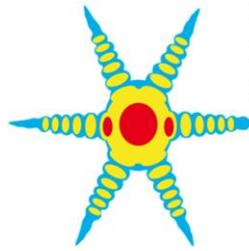


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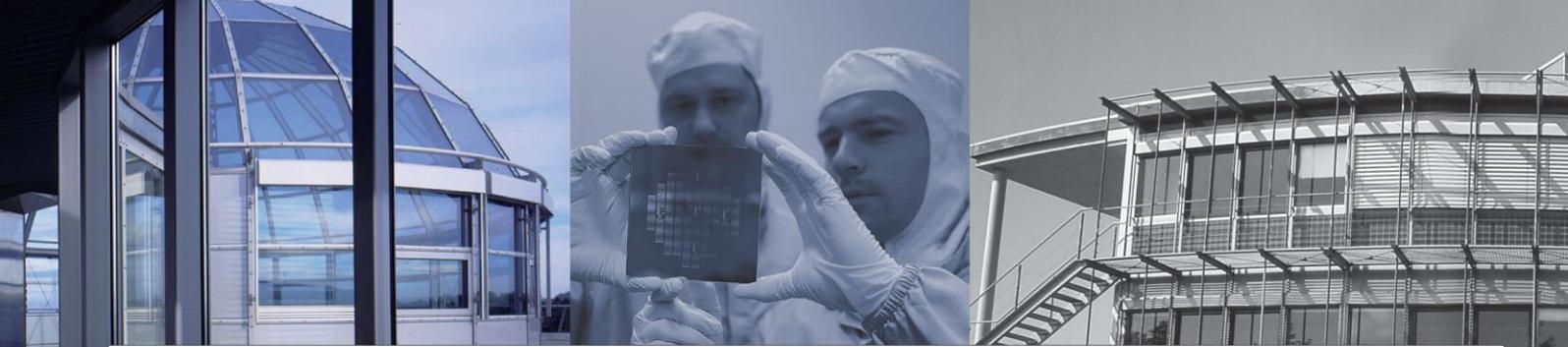
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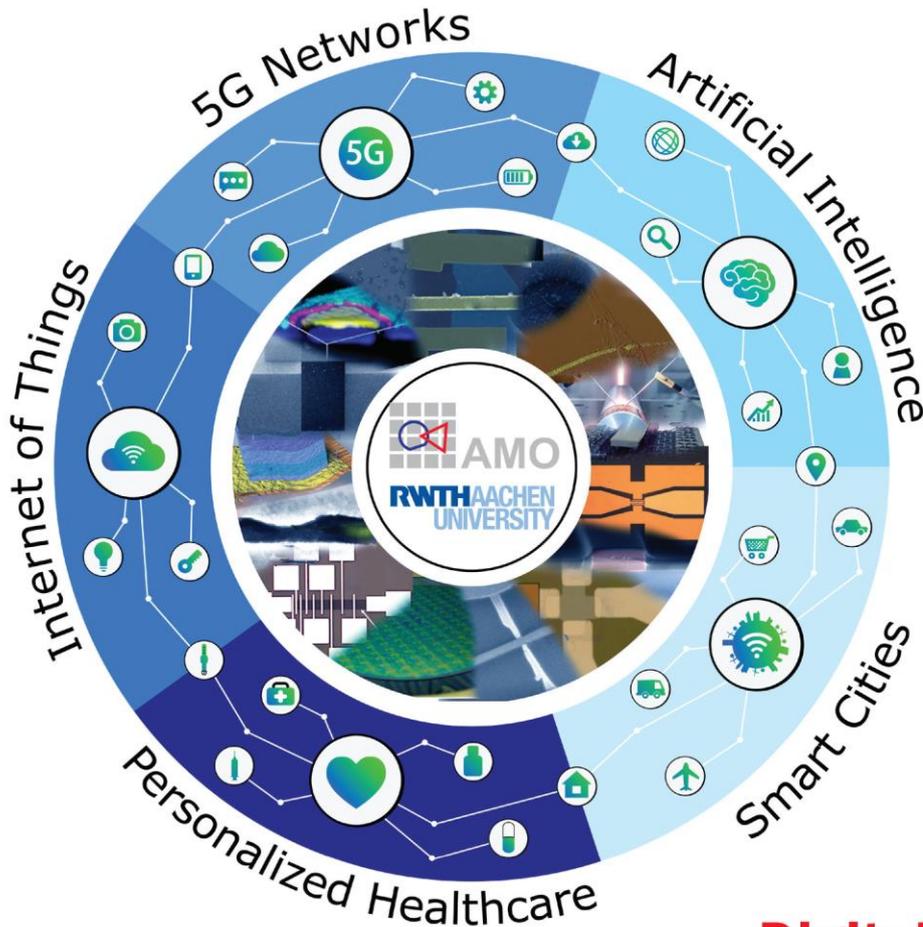
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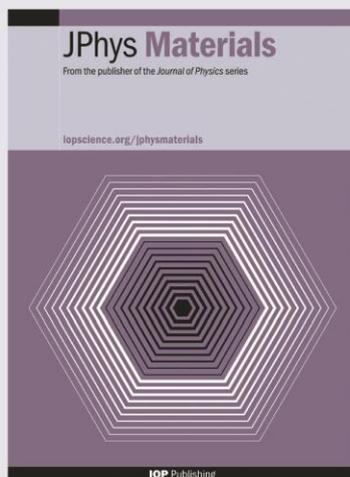
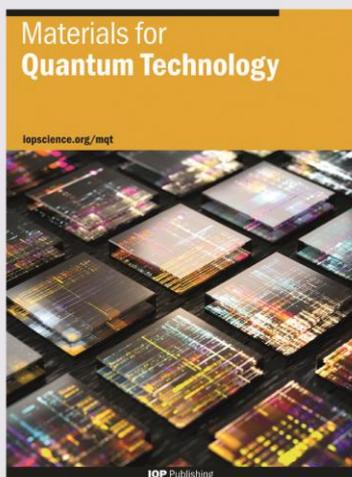
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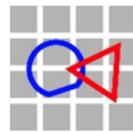
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Molecular optical spectroscopy and microscopy at the atomic scale

Javier Aizpurua

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Scanning Tunneling Microscopy (STM) provides a perfect configuration to explore light emission from single organic molecules [1,2].

A general theoretical framework which describes the coupling of an exciton and a plasmonic picocavity serves to reproduce and interpret the spectral information of light emission in STM, as well as the intensity emission maps with intramolecular resolution [3].

On top of the intensity maps, the control of the plasmonic cavity within the STM configuration allows for tracing the Purcell factor (broadening of emission), Lamb shift (energy shift of emission), and Stark effect (static shift of emission) in the emission of a free-base phthalocyanine (H_2Pc) in an STM cavity [4].

Light emission from organic chromophores in picocavities allows for bringing cavity quantum electrodynamics (c-QED) to the realm of the nanoscale, opening avenues to control excitonic states of matter at the single molecule level, and use light-matter polaritonic states associated with molecules in engineering of chemical reactivity and in tailoring quantum information technologies with polaritonic q-bits.

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Figures

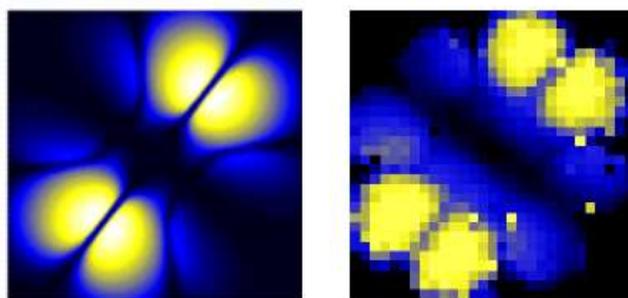


Figure 1: Left: Experimental map of light emission from a free-base phthalocyanine (H_2Pc) deposited on an NaCl-Ag(111) surface when scanned in a STM cavity. The area of scanning is $2.5 \times 2.5 \text{ nm}^2$, and the bias voltage applied is $V = -2.5 \text{ V}$, with a current $I = 100 \text{ pA}$. Right: Theoretical map of light emission under the same circumstances as in the experiment. The spectral range of light emission considered is at the excitonic emission line around 1.975 eV .

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Controlling light and heat at the nanoscale

Pablo Albella

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F. González, F. Bresme, F. Moreno

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Abstract

Plasmonic nanoantennas have shown as a versatile tool to control light beyond the conventional diffraction limit. They are known to suffer relatively large losses and consequently the Joule heating of the structure and its local environment. On the other hand, nanoparticles made of High Refractive Index (HRI) dielectric materials, such as Si, or other semiconductors have been proposed recently as an alternative to metals, driven by their low-losses and presence of magnetic response in spite of being non-magnetic materials [1]. In the first part of the talk I will introduce the basics and some advances of this recent topic of dielectric nanophotonics, discussing some recent results[1] that are boosting its progress. Special attention will be paid to applications like directional control or light diodes [2-4] (see fig 1), optical switching [5], beam steering [6], surface enhanced spectroscopies [7], enhancing chiral sensing (see fig. 2) [8,9] or non-linear optics[10]. The second part of the talk will be framed in the topic of thermoplasmonics and dedicated to the design of efficient nanoheaters and to discuss novel ways to control the heat delivery in photothermal applications [11].

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Figures

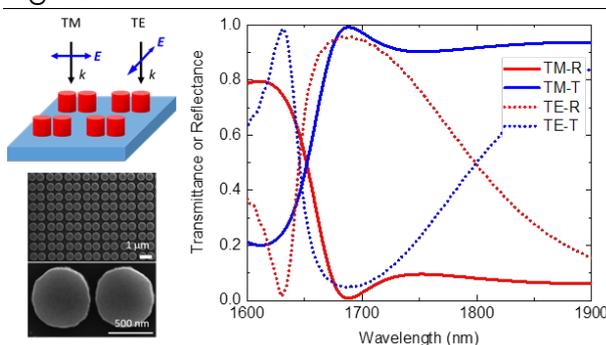


Fig. 1: Transmission and reflection of a Si dimer array for TM (solid) and TE (broken) polarization.

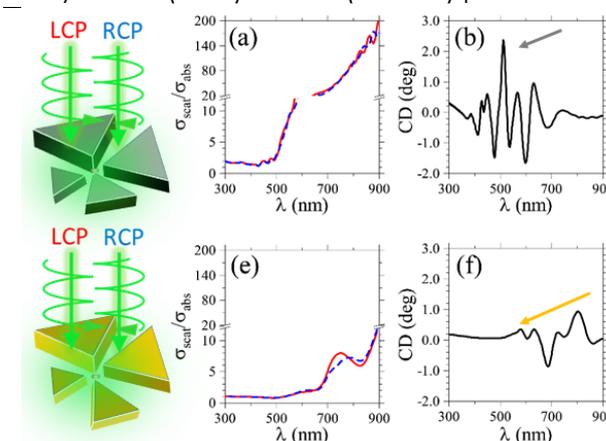


Figure 2: Scattering to absorption ratio and CD spectrum, for systems made of Si and Au.

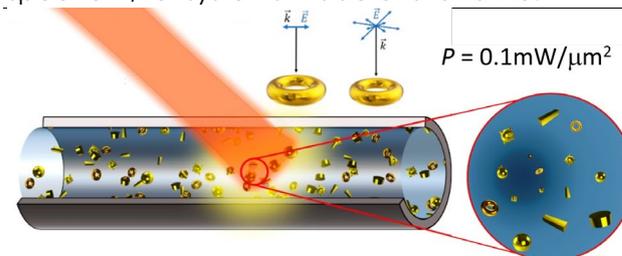


Figure 3: Standard thermofluidic application. Upon excitation by light, an effective local temperature increase will correspond to an average temperature of all possible orientations [see ref. 11]

Strong coupling in nanophotonics

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Electromagnetic resonances at visible and infrared frequencies can be strongly coupled to the excitations in a material, such as molecular excitons and vibrations, leading to the formation of new polaritonic states of matter that present novel characteristics and may allow to control the properties of the material [1,2].

We present the experimental and theoretical response of strongly-coupled micro and nanophotonic systems ranging from Fabry-Pérot microcavities to plasmonic nanoresonators that confine light to a gap of just a few nanometres. We use these systems to discuss effects such as the limitations imposed by electronic quenching on the coupling strength [3], or the different strong coupling signature that is observed in the inelastic molecular photoluminescence signal and in the elastic extinction spectra [4]. We show that the photoluminescence can be sensitive to the full dynamics of the system [5,6].

We also stress the potential of phonons in Van der Waals materials, e. g. hexagonal hBN, for strong coupling. These phonons can directly interact with electromagnetic modes (Figure 1) [7] or, in a different scheme, the localized phonon polaritons supported by hBN nanostructures can be strongly coupled with molecular vibrations [8,9]. The direct interaction with phonons notably allows for reaching the regime of ultrastrong coupling, where the coupling strength is comparable with the resonant frequencies and new phenomena appear, including the modification of the ground state [10]. We further discuss how the ultimate coupling strength is set only by the

material, not by the electromagnetic resonance, and is given by the splitting present in the bulk polariton dispersion.

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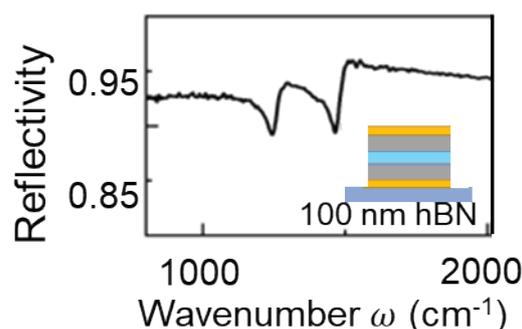


Figure 1: Example of strongly coupled system, consisting in a $\sim 100\text{nm}$ hBN layer inserted in the centre of a Fabry-Pérot microcavity. The reflection dips in the spectra indicate the two polaritonic states.

Chiral semiconductor nanophotonics

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Chirality plays a pivotal role in the functionality of biomolecules such as proteins, amino acids, and carbohydrates. Circular dichroism can distinguish enantiomers thanks to a small difference in the absorption of circularly polarized light. However, chiral sensing faces significant limitations due to inherently weak chiroptical signals. It is thus severely limited by low sensitivity and low spatial resolution. As a result, it is challenging to resolve the chirality of individual nanoscale objects using light for critical applications such as detecting protein aggregates linked to various diseases.

In this presentation, I will discuss our progress to push the limits of optically resolvable chirality through new concepts in semiconductor nanophotonics. First, I will show several strategies to optimize chiral molecular sensors based on silicon metasurfaces to detect low molecular concentrations. Specifically, I will present our recent results on tailoring silicon nanostructures to enhance polarized fluorescence and Raman spectroscopies, increase optical chirality, and maximize chirality transfer [1,2].

Second, I will introduce an approach to molecular sensing based on excitons in atomically thin semiconductors. I will show how monolayer semiconductors can exhibit strong fluorescence fluctuations [3] that report on charge transfer events to nearby nano-objects.

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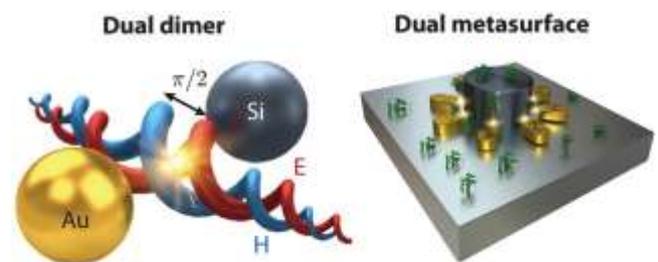


Figure 1: Dual metal-dielectric nanoresonators for chiral sensing

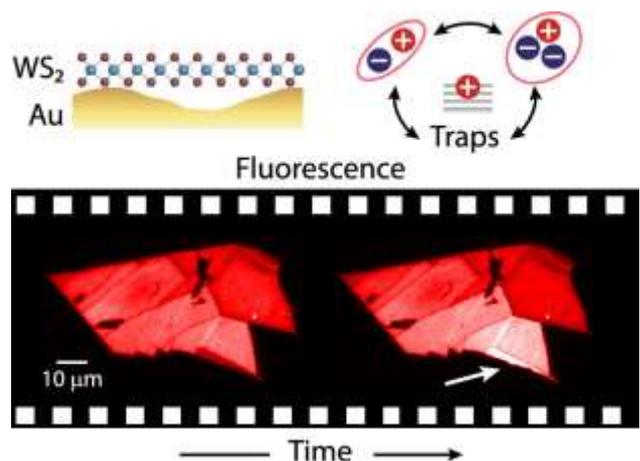


Figure 2: Exciton fluctuations at the interface between a 2D semiconductor and a metal.

Control of Light at the Atomic Scale: Fundamentals and Applications

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Two-dimensional materials have been recently shown to host robust polaritonic modes, ranging from plasmons in highly doped graphene to excitations in transition metal dichalcogenides. The electromagnetic behavior of these modes can be well understood in terms of an effective surface conductivity, in which we 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 we can understand in terms of simple theoretical descriptions. We will also cover more sophisticated descriptions, aiming at exploring genuinely quantum-mechanical effects. We will further overview recent advances in ultrafast optical response and nonlinear optics, as well as the potential application of these materials for light modulation, quantum-optics, and optical sensing.

This work has been supported in part by the Spanish MINECO (MAT2017-88492- R and SEV2015-0522), the ERC (Advanced Grant 789104- eNANO), the Catalan CERCA Program, and Fundació Privada Cellex.

Fine-tuning of the nanoporous alumina photonic stopbands by using non-conventional pulse anodization

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Photonic stopbands structures (PSB) based on nanoporous anodic alumina (NAA) are engineered by different periodic anodization of aluminum under specific conditions. The optical properties of NAA structures rely intrinsically upon its nanoporous architecture, and on the geometry and distribution of its nanopores, which can be precisely engineered during the anodization process [1, 2, 3]. Some intrinsic properties such as photoluminescence can also be modified during and after their anodization.

Here we present a novel pulse anodization approach using a gaussian current density profile to engineer NAA photonic crystals with tuneable photonic bands in the UV-Vis-NIR spectral range.

The relationship between the technological parameters and the characteristic features of the photonic stopbands were successfully assessed.

The results provide insights onto optimal anodization conditions to fabricate high-quality NAA-based photonic crystals, opening new opportunities to engineer high-quality structures for light-based technologies such optical sensing, photocatalysts for green energy generation and environmental remediation, optical encoding and lasing [4, 5, 6].

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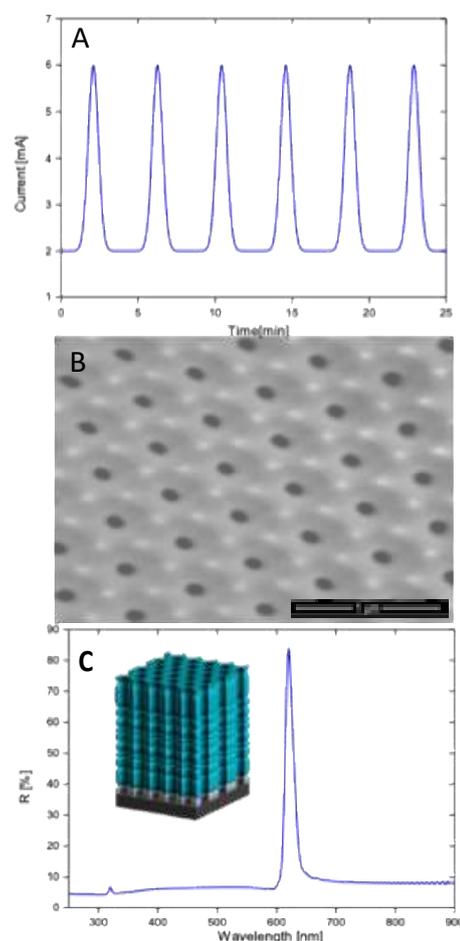


Figure 1: **A.** Gaussian pulse anodization current density profile. **B.** Top view ESEM image of NAA. **C.** Reflection spectrum of a gaussian NAA photonic crystal. Inset a schematic cross-section of the photonic structure.

The Chiral Induced Spin Selectivity(CISS) Effect

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Utilizing the Chiral Induced Spin Selectivity (CISS) effect we demonstrated a simple spin based nano magnet-less optical and electrical memory. The CISS effect also enables the developing of local spin qubit. The presented technology has the potential to overcome the limitations of other magnetic memory technologies allowing the fabricating of inexpensive, high-density universal and dense quantum and classical memory devices.

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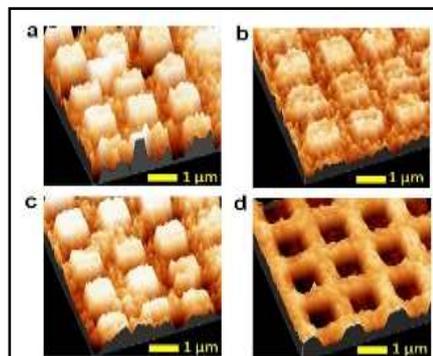


Figure 1: Topography and magnetic phase MFM images of a molecular-induced magnetization orientation.

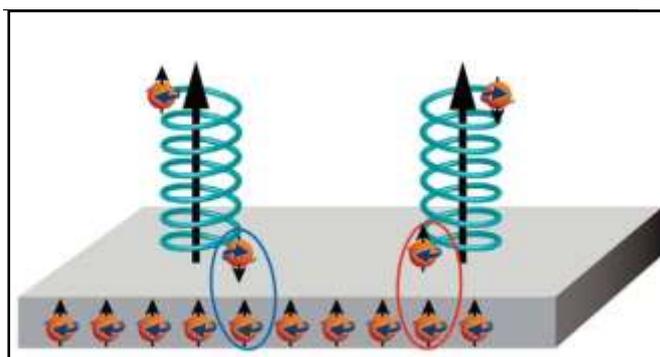


Figure 2: When an electric field E (black arrows) acts on a chiral molecule, due to its interaction with other molecules or with surfaces, charge reorganization occurs in the molecule and is accompanied by spin polarization.

Quantum nanophotonics with 2d materials

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The field of two-dimensional (2D) materials-based nanophotonics has been growing at a rapid pace, triggered by the ability to design nanophotonic systems with in situ control, unprecedented number of degrees of freedom, and to build material heterostructures from the bottom up with atomic precision [1]. A wide palette of polaritonic classes have been identified, comprising ultraconfined optical fields, even approaching characteristic length-scales of a single atom. These advances have been a real boost for the emerging field of quantum nanophotonics, enabling quantum technologies harnessing single-photon generation, manipulation, and detection using 2D materials. In my talk, I will show several hybrid systems consisting in lifetime-limited single emitters [2, 3] (linewidth ~ 40 MHz) and 2D materials at sub-wavelength separation without degradation of the emission properties [4]. We have demonstrated that their nanoscale dimensions enable ultra-broadband tuning (tuning range > 400 GHz) and fast modulation (frequency ~ 100 MHz) of the emission energy [5], which renders it an integrated, ultra-compact tuneable SPS. I will also present recent results on unusual Stark tuning of ultra-narrow quantum emitter located at the edge of a graphene transistor and electrostatic engineering of excitons in 2D semiconductors.

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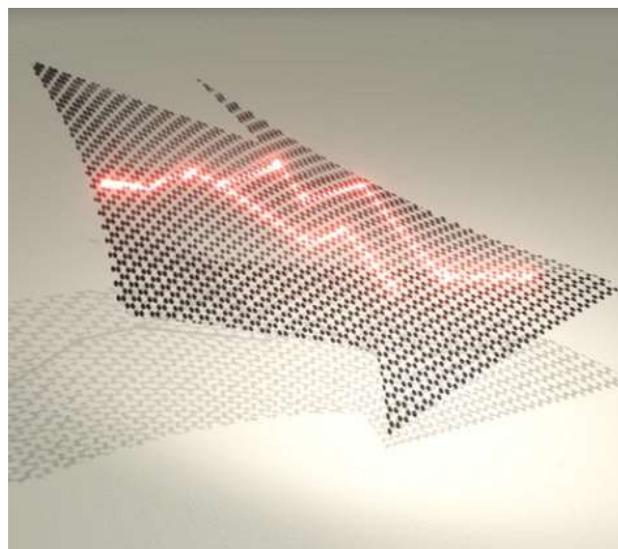
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Figures



Bound States in the Continuum in all-dielectric resonant metasurfaces

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Bound states in the continuum (BICs) have attracted much interest lately in photonics for their (theoretically) infinite Q factor. These states are leaky modes that in a certain limit of some parameter space cannot couple to any radiation channel [1]. In order to trap light in such nearly-zero-linewidth electromagnetic modes, a common approach is to exploit metasurfaces: outgoing specular channels can be suppressed by tuning the parameters of the system in various manners, leading to symmetry-protected BICs.

Here we will show that simple metasurface configurations may support robust, symmetry-protected BICs. On the basis of a generalized coupled electric/magnetic dipole theory for infinite arrays [2], a variety of scenarios are investigated where single/double meta-atoms can be simply described by a combination of various electric (ED) and/or magnetic dipoles (MD). First, a dipole-dimer array is shown to yield a BIC at normal incidence as the dipole detuning parameter vanishes; this has been experimentally verified through Au-rod dimer metasurface in the THz domain [3], unveiling the symmetry-protection mechanism through near-field excitation and detection [4]. Second, an array of single perpendicular

MDs exhibits a so-called Brewster BIC at normal incidence, which evolves into a quasi-BIC at oblique incidence with a rich phenomenology as the (non-degenerate) MD is tilted. We will show that a high-refractive-index disk metasurface in the GHz domain in turn provides clear experimental evidence of such Brewster quasi-BICs [5].

All these configurations can be exploited at the nanoscale on the basis of all-dielectric resonant metasurfaces, allowing for robust BICs in the visible domain with Si nanodisk metasurfaces [6], in turn leading to e.g. lasing action demonstrated in TiO₂ disk metasurfaces [7], and magneto-optical BIC tuning and switching, theoretically proposed in Ref. [8].

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Phononic Circuits: an Optomechanics-Mediated Demonstration

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In the quest to find low power information processes, phonons offers an attractive possibility. The key mechanism explored here is an strong interaction of coupled photons and phonons: optomechanics. We designed, simulated, realised and characterised a phononic circuit built in a nc-silicon-AlN platform consisting of a phonon source [1], an interaction region of phonons and photons in the form of a waveguide which exhibits switching behaviour [2] and a detector. The circuit operates at room temperature and at low power (< 1 mW) [3] and is based on 1-dimensional nanobeams which exhibit simultaneously a band gap for photons in the THz regime and for phonons in the GHz range. State-of-the-art optical and mechanical Q-factors are reached at 300 K.

The experimental realization has been described [4] and we demonstrated that optomechanical crystal cavities of Si and nc-Si have optical and mechanical properties that enable non-linear dynamical behaviour based on effects such as thermo-optic/free-carrier-dispersion self-pulsing,

phonon lasing 0.3 GHz in self-pulsing mode and at 5.5 GHz in back-action mode. Recently synchronization between two nanobeams has been obtained [5]. Our results open the door to advanced NOEMS as well as quantum applications. This work was funded by the EU H2020 FET Open project [PHENOMEN](#) (All-Phononic Circuits Enabled by Opto-mechanics), GA Nr. 713450.

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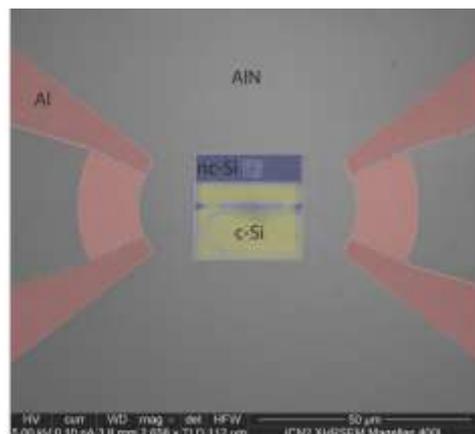


Figure 1: The core of the PHENOMEN platform. Left and right in orange depict the interdigitated electrodes for launching and detecting surface acoustic waves. The central square is the heart of the circuit. It includes a nanobeam actuated by a waveguide running in parallel and with a photonic crystal mirror termination This platform was demonstrated at 1 GHz [6] and 2 GHz (To be published).

Recognizing the face of wave functions

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Electromagnetic wave transport through disorder and correlated media shows a rich phenomenology. Depending on the degree of correlation and frequency, different light transport regimes emerge, while keeping other relevant parameters constant such as density and optical properties of scatters.

In this context, two dimensional stealthy hyperuniform structures (SHU) [1] have been recognized to present at least five different transport regimes, namely transparency [2], diffusion, Anderson localization, pseudo-tunneling [3] and tunneling through complete stop bands [4].

Determining the transport regime associated to an individual wave function has been traditionally addressed by computing the inverse participation ratio (IPR) [5] associated to the wave function. IPR estimates the number of scattering units involved in the build up of a particular wave function. Small values of IPR are associated to delocalized modes, while a large IPR corresponds to localized modes. Nevertheless, IPR and its generalized versions can not account for the variety of transport regimes found in SHU media.

Motivated by the successful application of machine learning techniques in this field [6], in this work we take a machine learning approach based on artificial neural networks to determine the transport regime associated to a particular wave function. A convolutional neural network (CNN), called EfficientNet [7], is trained with field maps of

more than 200'000 wave functions, labeled with its corresponding transport regime.

In our implementation we take advantage of most of the architecture of the CNN, designed to perform high accuracy image recognition, and adapt it by knowledge-transfer to perform wave function recognition.

When new field maps are shown to the neural network, we obtain a high accuracy (>92%) on the predictions on the transport regimes.

It is worth noting that no information about the frequency or structure of the underlying scattering system is presented to the artificial neural network.

In this work we discuss the ability of the neural network to successfully generalize to new data sets without re-training in cases where IPR is not a good indicator.

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ThermoMechanical Imaging and Characterization of Single Bacteria Cells

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We have developed a high throughput spectrometric technique addressing single biological entity (bacteria) resolution. This novel technique will be used to develop a novel imaging technique based on mechanical frequency shift of a nanomechanical resonator to generate a mechanical image of single particles and bacteria. The physical principle behind this technique is the modulation of the light absorption by the particle, which is translated into a thermo-mechanical effect on the nanomechanical resonator. This idea was recently demonstrated by using plasmonic gold particles of 100 nm in diameter [1] and to mechanically image viruses and bacteria cells (in press) of 700 nm in diameter. We will show not only the optomechanical coupling that emerges in the cavity formed by a plasmonic nanoparticle onto a free-standing silicon nitride membrane, but also the thermomechanical coupling by using dielectric particles. The optical absorption depends on the scatterer material; therefore, it is possible to unambiguously discern in between different dielectric particles and bacteria cells of the same size by simply analyzing the mechanical frequency shift while shining with a laser [2]. The optical absorption can also be tuned by playing around with the optical resonances of a photonic crystal, which allows to actively tune the mechanical resonance frequency [3].

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Figures

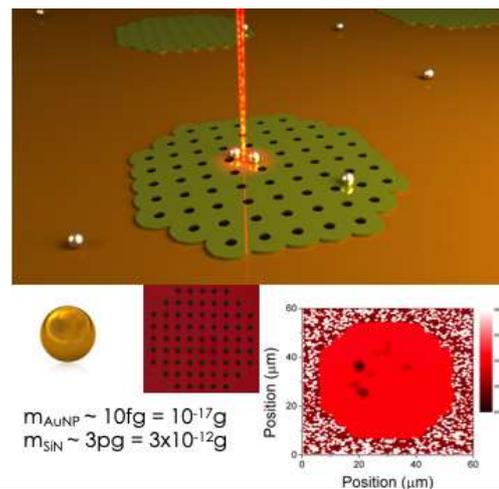


Figure 1: Figure. a. Schematic depiction of the system used for the mechanical imaging of gold nanoparticles of 100 nm in diameter and the experimental mechanical image of the whole membrane (adapted from [D. Ramos et al., *Nano Letters* **18**, 7165-7170 (2018)]).

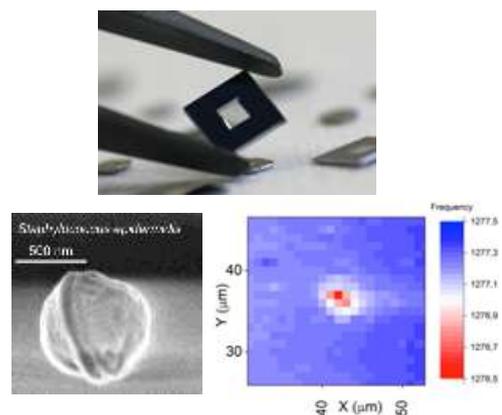


Figure 2: Membranes used for the mechanical image of *Staphylococcus epidermidis* (SEM image taken at CSIC) and the first experimental mechanical image (in press).

Advanced FIB-Patterning Strategies for seamless Photonic Devices

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Focused ion beam (FIB) systems are valuable tools for nanofabrication and rapid prototyping in R&D by providing direct and resistless patterning [1, 2]. Although FIB milling is typically slower than a resist process, the simplification of the nanofabrication approach can help to achieve faster results. However, FIB milling of large nano or micro devices is always a trade-off between resolution (low ion beam current for small beam size) and short process time (high beam current for large volume removal). Furthermore, patterning with conventional FIB-SEM tools is usually limited to a single field of view.

The system used here (Raith VELION) overcomes nanofabrication specific limitations of analytical FIB instruments by a dedicated lithography architecture.

Here we present FIB patterning of extended photonic crystals and mm-long waveguides. To overcome FIB milling related patterning artifacts like edge effects at stitch field boundaries, caused by higher sputter rates during milling of elements at an existing edge, advanced strategies have been developed. In case of hexagonal photonic crystals (Figure 1) dividing single circle elements at stitch field borders can be simply avoided by overlapping write fields and stage movement steps with the size of a base cell of the photonic crystal. For the fabrication of waveguides, which do not fit into a single field, more complex strategies like a combination of repetitive stage movements and field boundary shifts need to be applied to completely remove edge effect artifacts (Figure 2). Thus, mm-long waveguides can be accurately fabricated during an unattended automatic over-night process (Figure 3).

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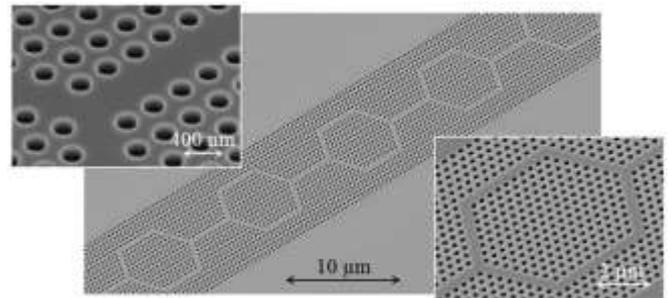


Figure 1: SEM micrograph of a 2 mm long photonic crystal device employing field stitching with overlapping write fields



Figure 2: Etch effect milling artifacts at stitch field boundaries: a) if no further strategies are

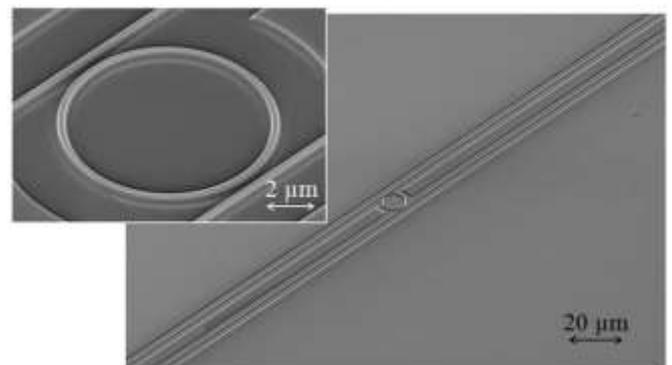


Figure 3: Extract of 3 mm long waveguide device including ring resonator (45° sample tilt).

Controlling Light Generation in Disordered Fractal Networks of Nanostructures

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Abstract

The use of light as a way to communicate and process information to and from the nanoscale is one of the technological milestones that is advancing innovation in modern times across many areas, from computer sciences and renewable energies to personalized healthcare and sensing technologies. While, to date, control of light flow at the nanoscale has mainly been achieved with periodically structured materials, disordered photonic nanostructures are slowly emerging as suitable easy-to-fabricate designs that can lead to performances superior to those offered by conventional photonic structures in, e.g., imaging and photovoltaics. Here, I will present recent experimental results where disordered fractal nanostructures allow us to generate and control light at the nanoscale. These novel disordered arrays of nanostructures can play a key role in controlling the light transport properties of complex photonic systems and, thus, in controlling their final optical properties, which are ultimately of interest to develop next generation optical devices.

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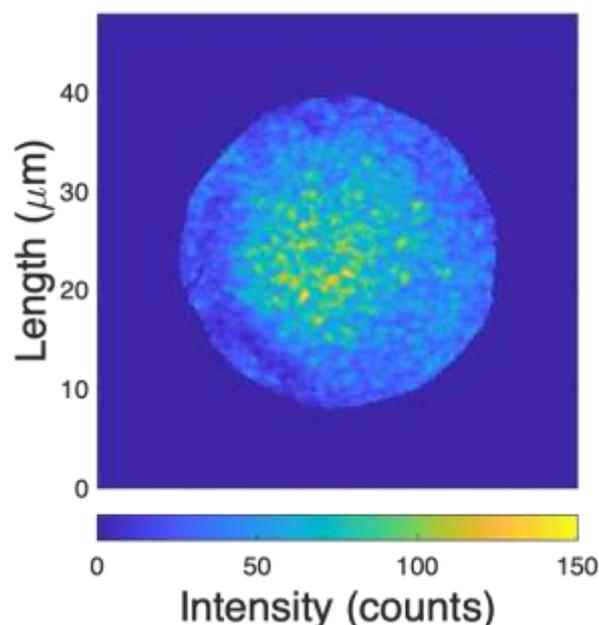


Figure 1: Optical microscopy image of the intensity of inelastic light generated by a fractal network of silicon nanostructures.

Planar refraction and lensing of nano-light in an anisotropic van der Waals material

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Refraction between isotropic media is characterized by light bending towards the normal to the boundary when passing from a low- to a high-refractive-index medium. However, refraction between anisotropic media is a more exotic phenomenon which remains barely investigated, particularly at the nanoscale. Here [1], we visualize and comprehensively study the general case of refraction of electromagnetic waves between two strongly anisotropic (hyperbolic) media, and we do it with the use of nanoscale-confined polaritons (hybrid light-matter waves) in a natural medium: α - MoO_3 [2-4]. The refracted polaritons exhibit non-intuitive directions of propagation as they traverse planar nanoprisms, enabling to unveil an exotic optical effect: bending-free refraction. Furthermore, we develop an in-plane refractive hyperlens, yielding foci as small as $\lambda_p/6$, being λ_p the polariton wavelength ($\lambda_0/50$ compared to the wavelength of free-space light). Our results set the grounds for planar nano-optics in strongly anisotropic media, with potential for effective control of the flow of energy at the nanoscale.

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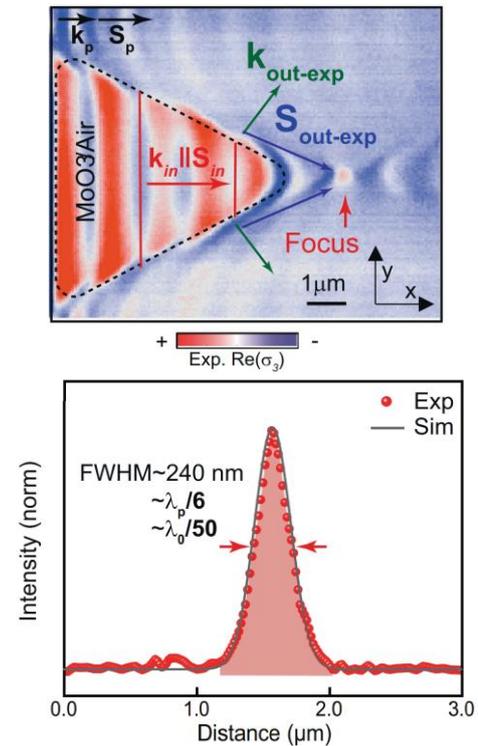


Figure 1: Sub-diffractional planar lens based on refraction of nano-light in an anisotropic vdW material.

Effect of Electronic Coupling in the Optical Response of Plasmon-Exciton Systems

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We use time-dependent density functional theory (TDDFT) to explore the influence of electronic coupling in the optical response of quantum emitters (QEs) interacting with metallic nanoparticles (MNPs) that support plasmonic resonances. Our model system consists of a QE, with a HOMO-LUMO transition, placed at the center of a (sub)-nanometric gap nanoantenna formed by two spherical MNPs (Figure 1a). The TDDFT calculations address, without any *a priori* parameter, the dynamics of the electronic states of both the QE and the MNPs under external illumination, thus going beyond the electromagnetic interaction picture that is typically considered when describing the optical response of such narrow junctions.

Our study reveals the importance of the electronic coupling between the QE and the plasmonic gap nanoantenna for separations of the order of 1 nm. First, the electronic states localized at the MNPs hybridize with the states localized at the QE, which produces a drastic quenching of the LUMO state that standard classical models cannot capture. This *electronic quenching* strongly modifies the energy and width of the optical resonances of the coupled system (Figure 1b). Moreover, for subnanometric cavities, the presence of the QE triggers a net electron transfer between the two MNPs at low illumination frequencies

[2,3]. The results in this work thus demonstrate that the electronic QE-MNPs coupling and charge-transfer processes between the emitter and the nanoantenna play a crucial role in the optical response of (sub)-nanometric junctions.

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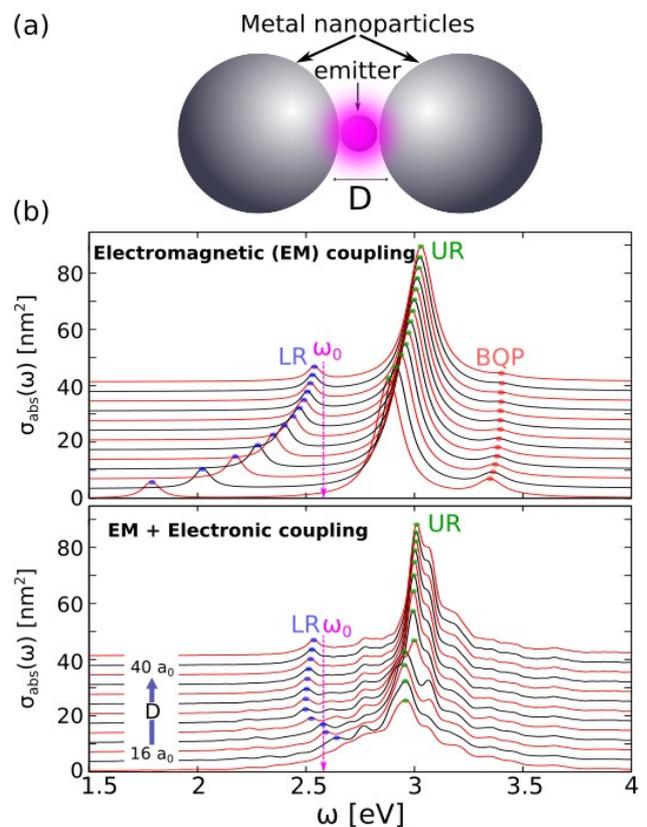


Figure 1: (a) Sketch of the studied system, consisting of a QE placed at the nanogap of size D formed by two MNPs. (b) Absorption cross-section spectra of the coupled QE-MNPs system calculated within a classical model only accounting for the electromagnetic (EM) interaction (top) and within a TDDFT description that includes both the EM and the electronic coupling (bottom).

Plasmon-exciton coupling and charge transfer plasmons in metallic cluster dimers

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Recent results have shown that atomistic ab-initio methods allow for an accurate description of the coupling between metallic clusters and molecules, capturing the physics of their plasmonic response [1]. We have studied the optical spectra of porphyrin molecules coupled to silver cluster dimers, using the ab-initio SIESTA [2] software to obtain the ground-state of the system, and the linear-response TDDFT code PyNAO [3] to compute the optical excitations. The results show the emergence of a Fano spectral line in the absorption spectra near the plasmonic resonance, a characteristic feature of the weak light-matter coupling regime. The strength of this coupling is very dependent on the geometric features of the system, such as the separation between the clusters or the orientation of the molecule. Our simulations also reveal the existence of a Charge Transfer Plasmon at lower frequencies for the smallest nanocavity sizes. These states are predicted to emerge when a molecular energy level is resonant with the Fermi level of the clusters [4]. By applying a shift of the local potential around the atoms belonging to the molecule, we control the energies of the molecular level relative to those of the metal clusters, allowing for the tuning of the strength of the charge transfer modes.

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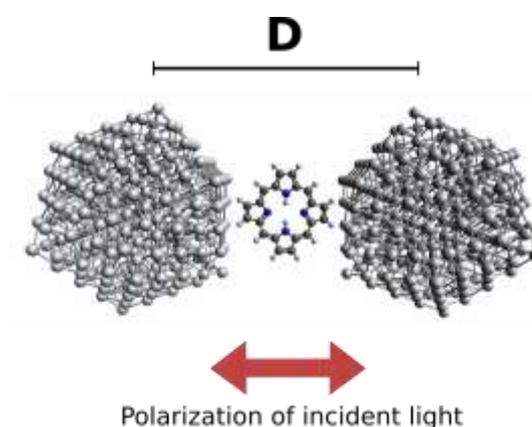


Figure 1: Atomistic structure of the studied system. It is composed of two Ag_{309} clusters with a 2,3-Dihydroporphyrin molecule placed in the gap between them.

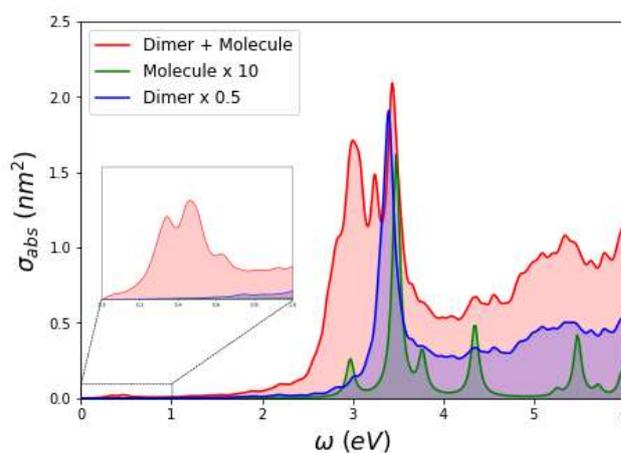


Figure 2: Absorption spectra of the bare silver dimer (blue), the bare single molecule (green), and the complete dimer and molecule system (red). The inset shows the Charge Transfer Plasmon emerging at low frequencies.

Plasmonic nanorattles for in situ SERS imaging of pH in microbial colonies

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It is well known that microbial populations and their interactions are largely influenced by their secreted metabolites. For instance, microbial fermentation processes often lead to the production of acids that can lower the local pH significantly, thus affecting the physiological state of resident microbes [1], promote resistance to antibiotics [2] or induce enamel demineralization and dental caries [3]. Therefore, non-invasive and simultaneous monitoring of extracellular bioactive metabolites and physicochemical factors (e.g. pH) can provide valuable information regarding the mechanisms that regulate the biogenesis, composition, and function of microbial communities. Herein, we report a SERS substrate consisting in plasmonic Au@Ag@mSiO₂ nanorattles embedded within an agar matrix for pH sensing in bacterial colonies. This multifunctional SERS substrate enabled us to efficiently perform spatiotemporal non-invasive detection and imaging of pH changes in colonies of *Escherichia coli*.

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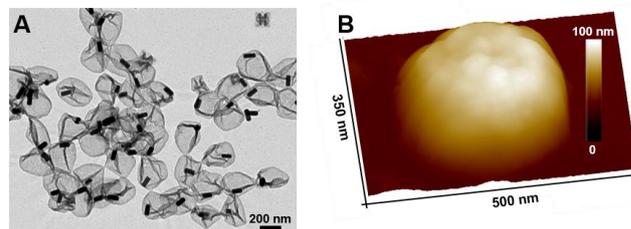


Figure 1: (A) TEM image of plasmonic mSiO₂ nanorattles. (B) AFM topographic 3D height image of a hydrated plasmonic mSiO₂ nanorattle.

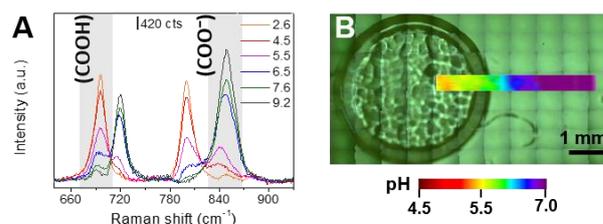


Figure 2: (A) SERS spectra of nanorattles@LB-agar substrates at different pHs. (B) Spatiotemporal pH distribution map of a selected area of the nanorattles@LB-agar substrate during the growth of a colony of *E. coli*.

Thermoplasmonic imaging: Free space propagation and confinement of 100-fs plasmons pulses in metallic structures

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Recent advances in nano-photonics lead to extreme light confinement (ELC) and light manipulation. This progress has spawned a variety of new important technological possibilities for the efficient delivery, control and manipulation of optical radiation on the nanoscale. Although the physical principles of ELC with plasmons i.e. nano-focusing has been clearly demonstrated in several studies, further fundamental studies are needed to optimise these processes and control losses in plasmonic devices for viable technological applications.

This talk will introduce the coupling of the ELC with the electron and lattice dynamics in metals. In one of our recent works, we have demonstrated the capability to image and film plasmon propagation in a metallic film. We probed the hot electrons heated by the plasmon dissipation via a Time Domain Thermoreflectance (TDTR). Figure 1 describes an appropriated designed plasmonic device where plasmons can be concentrated at the apex. TDTR is a unique tool to reveal and study energy transport processes induced by ELC in nanometric devices that have not been explored so far in low-dimensional systems.

We have detected and imaged the hot carriers generated in the hot spot and

exploited the mechanism of plasmon absorption in metals for the generation of hot carriers at femtosecond time scale, and this energy conversion was measured with femtosecond pump-probe technique. Femtosecond plasmon pulses will be launched and probed over hundreds of femtoseconds through the permittivity variations induced by the hot-carriers.

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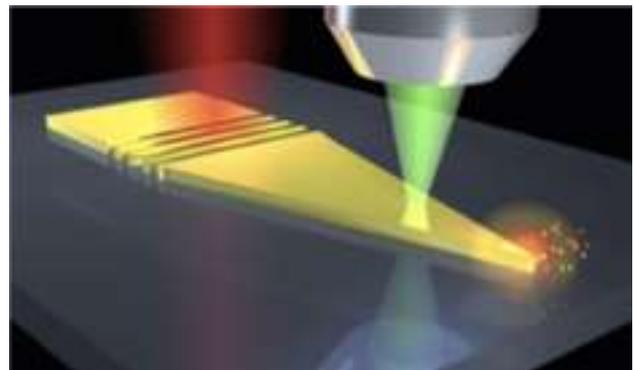


Figure 1: Schematic illustration of the experimental setup. The grating is shined by the red beam launches a plasmon focused at the tip apex producing hot carriers. The green laser probes the hot electrons during the focusing process.

Optical topological transition: enable canalization and control of highly confined polaritons

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Abstract

Recent discoveries have shown that when two layers of van der Waals (vdW) materials are superimposed with a relative twist angle between their respective in-plane principal axes, the electronic properties of the coupled system can be dramatically altered due to the topological transition of electronic band structure. Here, we demonstrate that a similar concept can be extended to the optics realm, particularly to propagating polaritons – hybrid light-matter interactions –. We demonstrate two types of optical topological transition: Type I (from closed to opened isofrequency curve)^[1] and Type II (isofrequency curve closes at one point)^[2]. These optical phenomena appear in either stacks composed of two twisted slabs of a polar vdW crystal (α -MoO₃) or heterostructures composed of in-plane anisotropic (α -MoO₃) and isotropic (4H-SiC) polaritonic crystals. Our nano-infrared images reveal that the propagation of polaritons can be strongly guided along predetermined directions (canalization

regime) with no geometrical spreading (Figure 1). Moreover, the topological transition enables propagation of anisotropic polaritons along forbidden directions. These results demonstrate new degrees of freedom (twist angle or heterostructure) for controlling the propagation of polaritons at the nanoscale with potential for nano-imaging, (bio)-sensing, quantum applications and heat management.

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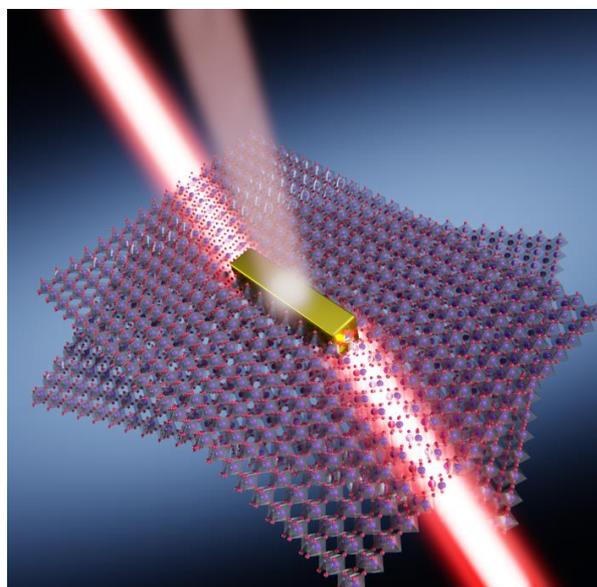


Figure 1: Canalized phonon polaritons in twisted van der Waals layers. Under a critical angle the polariton isofrequency curve undergoes a topological transition, generating the canalization of polaritons (i.e. propagation along one specific direction without diffraction).

Cove-edged nanographenes as a potential optical-gain media for lasing

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Abstract

The application of zigzag- and armchair-edged nanographenes (NGs) for the development of solid-state lasers has been compromised by their limited solubility and spontaneous aggregation^{[1][2]}. In this work, we study a cove-edged nanographene, hexa-peri-hexabenzobis-peri-octacene (**HBPO**), a novel material with improved solid-state solubility due to its contorted geometry and bulky substituents. In order to characterize the optical gain, we performed Transient Absorption Spectroscopy (TAS) measurements of **HBPO** in solution and in a **HBPO**-PS thin film composite. In solution, stimulated emission (SE) is revealed by a sharp feature at 613 nm coinciding with the 0-0 vibronic PL transition. TAS measurements of the composite confirmed an enhanced performance where SE outbalances excited-state absorption, paving the way for its application as solid-state optical gain medium. This conclusion was corroborated by Amplified Spontaneous Emission (ASE) measurements, obtaining an ASE threshold

around 2.4 mJ/cm² in **HBPO**/PS films upon pumping at 465 nm. Our results are indicative of the great potential of NGs with cove edges for solid-state lighting applications.

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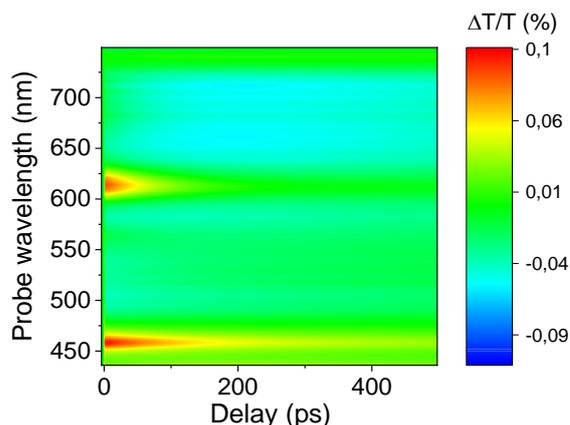


Figure 1: Fitted TA maps for **HBPO** in a **HBPO**/PS thin film taken at different pump-probe time delays.

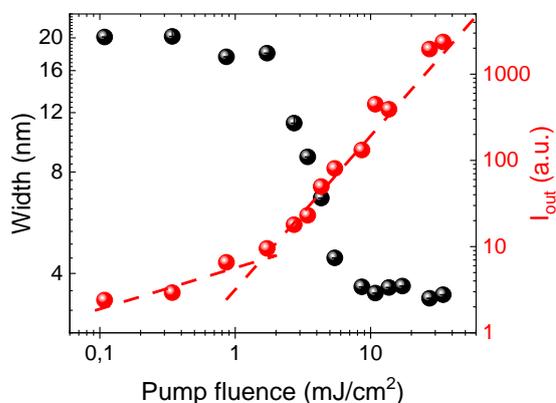


Figure 2: Input-output characteristics of ASE action and ASE threshold for the **HBPO**/PS thin film.

Towards Label-Free SERS Detection: Universal Fabrication of Highly Efficient Plasmonic Platforms

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Being characterized by high sensitiveness, surface-enhanced Raman scattering (SERS) spectroscopy currently represents one of the most promising analytical techniques in the field of sensors development. [1] Generally, identification of the analytes relies on the enhancement of the Raman active modes of target species or their Raman reporters when in close proximity to plasmonic substrates. High sensitivity can be ensured by the accurate design and engineering of the hot-spot size and density, i.e. the optimization of the interparticle distance. Herein, we report the fabrication of highly efficient plasmonic sensing platforms by electrostatic layer-by-layer deposition of metal nanoparticles (NPs). In order to optimize the response, we investigated the role of the NPs size and composition, as well as the hot-spot size and density. The analysis of the SERS performance of all the different platforms revealed that 89 nm Au@Ag core@shell NPs ensured the best SERS efficiency when electrostatically assembled using GO@PDDA (viz. poly(diallyldimethylammonium chloride) - functionalized graphene oxide) as interlayer. (Figure 1)

The high performance of this plasmonic platform as a label-free SERS substrate has been successfully tested by comparing the SERS and Raman spectra of methylene blue, methyl orange, and dopamine in Milli-Q water. Furthermore, we challenged our sensing platform to detect tamoxifen (TAM), a well-known anticancer cytotoxic drug, which has been found as

contaminant in hospital effluents. We were able to observe a linear variation of the signals of TAM as a function of its concentration (0.09 – 0.9 mg mL⁻¹) achieving a limit-of-detection of 3 × 10⁻² mg mL⁻¹, outperforming the previously reported SERS sensors (3.7 × 10⁻¹ mg mL⁻¹). [2] In addition, TAM detection was successfully achieved even in a complex matrix such as tap water. (Figure 2) [3]

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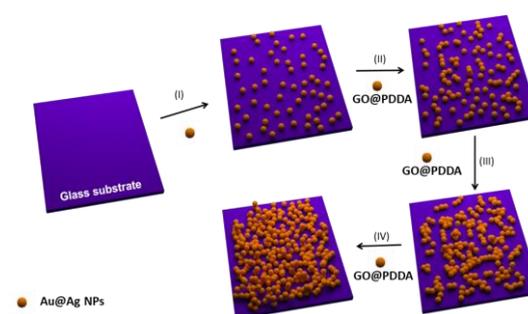


Figure 1: Schematic representation of assemblies of Au@Ag NPs fabricated by electrostatic LbL deposition using GO@PDDA as interlayer.

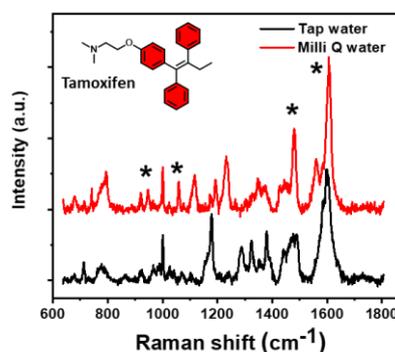


Figure 2: Representative SERS spectra of 0.9 mg mL⁻¹ TAM in Milli Q and tap water.

Surface Enhanced Raman Scattering, from labeling toward an analytical technique

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Surface Enhanced Raman Scattering arises when an exciting electromagnetic field resonates with localized plasmons of nanostructured surfaces. Molecular species experience enhanced local fields on the surfaces and the Raman scattering is amplified. Aggregated Gold nanoparticles,[1] as well as Gold nanostars,[2] are examples of some of the best candidates as colloidal SERS substrates. Labeling [3] or contrast agents [4], both in vitro or in vivo, have shown to be efficient when based on such substrates. The evolution of SERS toward a reliable quantitative technique is now attracting attention, due to the information present in the vibrational spectrum and the possibility to run measurements in aqueous samples. From previous experience on un-functionalized substrates,[5] a novel strategy for a quantitative assay based on a competitive approach was developed.[6] Janus magnetic/plasmonic FeO_x/Au nanostars (JNS) are here presented as successful colloidal SERS substrates for quantification of several analytes within the nano-micromolar range. Their magnetic behaviour was found useful for remote-controlled sample concentration within a microfluidic device, while their superior SERS activity was used for the quantification.

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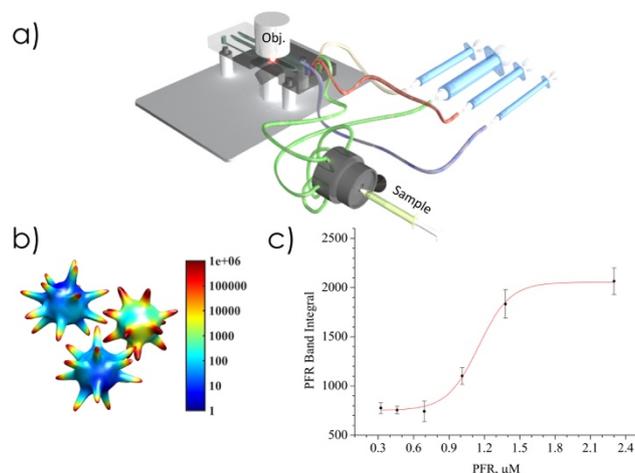


Figure 1: a) representation of the 3D printed microfluidic device in which the JNS interact with the analyte and concentrate at the magnet before the Raman measure; b) Boundary Element Method based simulations show high local field enhancements at the star tips; c) calibration curves of several analytes can be obtained within the nano-micromolar range.

Nonlinear Optics through optimal dye encapsulation into zeolitic nanochannels

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In this work, a styryl dye (trans-2-[4-[(dimethylamino)styryl] benzothiazole, DMASBT) is encapsulated into several aluminophosphates (MgAPO-11, MgAPO-5 and MgAPO-36, Figure 1) with different unidirectional nanochannel dimensions by the crystallization inclusion method. The synthesis of the hybrid material has been optimized through a systematic variation of the conditions in order to obtain the ideal optical properties for our hybrid system [1].

The tight-fitting between the molecular size of the guest dye and the pore dimensions of the host, favour a rigid planar conformation of the dye, restricting its inherent flexibility and boosting its fluorescence. The latter is crucial to enhance the non-linear optical (NLO) properties of the dye, which has successfully been achieved only in the particular case of MgAPO-11 framework.

Not only does the pore size of the framework lead to distinctive photophysical properties but also the formation of two different cationic species during the synthesis. In this sense, the pH plays a key role since allows us to adjust the amount of the desirable cationic species [2]. Therefore, the perfect alignment of DMASBT dye along the channels of MgAPO-11 has led to attractive non-linear optical

properties, proven through microscopy measurements (Figure 2).

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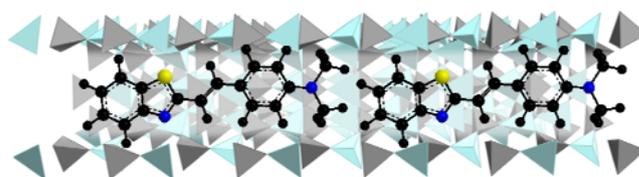


Figure 1: DMASBT dye molecules inside the unidirectional nanochannels of MgAPO-11 aluminophosphate.

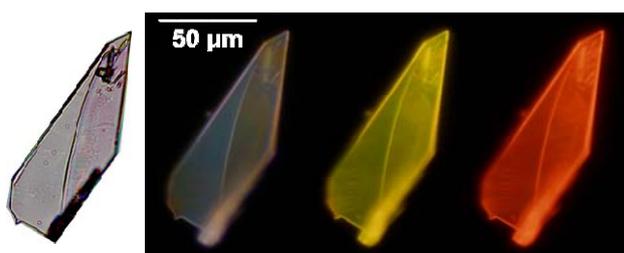


Figure 2: Transmission (white background) and emission images (black background) of a DMASBT-AEL single-crystal upon UV light, blue light and green light excitations.

On the helicity conservation: Detection of brand-new dipolar regimes

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In this Letter we demonstrate that the conservation of the electromagnetic (EM) helicity, signature of the EM duality restoration, can be used as a probe of single-polar spectral regions, particularly, electric and magnetic dipolar regimes. Our proof is solely based on a fundamental mathematical property of the Bessel functions that precludes the zero optical condition in a multipolar scattering process. Interestingly, we derive that the optimum forward light scattering, predicted for a diamond sphere, presents an infinite number of solutions for a fixed x size parameter, in striking contrast to previous findings.

Figure

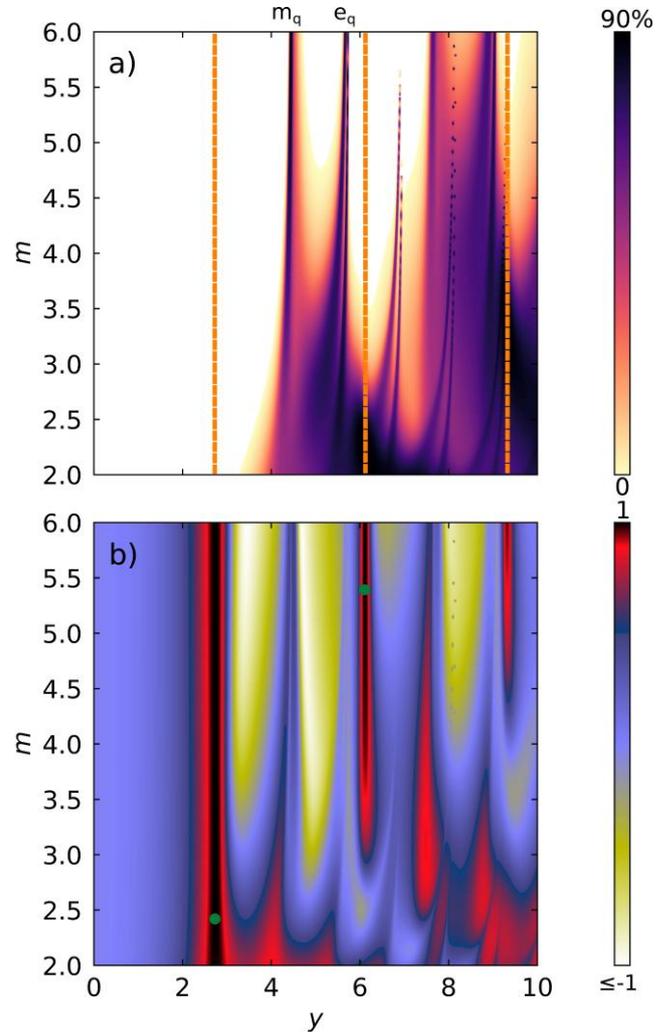


Figure 1: (a) Percentage error of presuming a dipolar response versus the $y = mx$ size parameter and contrast index m . The validity of assuming a dipolar regime, corresponding to the white region, extends beyond the magnetic and electric quadrupole for certain spectral regions. The first Kerker condition is as well illustrated for completeness by vertical dashed orange lines. (b) Color map of the expected value of the EM helicity after scattering by a Mie sphere under well-defined EM helicity plane wave illumination. As it is depicted in the attached color-bar, the EM helicity is preserved in the dipolar regime. The green circles illustrate two different materials fulfilling the optimum backward light scattering condition.

Unveiling atomic-scale features in plasmonic nanoparticles using electron beams

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Electron energy loss spectroscopy (EELS) in scanning transmission electron microscopy (STEM), together with optical spectroscopies, has played a crucial role in understanding the properties and dynamics of plasmons in nanoparticles (NPs). In particular, technical progress in the performance of STEM-EELS microscopy in the last two decades has enabled sub-nanometer resolution and sub-eV energy sensitivity in EELS, opening new opportunities for characterization of novel materials and nanostructures. In this sense, *ab initio* atomistic methods such as Time-Dependent Density Functional Theory (TDDFT) provide an appropriate quantum framework to consider sub-nanometric atomistic features in plasmonic NPs [1,2]. Indeed, most of the classical and semiclassical theories rely on spherical descriptions of the NP's geometry, disregarding any dependence of the NP's shape and orientation on the electron energy loss (EEL) spectra. We have studied within atomistic TDDFT the influence of atomic-scale features in small plasmonic NPs on the EEL spectra of electron beams passing nearby and through atomistic NPs, revealing that EEL spectra strongly depend

on the orientation of the atomistic clusters (Fig 1). Moreover, we show that classical electrodynamics (Boundary Element Method, BEM) models are able to reproduce the EEL spectra corresponding to localized surface plasmons (LSPs), if the shape of the NP is properly described, but fail to address the confined bulk plasmons (CBPs) observed in the TDDFT calculations [3]. The CBPs show correspondence with those observed within hydrodynamical models for spherical NPs [4]. These findings bear out the importance of a proper consideration of the atomic-scale shape of nanoparticles in EEL spectroscopy.

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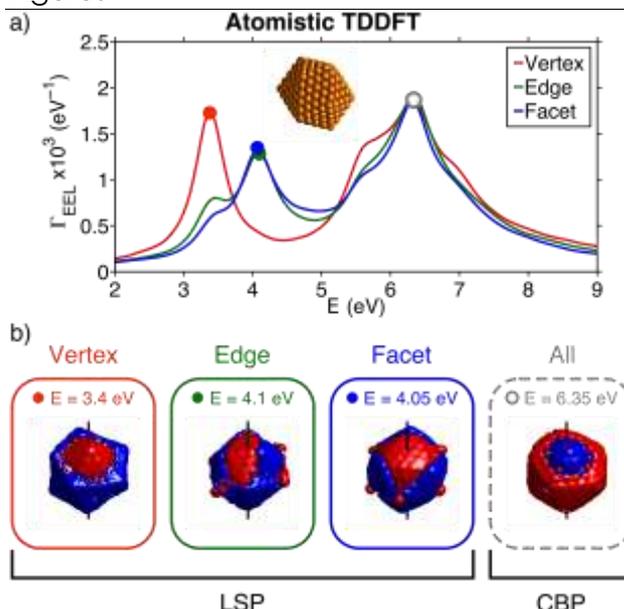


Figure 2: Electron energy loss spectra for the Na icosahedral cluster calculated using TDDFT for electron trajectories penetrating the NP through different atomic-scale features. b) Charge density isosurfaces corresponding to the main excited plasmon modes for each trajectory.

Spintronic-plasmonic antenna metasurfaces for molecular sensing

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Hybrid platforms merging metallic antennas and materials with specific functionalities offer excellent technological opportunities for active plasmonics, as they provide large changes in their optical response, which can be activated by external stimuli. In this talk I will focus on the magnetic modulation of the optical response of spintronic metasurfaces composed of microantenna arrays fabricated out of giant magnetoresistance Ni₈₁Fe₁₉/Au multilayers [1,2]. In this case, the plasmonic response of the antenna, combined with the Magneto-Refractive Effect (MRE) of the multilayer, allows for low magnetic-field control of the modulation of the optical response in the mid-infrared. Moreover, the relative difference in optical transmission (MR signal) with (T_P) and without (T_{AP}) magnetic field shows the presence of molecules (PMMA deposition in Fig. 1). Our experimental and theoretical results suggest that these GMR plasmonic metasurfaces are excellent candidates to improve the molecular detection capabilities of traditional Surface-Enhanced Infrared Absorption (SEIRA) Spectroscopy platforms and develop a novel type of infrared sensing technique based on spintronic antennas [3].

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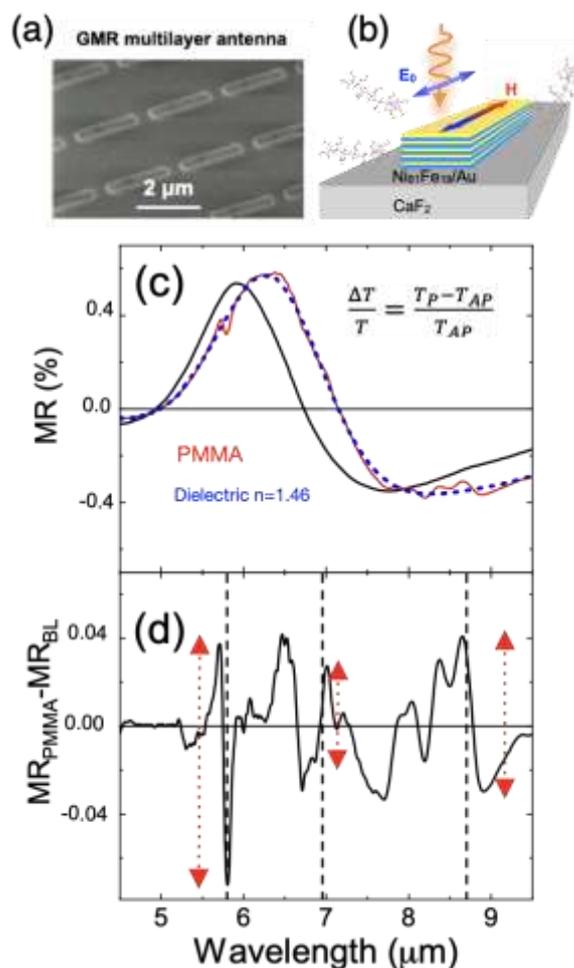


Figure 1: a) SEM image of the rod antenna array. b) Sketch of the GMR antenna structure, applied magnetic field and polarization of incident light. c) MR signal of the bare antenna array (black), covered with PMMA (red) and covered with a dielectric of $n=1.46$ (blue). d) Difference between the MR transmission spectra after PMMA deposition and the baseline spectrum. Red arrows show the contrast of vibrational features (dashed black) of PMMA.

Designing plasmonic structures for an optimized local thermo-optical response

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Abstract

Nowadays, efficient local heat generation using metallic nanoparticles is a research field of growing interest. These optically excited plasmonic structures often show large resistive losses and are therefore capable of converting optical energy into heat efficiently [1]. The spatial distribution and the magnitude of the obtained temperature mainly depends on the geometry and composition of the nanoparticles; thus, different nanostructures can be designed to act as nanoheaters, opening a wide range of fascinating applications. Some examples are the degradation of micropollutants [2] or plasmonic photothermal therapy [3].

These applications often require biocompatible materials and a high particle tunability to resonate in the target wavelength domain. Here, we present our most recent investigation on this topic, showing an exhaustive comparison of different efficient and tunable nanoheater prototypes.

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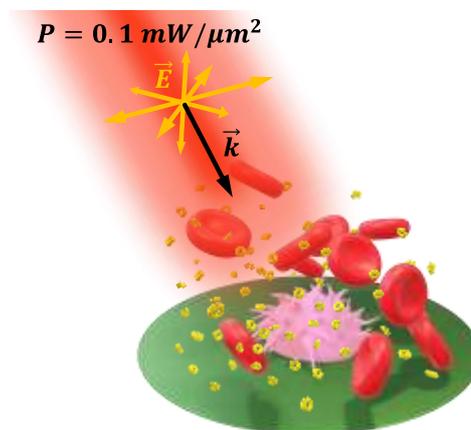


Figure 1: Illustration of blood red cells and cancerous cells being treated with toroidal particle-assisted photothermal therapy.

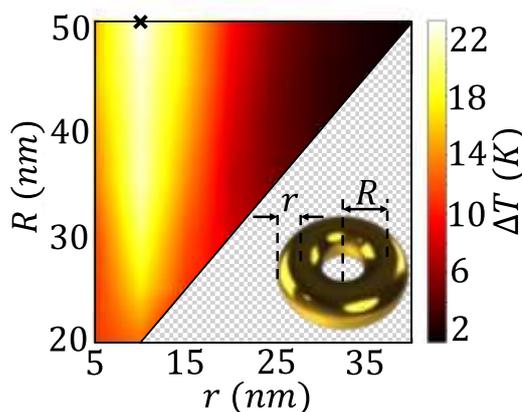


Figure 2: Colormap of the maximum temperature increment reached by a set of toroidal particle designs.

Photonic Band Structure Calculation of 3D-Finite Nanostructured Supercrystals

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Abstract

In the last decades, plasmonic nanostructures have made possible the miniaturisation of photonic devices such as photonic crystals for sensing, photonic integrated circuits, etc. [1,2] The light-matter interaction in such supercrystals (Figure 1) are dependent in the lattice type, particle size, shape, and composition, as well as microcrystal habit. With the countless superlattices now synthetically realizable, computational methods and theoretical models play a crucial role in identifying the supercrystals that exhibit the most exciting properties. To tackle this problem, two approaches are generally taken (i) an effective medium theory approach which neglects the nanoscale effects to focus on the overall optical properties of the supercrystal, and (ii) the use of a unit cell with periodic boundary conditions which neglect the overall habit of the supercrystal to focus on nanoscale behaviour. This second approach is used for the calculation of the photonic band structure of these periodic structures. However, it fails to describe the photonic properties arising from finite-size effects such as Fabry-Pérot resonances. Here, we developed a computational approach, based on FDTD electrodynamic method to accurately calculate the photonic band structures from finite, microscale 3D supercrystals of cubic, spherical, and rhombic dodecahedral habits.

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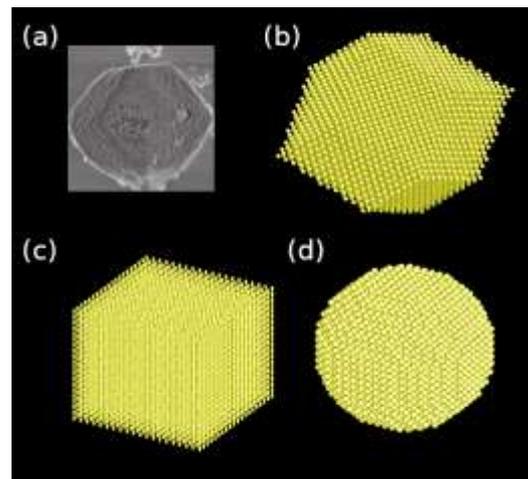


Figure 1: (a) SEM image of Au-nanoparticle rhombic-dodecahedral supercrystal [2]. (b)-(c) Au nanoparticle supercrystals with rhombic dodecahedral, cubic, and spheric habits

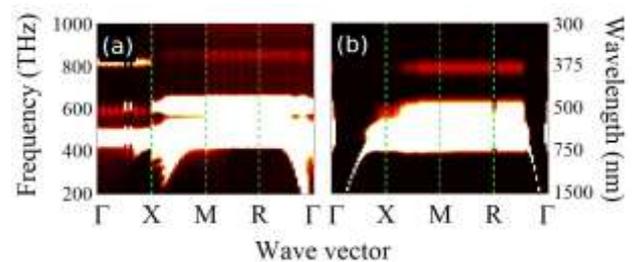


Figure 1: Photonic band structures of a (a) finite cubic lattice supercrystal with cubic habit of 2 μm in length and (b) infinite cubic lattice supercrystal both with distance of 20 nm between spherical 80-nm Au NPs

Grating metasurfaces as directional plasmon sources: applications in achiral and chiral sensing

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Optical biosensing is currently a highly active research area, particularly with respect to chiral compounds [1], where an increasing demand of highly selective, sensitivity-enhanced and low-cost devices has been noted. Plasmonic devices have risen as an alternative approach to conventional methods [2].

Our work focuses on the use of gold diffraction gratings to excite Surface Plasmon Polaritons (SPPs), using their intensity to characterize relevant features of the sample, such as their refractive index and their chirality.

We will present our current research on these devices. Using computational FDTD methods we demonstrate the capabilities of a gold grating to detect small changes in the refractive index of analytes upon wavelength interrogation, with a sensitivity up to 1500 nm/RIU [3]. Furthermore, we shall show its ability to excite plasmons directionally at non-normal incidences.

Then, using a hybrid dielectric-gold system, we demonstrate how the directional plasmon approach characterized in other works [4] can be applied to chiral sensing, with a structure that offers chiral enhancement in both the near and far field regimes, while presenting high-dissymmetry areas available to analytes.

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Figures

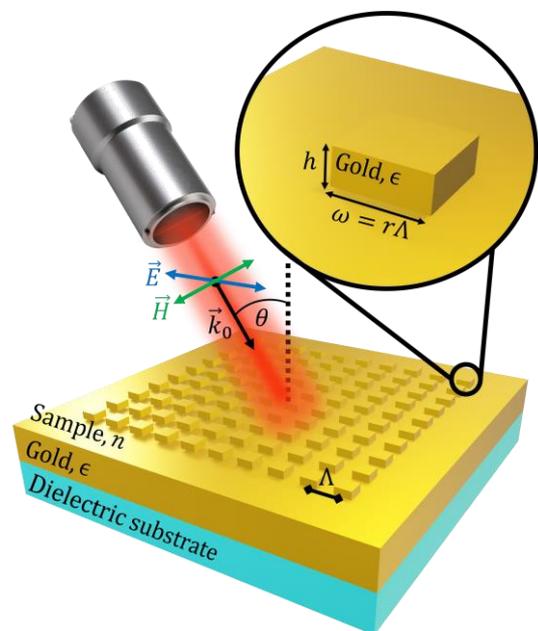


Figure 1: Operation diagram of the gold grating device for refractive index sensing.

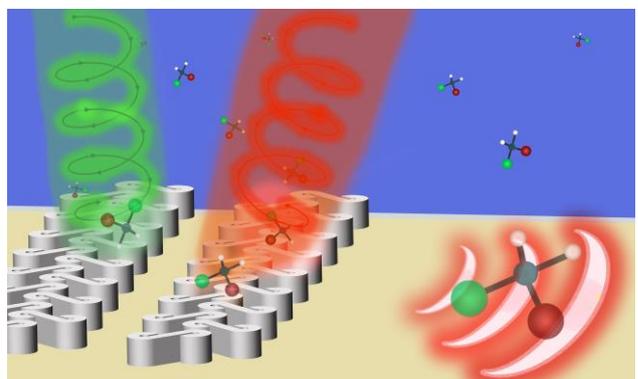


Figure 2: Operation diagram of the hybrid metasurface for chiral sensing.

Zero-to-full transmission switch with GST225-loaded all-dielectric metasurface

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Nonlinear and tunable nanostructures provide extraordinary abilities to control the optical response of modern functional photonic devices at the nanoscale. Recently emerged phase change materials demonstrate a dramatic and reversible change in dielectric properties under the influence of laser pulses or electric current and hence provide an enormous potential for integrated tunable nanophotonic devices [1,2].

In this work, we show that the use of phase-change material (GST225) allows us to design an all-dielectric metasurface with zero-to-full transmission switch occurring at the phase transition. The structure is designed to operate at the telecom wavelength (1.5 μm) vital for all-optical communications and data processing. Metasurface geometry is represented in fig. 1. It consists of silicon nanodiscs placed atop GST nanodiscs with the same diameter. Light transmission/reflection under normal incidence in the NIR range demonstrates a dramatic change in transmission (fig. 1). Figure 2 shows the effect of zero-to-full transmission switch in this structure occurring in the case marked with a cross in fig. 1. We believe that this metasurface can be used as a power limiter in nanophotonic devices.

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Figures

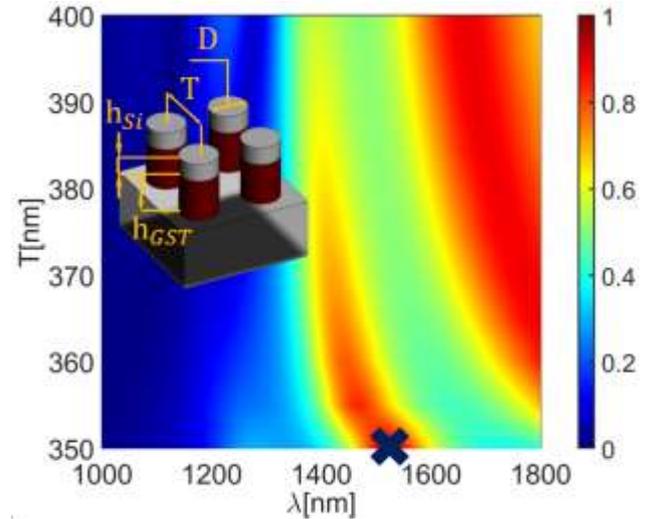


Figure 1: Absolute value of transmission difference between amorphous and crystalline states of GST nanocylinders vs period T and wavelength λ . Inset: schematic geometry of the metasurface. The regime of interest is marked with the cross.

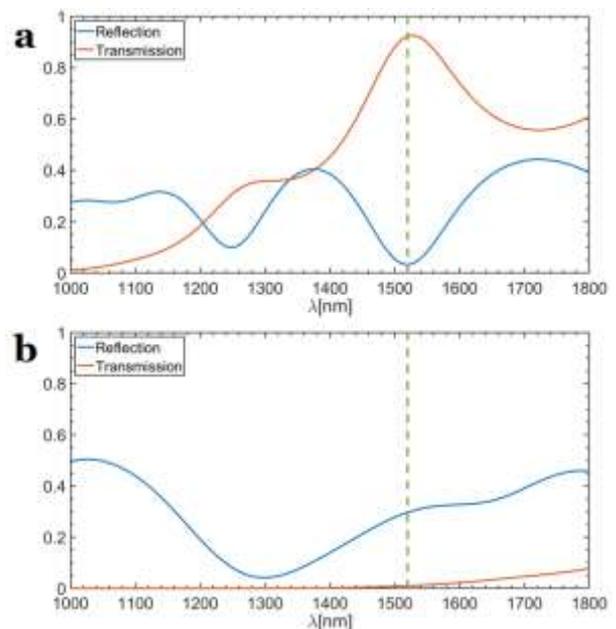


Figure 2: Reflection (red) and transmission (blue) for (a) amorphous and (b) crystalline states. The peak in transmission at 1520 nm in the amorphous state corresponds to the mark in fig. 1.

What it is 1



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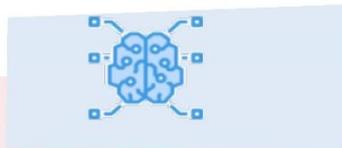
... with opportunities for industrial, business and institutional development

2 Its objectives



Flagship Areas 3

Neurobiosciences



Quantum Technologies (QT)



NeutriOnics



High Performance Computing & Artificial Intelligence (HPC-AI)



4 Expected impact

Research personnel



+400

Publications



+4.000

EPO Patents



+30

Spin offs



+20

Turnover



+350 M€

Employment



+3.200

5 Governance

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Biofisika Biokala Fundazioa
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Scientific Direction of IKUR's Flagship Areas