction

Strong coupling involving ensembles of organic molecules is an important light−matter interaction that has seen a dramatic increase in research activity in the last decade. Placing ensembles of organic molecules that possess an optically active transition inside a confined light field, such as an optical microcavity, may lead to a phenomenon known as strong coupling. Here, new hybrid states called polaritons are created that are part molecule, part light. [1, 2] However, directly access to the coupled molecules in such closed cavity system is a serious limitation. In contrast, as open cavities provide direct access to coupled molecules and are thus becoming more attractive. Recently, there have been reports showing strong coupling in planar open cavities. [3, 4] These studies of open cavities have mainly focused on exploring the reflection, transmission, and absorption of incident light. Here, we focus on examining modifications to photoluminescence (PL), a more stringent mechanism. To do so, we made use of three different cavity structures in our study. These were: (a) an open dielectric cavity: polymer dye molecules spun on a silicon substrate; (b) a semi-metallic cavity: a thin gold mirror was deposited onto silicon substrate and polymer dye molecules were spun on top of the gold, and (c) a closed metallic cavity: polymer dye molecules were placed inside two metallic mirrors.

To observe the modification in PL for all the three cases, we compared experimentally measured PL spectra of the different cavities. The figure shows PL spectra taken at zero incident angle for all three cases. Hardly any modification in the PL signal can be observed for the open dielectric cavity, shown in black. However, a modification in the PL spectrum is observed when a thin gold film was included between the substrate and the polymer, red. Finally, in the closed metallic cavity, emission from both the uncoupled and the lower polariton can be observed in blue.

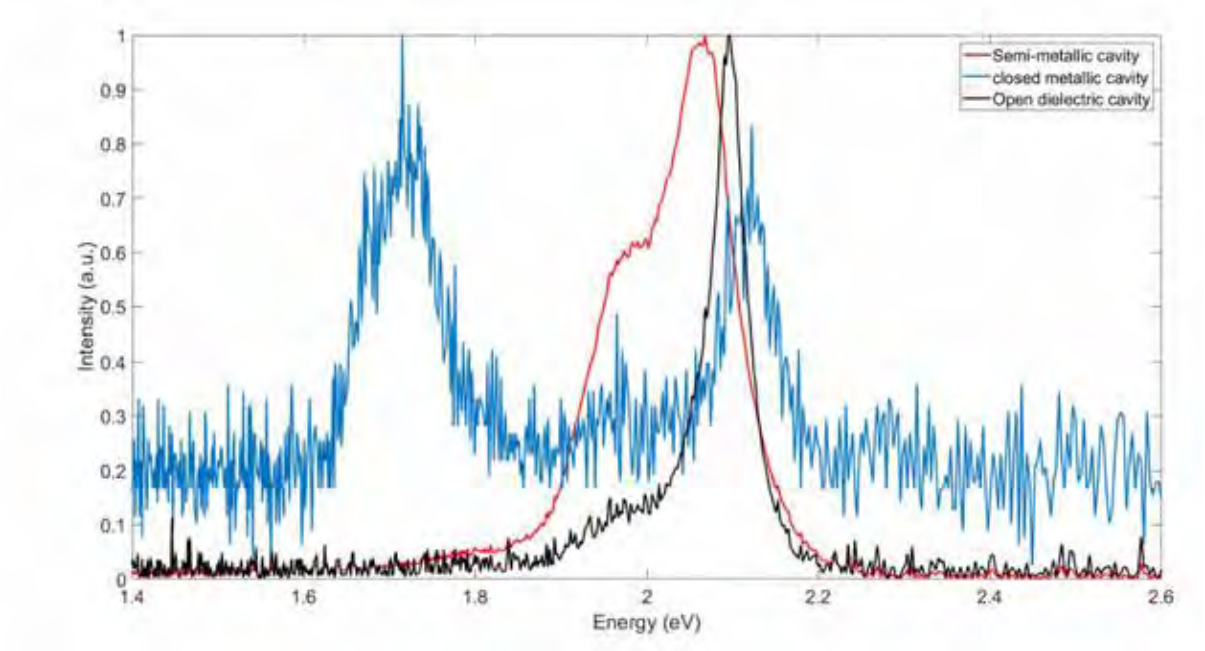

Figure: PL comparison between dielectric, semi-metallic and metallic cavity is shown at zero incident angle in black, blue and brown respectively. The resonance in the dielectric cavity occurs due to uncoupled TDBC molecules in black. However, the modification in the PL spectra is observed when thinner gold film was added underneath in red. In the closed cavity, both the uncoupled and the lower polariton is observed in blue.

Conclusion We have shown that modification of the photoluminescence coupled be achieved through open cavities by simply putting thin metallic film underneath. The modification in PL provides ample opportunities to understand behaviour of coupled molecules in open systems.

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Tunable Polymeric Metalenses

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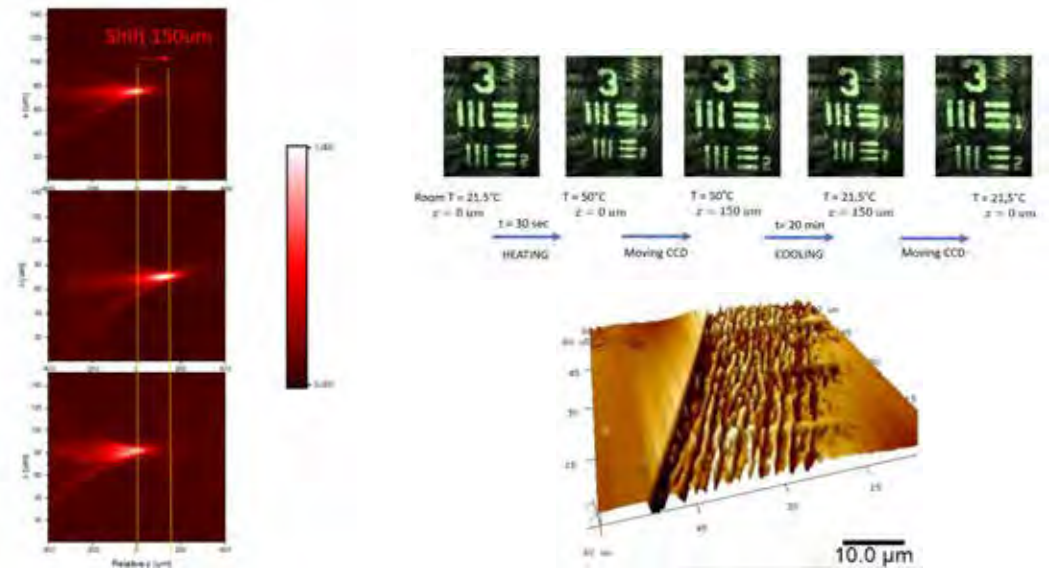
Metalenses have emerged as an innovative technology with functionalities that overperform conventional refractive lenses owing to compact footprint and subwavelength resolution. Despite the unique optical features, the fabrication of these nanostructured lenses is expensive and time consuming. Here, we report about the fabrication and the characterization of scalable, low-cost, flexible, and tunable metalenses for visible frequencies. The material of such metalens consists of a polymer matrix, easily fabricated via direct pattern replication from a master copy. In addition, thanks to liquid crystal (LC) inclusions, the lens shows tunable features. We report the all-optical reconfigurability of the dielectric metalens by the thermal induced changes of the local refractive index. The optical response of the metalenses undergoes a modification owing to the LC phase transition that can be easily induced by external stimuli. We have obtained a continuous and reversible tunability of the metalens focal length with a variation up to 150 μm.



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Ultrafast optical response of Titanium Nitride films: disclosing hot-electron relaxation processes

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Titanium nitride (TiN) has raised a lot of interest in the last decades due to its refractory nature, CMOS- and bio-compatibility and chemical stability. From the optical point of view, its intriguing properties of tunable permittivity when varying synthesis conditions and broadband plasmonic resonance, brought it in the spotlight as alternative plasmonic material. When compared to gold, it shows lower interband losses in the visible range [1], [2]. While the static optical response of TiN has been broadly investigated, a clear and complete description of the transient optical properties is under debate, especially for what concerns the timescale of carrier-phonon interactions following the excitation [3]–[6].

In this work, we studied the ultrafast dynamics of optically thick (200 nm), compact TiN films fabricated by pulsed laser deposition on soda lime glass, using pump-probe spectroscopy with temporal resolution down to ~15 fs; with tunable pump and broadband probe, extending over the plasmon resonance. The experimental analysis is supported by numerical simulations based on a two-temperature model (TTM) implemented with a dispersed contribution for the modulation of interband transitions. This complete proposed model allows to clarify the process of relaxation.

Specifically, by considering a low energy transition at the gamma point and an electron–phonon coupling coefficient 50 times higher than in gold [6], we were able to reproduce with a notably high degree of accuracy the pump-probe experimental data (*Figure 1a-d*). The electron–electron scattering takes place right after the interaction with the pump, for about 100 fs (initial signal rise). The ultrafast decay of the signal is the footprint of the high electron–phonon coupling in this material. The electron cooling through scattering with phonons is very rapid indeed, taking place in about 200 fs (~1 ps in gold [7]). The slow increase of the signal right after the initial peak is caused by the mechanical oscillations of the film's optical skin layer, induced by the heat generated during photoexcitation.

Our findings lead to a deeper understanding of the physics of this material, and appear to be of crucial importance in device design for plasmonics, thermophotovoltaics, hot carrier generation and nonlinear optics applications.

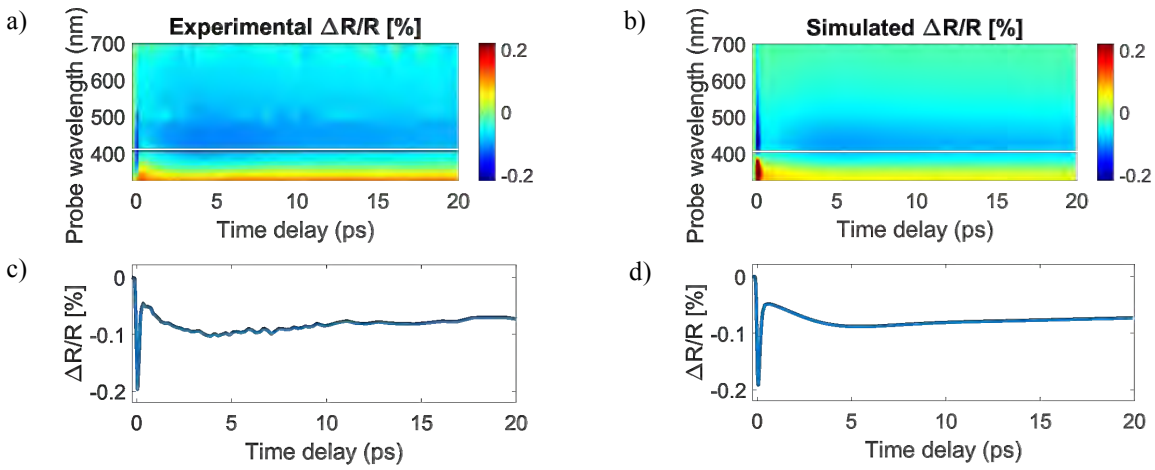

Figure 1: a) Experimental and b) simulated pump-probe map of a compact, 200 nm thick, TiN film on soda lime glass. ΔR/R is the normalized transient variation of reflection, $(R_{on}-R_{off})/R_{off}$, induced by the pump. Pump wavelength: 500 nm; experimental fluence: 105 μJ/cm^2; simulated fluence: 125 μJ/cm^2. c) Experimental and d) simulated ΔR/R dynamics at 410 nm probe wavelength.

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Posters

Photo Induced Enhanced Raman Scattering on hybrid 2D-MoS_2@Au Nanoparticles substrates

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Photo induced enhanced Raman Spectroscopy (PIERS) is a new characterization strategy developed on the basis of surface enhanced Raman scattering (SERS) carried out on hybrid substrates where plasmonic nanoparticles (NPs) are combined with photoactivated semiconductor materials such as TiO_2 or two-dimensional (2D) layered transition-metal dichalcogenides like WS_2, MoS_2, $MoSe_2$ etc, featuring high surface-to-volume ratio, and high carrier mobility [1,2]. PIERS takes advantages of the electron migration from semiconductors to NPs, which is triggered by UV light pre-irradiation of the substrate. The higher electron density boosts the chemical enhancement in the SERS amplification process of the Raman signal emitted by small molecules adsorbed on the hybrid substrates [1-3]. PIERS measurements on gold NPs mixed with few layer WS_2 nanosheets (chemically synthetized) have been proved to provide about 1.8% more signal compared to normal SERS effect [3]. In this work we study the PIERS enhancement of hybrid systems formed by SERS gold substrates fabricated by pulsed laser deposition (PLD) combined with 2D-MoS_2 few layered (~ 6) nanosheets (L ~ 150 nm) produced by liquid cascade centrifugation (LCC) [4] at different excitation wavelengths (561 nm, 638 nm and 785 nm). PIERS effect was evaluated using Rhodamine 6G at 1 μM, comparing the signal enhancement after UV irradiation with and without 2D-MoS_2 on PLD substrates. Results show variable intensity according to excitation energy employed.

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Optical Trapping of stained micro-plastics and particles in absorbing media

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Microplastics pollution has caused worldwide concerns due to its presence in the environment and the potential threat to human and animal health [1,2]. A lot of research (or a large number of studies) are focused on the identification and quantification of these particles on macro, micro or even nano-sized scale. In the macro regime, the efforts based on dye staining and visual observation are believed to be a better detection tool for large-sized particles [3,4]. For micro or nano-sized particles, techniques like microscopic spectroscopy and optical tweezers combined with Raman spectroscopy had been proposed as more accurate analytical methods [5]. The detection and identification in each scale with current methods have many highlights and challenges [6]. For example, simplicity in detection is one advantage of using dyes. However, this method has low accuracy in the identification of the particles. Besides, the high accuracy in identifying the type of micro and nano-particle is one of the merits of utilizing microscopic methods combined with spectroscopy. The idea of combining fluorescence dyes with Raman tweezers for faster and more accurate detection has not been presented yet due to the possible alteration of the trapping parameters induced by the absorption/emission properties of dyes.

In this work, we study the optical tweezers' parameters of polystyrene beads (2 μm in diameter) under controlled conditions and in the presence of fluorescent dyes. We have found out that the behavior of the optical forces is related to the optical properties of the fluorescent dyes dissolved inside the sample.

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Enhanced absorption in all-dielectric metasurfaces on large area

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The interest in metasurfaces has grown up in the last years because of the possibility to develop devices with optical properties that would be not possible to obtain with materials existing in nature. Moreover, metasurfaces grant a high control of these properties by engineering the nano structures constituting the device. [1]

When develop a metasurface, the first challenge is to understand the correlation between micro phenomena and macro behavior. This requires extensive parametric modeling to assess the impact of changes in geometry of the structure with the physical properties of the device. [2]

Another critical obstacle in the realization of devices based on metasurfaces is represented by fabrication. Changes in aspect ratio impact in a relevant way on the metasurface properties, which implies the needing for a highly controlled fabrication process. [3]

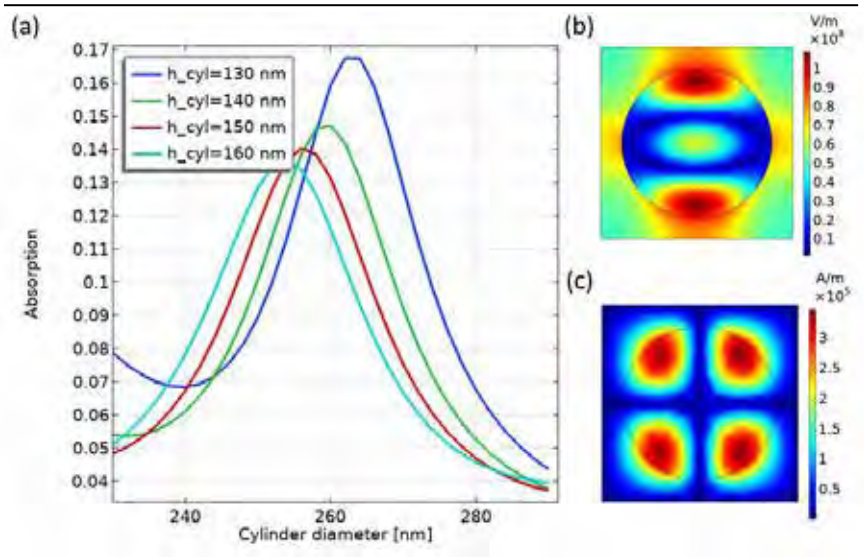

Figure 1. (a) Numerically calculated absorption of the a-Si nanopillars with diameters sweep between 230 nm and 290 nm and a height of 130 nm, 140 nm, 150 nm, and 160 nm. Simulated distribution of EM field on XY plane at 668 nm wavelength of (b) Displacement current density and (c) Magnetic flux.

In this work, we focused on individuate a simple geometry that enhances the light absorption. Major absorbance implies a stronger interaction of light with matter, and this represent an advantage in technologies as solar cells, sensors, non-linear optical devices etc.

Here we present results from a finite element method model that shows absorption of a metasurface composed of amorphous silicon nanopillars (**Figure 1a**). The electromagnetic field distribution reveals the role of resonant modes on the macroscopic properties of the metasurface. (**Figure 1b, 1c)** The model shows that resonant modes exist for specific high and diameter combination and can result in up to 20% of the incident light absorbed.

Moreover, we show that nanosphere lithography is an effective method to fabricate the metasurface with the desired optical response. This technique allows to fabricate a wide area metasurface in short time and cost effective way with respect to traditional lithographic techniques.

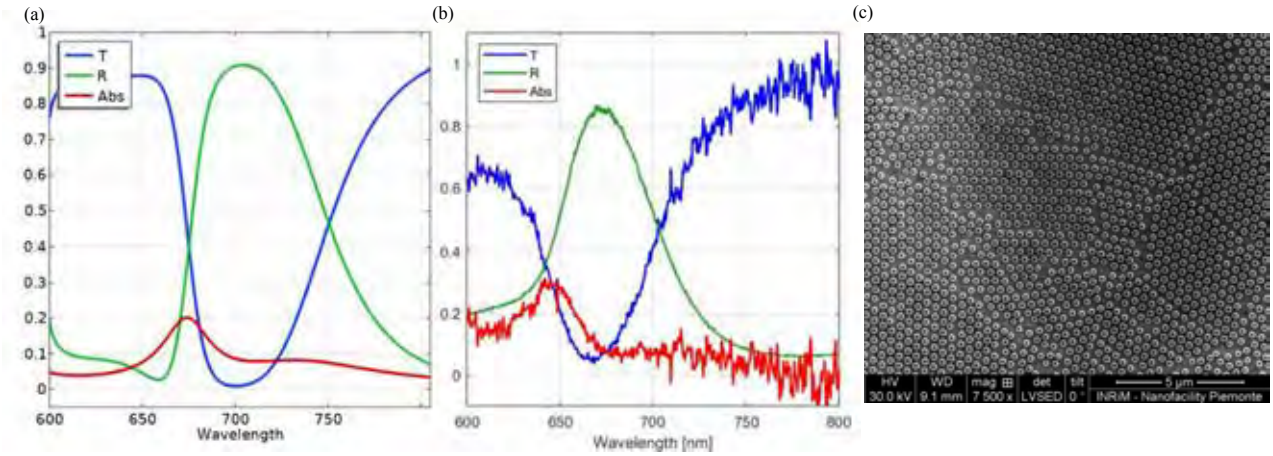

Figure 2. (a) Numerically calculated T, R, and abs of pillars with 290 nm of diameter and 154 nm of high. (b) T, R, and abs measured for pillars with 290 nm of diameter and 154 nm of high. (c) Scanning electron micrograph of the sample measured.

At the end we compare the simulation results with experimental data obtained by the characterization of our device **(Figure 2)**. The results show it is possible to reproduce the absorption enhancement. The shift in wavelength is due to a possible discrepancy in the geometry, that remarks the importance of having highly controlled fabrication processes.

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Elongated active particles in speckle fields

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Active particles, defined as units that autonomously extract energy from the environment to move or perform work, are ubiquitous in nature. Even though some interesting properties of these systems can be understood by approximating the particles as spheres, the shape of these particles has recently been highlighted as a key property to engineer their active motion [1,2,3]. The motion of these non-spherical active particles has been well characterized in homogeneous energy landscapes; however, real life active systems often find themselves in much more complex environments. Light speckle patterns can generate random energy landscapes introducing part of the complexity of real-life situations [4,5].

The role that the shape plays in the dynamics of active particles in complex environments remains to be explored. In this work, we study the dynamics of 3D printed elongated active particles in a speckle light field. The particles are coated with platinum on one end, creating an asymmetry that is exploited for activating the particles when illuminated with laser light. In this way, light plays a double role. It does not only generate a potential energy landscape, but also induces activation by thermophoresis when heating the platinum. We find that the properties of the particles' dynamics are strongly affected by parameters like their aspect ratio, the speckle grain size, and the intensity of the light. Furthermore, the particles' trajectories tend to generate a network where particles starting from different positions can end up following very similar paths (Fig 1).

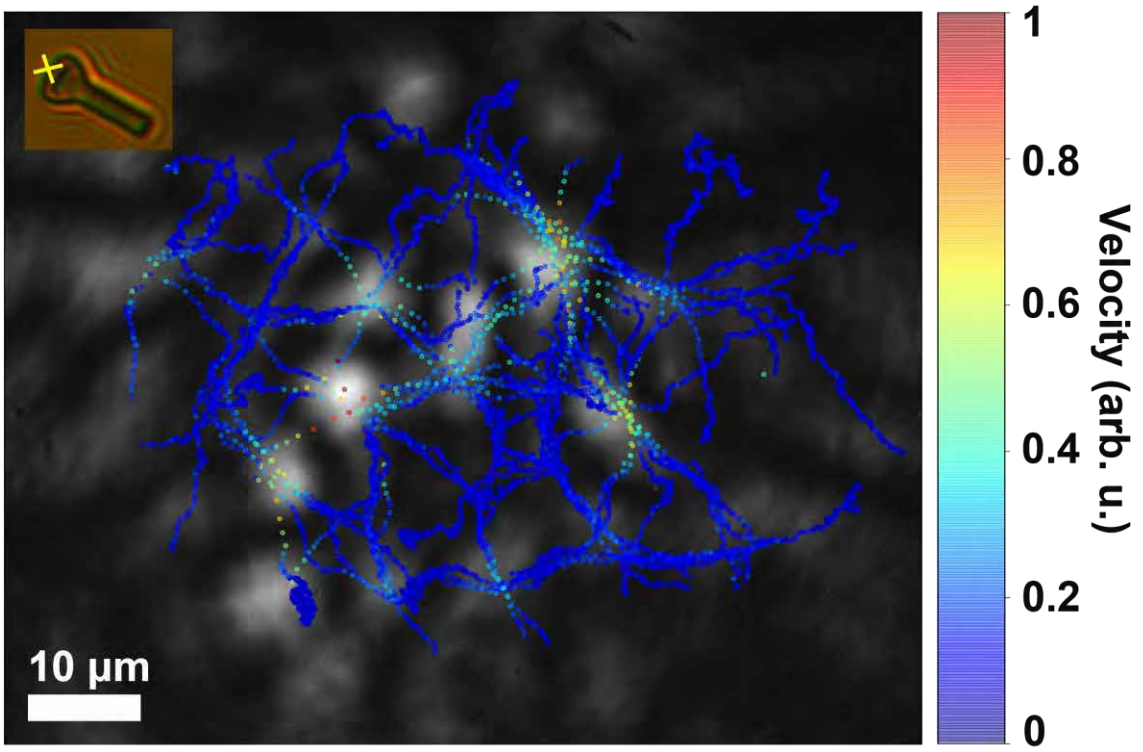

Fig 1: Trajectories of elongated active particles in a speckle field. The image in the top left corner shows the particle studied in this figure. The yellow cross corresponds to the platinum coated part. The positions of the center of this coated part are plotted for different initial positions. The color of the points represents the normalized velocity of the particle at that given point.

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Directivity of photon pairs from a resonant metasurface

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Metasurfaces, i.e., 2D arrangements of subwavelength meta-atoms, are quickly conquering linear and nonlinear photonics. By using high-index dielectric metasurfaces with Fano-type resonances, several orders of magnitude enhancement of second harmonic generation (SHG) has been already demonstrated [1]. Recently, metasurfaces are also used for the reverse process, spontaneous parametric down-conversion (SPDC), which is a convenient source of entangled photons, ubiquitous for quantum technologies. Metasurfaces are attractive for SPDC because they resonantly enhance the vacuum field and therefore stimulate the emission of photon pairs. Besides, due to the relaxed phase matching condition, metasurfaces can be made of any materials, including strongly nonlinear ones [2]. Finally, a strong advantage of metasurfaces is their multifunctional operation, combining, for instance, linear and nonlinear optical functionalities.

In this work we demonstrate, for the first time, how the design of a metasurface affects the directivity of photon pair emission. Our GaP metasurface, where a quasi-bound state in the continuum (BIC) structure is formed by pairs of tilted ellipsoids, has a high-Q resonance at 1196 nm. The resonance is seen as a peak in reflection, preventing the photons emitted at resonance from leaving the metasurface through the transmission channel. Accordingly, by pumping at 594 nm, we observe photon pairs emitted bi-directionally: the photon at the resonant wavelength is emitted in the backward direction and its match, at the wavelength 1180 nm, is emitted in the forward direction. This bi-directional emission is caused by the design of the metasurface and is not observed in bulk crystals or uniform films. Note that the metasurface separates photons whose wavelengths differ by only 16 nm. This is another example of the multifunctional operation of metasurfaces.

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Effect of gold nanoparticles size and buffer solution on their interaction with fixed cells

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Fluorescence imaging is one of the most commonly used diagnostic techniques which allows the localization of biomolecules inside cells. However, the strong overlap between intrinsic fluorescence and emitted signal from the protein of interests is one of the most important limits related to this technique. In this context, numerous studies have confirmed the strong ability of plasmonic nanoparticles to alter the fluorescence signal, making them good candidates for the optical contrast enhancement [1,2]. Nevertheless, there have been conflicting reports on the role of AuNP size and buffer solution on their effect on the fluorescence and the interaction with cells. We therefore thought to extensively investigate the AuNP size/buffer solution-fluorescence bioimaging relationship. Here we review the effects of nanoparticle size variation on the contrast enhancement in laser scanning confocal microscopy imaging modality, to detect the endothelial nitric oxide synthase proteins in lungfish gills. we used AuNPS with sizes ranging from 5 to 100 nm. We demonstrate that smaller AuNPs show greater optical contrast enhancement and easier biodistribution than larger one [3]. we have also demonstrated an increase of the affinity between nanoparticles and cells thanks to the optimization of the optical contrast by 3-fold, simply by using a different buffer [4].

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Hot electron relaxation and transport in Hyperbolic Meta-antennas

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Light generated energetic electrons, also known as hot electrons (HE) in plasmonic systems are crucial for future applications in the areas such as opto-electronics, solar energy converters and photocatalysis [1].

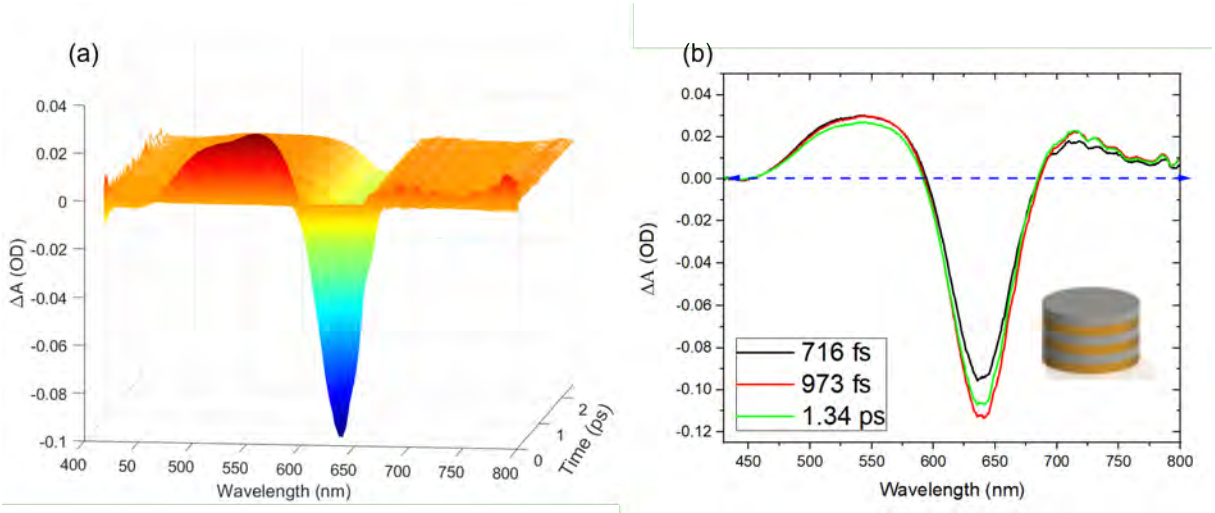

Figure 1: (a) Differential absorptivity spectra of hyperbolic meta-antennas (HMA) as a function of pump-probe delay time, when excited at 400nm wavelength. (b) The extracted transient absorption spectra of HMA for for specific delay times.

However, spatial and temporal understanding of HE generation and transportation processes holds the key to optimize plasmon-induced HE effects for the next generation applications. In particular, ultra-short life, low yield and short mean free path of HEs significantly hinder their hot-carrier based applications. To improve the life time and extraction of HEs, hyperbolic meta-antennas (HMA) based on gold/dielectric stacking disks have been designed and fabricated. While, nanodisks antennas (NDA) of thickness equivalent to that of total metal layers in HMA were also investigated for the reference purpose. HMA have shown the characteristic to exhibit separate and tuneable wavelength regions of absorption and scattering resonances in the same architecture [2]. The elognated HE's life time of HMA have been observed in comparison

to that of NDA at interband transition excitation. This can be further enhanced when HMA are excited at its distinguished absorption band, confirming the role of absorption efficiency in the increased relaxation of HE's life time. Our findings can provide new insights to the fundamental understanding of hot electron generation and transport in such particular metallo-dielectric meta-antennas and can lead to design efficient hot carrier devices.

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Micro- and Nano-structuring of biodegradable and biocompatible polymers for optical applications

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In recent decades, biodegradable and biocompatible polymers have emerged as eco-friendly materials for a wide range of applications including food packaging, textile fiber manufacturing, drug delivery, foldable electronics and additive manufacturing. We faced this important technological challenge in a photonic framework, by implementing a novel fabrication method and using sustainable materials for breakthrough systems. We demonstrate the possibility of replicating micro- and nano- scale photonic structures by using a bacteria growth medium (Luria Bertani Agar - LBA) *via* a replica-molding technique. Following this process, we used several masters, fabricated by two-photon lithography, made by lattice arrays with micro- and nano-scale periodicity. Starting from a standard LBA solution, we increased the w/v concentration of the agar polymer in order to allow the films formation by employing different thermal profiles. An Atomic Force Microscopy (AFM) investigation of masters and replica confirm the patterns molding. The high-quality morphological and optical properties of the LBA replica have been verified by acquiring the diffraction patterns, that resulted very similar to the ones of the masters. The obtained gratings lead to high quality diffraction patterns with good efficiency performances, resulting in structural dielectric colours. Although there are still further investigations to be conducted, we believe that the presented approach may represent a potential platform to incorporate various biological entities, such as cells, DNA or proteins, in order to enable biosensing applications.

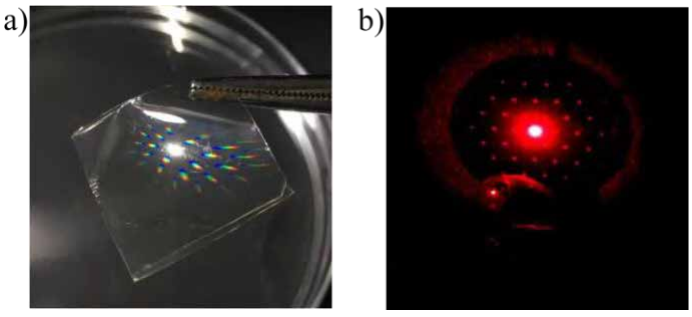

Figure 1. a) A micropatterned LBA film after it was peeled of the master; **b)** Diffraction micropattern recorded in transmission from LBA under illumination laser light at 633nm wavelength.

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Photonic Plasmonic Metamaterials based on Ta-doped TiO_2 Transparent Conducting Films with Tunable Electrical and Optical Properties

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The raising need to extend the excitation range outside the visible and to tailor the optical characteristics of plasmonic metals has stimulated the research on low-dimensional structures based on Transparent Conductive Oxides TCOs, where novel functionalities can be met in addition to visible transparency and electrical conductivity [1]. TCOs possess plasmonic characters tunable from VIS to mid-IR through the carrier concentration (extrinsic doping/stoichiometry) or by applying external stimuli (electrical bias). Hence these materials have been proposed as alternatives to metals, whose main limitation is the non-modifiable carrier density (i.e. fixed plasma frequency). For instance, Tantalum-doped TiO_2 (Ta:TiO_2) has been already demonstrated to be a performing TCO, while ensuring the advantages of TiO_2 such as low-cost, non-toxicity and chemical stability [2]. In this framework, a new class of artificial metamaterials exhibiting peculiar functionalities is rapidly emerging. Hyperbolic multilayer metamaterials especially present unique electromagnetic phenomena (high-k modes) activated by the anisotropic permittivity resulting from the periodic alternation of conductors and dielectrics of subwavelength thicknesses [3]. When increasing the characteristic dimension of layers, one-dimensional (1D) photonic crystals can be achieved straightforward [4]. By selecting materials and device geometry, plasmonic behavior and light propagation can be engineered successfully, while control over nanoscale morphology, structure and electrical performance looks promising for targeted applications in optoelectronics, nanophotonics and bio-sensing [3].

In this work, nanocomposites and multilayer meta-structures have been developed starting from Ta:TiO_2 TCO thin films, involving conventional metals (Au) and original design routes (via pulsed laser deposition). The final goal is to explore and modulate electrical/optical responses resulting from unusual combinations of materials and properties (transparency, conductivity, tunable VIS-IR plasmonics, active modulation).

First, optical and electronic properties of Ta:TiO_2 films have been studied in detail as thickness is decreased below 10 nm and as a function of Ta doping (from undoped TiO_2 to 10% at. Ta), while keeping attention to the correlation with morphological and structural properties, showing a high degree of tunability [5]. Then, Au nanoparticles have been integrated in Ta:TiO_2 thin films to form a metal-TCO nanocomposite (**Figure 1a**). Indeed, the Localized Surface Plasmon Resonance of gold can be tailored experimentally of more than 100 nm by changing the permittivity of the matrix, i.e. by exploiting the control over carrier concentration in the TCO through the Ta doping content [6] (**Figure 1b**). As a clear demonstration of its versatility, Ta:TiO_2 TCO has been also exploited for the fabrication of original multilayers (**Figure 1c**), obtained directly in a simple one-step synthesis, through which the alternation of conductive

(compact) and dielectric (nanoporous) layers can be realized by varying the deposition pressure (1-6 Pa O_2). Following a material science approach, structural and electrical properties have been optimized to rule optical/plasmonic outputs as a function of deposition conditions, doping content and geometrical parameters. Hyperbolic platforms in the IR are one of the most promising applications, while 1D photonic crystals can be accomplished by customizing compact/porous fraction and dimension of the layers. The proper optimization of material properties leads to an intense photonic band gap, spanning from green to red wavelengths in the visible, which in turn can be actively modulated with an external bias (**Figure 1d**).

All the systems investigated show an inherent tunability in optical/plasmonic properties, unattainable in metals, enabled by the possibility to adjust the carrier concentration in alternative conductors (indirectly through the surrounding matrix, directly at synthesis or with an "active" approach). Moreover, these multifunctional meta-devices are application-oriented and possess a cross-disciplinary attitude, thus are appealing as plasmonic-functionalized TCO-based electrodes, nano-architectures for sensing or optoelectronic elements for active manipulation of colors.

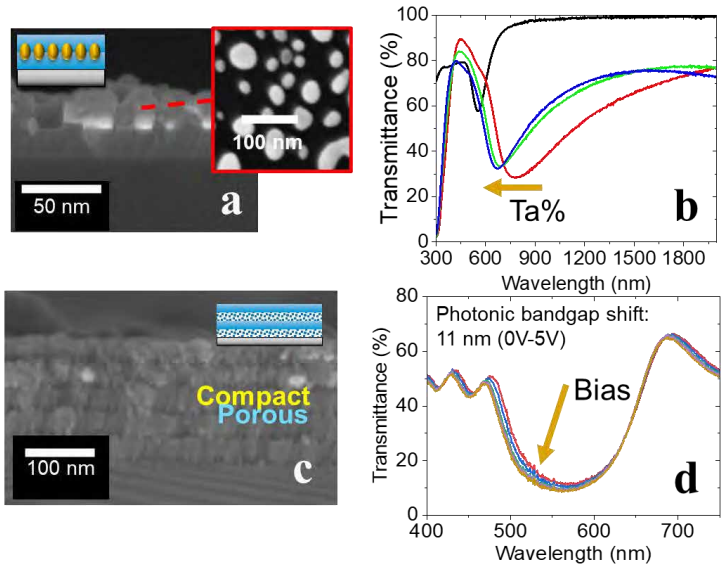

Figure 1: SEM images of Au nanoparticles embedded in Ta(5%):TiO_2 in "sandwich" configuration (a) and related transmittance spectra with different Ta doping of the TCO matrix (b). SEM image of Ta(5%):TiO_2 porous/compact multilayer (c) and transmittance spectra of a Ta:TiO_2-based photonic crystal with external bias applied (d).

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Optical characterization of holographic acoustic tweezers

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Acoustic tweezers use sound waves to trap solid, liquid or biological samples [1, 2]. Ultrasonic emitters, arranged in space as an ordered array, can be used to realize acoustic tweezers in air, provided that the phase of the signal driving each transducer is assigned according to a suitable phase pattern, giving a trapping point at a target position [3]. This approach, similar to the working principle of spatial light modulators [4], allows to trap and manipulate single or multiple particles. Different phase masks can be generated, giving the *twin trap*, the *vortex* trap and the *bottle* trap, each one with peculiar properties [3]. Remarkably, the setup is based on low-cost components, leading to a "democratization" of acoustic levitation [5].

In this work, we report on the use of the twin trap for the acoustic trapping of millimeter particles in air. Our setup is based on a flat array of 8×8 ultrasonic emitters (Fig. 1 a), each one having 1 cm diameter [6]. Trapped particles are styrofoam beads with 1.6 mm radius (Fig. 1 b). To study their dynamics in the acoustic trap, we adopt two optical detection methods typical of optical tweezers. In the first method, we shine a laser beam on the trapped particle and we detect the fluctuations of its shadow on a Quarter Photodiode (QPD). In the second method, we acquire the silhouette of the trapped particle by a CCD camera and a telescope, then we extract data on the particle position using a video tracking software. With both methods we collect data on particle displacement in x, y, and z directions. We analyze these data by calculating their power spectra, since periodic oscillations of the particle give rise to a peak in the power spectrum at the corresponding frequency. The spectra obtained with both CCD and QPD signals exhibit peaks at the same frequency. This allows us to evaluate the force constant of the acoustic trap on the basis of the simple harmonic oscillator model $f = \frac{1}{2\pi}\sqrt{\frac{k}{m}}$. For instance, we obtain a force constant $k_z \simeq 10^{-3} N/m$ for the z direction, perpendicular to the ultrasonic array.

This study allows us to conclude that the two detection methods give consistent results on millimeter particles. The choice of which detection system to use depends on the range of oscillation frequencies of the particles under study.

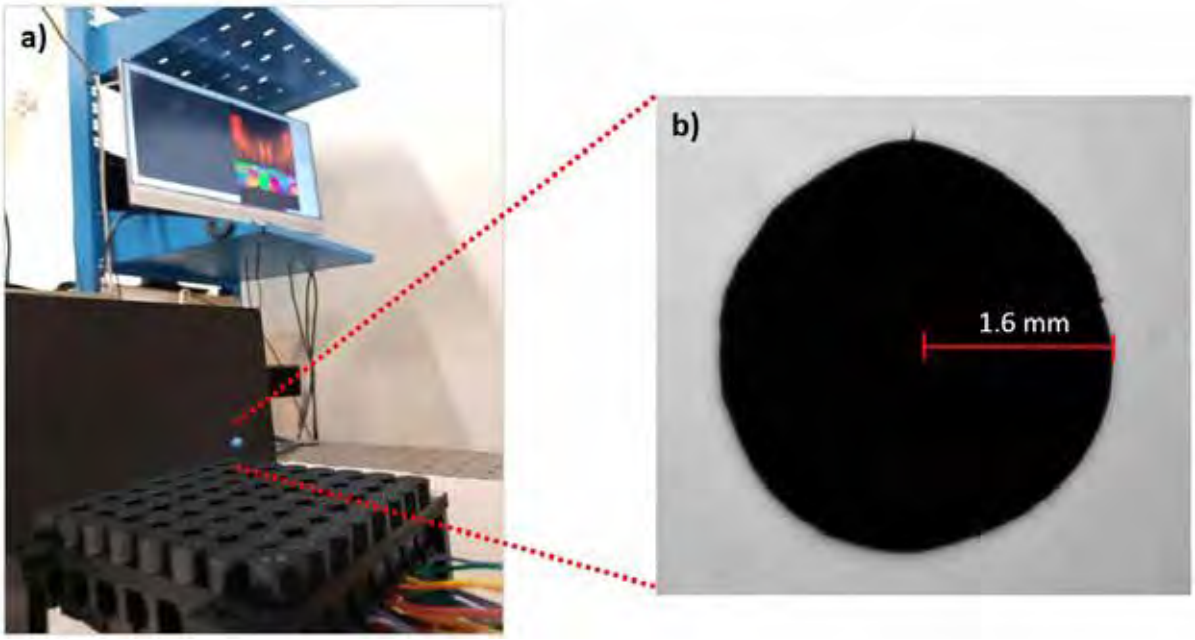

Figure 1: a) 8×8 ultrasonic array used to trap a styrofoam millimeter sphere; b) Silhouette collected with the CCD camera, showing a trapped levitating particle.

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Plasmonic metasurfaces based on pyramidal nanoholes for high efficiency SERS Biosensing

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An inverted pyramidal metasurface was designed, fabricated and studied at the nanoscale level for the development of a label-free pathogen detection on a chip platform that merges nanotechnology and surface enhanced Raman scattering (SERS). Based on the integration and synergy of these ingredients, a virus immuno-assay was proposed as a relevant proof of concept for very sensitive detection of hepatitis A virus, for the first time to our best knowledge, in a very small volume (2 μl), without complex signal amplification, allowing to detect a minimal virus concentration of 13 pg/ml. The proposed work aims to develop a high flux and high accuracy Surface Enhanced Raman Spectroscopy (SERS) nano-biosensor for the detection of pathogens in order to provide an effective method for early and easy water-monitoring which can be fast and convenient. [1]

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All-Optical Reconfigurable Metalenses

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Nanostructured metalenses have been engineered to generate a wide range of optical phenomena, allowing an unprecedented control over the propagation of light. However, they are generally designed as single-purpose devices without a modifiable optical response, which can be a barrier to applications. In this work, we report the nontrivial infiltration of a nanostructured planar silica metalens with nematic liquid crystals [1]. LCs are particularly useful because they can be manipulated thermally, electrically, magnetically, or optically, which creates the potential for flexible or reconfigurable lenses. We present and discuss the all-optical reconfigurability of metalens optical response as a consequence of thermo-plasmonic induced changes of the local refractive index. Since the orientation-dependent optical properties of liquid crystals can be controlled with external stimuli, this technology can enable dynamic control of the metalens optical response.

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Detecting and inhibiting the generation of reactive oxygen species for biomedical applications of gold nanorods under ultrashort laser pulses

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Individual gold nanoparticles can produce reactive oxygen species (ROS) over a micrometer range under ultrashort pulsed light [1]. While beneficial for photodynamic therapy, it is prohibitive for other biomedical applications such as imaging or targeted gene or drug delivery by plasmonic photothermal conversion. ROS are usually detected with specific fluorescent molecules. SOSG is widely used to probe singlet oxygen ($^1O_2$). However, the nanoparticle chemical environment may interact with these probes and disturb the determination of ROS production. It is then needed to control the conditions in which the latter is carried out. Besides, gold nanorods (AuNRs) are an iconic choice for biomedical applications as they can exhibit an intense longitudinal plasmon mode in the transparency windows of biological tissues. A surfactant molecule, CTAB, ensures their growth and stability in solution, but it is cytotoxic. Coating AuNRs with a silica layer makes AuNRs biocompatible by removing CTAB from their surface, prevents them from reshaping under high laser intensity, and avoids the formation of a protein corona. In addition, a mesoporous silica shell can be loaded with drugs, dyes, or imaging agents.

We first investigate the effect of the CTAB-SOSG interaction for reliable $^1O_2$ detection when produced by ultrashort laser-pulse irradiation of AuNRs, and suggest proper AuNR surface chemistry to assess the ROS production. Then, we study the effect of coating AuNRs with silica on their ROS generation.

ROS produced in water by irradiating AuNRs with 400-fs laser pulses, tuned to their plasmon resonance, are detected by fluorescence spectroscopy. SOSG is used to detect $^1O_2$ while DRH probes $^1O_2$ and •OH. Calculations enable us to determine the ultrafast transient optical near-field (Fig. 2), a key parameter to interpret ROS production properties. We first demonstrate that the SOSG emission properties and sensitivity to $^1O_2$ strongly depend on the CTAB concentration due to CTAB micelles. With AuNRs acting as $^1O_2$ photosensitizers, we show that $^1O_2$ detection is not possible in CTAB, whereas a high sensitivity of SOSG for $^1O_2$ is obtained in citrate and PEG [2]. We then evidence that a dense silica shell added onto AuNRs inhibits the formation of $^1O_2$ and •OH efficiently. The plasmonic field enhancement at the AuNR tips is reduced [3]. With the multiphotonic ejection of electrons and the Dexter energy transfer to $^3O_2$ being also blocked, the silica coating hinders all the possible pathways for ROS production. Silica-coated AuNRs are then not suitable for use as direct photosensitizers but are safer than pure AuNRs for a range of other applications in biomedicine.

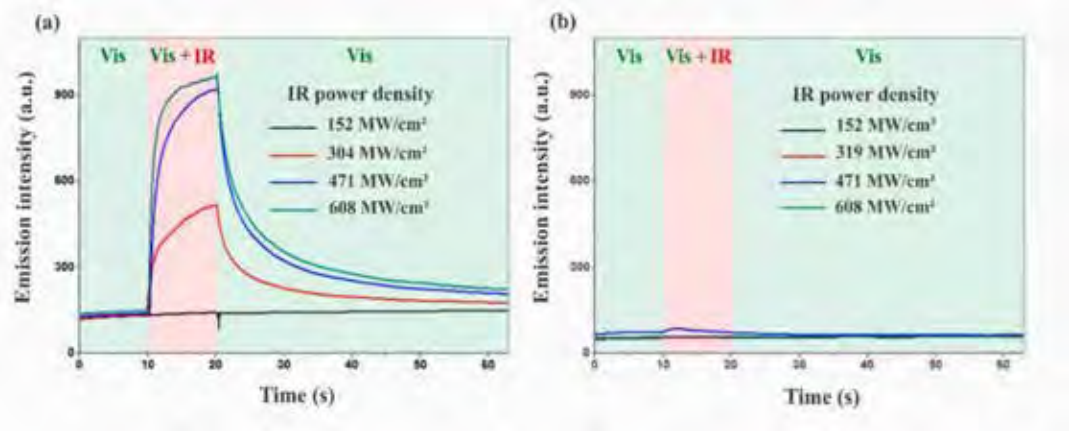

Fig. 1. Time evolution ot the SOSG emission intensity in the presence of (a) PEGylated AuNRs and (b) AuNRs coated with a 8.6-nm thick silica shell, acting as ROS photosensitizers, in water. SOSG fluorescence under visible light (Vis) is recorded before, during and after the excitation of the longitudinal plasmon mode of the nanoparticles by 1030-nm fs laser pulses (IR) with different peak intensities.

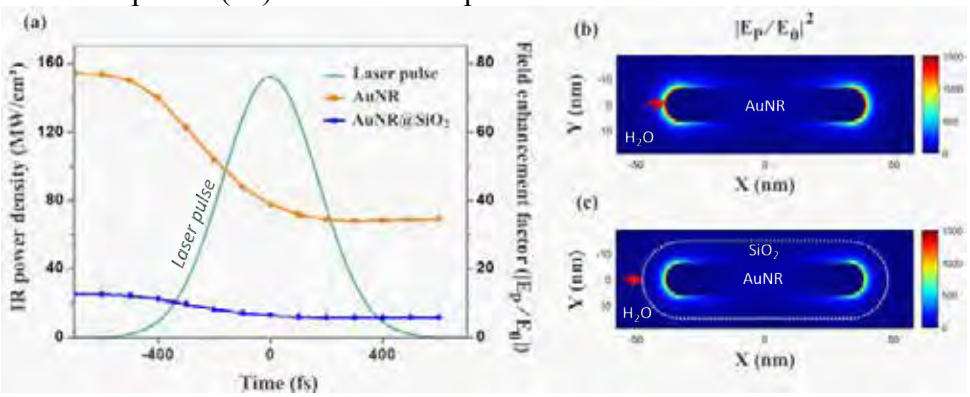

Fig. 2. Transient optical near-field enhancement at the AuNR tip (right scale) evolving along the laser pulse (left scale). (b) Optical near-field intensity collected at the pulse peak. Simulations combining the BEM and Boltzmann's equation for the photo-induced hot electron dynamics. It can be observed that (ii) the plasmonic field is transiently attenuated by the hot electron gas generation, and (ii) the presence of the silica layer strongly hinders the field enhancement.

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Room-temperature lasing action in hybrid semiconductor nanowire - metal grating plasmonic nanolasers

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Nanowire(NW)-based semiconductor-insulator-metal (SIM) plasmonic structures represent a benchmark platform for the next generation of hybrid nanolasers, capable of sustaining sub-wavelength lasing action supported by hybrid modes [1] , larger Purcell-enhanced stimulated [2], leading to an overall speedup of the lasing dynamics [3], when compared to their photonic counterparts [4]. In this work, we report on the realization and optical investigation of modified SIM platforms [5], in which single ZnO NWs are deterministically placed onto metal gratings (MGs), FIB-milled into a 70 nm Al layer with a nanometric Al_2O_3 spacer on top. By performing ns-excitation steady-state and fs-excitation time-resolved μ-PL measurements, we study the spectral and temporal properties of such hybrid platforms, as a function of the cavity geometry (i.e. NW size, grating period and NW-trench orientation) and temperature. We report about room-temperature lasing in most of these hybrid NW-MG structures and lowering of the lasing threshold compared to the planar SIM plasmonic nanolasers.

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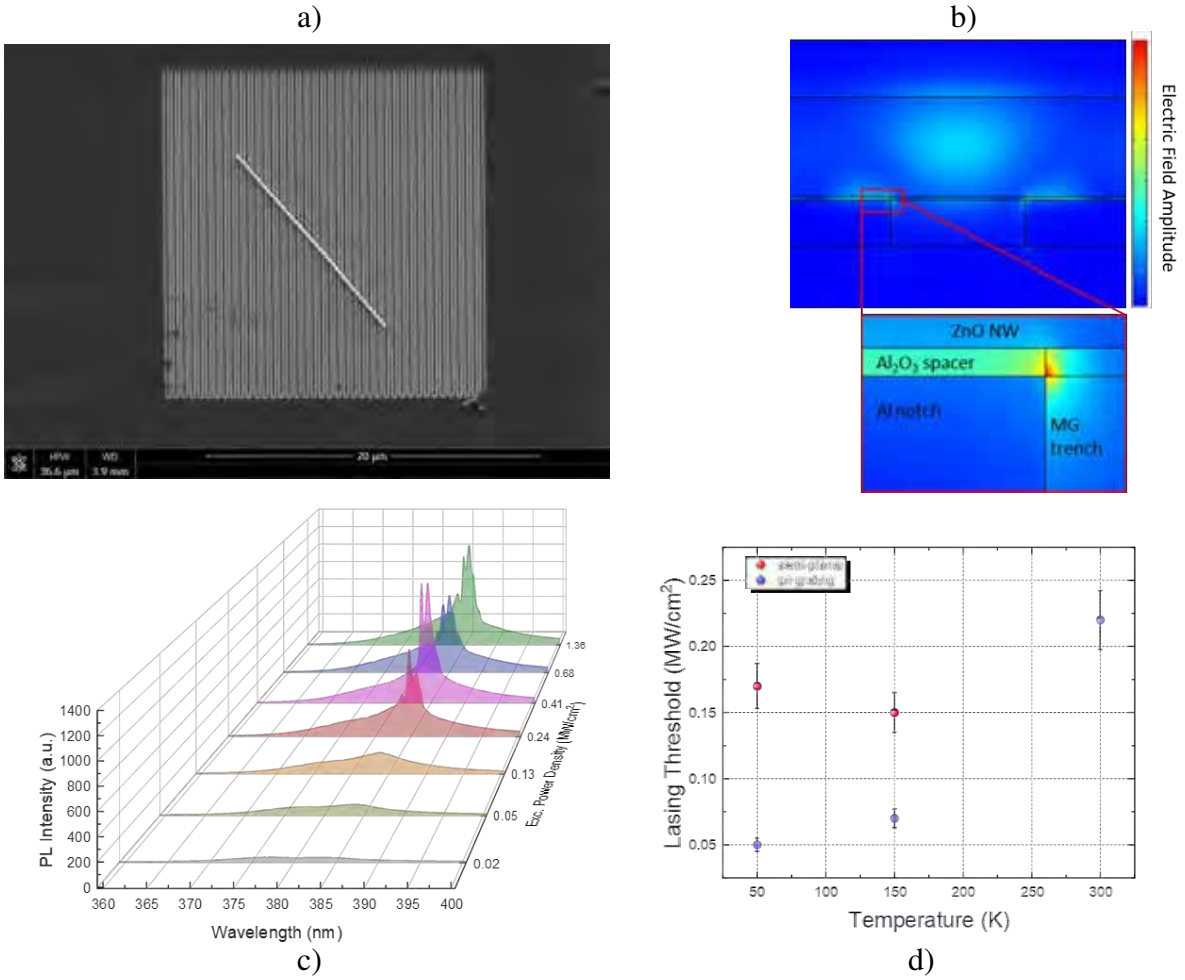

Figure 1: a) SEM image of a representative NW-MG cavity, featuring a 14µm-long ZnO nanowire deterministically oriented at an angle of $\approx 42°$ with respect to the trench direction of an Al grating with period 500 nm. b) Simulation of the field distribution for the hybrid "hopping" mode sustaining the lasing action in the hybrid cavity. c) Room-temperature photoluminescence spectra for the on-grating configuration of a NW moved onto the metal grating from an initial planar configuration. d) Comparison of the lasing thresholds at 3 different temperatures for the planar and on.grating configurations.

Wearable Lab-in-a-watch Plasmonic Biosensor of Sweat Molecules for Non-invasive Glucose Monitoring

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Wearable molecular sensing technology capable of real-time monitoring and online analysis of biomarkers in biofluid is an essential part for future personalized medicine. In particular, wearable optical, colorimetric and electrochemical biosensors on skin allow analysis of the rich composition of solutes and metabolites in sweat, which are secreted from skin pores, for real-time monitoring of sport performance as well as health diagnosis. The correlation between sweat glucose and blood glucose level shows the potential of the optical skin sensors as non-invasive monitoring tool for diabetes patients. However, the state-of-the-art optical skin sensors still need a separate step of detection by portable detectors [1], which hinders their development toward continuous measurement of sweat glucose.

In this work, we report a wearable optical sensing system on skin by integrating an electro-plasmonic chip into an optical watch that together enables controllable sweat stimulation and on-watch detection of the sweat glucose, as shown in Figure 1. The chip includes iontophoretic electrodes for local stimulated excretion of sweat and gold nanohole array for subsequent localize surface plasmon resonance (LSPR) detection of the sweat glucose. The optical watch will work as an integrated electro-optical platform of an optical sensor (for LSPR detection) with an electric DC power supply (for sweat stimulation). By fine controlling gold nanogaps in the nanohole array, we have achieved LSPR detection of molecule monolayer in water with a sensitivity of 170 RIU.

The sensor system as a whole can be a universal molecular sensing platform potentially to detect an array of biomarkers molecules in sweat, though it targets sweat glucoses firstly. The microfluidic electro-plasmonic chips capable of on-demand sweat stimulation would allow enough amount of sweat collected from patient skin for detection without the requirement that the patients do exercise to sweat. Therefore, the system can find broad applications in many diseases and may trigger the wearable optical sensor as a standard technological platform for personalized healthcare.

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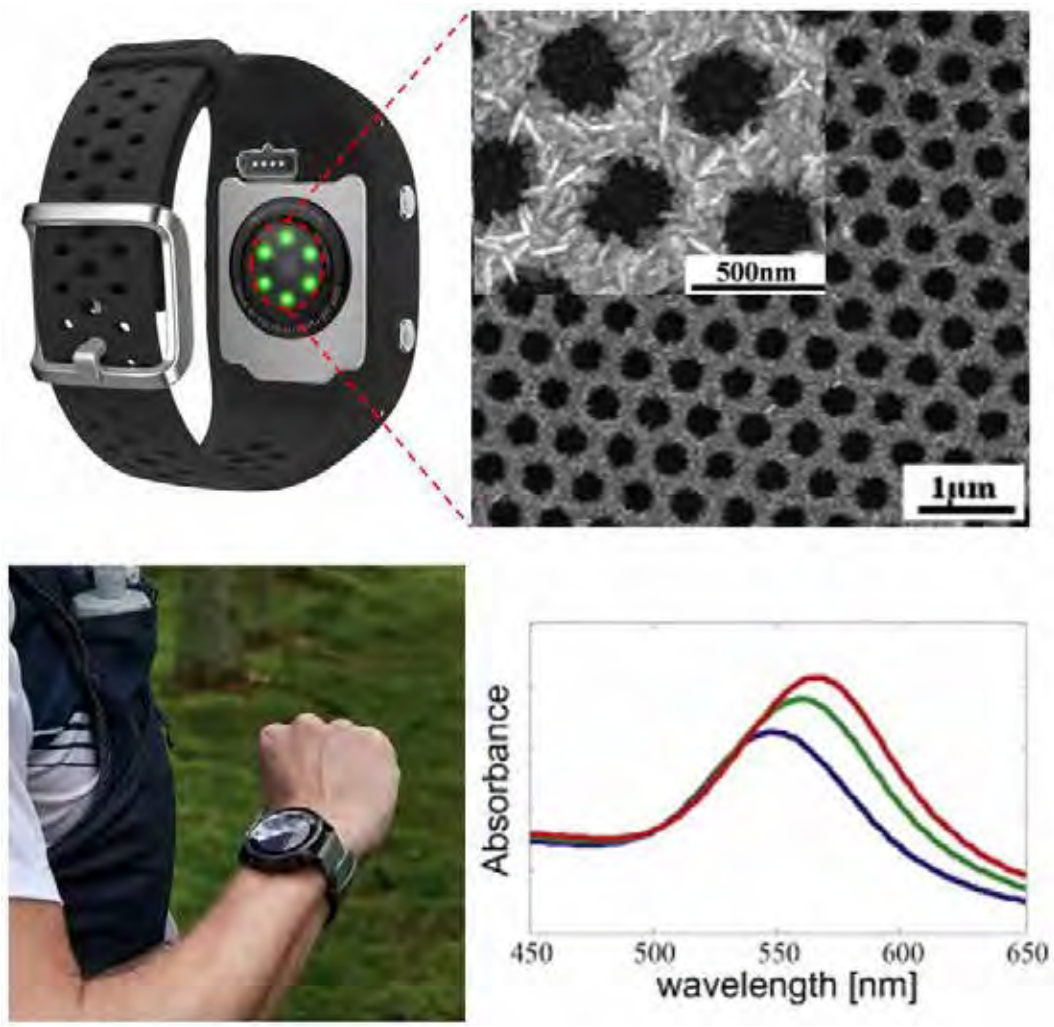

Figure 1. Schematic of the Lab-in-a-watch plasmonic biosensor with a secondary electronic microscopic image of the nanohole array for non-invasive glucose monitoring.

Dedicated to the memory of
"Prof. Daniela Pucci"

Daniela was an outstanding chemist who made possible significant advancements in materials science and synthetic chemistry. She was also an extraordinary human being and a wonderful friend,
who passed away on April 24 - 2014