

MOLECULAR PLASMONICS 2025

MAY 15 - 17, 2025

Leibniz IPHT // Campus Beutenberg // Jena

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THURSDAY, MAY 15, 2025

Leibniz IPHT, Campus Beutenberg Bus no. 10, 11, 12

Satellite-Workshop "DNA Mitteldeutschland" (free to join)

12:00 Get-Together & Lunch Buffet

13:00 Presentations

17:00 END

Bus to downtown + 10 min walk, or appr. 40 min walk

Botanical Garden, Downtown Entrance Fürstengraben 26

18:00 Get-Together // Guided Tours

19:30 **DINNER**

FRIDAY, MAY 16, 2025

Leibniz IPHT, Campus Beutenberg Bus no. 10, 11, 12

8:00 Registration

9:00 Opening & Introduction // Wolfgang Fritzsche

9:05 Session 1

Catalytic Plasmonic Nanoparticles and Their Applications // Itamar Willner (Jerusalem)

The Importance of Surface Sensitivity in the Optimization of Plasmonic Sensors // Attila Bonyár (Budapest)

Challenges in Fabrication of LSPR Sensors for Molecular Detection // Tomáš Lednický (Jena)

10:20 COFFEE BREAK

11:00 Session 2 // Itamar Willner

Organic Excitonic Nanostructures for Metal-free Plasmonics // Jussi Toppari (Jyväskylä)

Stark Control of Plexcitonic States in Incoherent Quantum Systems // Hira Asif (Antalya)

Quantitative Photocurrent Scanning Probe Microscopy of Plasmon-Coupled Quantum Dots // Joachim Krenn (Graz)

Molecular Vibrations for Quantum-Enhanced Imaging and Spectroscopy // Markus Raschke (Boulder CO)

12:40 LUNCH

14:00 Session 3 // Markus Raschke

Surface Lattice Resonances for High-Performance Sensing: A Study on Gold Nanoparticle Arrays // Cesar Herreno (Bogota)

Self-Assembled Plasmonic Lattices and Their Applications // Tomas Tamulevičius (Kaunas)

CuFeS, **Semiconductor Nanocrystals: An Alternate Plasmonic Catalyst** // Biswajit Bhattacharyya (Potsdam)

Metal Photoluminescence as Probe for Hot Electrons // Wouter Koopman (Potsdam)

Plasmon-Induced Regioselective Grafting of Colloidal Gold at Hot Spots: Boosting Light Emission Efficiency // Nordin Felidj (Paris)

Poster Pitch Talks // Menbere Mekonnen

16:20 COFFEE BREAK & POSTER SESSION

17:00 Excursion // Hike to Ernst Haeckel Monument

19:00 POSTER & BEER (& BARBECUE)

SATURDAY, MAY 17, 2025

Leibniz IPHT, Campus Beutenberg Bus 10 leaves 8:35 a.m. downtown

9:00 Session 4 // Joachim Krenn Plasmonic Actuating and Continuous Monitoring Biosensing // Jakub Dostalek (Prague)

Plasmonic Single Molecule Detection Enhanced by DNA Nanostructure // Naoto Asai (Wiener Neustadt)

Optimizing Light for Plasmon Enhancement // Volker Deckert (Jena)

Altering the SERS Spectra of Molecules on Nanostructured Surfaces by Laser Focusing / Defocusing // Cosmin Farcău (Cluj-Napoca)

10:40 COFFEE BREAK

11:00 Session 5 // Jussi Toppari

Morphological Adjustment of Gold Nanoparticles AuNPs Through Polydispersants // Daniel Llamosa (Bogota)

From Development to Detection: Dendritic Nanostructures in SERS for Advanced Biomolecular Analysis // Aradhana Dwivedi (Jena)

Large-Scaled Plasmonic Assemblies for Efficient SERS Enhancements // Swagato Sarkar (Dresden)

Molecular Effects of NBD-Cl and NBD-Ceramide on Living Cells Using SERS // Yiqing Feng (Berlin)

12:40 END OF THE SESSIONS & LUNCH

LOCATION



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www.leibniz-ipht.de



HIKE LEIBNIZ IPHT TO HAECKEL STONE

2.7 km roundtour, height differenc 140 m



- A Höchster Punkt 330 m
- ▼ Niedrigster Punkt 190 m



↔ 2,36 km ↗ 140 m



Itamar Willner

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Nucleic acid-modified Au plasmonic nanoparticles (NPs) exhibit unique catalytic and optical properties guided by the hybrid nucleic acid/NPs conjugates. The hybrid DNA/NPs emerging functions find diverse applications reflected by controlling materials properties, sensing and biomedical therapies. These functions will be highlighted by several examples:

- 1. Au NPs play a key role in the area of nanozymes. Au NPs reveal peroxidase-like activities or oxidase activities, yet, these catalytic activities are substantially lower as compared to the analog native enzymes. Conjugation of nucleic acids to the Au NPs conjugates can significantly improve the catalytic functions of Au NPs nanozymes.
 - (a) Stabilization of Au NPs with poly A (pA)-aptamer units yields effective peroxidase or oxidase "aptananozymes" catalyzing the oxidation of dopamine to aminochrome or the cascade oxidase/peroxidase oxidation of dopamine by glucose using reactive oxygen species (ROS) as reactive agents¹.

By stabilization of Au NPs with aptamer sequences recognizing cancer-cell receptors (nucleoline of MUCI), AS 1411-aptamer or MUCI aptamer-modified Au NPs acting as aptananozymes for *in vitro* and *in vivo* chemodynamic treatment of breast cancer cell/tumors were demonstrated.

- (b) Polyadenine (PA) hairpin frameworks (bi-arm, tri-arm, four-arm) were prepared and used as template-molds for stabilization of programmed PA-stabilized Au NPs clustered nanozyme equivalents. The clustered Au clustered assemblies revealed peroxidase-like catalytic activities².
- Dynamic, transient aggregation/deaggregation of plasmonic Au NPs frameworks and 2. programmable Right/Left-handed chiroplasmonic Au NPs frameworks were demonstrated. By employing two kinds of nucleic acid-modified Au NPs in the presence of DNA-reaction module controller. and nickina enzvme dynamic dissipative transient а as the aggregation/deaggregation of the plasmonic NPs has been demonstrated. The transient aggregation/deaggregation process is followed by temporal interparticle coupled plasmon exciton, dynamic light-scattering and temporal TEM analyses.³

^{[1].} Y. Ouyang (2022). ACS Nano, 16, 18232-18243.

^{[2].} X.Chen et al. (2022). J. Am. Chem. Soc., 144, 6311-6320.

^{[3].} Y. Ouyang et al. (2021). J. Am. Chem. Soc., 2021, 143, 17622-17632.

The Importance of Surface Sensitivity in the Optimization of Plasmonic Sensors

<u>Attila Bonyár^{1,2}</u>, Tomáš Lednický³, Rebeka Kovács¹, Nóra Tarpataki¹, Manuela Proença⁴, Joel Borges⁴, Andrea Csáki³, Wolfgang Fritzsche³

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One of the main challenges of fabricating plasmonic nanostructures for chemical and biosensing applications is optimization of the structures in order to maximize their sensitivity. The most widely used parameter for benchmarking the fabricated sensors is the bulk refractive index sensitivity (*RIS*), which relates the spectral response to the RI changes in the medium surrounding the nanoparticles. However, for small molecule sensing applications the surface sensitivity, that characterizes the spectral response in the function of the deposited layer thickness can be more meaningful. Surface sensitivity is primarily depending on the plasmon decay length around the particles, affected by their size and shape and strongly influenced also by plasmonic coupling.

In this talk, the importance of optimizing the surface sensitivity of plasmonic nanostructures will be highlighted. It will be demonstrated through numerical simulations that if the size of the receptor-target molecules is known, the field decay length can be optimized to maximize the signal of target binding, trough the interparticle gap engineering of densely-packed nanoparticle arrangements. Two case studies will also be presented to illustrate the effect of layer formation on the interpreted signal during gas sensing and fluid refractive index sensing applications [1].



Fig. 1

Fig. 1/a: The structure and mesh of the hexagonal (periodic) unit cell for modeling the response and surface sensitivity of densely-packed gold nanoparticle arrays. In the simulations a growing thickness of layers with known refractive index (n=1.5) is deposited onto the nanoparticles in water medium (n=1.33) [1].

Fig. 1/b: The calculated peak shift values as a function of the deposited layer thickness [1].

[1] Proenca et al. (2024). ACS Appl Mat Int, 16 (42), 57832-57842.

Challenges in Fabrication of LSPR Sensors for Molecular Detection

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Building on A. Bonyár's submission, 'The Importance of Surface Sensitivity in the Optimization of Plasmonic Sensors', this talk will focus on the key practical challenges of fabricating plasmonic nanostructures for biosensing applications. Specifically, it will explore synthesis and characterization strategies to maximize sensors surface sensitivity.

Similar to the bulk refractive index sensitivity (RIS), the surface sensitivity strongly depends on the electric field enhancement. Yet, to pronounce the surface effect it is essential to promote localization of field enhancement by plasmonic structures engineering. One possibility utilizes sharp morphologies (e.g., nano-stars/triangles) that drastically increase the field intensity in tiny hotspots. A major drawback, however, is the reduced density of these hotspots, which leads to lower signal levels and, consequently, poorer sensor performance. Alternatively, field enhancement can be achieved through plasmon coupling between nanoparticles in ordered periodic lattices, where both hotspot size and density can be independently tuned. Despite these advantages, the real-world application of such plasmonic structures remains limited by their high fabrication costs.

This talk will present a comparative analysis of various fabrication approaches for plasmonic sensors, evaluating not only their surface sensitivity performance but also their fabrication complexity and other practical factors critical for biosensing applications. In addition, methods for benchmarking and estimating surface sensitivity will be discussed. Finally, strategies to enhance sensor performance through improved plasmon coupling—specifically via interparticle gap engineering—will be explored, offering insights into optimizing plasmonic structures for advanced biosensing.



Fig. 1: Experimental comparison of the LSPR response between two fabricated nanoparticle arrangements.

Arpan Dutta and J. Jussi Toppari

Nanoscience Center and Department of Physics, P.O. Box 35, 40014 Univ. of Jyväskylä, Finland

Organic thin-film-based excitonic nanostructures are a promising alternative for plasmonic systems. Such organic films exhibit Frenkel excitons at room temperature with high absorption coefficient. These excitonic materials are derived from common organic dye-doped polymer films by increasing the doping concentration so much, that a real part of the permittivity goes negative, $Re{\epsilon(\omega)} < 0$, within an energy range just above their material absorption. Within this narrow band of negative-real-permittivity, the organic film supports surface exciton (SE) modes analogous to the surface plasmon (SP) modes in metals. These modes exist as propagating ones on top of a film or/and localized SE modes in organic excitonic nanostructures [1,2]. Like SP modes, SE modes can be exploited in refractive index sensing and near-field enhanced spectroscopy in the weak coupling regime [3] and as an optical cavity in the strong coupling regime [4].

In this work, we report how to enhance the optical performance of the SE modes by optimizing the choice of excitonic dyes as well as their doping concentration and show how they can be utilized as the resonator mode for the strong coupling applications. In the case of weak coupling, we study the effect of molecular concentration in terms of oscillator strength and Lorentzian broadening on various SE modes when employed in sensing and spectroscopy. The optical performance of the SE modes is evaluated in terms of sensing, like sensitivity and figure of merit, as well as near-field enhancement, like enhancement factor and field confinement. In the case of strong coupling, we study whether SE modes can strongly couple to the photoactive molecules and SP modes or not, and whether the polariton modes are visible.

Our simulations reveal that, in the weak coupling regime, in general, an increase in oscillator strength of the excitonic dye enhances the performance of the SE modes while a broadening in the dye absorption linewidth degrades that as a counteracting effect. This demonstrates that the optical performance of an excitonic system is tunable via molecular concentration, which is a clear advantage over the plasmonic systems. In addition, different SE modes show different degrees of tunability and equivalency in performance when compared to SP modes in silver and gold.

In the strong coupling regime, our numerical findings reveal that SE modes can facilitate strong coupling by sustaining the energy-splitting-induced transparency. However, the polaritons may not be visible in the absorption since they can easily be located outside of the narrow energy-range of negative-real-permittivity. Also, it seems that the SE modes cannot couple strongly with the SP modes.

Our findings shed light on the weak and strong coupling properties of SE modes and provide crucial information for developing and optimizing novel excitonic nanodevices for contemporary organic nanophotonics.

- [1] L. Gu et al. (2013). Appl Phys Lett, 103(2), 021104
- [2] M.J. Gentile et al. (2014). Nano Letters, 14(5), 2339–2344
- [3] A. Dutta and J.J. Toppari, (2023). Opt Mater Express, 13, 2426-2445
- [4] A. Dutta and J.J. Toppari, (2024). Phys Rev B, 109(16), 165117

Hira Asif and Ramazan Sahin

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Active control of quantum states in incoherent quantum system is crucial for in situ programmable and multifunctional photonic integrated circuits (PICs) [1]. Quantum plasmonics provides an efficient pathway to achieve this control by leveraging the quantum properties of intense plasmon modes and the excitonic states of quantum emitters (QEs).

Here, we theoretically demonstrate the active tuning of plexcitonic modes in both (i) off-resonant and (ii) resonantly coupled plasmon-emitter systems through optical Stark effect (OSE) [2]. Using the Heisenberg-Langevin approach, we evaluate the dynamics of the intense plasmonic field generated by a bow-tie nanoantenna coupled to QE. In the off-resonant systems, the Stark field shifts the degeneracy of a three-level QE and coherently drives the off-resonant plexcitonic states closer to resonance, leading to a path interference effect and the formation of a transparency window, which we refer as Stark-induced transparency (SIT). Furthermore, even small perturbations in the Stark field yield significant changes in Rabi splitting, reaching values up to $\Omega \leq 350$ meV. These pronounced resonant shifts in the excitonic levels also enables the tunable photoluminescence (PL) in the visible regime, with on/off modulation of PL intensity. In resonantly coupled systems, the Stark field lifts the degeneracy by splitting the excited state of a two-level QE, inducing path interference in the hybrid plexcitonic modes. The Stark-induced splitting also shows the signature of Mollow triplets in the plexcitonic modes, with a maximum energy splitting up to 491 meV between the upper (UP) and lower (LP) plexcitons. We validate these plexcitonic shifts through PL spectra and evaluate the optical response of the system for varying Stark field strengths [Fig.1(d)]. With increasing field strength, the splitting between UP and LP polaritonic states increases, while the PL intensity gradually decreases, indicating a transition of spontaneous photon emission from high (on) to low (off) as a function of probe field. Our proposed method of Stark tuning of plexcitonic modes provides a pathway for coherent control of quantum devices, offering a means to mitigate decoherence in quantum systems.



Fig. 1

Fig. 1: (a) Hybrid quantum plasmonic system of Au bow-tie nanoantenna and voltage-tunable QE. (b) Stark-induced SIT in the off-resonant coupled system. (c) Stark-induced Rabi splitting in resonantly coupled plexcitonic states. (d) PL spectra of Stark tuned plexcitons as a function of detuning for different strength of Stark field in the weak (f : $0.02\omega o$) and (e) strong coupling (f : $0.05\omega o$)

T.Giordani et al (2023). Riv. Nuovo Cim, 46, 71.
 H.Asif and R. Sahin (2024). Phys. Rev. A., 110, 023713.

Quantitative photocurrent scanning probe microscopy of plasmoncoupled quantum dots

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Plasmonic nanoparticles can strongly modify the optical response of light emitters as molecules or quantum dots (QDs) via either their optical fields or by providing hot carriers. As a versatile tool to track the involved processes with high spatial resolution, we apply photoconductive atomic force microscopy (pcAFM), relying on a standard cantilevered metalized silicon tip.

First, as a model system we probe monolayers of PbS QDs with a perovskite-type ligand shell on an indium tin oxide (ITO) substrate. We show stable and reproducible photocurrent mapping and I/V measurements with a spatial resolution of a few QDs (Fig.1). From the data, quantitative values for energy barrier height, built-in voltage, diffusion constant and ideality factor are deduced with high precision. Second, we couple the PbS QDs to plasmonic gold nanoparticles. The photoresponse of the QDs is strongly modified, e.g., the intrinsic power law exponent of the QD irradiance-dependent photocurrent of 0.64 is increased to a value close to unity (Fig.2). Due to the quantitative character of the pcAFM measurements, the involved field and charge effects can finally be disentangled (Fig.3) [1].

We expect the pcAFM platform to be useful for a variety of plasmonic systems, in particular as it enables to directly observe with high spatial resolution effects as hot carrier generation. In addition, the measurement scheme can be extended to, e.g., optical near field functionality by including the detection of light scattered by the probe tip.



Fig. 1: pcAFM I/V curve from a PbS QD monolayer. The inset shows four exemplary photocurrent maps of a QD monolayer (upper right) and the bare ITO surface (lower left in each map). Fig. 2: Irradiance-dependent currents from two plasmonic gold nanoparticles and the bare ITO.

Fig. 3: Charge energy level model deduced from pcAFM data.

[1] F. Küstner et al., Nanoscale 16, 16664 (2024)

Markus B. Raschke

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Properties and functions of molecular materials often emerge from intermolecular interactions and associated nanoscale structure and morphology. However, defects and disorder disturb from energy conversion to carrier transport. Conventional spectroscopy and imaging techniques lack spatial resolution and sensitivity to the low-energy scales of the underlying both intra- and intermolecular interactions. We address these outstanding challenges in novel combinations of spatio-spectral and spatio-temporal infrared vibrational coupling nano-spectroscopy and -imaging. Here, coupling between molecular vibrations in nano-scale ensembles leads to hybridization, entanglement, and collective modes, with vibrational wavefunction delocalization giving distinct spectral features sensitive to local electric fields (Stark effect), intermolecular distance, and relative molecular orientation. Resolving this vibrational exciton formation as a molecular ruler in IR nano-spectroscopy, we image competing phases and local disorder in molecular solids - information inaccessible by conventional X-ray or electron-based crystallography. Specifically, in the application to the growth of porphyrin model organic electronic nanocrystals we observe the evolution of defects in competing amorphous and crystalline phases with nanometer spatial resolution [1,5]. Similarly, imaging vibrational coupling in polymers [2] and molecular monolayer [3], we resolve domain formation from the molecular to nano-scale. Further, through mode selective coupling of vibrational resonances to IR nano-antennas and associated Purcell-enhanced modification of vibrational lifetimes, we resolve intramolecular vibrational interaction and vibrational energy redistribution (IVR) [4]. In the extension to full spatio-spectral-temporal imaging we use molecular vibrations as sensors for the correlation between disorder and polaron formation in photovoltaic perovskites [6]. I will summarize with a perspective for nm-fs resolved precision vibrational nano-spectroscopy for functional imaging of molecular materials.



Fig. 1. Infrared nano-imaging of spatial delocalization of the vibrational wavefunction, serving as quantum sensor and molecular ruler of molecular disorder, crystallinity, and intermolecular coupling that control the properties of functional molecular materials at their elementary level.

[1] Muller, et al., PNAS **117**, 7030 (2020)

[2] Gray, et al., Nano Lett. **21**, 5754 (2021)

[3] Dönges, et al. Nano Lett. **21**, 6463 (2021) [4] Wilcken, et al. PNAS **120**, e2220852120 (2023);

[5] Puro, et al. Nano Lett. **24**, 1909 (2024).

[6] Wilcken et al. Science Advances **11**, eads3706 (2025)

Surface Lattice Resonances for High-Performance Sensing: A Study on Gold Nanoparticle Arrays

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The strong confinement of the electric field in nanostructures due to localized surface plasmon resonances (LSPR) makes them excellent transducers for sensing applications. However, LSPR biosensors often suffer from a low-quality factor. Periodic arrays of metallic nanoparticles (NPs) offer a promising solution by enabling interactions between LSPR modes and diffractive effects, leading to the formation of surface lattice resonances (SLRs). Extensive research has shown that optimizing NP size, shape, and lattice constant enhances the optical response of these arrays, making them ideal for high-sensitivity and high-resolution applications [1-5].

In this work, we investigate a variety of structural parameters governing the spectral position, Q factor, and sensitivity of SLRs in periodic arrays of gold NPs. Using finite element method (FEM) simulations implemented in COMSOL Multiphysics®, we examine the influence of the NP geometry and the lattice constant on the optical response of these arrays while comparing them to their isolated NP counterparts. Our results reveal that periodic NP arrays with optimized geometries can achieve Q factors one order of magnitude higher than those of isolated NPs, with bulk sensitivity improvements higher than 100 nm/RIU compared to LSPRs. Additionally, we validate our theoretical findings experimentally by fabricating a proof-of-concept sensor based on a periodic array of gold nanodisks by using electron beam lithography. The fabricated array demonstrates a competitive performance against previously reported works.







Fig. 3

- Fig. 1: Influence of lattice effects on the optical response of different gold NP geometries.
- Fig. 2: Sensing performance of isolated and arrayed gold nanoparticles.
- Fig. 3: Experimental results of the 600-nm periodic array of nanodisks as a plasmonic sensor.

[2] J. Černigoj, et.al (2018). Sci Rep 8(1), 15706.

^[1] E. S. A. Goerlitzer, et.al (2020). Advanced Materials, 32(22).

^[3] M. Kataja, et.al (2015) Nat Commun 6(1), 7072.

^[4] X. Yang, et.al (2019) Opt Express 27(18), 25384.

^[5] Lasa-Alonso, Jon, et al. (2020) Acs Photonics, 7(11), 2978-2986.

Self-Assembled Plasmonic Lattices and Their Applications

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Noble metal plasmonic nanostructures attract much attention due to their unique material and shapedetermined light scattering and absorption properties, delivering strong wavelength-dependent responses. Arranging the individual or clustered nanoparticles (NPs) into regular sub-wavelength arrays can diffractively couple the scattered light from broad localized surface plasmon (LSP) into narrow and high-Q resonances termed plasmonic lattices or surface lattice resonances (SLRs) [1-6]. Precise average size, high monodispersity, shape control, and reproducibility of wet-chemically synthesized plasmonic NPs [6-7], along with the template-defined NP self-assembly, enable their applications for photonic devices, which are originally made employing top-down methods [1-6]. Capillarity-assisted particle assembly from concentrated NP colloids into soft lithography-replicated elastomer templates (Fig. 1) is an established alternative for the manipulation and deposition of NPs into lithography-defined positions originating desired photonic coupling and controlled local field enhancements [1-6]. NP particle arrays in the polydimethylsiloxane elastomer deposition templates act as a plasmonic metasurface for direct application or as a transfer carrier for integration into more complex photonic structures. The ability to tailor SLR positions rises from the size-dependent LSP coupling with the pitch-determined Rayleigh's anomaly [1-6]. Regular plasmonic hybrid structures in wide band gap semiconductor layers serve as efficient hot carrier injectors, while NP arrays in organic days sustain resonance modes amplifying the stimulated emission. Therefore, the laterally confined light can act as an optical resonator for a dye nanolaser [5], can confine the electromagnetic radiation for desired wavelength Raman enhancement [3-4], and can contribute to efficient solar water splitting by metasurface photoelectrodes [8].



Fig. 1 Capillarity-assisted nanoparticle deposition method [9]

[1] Juodėnas M., et al. (2019) ACS Nano 13 (8), 9038–9047.

[2] Juodénas M., et al. (2020) ACS Photonics. 7 (11), 3130–3140.

[3] Khinevich N., et al. (2023) Sens. Actuator B-Chem. 394, 134418

[4] W. Andrzejewska, et al. (2025) Biosensors and Bioelectronics, 274, 117143

[8] Hüttenhofer L., et al. (2024) J. Phys. Chem. C 126 (23), 9002–9073. [8] Hüttenhofer L., et al. (2021) Adv. Energy Mater., 11 (46), 2102877.

 ^[5] M. Juodénas, et al. (2025) Nanoscale Horiz., 10, 142-149
 [6] Khinevich N., et al. (2024) J. Materiomics 10 (3), 594-6002023.

^[7] Klinavičius T. et al. (2024) J. Phys. Chem. C 128 (23), 9662–9675.

^[9] http://nanoversa.lt

CuFeS₂ Semiconductor Nanocrystals: An Alternate Plasmonic Catalyst

<u>Biswajit Bhattacharyya¹</u>, Shashank Gahlaut¹, Felix Stete², Andreas Taubert¹, Matias Bargheer², Ilko Bald ¹Institute of Chemistry, ²Institute of Physics and Astronomy, University of Potsdam, Potsdam-14476, Germany

bhattacharyya@uni-potsdam.de

Abstract:

Plasmonic photocatalysts enhance light-driven reactions via surface plasmon resonances, improving light absorption and charge carrier generation. While noble metals dominate this field, recent efforts focus on alternative materials. In this talk, I present CuFeS₂ nanocrystals as efficient non-noble plasmonic photocatalysts. Their structural and optical properties, photocatalytic performance (monitored via SERS), and underlying mechanisms (studied using ultrafast spectroscopy) will be discussed.

References:

- 1. Bhattacharyya, B., Gahlaut, S. et al. CuFeS₂: An Alternate Plasmonic Photocatalyst with Visible Light, 2024 (Manuscript under revision).
- Bhattacharyya, B.*, Balischewski, C., Pacholski, C., Pandey, A., Bald, I., Taubert, A., Copper Iron Chalcogenide Semiconductor Nanocrystals in Energy and Optoelectronics Applications—State of the Art, Challenges, and Future Potential. *Adv. Optical Mater.* 2023, 2202411.
- Sugathan, A.[#]; Bhattacharyya, B[#].; Kishore, V. V. R.; Kumar, A.; Rajasekar, G. P.; Sarma, D. D.; Pandey, A., Why Does CuFeS₂ Resemble Gold? *J. Phys. Chem. Lett.* 2018, *9*, 696-701 ([#] both the authors contributed equally).
- Bhattacharyya, B.; Pandey, A., CuFeS₂ Quantum Dots and Highly Luminescent CuFeS₂ Based Core/Shell Structures: Synthesis, Tunability, and Photophysics. *J. Am. Chem. Soc.* 2016, *138*, 10207-10213.

Metal photoluminescence as probe for hot electrons

Wouter Koopman, Jan Kutschera, Felix Stete, Athena Maljesi, Matias Bargheer

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Plasmonic noble-metal nanoparticles have been demonstrated to exhibit substantial photoluminescence, despite the absence of a bandgap in metals [1]. This emission is related to the density of excited charges in the particle and therefore currently being explored as a promising probe for investigating the latter. The intensity and spectrum of the emitted light can be used to deduce conclusions about the quantity and energy of the excited charge carriers. At present, however, the exact emission mechanism has not yet been conclusively clarified and various excitation scenarios are being discussed [1,2] (Fig. 1). The acquisition of experimental data, crucial for a comprehensive understanding of the recombination process, is hindered by the presence of plasmon resonance. Eliminating the influence of the plasmon resonance is therefore of great importance in order to be able to investigate the recombination process directly.

In this presentation, we show how photoluminescence spectroscopy with various excitation wavelengths can be used to investigate the excitation pathway in films of gold nanoflowers The spectroscopic evidence indicates that the luminescence is predominantly attributable to Auger-excited intraband emission from gold nanoflowers. While the emission spectra could be unequivocally assigned to intraband recombination, the excitation spectrum clearly demonstrates absorption by interband transitions (Fig 2 & 3). The results suggest that Auger excitation is a promising route to generate energetic "hot" electrons with energies substantially above the Fermi level. The exploitation of this effect could significantly benefit applications in nano-luminescent probes and the advancement of plasmon catalysis.



Fig. 1 : Proposed excitation scenarios to explain the PL form plasmonic nanoparticles.



Fig. 2 : PL spectra of gold nanoflowers for different excitation wavelength.



Fig. 3: The relative change of emission eliminates the influence of the plasmon resonance.

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Plasmon-Induced Regioselective Grafting of Colloidal Gold at Hot Spots: Boosting Light Emission Efficiency

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In this study, we present a novel approach that combines diazonium salt chemistry with plasmonic properties of gold nanoparticles to achieve regioselective grafting of small emitters precisely at regions of high field enhancement created by plasmon excitation. Our process begins with the fabrication of well-ordered arrays of gold nanostructures (NSs) via electron beam lithography. Next, we take advantage of plasmon excitation to selectively graft aryl films derived from 4-(aminomethyl)-benzene-diazonium-tetrafluoroborate, resulting in the aryl film being concentrated in regions of intensified electromagnetic fields of the NSs (see figure 1a) [1,2]. Since these aryl films are positively charged, we complete the process by anchoring negatively charged gold colloidal particles, serving as model emitters, specifically to the areas coated with the aryl layer (see figure 1b). This approach allows precise placement of emitters at plasmonic hot spots, paving the way for enhanced light emission that benefit from controlled nanoparticle positioning, particularly useful for applications in light-emitting devices, LEDs, and display technologies.



Figure 1: (a) SEM image of a square array of gold nanostructures (NSs) deposited on an indium tin oxide (ITO) substrate, after a plasmon-induced grafting of aryl film derived from 4- (aminomethyl)-benzene-diazonium-tetrafluoroborate; SEM image of a square array of gold NSs, after anchoring negatively charged gold colloidal particles on the NSs.

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Plasmonic actuating and continuous monitoring biosensing

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Metallic nanostructures allow for tight confinement of electromagnetic field through the coupling to surface plasmons. These optical resonances originate from collective oscillations of charge density at the metallic surface and they are associated with a strongly enhanced electromagnetic field intensity and local density of optical states. Therefore, metallic nanostructures are excellently suited for amplification of weak optical signals in various methods including fluorescence spectroscopy. Plasmon-enhanced fluorescence spectroscopy increasingly serves in numerous types of bioanalytical tools and bioimaging methods that were developed for ultrasensitive detection of molecular species, in particular biomarkers relevant to medical diagnostics [1]. This paper will report on plasmon-enhanced fluorescence biosensors that take advantage of responsive hydrogel materials providing means to actively control localized surface plasmon resonances [2] and that allow for a preparation of miniature elements serving as actuators [3]. Dedicated thermo-responsive polymers that exhibit lower critical solution temperature will be employed with the use of plasmonic heating [4]. In addition, these materials will be combined with biofunctionalization strategies and reversible types of affinity bioassays. Such developed building blocks are currently pursued for a construction of miniature sensing elements that allow for continuous monitoring of molecular species and that can be optically positioned at desired locations. This research will be put in context with a future implementation in cell-on-chip systems enabling accurate spatially resolved monitoring of molecular species secreted by stimulated cells.



Fig.1: Schematics hydrogel element with biofunctionalized metallic nanostructures for a miniature sensing element detecting cellular-secreted species.

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Plasmonic single molecule detection enhanced by DNA nanostructure

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Bioassays with a digital readout format have become an established approach for the ultra-sensitive analysis of the most important types of biomolecules used in biomarker analysis including nucleic acids and proteins [1]. These methods allow for single molecule detection (SMD), and they rely on the counting of individual target molecules present in an analyzed liquid sample that is partitioned to a series of miniature compartments (microwells or droplets) [2]. The loading of target molecules into such compartments is governed by Poisson statistics, and it is detected based on target molecule-induced enzymatic amplification. In most previous works, a fluorescence output signal in each compartment (serving as a reaction chamber) is generated by using polymerase with fluorescent oligos. Such reliance on enzymes and compartmenting method adds a layer of complexity and lowers robustness and reproducibility of digital assays, especially in non-laboratory settings.

Herein, we report on an alternative approach that allows to overcome the necessity to use of enzymes and sample partitioning in assays with digital readout. This method is based on a combination of plasmon-enhanced fluorescence (PEF) [3] and a newly developed non-enzymatic DNA amplification, termed as tethered catalytic hairpin assembly (tCHA) [4], using a long double-stranded DNA architecture assembled at nanoscale length, 15-135 nm, depending on the combination of strands used. This long DNA architecture acts as a flexible polymer linker (FPL) to tether an entire DNA amplification process at the interface where target analyte molecules are affinity captured on the sensor surface. After being initiated by a specific sequence T at FPL, tCHA catalytically and cyclically produces the fluorescent amplicons HA hybridizing with surface-immobilized hairpin B (HB) at a defined area governed by the length of FPL. Such tCHA takes advantage of PEF, which provides an additional optical enhancement of fluorescence signal from photoactive compounds present in a close proximity to the sensor surface. This allows for the real-time wide-field fluorescence microscopy-based monitoring of the amplification reaction on the sensor surface that is not masked by the parasitic fluorescence signal originating from assay constituents dissolved in the contacted bulk aqueous solution.



Fig. 1: Design of FPL composed of NS segments of dsDNA connected by ssDNA hinges. b) Schematic of a biointerface configuration for tCHA carrying HB sequences that are co-immobilized with T coupled to an FPL for spatial confining of CHA cyclic reaction.

Fig. 2: Schematic of the sandwich immunoassay with PEF readout of fluorescent spots generated by tCHA at locations where individual target analyte molecules were affinity captured.

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Optimizing Light for Plasmon Enhancement

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Tip-enhanced Raman scattering (TERS) is a technique that utiliz the highly confined plasmonic enhancement at the apex of a metallized tip. The optimal tip shape remains a topic of debate since the advent of TERS, with studies indicating that a granular structure may be advantageous and that the atomic structure of the apex also influences the response. So far the impact of the incoming light on the enhancement remains less well investigated and will be discussed in this presentation.

The underlying hypothesis is that minor nanoscale changes at the tip significantly impact the signal of TERS, suggesting that these changes also have a pronounced effect on electric field coupling with a specific tip. In this investigation, the focus is on the light-tip coupling mechanism in an epiillumination TERS configuration. This configuration is regarded as the most efficient for signal collection, as it utilizes high numerical aperture objectives (e.g., liquid immersion) characteristic of advanced microscope. While the signal collection is very efficient, tip illumination presents a significant challenge, often necessitating the use of radially polarized beams to ensure optimal coupling of the electric field components to the antenna. The employment of a spatial light modulator enables the optimization of the beam, thereby amplifying the TERS signals by a factor of 5-10 in comparison to the conventional manual alignment approach. Subsequent analysis of the optimized beam reveals a complex pattern, which ultimately results in a highly efficient coupling to the TERS probe.

The implementation of the general scheme to optimize and increase the long-term stability of TERS experiments will be presented, as well as the implications of using such a system to test and create novel tip-enhanced geometries by analyzing the final illumination beam, i.e., the spatial light modulator phase pattern.

Altering the SERS spectra of molecules on nanostructured surfaces by laser focusing/ defocusing

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We present an analysis of SERS signal intensity and spectral profiles along Z-axial linescans, unveiling some insights into the SERS response of molecules on SERS substrates. The signal strength profile of 4-aminobenzenethiol (ABT) adsorbed on gold films over nanospheres (AuFoN), recorded through a high-NA objective, demonstrates a distinctive shape along the axial Z-direction scans, with a frequency-dependent offset that peaks above the sample surface. Notably, the intensity ratios of various spectral regions, including SERS bands and background, display non-uniform variations along the Z-axis. By analysing reflectance spectra obtained through high-NA objective at different Z positions and FDTD simulations, we established that the near-field spectra reveal different profiles and maxima at varying wavelengths depending on the focus position. This correlation suggests that the trends identified in the SERS experimental data are intrinsically linked to the specific plasmonic near-field response induced by the focused beam on the SERS substrate surface.

Given these insights, we suggest to consider this phenomenon in future SERS analyses, as it enriches our knowledge of analyte-plasmonic substrate interactions. Furthermore, understanding the relation between the morphology of SERS substrates and their spectral profiles is crucial before undertaking more complex analyses, such as evaluating molecular orientation or performing quantitative assessments based on SERS bands intensity ratios. These findings are expected to have implications for the development of SERS substrates with morphological features comparable to the excitation wavelength, contributing to more refined and accurate analytical applications.

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Fig. 1: Schematics of focused laser beam on AuFoN SERS substrate

Fig. 2: SERS spectra of ABT on AuFoN for different values of surface-focus distance

Fig. 3: Near-field spectra at the AuFoN surface for different values of surface-focus distance

Morphological adjustment of gold nanoparticles AuNPs through polydispersants

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Through an experimental design 3³, the aggregation of gold nanoparticles (AuNPs) was evaluated by ultrasound (US), microwave radiation (MIC) mechanical agitation (MEC) and different anionic (sodium dodecyl sulfate, SDS), cationic (hexadecyltrimethylammonium bromide, CTAB) and neutral (polyvinylpyrrolidone, PVP) polydispersants, where the best synthesized system was functionalized with MHDA, to support a monoclonal antibody (Ab). Statistical analysis found with 95% confidence that the assistance by ultrasound and microwave radiation are not significant against the concentration and type of polydispersant.

Experimental techniques such as UV-Vis spectroscopy, scanning/transmission electron microscopy (STEM), Shift Raman spectroscopy, Z Potential were used to characterize the AuNps, some of the results are presented below.



The experimental results show that ionic polydispersants (CTAB; SDS) control the morphology and size better than a neutral polydispersant (PVP), therefore the synthesis of AuNPs with SDS-0.01M was used to functionalize with MHDA obtaining a shift of 16 nm which was attributed to the Au-MHDA interaction, which was used as Au-MHDA-Ab support.

Fig. 1: Surface absorbance graph vs.polydispersant type and concentration

- Fig. 2: Effect of energy assistance
- Fig. 3: Effect of polydispersant concentration
- Fig. 4: AuNPs with Ionic polydispersant
- Fig. 5: AuNPs with neutral polydispersant
- Fig. 6: Functionalized AuNPs

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Surface-enhanced Raman spectroscopy (SERS) is a powerful technique that has shown great promise in the field of biomolecular detection for various applications in medical diagnostics and research [1, 2]. Conventional SERS substrates, typically fabricated from silver nitrate, suffer from oxidation and degradation, restricting their long-term utility. By using silver sulfate as a precursor instead of silver nitrate, we developed dendritic nanostructures with sensitive SERS detection capabilities. Silver sulfate acts as a growth agent and a mild capping agent, which simplifies the fabrication process and improves the stability of the substrates. The substrates demonstrated excellent sensitivity and stability, effectively detecting 4-mercapto benzoic acid (4-MBA) at subfemtomolar concentrations. Thus, these findings highlight their potential for use in tracing small concentrations of biomolecules. The newly developed SERS substrate was also tested with a range of drugs, including 6-thioguanine, methotrexate, erlotinib, doxorubicin, and moxifloxacin, with detection limits down to the sub nanomolar range, demonstrating its potential in drug monitoring. Furthermore, to simulate clinical applications in relevant matrices, the drugs were spiked into human plasma from healthy donors. Despite the complexity and interference of the plasma matrix, the drugs were detected in the concentration range required for therapeutic monitoring. These results demonstrate the significant potential of sulfate ion-directed dendritic structures for therapeutic drug monitoring in clinical diagnostics.

Acknowledgement: The Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) supported this work under grant 465289819.

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Large-scaled plasmonic assemblies for efficient SERS enhancements

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Plasmonic nanoparticle assemblies are widely recognized for their remarkable ability to significantly amplify Surface-Enhanced Raman Spectroscopy (SERS) signals through the generation of plasmonic hotspots. Building upon previous studies of 1D-plasmonic lines,^[1] we have now successfully explored 2D-zigzag plasmonic chains, fabricated using wrinkle-assisted colloidal self-assembly techniques. Such zig-zag chains exhibit isotropic enhancement behavior, overcoming the polarization-dependence limitations of earlier 1D configurations.^[2] Detailed optical characterization and finite-difference time-domain (FDTD) simulations confirm significantly intensified hotspots arising from these novel configurations, that support the experimental findings.

Additionally, this talk introduces the concept of coupling plasmonic chain modes with guided photonic modes in thin dielectric waveguide systems. Initial studies indicate that hybrid modes resulting from plasmonic-waveguide coupling can surpass diffraction-coupled modes in enhancement performance, demonstrating considerable potential for efficient SERS measurements. The talk concludes by outlining future prospects for developing advanced hybrid plasmonic-waveguide systems tailored for versatile and high-Q sensing applications.



Fig. 1: SEM image showing AuNP@PANI assembly as zigzag plasmonic chains

Fig. 2: Raman shifts as a function of sample stage rotation

Fig. 3: Plasmonic-waveguide systems for comparison of SERS enhancements

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Molecular Effects of NBD-CI and NBD-ceramide on Living Cells using SERS

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7-Nitrobenz-2-oxa-1,3-diazole (NBD) is a popular fluorescent label for proteins, peptides, and lipids. Its chloride derivative, NBD-CI, can form stable fluorescent adducts by reacting with thiol and amine groups, while ceramide, that is a crucial structural component and potent regulator in the physiological responses, labeled by NBD (NBDCER) is frequently employed to study sphingolipid metabolism^{1,2}. Previous studies in molecular models containing ceramide and other sphingolipids have demonstrated the potential of surface-enhanced Raman scattering (SERS) as a local vibrational probe of lipid-nanostructure interactions^{3,4}.

In this study, we investigated the effects of NBD-CI and NBDCER on intracellular molecular changes in living cells using SERS with gold nanoparticles as the probe. SERS spectra obtained from 3T3 fibroblast and J774 macrophage cells reveal significant changes in the molecular composition and interactions with gold nanoparticles under different incubation conditions. The random forest-based surrogate minimal depth (SMD) algorithm was applied to identify important spectral features and evaluate their relations, revealing the colocalization of molecules. NBD-CI and NBDCER were found to affect the biochemical makeup and protein-lipid interactions in the endolysosomal compartments.

These findings demonstrate the potential of SERS with gold nanoparticles in probing intracellular physiological processes and provide insights into the molecular impact of NBD labels on cellular metabolism. Integrating spectroscopic and computational approaches offers valuable insights into understanding cellular responses and enzyme functions.



Fig. 1: relative band occurrence of the respective SERS data sets of 3T3 fibroblast cells incubated with (A) NBDCER and (B) NBD-CI after incubation with gold nanoparticles for 3 h. (C) Spectral variables co-occurring with the important bands (left) selected by SMD analysis of SERS spectra from 3T3 cells treated by NBDCER and NBD-CI after incubation with gold nanoparticles for 3h. ⁵



POSTER PRESENTATIONS

- 01 Towards better characterization and control of protein-gold nanoparticle interactions // Shrobona Banerjee (Berlin)
- 02 Evaluation of health risks of Whole-Body Electromyostimulation (WB-EMS) based on electrophysiological and biochemical characterization // Steffen Derlien (Jena)
- 03 Tunable Dielectric-Loaded Metasurface for Directional Excitation of Surface Plasmon Polaritons with Linear or Circular Polarization // Abhrodeep Dey (Jena)
- 04 Raman-based Detection of Natural Products in Microbial Communication // Tony Dib (Jena)
- 05 Tailored Optical System for Advanced Characterization and Analysis of LSPR Biosensors // Laszlo Domjan (Budapest)
- 06 Water contaminants probed by plasmonic nanoarray using aptamer receptors // Lukas Dubbert (Jena)
- 07 Point-of-care diagnostics potential of cerumen: vibrational spectroscopy meets machine learning // Edoardo Farnesi (Jena)
- 08 Aptamer-conjugated SERS detection scheme for wastewater treatment // Arna Ghosh (Jena)
- 09 Determination of in vivo and in vitro DNA Binding Affinities of Human BCL6 protein // Dhaarna Gobin (Jena)
- 10 Nanocube Quadromer for Efficient LSPR Biosensing // Hovhannes Haroyan (Yerewan)
- 11 Tip enhanced luminescence of gold nanoparticles // Andreas Hohenau (Graz)
- 12 Broadband optical chirality enhancement in 3D plasmonic Archimedean spiral for chiral sensing // Min Jiang (Jena)
- 13 Flexible Silver-Coated Gold Nanoparticles Loaded Graphene textiles for Direct Detection of Imidacloprid Pesticide via Label-Free SERS // Sujia Jiao (Jena)
- 14 SERS detection of benzotriazole for wastewater treatment // Klaudzia Kamiak (Jena)
- 15 Plasmon-Induced Photopolymerization of Molecularly Imprinted Polymers for Nanosensor Applications // Amine Khitous (Mulhouse)
- 16 Conformational Dynamics of Hsp90 // Florian Kofler (Mainz)
- 17 Interaction between DNA and gold nanoparticles investigated by fluorescent probes // Wouter Koopman (Potsdam)
- 18 Hybrid Nanostructures // Rasheequddin Kuzhimpadath (Jena)
- 19 Gold nanoparticle-facilitated assembly of supernatant transfer enables the rapid determination in the discovery of ssDNA aptamers specific to steroid hormones // Jeong Min Lee (Seoul)
- 20 Label-free detection of SARS-CoV-2 (RBD) by surface plasmon resonance // Mary Carmen Licuona Puma (Jena)
- 21 Immobilised gold and silver nanoparticles for an optofluidic SERS platform // Anant Mantha (Berlin)
- 22 Development of a low-cost filter membrane based on cannabis sativa for the reduction of bacterial loads in non-potable domestic water in certain regions of the capital through green synthesis // Yehidi Julieth Medina Castillo (Bogota)
- 23 Synthesis of plasmonic nanoparticles and bulk sensitivity investigation for advancing LSPR sensing technology // Olesia Petrova (Jena)
- 24 Development of a Lateral Flow Assay Biosensor for miRNA-34a and miRNA-155 Detection Utilizing the Rolling Circle Amplification // Farzad Rahbar Kouibaran (Bologna)
- 25 Investigation of plasmonic enhancement of hyper Raman and Raman scattering // Fariha Binte Rahman (Berlin)
- **26 LSPR studies of protein-DNA interactions** // Florian Seier (Jena)
- 27 Probing intracellular interactions with SERS in the presence of a fluorescent dye // Cecilia Spedalieri (Berlin)
- 28 Numerical Demonstration of Archimedean spiral with highest dissymmetry factor // Karthika Sunil (Jena)
- 29 SERS for cytosine methylation detection: a step toward epigenetic biomarkers // Gennadii Ustinov (Jena)
- 30 Silver nanoparticle decorated titania and zinc oxide nanowires by MS-ALD for Photocatalytic dye degradation // Mario Ziegler (Jena)

Towards better characterization and control of protein-gold nanoparticle interactions

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Surface enhanced Raman scattering (SERS) of protein molecules is extremely sensitive to minuscule changes in their interaction with a plasmonic substrate and therefore can vary even with small changes in experimental conditions, such as altered molecular concentration¹. It is compatible with aqueous environments and can be performed in the absence of additional labels and at high enhancement. This makes SERS an excellent technique to probe nanoscale events in biological and biomimetic systems.

Proteins are structurally diverse biomolecules whose interaction with nanoparticles is important in many applications and must be understood in order to be eventually controlled. SERS spectra of single proteins/protein mixtures can show great variations². Thus, the high sensitivity makes SERS ideal to investigate protein structure and composition.

In the wider context of using SERS to probe biomolecules and/or biological systems³, it is crucial to understand the different spectral responses from a biomolecule occurring at different concentrations or under different conditions in nature. Thus, we analyzed a wide range of conditions and their effects on the SERS spectra of proteins, which can be utilized in interpreting more complex systems. On the other hand, we have also identified reproducible experimental conditions which produce more homogeneous SERS spectra.

In the wider context of using SERS to probe biomolecules and/or biological systems, it is crucial to understand the different spectral responses from a biomolecule occurring at different concentrations or under different conditions in nature^{3,4}. Thus, we analyzed a wide range of conditions and their effects on the SERS spectra of proteins, which can be utilized in interpreting more complex systems. On the other hand, we have also identified reproducible experimental conditions which produce more homogeneous SERS spectra.

SERS spectra have a high degree of variability, particularly in solution. To understand these variations, we identified patterns using unsupervised and supervised machine learning algorithms. These patterns explain the intrinsic variations in SERS of proteins. As SERS continues to be employed in analyzing increasingly heterogeneous systems, these tools will be valuable in ensuring reproducibility of interpretation.

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Evaluation of health risks of Whole-Body Electromyostimulation (WB-EMS) based on electrophysiological and biochemical characterization

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Whole-body electromyostimulation (WB-EMS) is a training method that delivers electrical impulses to stimulate all major muscle groups simultaneously, with adjustable intensity for each electrode. Its efficiency and low impact on joints make it a potentially appealing option for individuals who are either unwilling or unable to participate in traditional high-intensity exercise routines.

EMS can therefore have very positive effects in these cases, but its use is not without risk. This is due to the non-physiological and supramaximal stimulation of the muscles. The muscle fatigue that sets in during conventional sporting activities normally limits the extent of exercise intensity – both in terms of its intensity and duration. This natural boundary is blurred with EMS: the duration is predetermined within the programs, and its intensity is not limited by physiologically induced muscular failure. On the contrary: the intensity is regulated based on the subjective assessment of the user. They are encouraged to adjust the selected intensity up to the limit of what is tolerable. This is a combination of pain and muscular tension. The tolerance limit therefore varies greatly from individual to individual and increases a) during a specific application and b) over repeated applications. In particular, increasing the tolerance limit leads to the stimulation intensity sometimes being significantly increased during training. In addition to the advertised training effect, this has consequences for the integrity of the affected muscles. Significant increases in markers associated with the occurrence of muscle damage have been observed. This particularly applies to creatine kinase (CK), where increases of up to a thousandfold have been observed. This poses a significant health risk, which the proposed study aims to evaluate and minimize.

Therefore, a LSPR biosensor array is planned to monitor certain related biomarkers simultaneously with a time resolution in the s ... min range. A panel of biomarkers will be selected, and respective aptamers or antibodies as highly specific receptors will be identified in the literature, and spotted in an array pattern with repeats. Then, assay protocols will be tested for a simultaneous or group-wise detection utilizing the array detection setup established at the Leibniz IPHT.

Tunable Dielectric-Loaded Metasurface for Directional Excitation of Surface Plasmon Polaritons with Linear or Circular Polarization

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Dedicated optimization of plasmonic geometries in the nanoscale can be employed to efficiently host strongly localized electromagnetic modes in the near field via the satisfaction of momentummatching condition. The refractive index contrast at the interface, that is pivotal to attain strongly localized mode confinement, can be tailored by employing appropriate dielectric-loaded plasmonic system. In this work, we propose such a system that comprises of a dielectric grating on top of a thin gold film using a phase change material (PCM) known as Stibnite (Sb_2S_3). The usage of PCMs provides useful advantage of drastic optical property change once tuned from their amorphous to crystalline state [1]. Sb_2S_3 can be easily tuned from amorphous to crystalline state at an ultrafast rate using an optical beam or via thermal annealing process [1, 2]. The dielectric grating is designed in a fishbone configuration [3] to exhibit a circular polarization-dependent directional coupling of SPPs. Finite-difference time-domain (FDTD) simulations based on amorphous Sb₂S₃ fishbone grating exhibit strong directional propagation of SPPs, the directionality of which changes when the handedness of circular polarization is changed. Interestingly, when the phase of the material is switched to crystalline Sb_2S_3 , the aforementioned handedness-dependent response flips its direction. Furthermore, using the same principle, we also demonstrate unidirectionality and material phase dependent directional switch based on linearly polarized light.



Fig. 1



Fig. 1: Directional propagation of SPPs excited by incident right handed circularly polarized light on amorphous Sb_2S_3 fishbone metasurface

Fig. 2: Propagation of SPPs switches its direction on changing the material phase of Sb_2S_3 fishbone metasurface from its amorphous to crystalline state while keeping the handedness of incident light same

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Raman-based Detection of Natural Products in Microbial Communication

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The interaction between prokaryotic and eukaryotic microorganisms in has been proven to play a critical role in the functioning of ecosystems. The polyketides, for example, that are derived from arginine and known as arginoketides, are produced by *Streptomyces* species to play a key role in mediating cross-kingdom microbial interactions with the *Aspergillus* fungi, leading to the production of natural products. These arginoketides can either be cyclic, such as monazomycin and desertomycin A, or linear, like lydicamycin and linearmycin A, all of which produced by *Streptomyces iranensis* and have been found to stimulate the orsellinic acid gene cluster in *Aspergillus nidulans* [1]. The characterization and detection of natural products by surface-

enhanced Raman spectroscopy (SERS) is expected to shed light on how they are released and transported through the environment, and how theytrigger responses in other microorganisms [2].

For that purpose, a special silver substrate for SERS measurements was fabricated on a silicon wafer by means of galvanic replacement of silver and sulfate ions that resulted in dendritic structure. At the а nanoscale, the branches of the treelike structure form sharp angles and narrow gaps that are supposed to turn into "hot spots" and enhance the Raman signal of the target molecules. The Raman and SERS spectra of both S. iranensis and A. nidulans products exhibit marker modes that should enable their detection and identification in the microbial culture



Figure 1 (A) SEM image of the SDS on the left and its spectrum on the right. (B) Spectra of *A. nidulans* and *S. iranensis* agar co-culture with SDS.

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This paper presents the design and implementation of a customized hyperspectral readout system tailored for the rapid characterization of transmission-based localized surface plasmon resonance (LSPR) biosensors. By addressing a critical gap in plasmonic research, the system provides high flexibility for both chip fabrication and readout methodologies. The results outlined in this study contribute to the advancement of multiplexed LSPR array sensors with high sensitivity (120 nm/RIU), compact dimensions (2 × 3 mm), and rapid data acquisition. The system enables optimization of synthesis parameters, such as nanoparticle density and size, while establishing a robust platform for designing an effective optical readout mechanism.

A key feature of the setup is a digitally controlled monochromator, which allows for precise adjustments in illumination wavelength, resolution, and bandwidth. The use of a laser-excited phosphor source significantly reduces detector integration times compared to conventional Tungsten-Halogen light sources, leading to improved signal-to-noise ratio and faster measurement speeds. The fully automated, software-driven system streamlines the optimization process for key chip parameters, including nanoparticle density and sensing spot dimensions.

Additionally, to minimize errors caused by source wavelength drift, the system incorporates a reference optical path with a spectrometer for real-time illumination spectrum monitoring. This feature enhances measurement accuracy and reliability. Furthermore, the system facilitates efficient detection of manufacturing defects, such as misalignment or non-uniform nanoparticle distributions, thereby improving overall biosensor evaluation and quality control.

Point-of-care diagnostics potential of cerumen: vibrational spectroscopy meets machine learning

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The application of point-of-care (POC) testing in diagnostics holds the potential to transform the field by enabling early detection and real-time monitoring of the disease, allowing timely interventions and improving patient outcomes. Being minimally invasive diagnostic technique, liquid biopsy results in valid POC test, involving the analysis of human body-fluids.

The diagnostic potential of cerumen remains largely unexplored, despite its rich molecular composition and proximity to critical anatomical sites. The non-invasive, stable, and easily accessible nature of earwax collection presents significant advantages over traditional liquid biopsies such as saliva or blood, which are susceptible to external contamination and variability. Initially, integrating complementary vibrational spectroscopic methods, including Raman scattering, surface-enhanced Raman spectroscopy (SERS), broadband coherent anti-Stokes Raman scattering (BCARS), stimulated Raman scattering (SRS), and optical photothermal infrared (OPTIR) spectroscopy, has enhanced the molecular characterization. [1] Comparative analyses reveal that while conventional Raman spectroscopy provides critical molecular fingerprints, the enhanced sensitivity of SERS and nonlinear coherent Raman techniques (CARS and SRS) facilitate the detection of disease-associated lipid and protein alterations. OPTIR spectroscopy further enriches spectral interpretation, supporting the robustness of cerumen-based diagnostics. These findings underscore the transformative potential of cerumen spectroscopy for early-stage cancer diagnostics and broader disease monitoring.

Subsequently, a study leverages SERS in combination with machine learning (ML) to develop a non-invasive POC diagnostic tool for head and neck cancer (HNC). [2] By analyzing cerumen samples from HNC patients and healthy individuals, we demonstrate that SERS-based spectral profiling, coupled with principal component analysis and linear discriminant analysis (PCA-LDA), enables the identification of spectral biomarkers indicative of malignancy. Our approach achieved 87.2% accuracy, 87.3% specificity, and 87% sensitivity, reinforcing cerumen's viability as a body-fluid for cancer screening, paving the way for innovative, rapid, and cost-effective POC diagnostics, positioning earwax as a promising biofluid for translational medicine.

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Aptamer-conjugated SERS detection scheme for wastewater treatment

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Aptamers, which are single stranded DNA, is a cutting-edge innovation of DNA technology. Many researches have been shown that aptamers can be used in many detection systems. We are combining this DNA technology with surface-enhanced Raman spectroscopy (SERS) to allow for a very sensitive detection down to the trace level regime. SERS is known as powerful tool to address the analytical need in water and environmental analysis. [1] We aim the development of an innovative detection method for pollutants in water using SERS detection schemes by combining the SERS method with aptamer assisted microfluidic sensors. Due to optical features, aptamer-conjugated AuNPs gained huge attention for designing of biosensing systems.

Three chemicals serving as indicator parameter for water quality are under investigation, i.e. Carbamazepine, Diclofenac and Benzotriazole, which represent typical contaminations in wastewater. In order to target these chemicals, SERS-active surfaces are modified with selected aptamers which are specific toward their respected target. Thus, these molecules can be identified in low concentrations accompanied by a high specificity. Within this presentation, we will introduce our SERS platform to be applied in water analytics, which is a microfluidic platform with incorporated gold nanoparticle arrays. The recorded SERS spectra are dominated by the contribution of the aptamer molecules, i.e. the DNA bases, and it is expected that their SERS response is altered upon interaction with their target due to changed orientation towards the metallic surface. [2]



Fig. 1: Schematic representation of aptamer-gold nanoparticles on microfluidic chip.

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Analysis of in vivo and in vitro DNA binding affinities of human BCL6 protein

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The transcriptional repressor B-cell lymphoma 6 (BCL6) plays a pivotal role in regulating germinal center B-cell functions and is critically involved in lymphomagenesis. Precise mapping and quantification of its DNA-binding activity are essential for understanding its physiological roles and pathological dysregulation. To investigate direct DNA-binding of BCL6 to selected genomic targets, we applied a combination of established biochemical assays and advanced plasmonic sensing.

Initially, DNA-affinity purification was performed using biotinylated DNA probes representing BCL6 ChIP-seq target sites [1]. Wild-type and DNA-binding-deficient BCL6 variants were expressed in model cell systems. Pull-down experiments followed by Western blot detection confirmed direct binding of BCL6 to several target sequences, with binding strongly correlating to the presence of canonical BCL6 DNA motifs. Parallel luciferase reporter assays assessed the functional impact of BCL6 on transcriptional regulation, supporting sequence-specific regulatory function.

To complement these biochemical approaches, localized surface plasmon resonance (LSPR) sensing [2] was employed as a label-free, real-time method for quantifying the binding interaction of recombinant BCL6 protein to immobilized DNA targets. Gold nanoparticle sensor transducers provided high sensitivity for detecting subtle binding events. LSPR analyses revealed differential binding affinities, with higher affinity binding correlating with target sequences containing conserved BCL6 consensus motifs.

Together, these complementary methods demonstrate that BCL6 binding to DNA is motif-dependent and varies across target sequences. Our results highlight the value of combining classical biochemical assays with modern plasmonic techniques for the detailed characterization of transcription factor-DNA interactions.

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Nanocube Quadromer for Efficient LSPR Biosensing

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Over the past two decades, advancements in the understanding of physical phenomena and fabrication techniques have motivated and guided plasmonic nanostructure research and development [1]. Nanostructures composed of noble metals show unique optical responses associated with the plasmon resonance - a phenomenon linked to the collective oscillations of the conduction electrons resulting in the nanoscale confinement. The plasmonic properties of the isolated individual subwavelength particles with various materials and morphology, including spheres, cubes, rods, discs, stars, triangles etc., as well as their oligomers and random aggregates, have been widely explored [2]. As a consequence, extremely high absorption and scattering of the incident light are observed, associated with the localized surface plasmon resonance (LSPR). Compared to the individual particles, nanoparticle arrays can produce even more enhanced optical near fields in the nanometer-scale separation gaps between particles acting as subwavelength resonators for surface plasmons [Fig.2 (c)-(e)]. The operation principle of the LSPR sensors is based on the detection of a change in resonance wavelength upon a change in the ambient refractive index of the nanostructure caused by binding events of various biomolecules to receptor molecules attached to the nanosensor surface. This work investigates the structure of four nanocubes placed face to face as an effective plasmonic biosensor. Under a certain orientation of the nanoparticles against the incident field, a new narrower resonance arises near the conventional one due to the coupling of gap plasmons and localized surface plasmons. For the given dimensions and orientation of the incident field, the second peak exhibits a resonant shift of 40 nm for a refractive index change of $\Delta n=0.1$, whereas the primary resonance shifts by only 15 nm (see Fig.2 (b)). This phenomenon opens a new approach to building more sensitive LSPR biosensors based on structured nanoparticle aggregates.



Fig. 1. The side view of gold nanocube quadromer cluster on SiO_2 substrate. Nanocubes sides are 40 nm, and the gap g=5nm

Fig. 2. The extinction spectrum of nanocube quadromer (a) under oblique plane wave and (b) in different environments. (c-e) The electric field [V/m] distribution for different incident angles: 0, 45 and 90 degrees at the incident wavelength of 810 nm.

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In tip enhanced spectroscopy techniques, a metallic tip of some ten nm radius is brought into close proximity of a sample surface in an atomic force microscope. The tip is illuminated and, for example, the back-scattered light is collected and analyzed to give spectra of sub-diffraction limited spots of the sample surface. The spot size is largely defined by the optical near field enhancement of the tip and is typically in the range of or below about 10 nm.

In this work we investigate the signal generated when a gold-coated silicon nitride tip is scanned across lithographically fabricated gold nanoparticles on a dielectric substrate. Even without any molecules on the surface, we find a clear signal in the Stokes-shifted, backscattered light, that can be largely attributed to the spectrally broad photoluminescence of gold. The strongest signal appears at the edges of resonantly excited gold nanoparticles and thus resembles the particles' optical near field distribution. When adding molecules to the surface, their Raman signal is found on top of the photoluminescence signal.

The results indicate the possibly beneficial application of structured plasmonic surfaces in tip enhanced measurements.



Fig. 1

Fig. 1: a) Tip enhanced fluorescence map and b) scanning electron micrograph of rectangular gold nanoparticles with different aspect ratios. The fluorescence is excited by a 632.8 nm HeNe laser with horizontal polarization with respect to the figures.

Broadband optical chirality enhancement in 3D plasmonic Archimedean spiral for chiral sensing

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Molecular chirality plays a pivotal role at every scale in the natural world. Chiral enantiomers with identical physical and chemical properties can govern different behavior in biochemical processes. Simply the difference in the geometrical handedness of enantiomers could be the difference between a drug and a toxin. Therefore, chiro-sensitive molecular detection is of strong interest to the emerging fields of chiral sensing and analysis. Circular dichroism is conventionally used to detect optical chirality, but it is constrained by the weak and narrowband chiroptical effects of the molecules, thereby, necessitating high sample concentrations and extended measurement times. Tseng et al showed that using a metasurface of 3D plasmonic single-arm Archimedean spirals makes it possible to get strongly enhanced and stably localized broadband near-field optical chirality in the middle infrared regime [1]. However, the fabricated structures have proved to be structurally unstable. Furthermore, the spectral regime of operation used in this work is not friendly to easily available and highly sensitive optical equipment. This limits the potential applicability in detecting the targeted molecules at extremely low concentrations. To circumvent this, we demonstrate a 3D plasmonic twoarm Archimedean spiral that achieves large broadband optical chirality enhancement in visible to near infrared region and robust structural stability. Numerical calculations of the near-field optical chirality enhancement and transmission have been systematically investigated. Moreover, fabrication of the simulated structures achieved by using focus ion beam milling has been demonstrated.

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Flexible Silver-Coated Gold Nanoparticles Loaded Graphene textiles for Direct Detection of Imidacloprid Pesticide via Label-Free SERS

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Imidacloprid (IMI) is a widely used pesticide on crops, fruits and vegetables, but its residues pose a threat to human health [1]. Surface-enhanced Raman scattering (SERS) technology has become a widely used spectroscopic tool for identifying and detecting chemical and biological species because it provides rich fingerprint information at trace or even single molecule levels [2]. Noble metal nanomaterials (gold and silver) are often used as SERS substrates due to their unique physical and chemical properties. Compared with single-component AuNPs or AgNPs, gold-silver core-shell nanoparticles (Au@AgNPs) exhibit more excellent physical and chemical properties due to their multiple components [3]. Graphene (G), as a two-dimensional material, is widely used in

the manufacture of SERS substrates. It not only brings chemical enhancement and target molecule interaction, but more importantly, it improves the sensitivity, stability and reliability of SERS substrates [4]. Compared with rigid substrates (such as silicon wafers and glass slides), flexible substrates can directly collect analvtes from complex surfaces through "adhesion" and "wiping" methods to improve detection efficiency [5].

In this study, a multilayer mesh-structured graphene flexible material (GF) was used as a scaffold to adsorb Au@AgNPs, so that the Au@AgNPs were close to each other and "hot spots" were formed between adjacent Au@AgNPs to prepare a SERS substrate called Au@Ag-GF (see Fig. 1). Based on this, a label-SERS free sensor for the detection of imidacloprid pesticide was developed. Flexible, reliable, and stable SERS sensors are expected to be used for direct, sensitive, and rapid on-site detection of pesticides in apples.



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SERS detection of benzotriazole for wastewater treatment

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Benzotriazole is an organic compound with broad applications in various industrial processes. It is widely used as a corrosion inhibitor, antifreeze and dishwashing detergent. Benzotriazole, see the molecular structure in Fig. 1, is water soluble and quite persistent against biological and photochemical degradation, which makes is only partly removable from the wastewater. At the same time, it is classified as a toxic compound, which can lead to cell cycle disruption, cause adverse effects in aquatic organisms, poses risks to human health. Hence, there is an urgent demand for on-site benzotriazole detection in the wastewater. [1]

Surface enhanced Raman spectroscopy (SERS) is a powerful spectroscopic method enabling the detection of molecular analytes with concentrations down to nanomoles. [2] In conventional Raman spectroscopy signal has low intensity, while in SERS it is enhanced with the localized surface plasmon resonance, which is achieved by utilizing metal nanoparticles. Here, silver nanoparticles were deposited on top of the glass substrate to create local enhanced electromagnetic field. A sensor, consisting of microfluidic chip, as depicted in Fig. 2, with incorporated SERS-active nanoparticles, is presented. Fig. 3 shows the schematic illustration of the SERS-based detection. This detection method allows rapid *in-situ* trace detection of benzotriazole in water, using small volume of the samples.



Fig. 1: Molecular structure of benzotriazole

Fig. 2: Microfluidic chip applied in Fig. 3: Schematic illustration of the SERS detection SERS experiments

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Plasmon-Induced Photopolymerization of Molecularly Imprinted Polymers on arrayed Nanopartciles for Nanosensor Applications

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A new simple, fast, and versatile method is developed to functionalize gold nanoparticles (AuNPs) via nanoscale layers of molecularly imprinted polymers (MIPs). The key step is based on near-field radical photopolymerization of the MIP prepolymerization mixture [1]. This enables the fabrication of AuNPs@MIPs hybrid nanoparticles, which are used as substrates for localized surface plasmon resonance (LSPR) and surface-enhanced Raman spectroscopy (SERS) analysis with excellent sensitivity and specificity. This fabrication method allows to obtain robust and reusable sensor surfaces with high sensitivity and selectivity. The nanometric thickness of the MIP allows for shorter analysis times (10 min), thereby improving the performance of MIP-based sensors and opening up new perspectives for the detection of molecules at very low concentrations.

In order to realize multiplex LSPR sensing, gold nanoparticle arrays were prepared by spotting, and such substrates functionalized with MIPs using plasmon-induced photopolymerization. The poster presents the characterization of these MIPs-functionalized gold nanoparticle arrays.

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The function of proteins is determined by their structure and molecular dynamics. Advances in electron microscopy have radically improved the access to structural information. Conversely, the acquisition of protein dynamics over long timescales remains challenging^[1].

Here, we present a plasmon ruler-based single-molecule approach to investigate conformational dynamics over 24 hours in and out of equilibrium (i.e. in the absence and presence of ATP). This method does not impose external forces and enables the measurement of protein dynamics over six orders of magnitude – which cannot be achieved by established single molecule techniques ^[2-5]. We applied this technique to study the dynamics of the heat shock protein 90 (Hsp90). We identified states with long-term dwell times in the minute timescale (linked to rarely visited states) beside the well-known fast dynamics in the 0.1-10 s range.

Our technique enables us to access the complex local and global conformation dynamics at the level of individual proteins. Consequently, the conventional "structure-function" paradigm is expanded to encompass the "structure-timescale-function" framework. By accessing the dynamics of single proteins, we are able to address important questions concerning conformational heterogeneity among proteins, ergodic behavior, and non-Markovian dynamics ^[6]. For the dynamics of Hsp 90, we confirmed memory in the conformational dynamics for up to 50 s^[7]. This timescale is similar to processes such as the translation of proteins and could be a significant factor in Hsp90's role as a chaperone in the folding of proteins.

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Complexes of plasmonic noble-metal nanoparticles with nucleic acid molecules represent an interesting material for various applications, mainly in biosensing and plasmonics. When excited with light, emission of hot electrons can be observed at such particles. This process is assumed to cause observed effects like local degradation of PMMA [1], but also excitation transfer over a few micrometers along extended double-stranded DNA [2] including fluorescence quenching [3].

In order to elucidate the mechanism of this process, we propose the use of charge-induced fluorescence quenching. The excitation-induced transfer of charges from a nanoparticle is expected to quench the emission of a dye that is attached to the nanoparticle via a DNA strand. In this contribution, we demonstrate a first system consisting of a fluorescent dye coupled to a gold nanoparticle via 55-base pair DNA strand. The emission of the dye was chosen to have low spectral overlap with the nanoparticle absorption, in order to minimize the quenching by radiative (Förster) transfer. Fist measurements indicate a very efficient quenching of the dye when it is excited directly, which could indicate a charge transfer from the dye to the nanoparticle. In further investigations we therefore plan to observe the charge of the nanoparticle via gold emission measurements [4].

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To achieve high sensitivity in localized surface plasmon resonance (LSPR)-based nano-biosensors [1], we investigated enhancing plasmonic coupling [2, 3] by optimizing nanoparticle immobilization into nanohole arrays. The fabrication approach of hybrid nanosensors relies on the directed assembly of 80 nm colloidal gold nanoparticles into a lithographically produced nanohole array patterned on a glass substrate. Following selective immobilization on a chemically functionalized surface, the PMMA mask is lifted off, removing unbound particles and retaining only those embedded within the nanoholes. This template-assisted method enables precise nanoparticle placement with high filling-efficiency and spatial order, crucial for strong plasmonic coupling. Single-particle extinction spectra in the visible range were acquired via a micro-spectrophotometer to evaluate spectral shifts related to nanostructure configurations. This scalable, cost-effective platform paves the way for robust, highly sensitive biosensors applicable in healthcare, drug discovery, environmental monitoring, toxin detection, and food safety.

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Gold nanoparticle-facilitated assembly of supernatant transfer enables the rapid determination in the discovery of ssDNA aptamers specific to steroid hormones

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Single-stranded DNA (ssDNA) aptamers have been demonstrated as bioreceptors which can bind to target molecules via systematic evolution of ligands by exponential enrichment (SELEX). We previously introduced gold nanoparticle-assisted SELEX (GNP-SELEX) to address the limitations of real-time monitoring during conventional round-by-round selection. This platform enabled rapid determination of target-specific ssDNA library enrichment without requiring target immobilization or modification. While GNP-SELEX offers advantages for straightforward, real-time screening in ssDNA aptamer discovery, it faces challenges related to ssDNA-dependent GNP adsorption and restricted target applicability. To overcome these issues, we developed GNP-facilitated assembly of supernatant transfer (GNP-FAST) as a pre-screening and pre-selection step prior to GNP-SELEX. GNP-FAST not only informs on target accessibility for GNP-SELEX but also aids in narrowing down target-specific ssDNA library pools through serial dilution of extensive ssDNA libraries and target molecule combinations facilitated by centrifugation and continuous GNP addition. This process allows for monitoring target binding via salt-induced color changes in pelleted GNPs. We applied cortisol, a steroid hormone, as a model target molecule in both GNP-FAST and GNP-SELEX, successfully identifying high-affinity cortisol-specific aptamers. We propose that this platform offers a rapid and convenient approach for discovering ssDNA aptamers against diverse target molecules.



Fig. 1: Schematic Illustration of GNP-FAST and GNP-SELEX for the discovery of target-bound ssDNA aptamer library

Label-free detection of SARS-CoV-2 (RBD) by surface plasmon resonance

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SARS-CoV-2, the virus responsible for the COVID-19 pandemic, infects uman cells through the highaffinity interaction between its receptor-binding domain (RBD) on the spike (S) protein and the angiotensin-converting enzyme 2 (ACE2) receptor. Understanding and detecting this interaction are critical for developing diagnostic tools and therapeutic interventions.¹ Surface Plasmon Resonance (SPR) is an advanced, label-free technique used to monitor biomolecular interactions in real-time and determine binding affinities.² In this study, we will present an SPR assay designed for detecting SARS-CoV-2 RBD using biotinylated ACE2-functionalized sensor surfaces. We will report the kinetic parameters, including the association rate constant (k_a), dissociation rate constant (k_d), and equilibrium dissociation constant (K_D) to characterize the RBD-ACE2 interaction. Additionally, different buffer media were evaluated to minimize non-specific binding, enhancing the assay's accuracy and reproducibility.



Fig. 1

Fig. 1: Scheme of SPR assay for measuring the binding of SARS-CoV-2 (RBD) to ACE2. Graph made with Biorender.

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Immobilised gold and silver nanoparticles for an optofluidic SERS platform

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The combination of SERS with microfluidics not only miniaturises the system but also introduces the possibility to control the environmental parameters of the analyte, which is not present when using either a droplet or cuvette. This allows for applications involving tiny sample volumes and control of sample environments. Direct usage of nanoparticles in solution with the analyte can cause the adsorption of the nanoparticles onto the surfaces of microfluidic systems, leading to undesirable memory effects. By using plasmonic nanoparticles of gold, silver or a mixture of the two, immobilised on removable glass or PDMS inserts, the life and reusability of the channels can be largely improved ^[1] because they can be inserted into the channel and replaced after use. Using immobilised nanoparticles also introduces a relatively large surface that can be scanned and can improve the reproducibility of the nanoparticle substrate properties and hence the measured spectra. Here, we will discuss strategies for the fabrication and characterization of immobilised plasmonic

Here, we will discuss strategies for the fabrication and characterization of immobilised plasmonic nanoparticles for bioanalytical applications in optofluidic systems. We will present the plasmonic properties as well as surface-enhanced Raman scattering (SERS) data of relevant compounds.

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Development of a low-cost filter membrane based on cannabis sativa for the reduction of bacterial loads in non-potable domestic water in certain regions of the capital through green synthesis

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Drinking water scarcity affects more than 40% of the global population, an alarming statistic that is expected to worsen with climate change. Therefore, Sustainable Development Goal 6 aims to ensure access to clean water and sanitation, contributing to health improvement by preventing the spread of infectious diseases associated with waterborne pathogens, which pose significant health risks [1,2]. Consequently, it is crucial to secure access to drinking water and reduce consumption-related diseases. Nanotechnology, particularly silver nanoparticles, offers excellent properties for biotechnological applications due to their antibacterial capabilities and selective toxicity towards microorganisms [3].

This study addresses bacterial contamination in water bodies, a serious public health challenge exacerbated by rising antibiotic resistance. The research proposes a hemp biofilter incorporating silver nanoparticles (AgNPs), synthesized from Cannabis sativa leaf extracts, to combat bacterial contamination in water [4]. AgNPs are effective against bacteria due to their antimicrobial properties, and their integration provides a sustainable and eco-friendly solution without generating toxic waste [5].

Silver nanoparticles (Bio-AgNPs) were synthesized using a green approach, employing silver nitrate (AgNO₃), sodium dodecyl sulfate (SDS), and aqueous Cannabis sativa leaf extract as the reducing agent. Gas Chromatography-Mass Spectrometry (GC-MS) was utilized to characterize the compounds extracted from Cannabis sativa. The physicochemical characterization of the nanoparticles was performed using Scanning Transmission Electron Microscopy (STEM), Energy Dispersive Spectroscopy (EDX), Fourier Transform Infrared Spectroscopy (FTIR), Raman spectroscopy, and UV-Visible Spectroscopy. The concentration of Bio-AgNPs was confirmed via Atomic Absorption Spectroscopy (AAS). The antimicrobial activity was evaluated against E. coli (ATCC 25923) through plate microdilution, diluting the Bio-AgNPs from 200 ppm to 6.25 ppm. Hemp fiber was processed into three layers impregnated with Bio-AgNPs, which were then dried under vacuum at 25°C for 24 hours, forming the membrane intended for reducing bacterial loads.

The GC-MS analysis revealed the bioactive compounds present in the Cannabis sativa extract as cannabidiol (CBD), cannabinol (CBN), and various phenolic compounds, which are responsible for the reduction of AgNO₃. FTIR analysis of the Bio-AgNPs confirmed the presence of organic groups characteristic of proteins, phenols, flavonoids, amino groups, alkanes, and alkenes from the Cannabis sativa leaf extract. The UV-Visible spectra showed the characteristic Surface Plasmon Resonance (SPR) signals of the AgNPs. UV-Vis spectroscopy was also employed to continuously monitor the nanoparticles and plant extracts over 15 days, tracking the evolution of the Bio-AgNPs. Although pure metallic silver does not exhibit active Raman modes in this region, the Bio-AgNPs displayed a band at approximately 260 cm⁻¹, attributed to the coupling effects of localized surface plasmons and the vibrational modes of the adsorbed molecules. This phenomenon is commonly observed in surface-enhanced Raman spectroscopy (SERS), where the Raman signal of the adsorbed molecules is significantly amplified due to their interaction with the surface plasmons of the AgNPs. EDX confirmed the presence of Ag in the Bio-AgNPs. SEM morphology results indicated that the Bio-AgNPs have an average size of 11.9 \pm 5.2 nm, which optimizes their interaction with Bio-

Synthesis of plasmonic nanoparticles and bulk sensitivity investigation for advancing LSPR sensing technology

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Localized surface plasmon resonance (LSPR) sensing is a powerful technique that exhibits the sensitivity of plasmonic nanostructures to changes in their surrounding refractive index. When light interacts with nanoparticles, their free electrons oscillate collectively, producing a resonance condition highly sensitive to the local environment [1,2]. This phenomenon makes LSPR-based sensors ideal for detecting analytes in biosensing, chemical analysis, and environmental monitoring.

To evaluate the optical properties and bulk refractive index sensitivity S_B , defined as the resonance wavelength shift of the extinction spectrum per unit change in the refractive index of the surrounding medium, we synthesized gold nanospheres, gold nanotriangles, and silver nanotriangles [3,4]. These nanostructures were characterized using several techniques, including SEM, TEM, UV-Vis spectroscopy, dynamic light scattering (DLS), and zeta potential measurements. In sensitivity tests silver nanotriangles showed the highest bulk refractive index sensitivity ($S_B = 300-700 \text{ nm/RIU}$). However, their susceptibility to oxidation in air limits their practical use in LSPR sensing. On the other hand, gold nanoparticles remain stable in air, and gold nanotriangles, being anisotropic, generally exhibit higher sensitivity. This was confirmed by our experiments, where gold nanotriangles of various sizes showed bulk sensitivity in the range of 200-330 nm/RIU, compared to 50-75 nm/RIU for gold nanospheres. The synthesis of gold nanotriangles, however, requires cetyltrimethylammonium chloride (CTAC) as a shape-directing and capping agent. The resulting dense bilayer of CTAC on the surface of gold nanotriangles significantly hinders the binding of biomolecules, limiting their application as nanotransducers in LSPR sensing. To address this limitation, we are working on developing effective ligand exchange strategies to replace CTAC with alternative ligands, such as citrate, alkylamines and -thiols, polyvinylpyrrolidone (PVP) or thiolated PEGs. As a first step in this direction, we propose methods for replacing the CTAC surfactant on the surface of gold nanotriangles with methoxypolyethylene glycol thiol (m-PEG-SH) and 2,5-dimercapto-1,3,4thiadiazole (DMT).



Fig. 1: Scanning electron microscopy (SEM) images of gold nanotriangles. Labels refer to the volume of intermediate seeds used in each synthesis. Scale bars are 100 nm

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Development of a Lateral Flow Assay Biosensor for miRNA-34a and miRNA-155 Detection Utilizing the Rolling Circle Amplification

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The rapid and accurate detection of microRNAs (miRNAs) holds immense promise for transforming disease diagnosis and management. MicroRNAs (miRNAs) are a class of small non-coding RNAs that play crucial roles in the regulation of gene expression and various biological processes. Aberrant expression levels of specific miRNAs have been implicated in the pathogenesis of numerous diseases, including cancer [1]. Among the various detection methods available, lateral flow assays (LFA) biosensors have emerged as powerful tools for rapid and point-of-care detection of biomolecules due to their simplicity, portability, and cost-effectiveness [2]. We present the development of a novel LFIA biosensor for the detection of two important oncogenic miRNAs, miRNA-34a and miRNA-155, using rolling circle amplification (RCA) as a signal amplification strategy. The integration of RCA with LFA offers several advantages, including enhanced sensitivity and specificity for target detection [1].

In our biosensor design, the amplification step is done in solution. Gold nanoparticle-detection probe conjugates specific to miRNA-34a and miRNA-155 are immobilized onto the conjugate pad of the LFA strip [3], and biotinylated DNA probes specific to miRNA-34a and miRNA-155 are immobilized onto the test lines of the LFA strip [4]. After the amplification step of the target sample, the product is applied to the LFA strip, allowing the semi-quantitative measurement of the target concentration in 15 minutes with an optical reader.

In the images, we show the performance of the lateral flow assay (LFA). The assay demonstrated the capability to detect miRNA-34a and miRNA-155 at a concentration of 20 pM and 2 pM respectively. Additionally, we have successfully integrated both test lines to enable the simultaneous detection of multiple miRNAs on a single strip, enhancing the assay's multiplexing capability.



Fig. 1: Example LFA strips for the detection of individual miRNAs. Fig. 2: Simultaneous detection of 2 targets by multiplexing.

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Investigation of plasmonic enhancement of hyper Raman and Raman scattering

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Similar to Raman scattering, its two-photon analogous Hyper Raman scattering (HRS) can also be significantly enhanced by utilizing the highly confined optical field in plasmonic structures. Because of its unique selection rules, HRS allows us access to silent and infrared active modes and can be used as a complimentary technique to SERS to provide a complete vibrational profile. However, finding a suitable substrate for Hyper Raman enhancement has been proven to be challenging, as the requirements are more stringent and physico-chemical properties play a more active role compared to their one-photon Raman counterpart.

In this work, we have focused on utilizing both gold and silver spherical nanoparticles for HRS enhancement. We investigated both electronically resonant and non-resonant systems and compared the enhanced HR vibrational modes with the enhanced Raman ones. We also report the effect of salt induced aggregation of the nanoparticles and how the aggregation mechanism translates in SERS and SEHRS spectral differences.



Fig. 1: Schematic representation of different vibrational scattering mechanisms Fig. 2: Representative SERS and SEHRS spectra of a non-resonant system

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Localized Surface Plasmon Resonance (LSPR) studies of protein-DNA interactions

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Interactions between nucleic acids and proteins are of upmost importance for cellular processes fundamental for cellular and higher life. There are, specific as well as unspecific, interactions between nucleic acids and proteins. These interactions are essential and play key roles during the replication, the transcription, and the translation of DNA. For example, single-stranded DNA-binding (SSB) proteins wrapping up single-stranded DNA (ssDNA) preventing the rehybridization and therefore stabilizing the single-stranded form ^[1]. Despite its importance the core mechanism and exact role of SSB proteins is still unknown. Therefore, studying SSB proteins is important, contributing to our understanding of cellular processes and presents new opportunities for handling cells. A suitable tool for studying these interactions is the localized surface plasmon resonance (LSPR) spectroscopy, which enables the real-time observation of the binding between ssDNA and SSB proteins under various different conditions like protein concentrations, salt concentration, mutated proteins, and aging effects by tracking the centroid position of the plasmonic absorption peak. This position is strongly influenced by the refractive index at the nanoparticles surface. When modified with a specific receptor (e.g. ssDNA), this local refractive index around the particle changes upon molecular binding or dissociation, and therefore allows to study these binding processes utilizing a time resolved spectrometer ^[2,3].



Fig. 1: LSPR-based detection of DNA binding proteins interacting with DNA.

(A) In the case of double-stranded DNA, it is immobilized on the surface of gold nanoparticles utilizing a terminal thiol group. The DNA can be hybridized either in advance or right before flushing the protein through the microfluidic chamber. The same principle applies to SSB proteins, here single-stranded DNA is utilized as capture. (B) The time resolved position of the LSPR peak shows the binding of SSB B proteins to DNA and its dissociation when PBST buffer is flushed over the sensor during an experiment.

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Probing intracellular interactions with SERS in the presence of a fluorescent dye

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The use of fluorescent dyes for distinguishing and targeting organelles is at the center of many biological studies at the single cell level. The optimization of fluorophore combinations for multichannel imaging and fluorophore concentration for live cell imaging are the starting point for analysing diverse metabolic processes, and the combination with other techniques for multimodal imaging opens the possibility for simultaneously characterizing structure, dynamics and molecular composition of different organelles or intracellular structures. In particular, surface enhanced Raman scattering (SERS) spectroscopy is a sensitive and powerful tool to characterize intracellular interactions with or near plasmonic nanoparticles.[1,2] Combining fluorescence and SERS microscopy requires careful consideration of the experimental conditions to avoid high backgrounds and signal hindering, but it is also necessary to understand how fluorophores and nanoparticles may interact in the intracellular space and what effect does that have regarding the cellular integrity.

In this work, we used gold nanoparticles for sensing the effect of a fluorescent dye in the intracellular interactions in vesicles along the endolysosomal pathway in vivo. We used LysoSensor Green DND-189 and evaluated the interactions with gold nanoparticles inside lysosomes via SERS spectroscopy. Depending on the concentration of LysoSensor used and where nanoparticles are located along the endolysosomal pathway, the SERS signals observed show differences in the interactions with different biomolecules like proteins and lipids. We characterized the Raman and SERS spectra of the fluorophore at different pH values to compare them to the spectra obtained in living cells. Gold nanoparticle distribution and aggregation in the cells and the cellular ultrastructure were characterized by soft X-ray tomography in combination with the distribution of lysosomes by fluorescence microscopy. Understanding how different probes interact in living cells can help the improvement of multimodal imaging and the interpretation of results.

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Numerical Demonstration of Archimedean spiral with highest dissymmetry

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Surface Plasmon Polariton s (SPPs) are collective oscillations of electrons at the metal/dielectric interface, where electromagnetic field is confined and enhanced [1]. Metallic nanostructures are therefore widely applied to manipulate light in nanoscale. Here we exploit a left handed Archimedean spiral (LH-AS) structure made on an aluminum film (Figure 1a) to control the emission of an emitter at the center of the AS by changing the handedness of circularly polarized excitation light (CPL). The working principle is based on the fact that the LH-AS can focus SPPs to a bright spot but generate a dark spot at the centre with right handed (RH) circular polarized (Figure 1b and c). Therefore, the local electric field and hence the excitation and emission of the emitter at the centre of AS depend on the illumination chirality by tuning the dissymmetry factor (g) from the center spot, i.e., $g = 2(L_L - L_R)/(L_L + L_R)$ [3], that has been optimized to 1.99, which is very close to the maximum possible value of 2 (Figure 1d). The absolute intensity difference of the field at the centre is also optimized by scanning the number of AS rings [4]. The map of the g factor of the final structure is shown in Figure 1f. This work presents aluminum AS that exhibits a dissymmetry factor at the center larger than any other reported structure



Fig. 1: Figure 1(a) The design of 8-ring LH-AS on a quartz substrate and its mathematical formulation in polar coordinate. (b,c) FDTD simulation results of 8-ring LH-AS with a bright and dark spot at the center, when excited with RH-CPL and LH-CPL (as shown in the inset). (d,e) the plot of g-factor and Intensity difference respectively with respect to the number of rings of AS. (f)g-factor map of the 8 rings AS with the maximum g-factor of 1.99 at the center.

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SERS for cytosine methylation detection: a step toward epigenetic biomarkers

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Cytosine methylation is one of the most common epigenetic modifications and frequently observed events in various diseases such as cancer. Despite the various methods for analyzing DNA methylation, the estimation of methylated cytosine remains a challenge due to the lack of rapid and highly sensitive detection techniques. We herein describe a sensitive and label-free surface-enhanced Raman spectroscopy (SERS) method [1] for determining the spectral profiles of cytosine and its methylated form.

By comparing the SERS spectra of a nucleotide base and its methylated counterpart, we identified the potential spectral markers of methylated cytosine. This method utilizes gold nanoparticledecorated crystalline silicon substrate with nucleotide base solutions deposited on the surface to obtain spectra. Then, we apply discriminative spectral analysis to evaluate the position and intensity of peaks and correlate them with different percentages of methylated cytosine in the solution. Our research has shown that the position of the peaks in the spectrum remains stable, but their intensity changes significantly depending on the fraction of methylated cytosine in solution. While the intensity of the in-plane bending C=O peak (600 cm⁻¹) decreases with increasing methylated cytosine abundance, there is a significant increase in the intensity of the ring breathing peak (790 cm⁻¹) with decreasing cytosine fraction in solution. The obtained results open prospects for quantitative evaluation of methylated cytosine levels in biological samples, which can be used in diagnostics for early detection of cancer processes.

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Silver nanoparticle decorated titania and zinc oxide nanowires by MS-ALD for photocatalytic dye degradation

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Because of their superior optical qualities, high oxidative power, high chemical and thermal stability, and non-toxicity, zinc oxide (ZnO) and titanium dioxide (titania, TiO_2) are the most researched photocatalytic materials for dye degradation. Unfortunately, both materials reveal a rather high band gap of 3.0-3.2 eV and 3.37 eV for ZnO and TiO_2 , respectively. The high band gap enables photocatalytic activity only under UV illumination [1].

This study presents a novel method for creating silver-nanoparticle-decorated titania and zinc oxide nanostructures with broad band absorption (200-2500 nm) that is based on well-established deposition techniques. Using a self-assembly technique, metastable ALD (MS-ALD) makes it possible to create intricate 3D nanostructures without the use of pre-structured or pre-defined 3D substrates [2]. Former, optimized structures based on silver decorated silica nanowires (Ag-NP@SiO₂) revealed superior optical absorption above 99 % from 220 nm up to 2500 nm [3]. By adjusting the corresponding MS-ALD process parameters, the morphology of the structures could be easily changed. Here, we introduce the growth behavior of two common precursors for the MS-ALD approach to 3D structure formation: diethylzinc (DEZ) and titanium (IV) isopropoxide (TTIP). The resulting structures consisted of silver nanoparticles (Ag-NP) and TiO₂ or ZnO nanostructures (see Fig. 1). A planar silver film will be used as the substrate and oxygen plasma as the co-reactant for the MS-ALD process. To find promising high absorption structures, dose times were adapted and formed structures were analyzed by cross-section SEM and their optical performance was determined (Fig. 2). X-ray diffraction was conducted to determine the crystal phases of TiO₂ and ZnO. Promising structures were tested for the degradation of a model dye AO7 (acid orange 7) in an aqueous solution under 3h UV illumination and compared with a model catalyst TiO₂ P25 (see Fig. 3) [4].



Fig. 1: Exemplary SEM images of structures formed using DEZ (A) and TTIP (B) as precursors for the formation of photocatalytic silver-decorated 3D nanostructures.

Fig. 2: Diffuse transmission, reflection and calculated absorption for an Ag-NP@ZnO nanostructure. Fig. 3: Norm. intensity of significant absorption peak of AO7 at 485 nm as a function of the UVexposure time for ZnO nanostructures. Suffix "E" and "O" stand for posttreatments using ethanol and oxygen-plasma, respectively.

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