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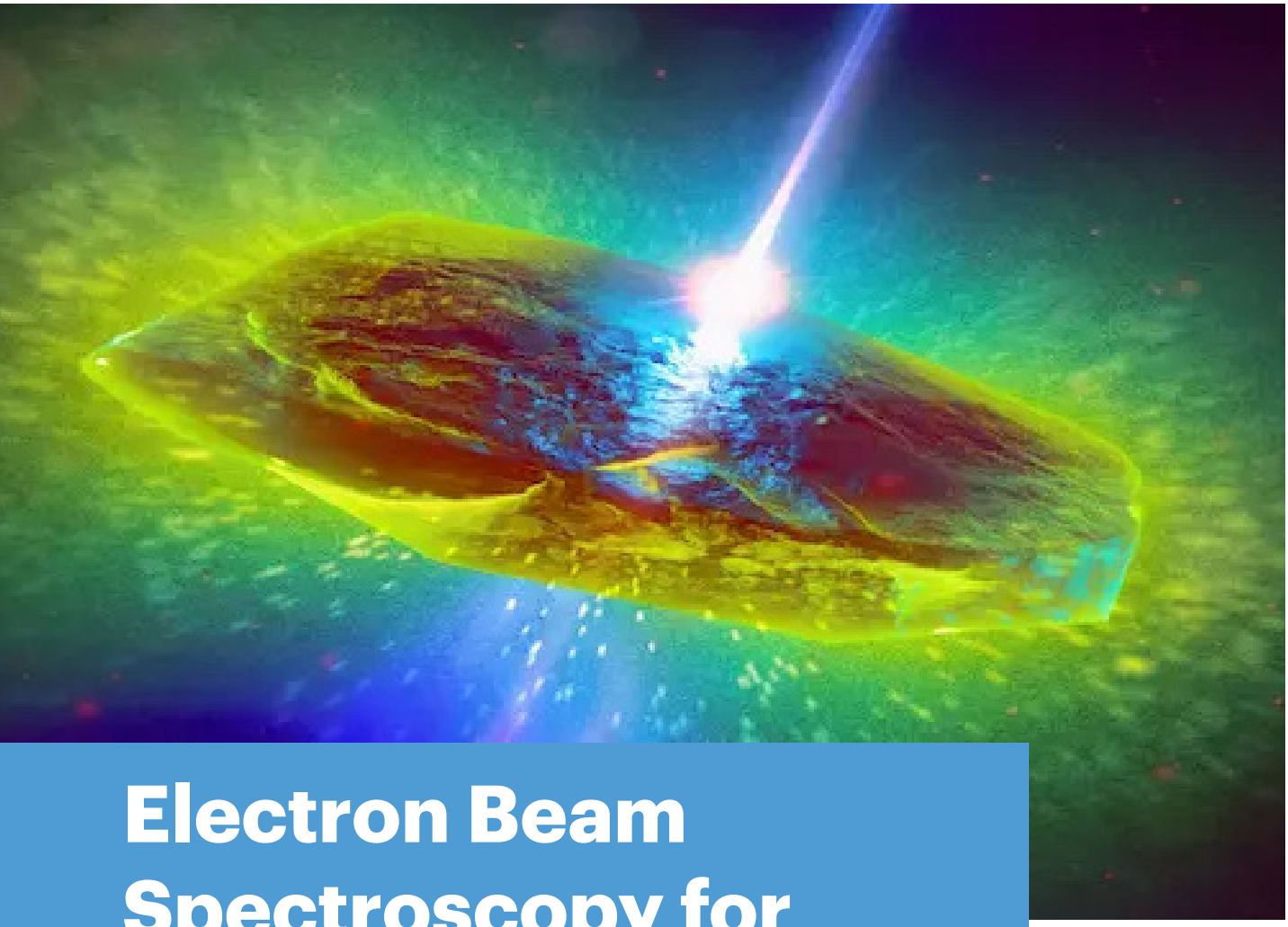

Electron Beam Spectroscopy for Nanophotonics 2025

29th - 31st October 2025
ICFO - Castelldefels (Barcelona), Spain

Book of Abstracts

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CONTRIBUTED TALKS

POSTERS

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Femtosecond and Attosecond Electron Microscopy with Single and Multiple Electrons

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All processes in materials, nanostructures and devices are on a fundamental level defined by electronic and atomic motion from initial to final conformations. Atoms realign in femtoseconds and electron densities respond in attoseconds when interacting with laser light. Our approach for a direct, real-space visualization is pump-probe electron diffraction and microscopy with single-electron wavepackets [1] under the control of laser light [2-9]. Terahertz waves are ideal to obtain isolated electron pulses [2-3] while modulation with the optical cycles of continuous laser light provides a train of electron pulses with attosecond shapes [4-5]. Using these concepts, we can visualize atomic motions and electron dynamics in space and time [6-7]. We will report selected results on magnetic materials [7], electronic circuitry [8], and light-wave propagation around metamaterials [4]. We will also report latest results on utilizing pulses with more than one electron to understand the quantum-mechanical basics of electron-electron interaction and scattering with materials [8].

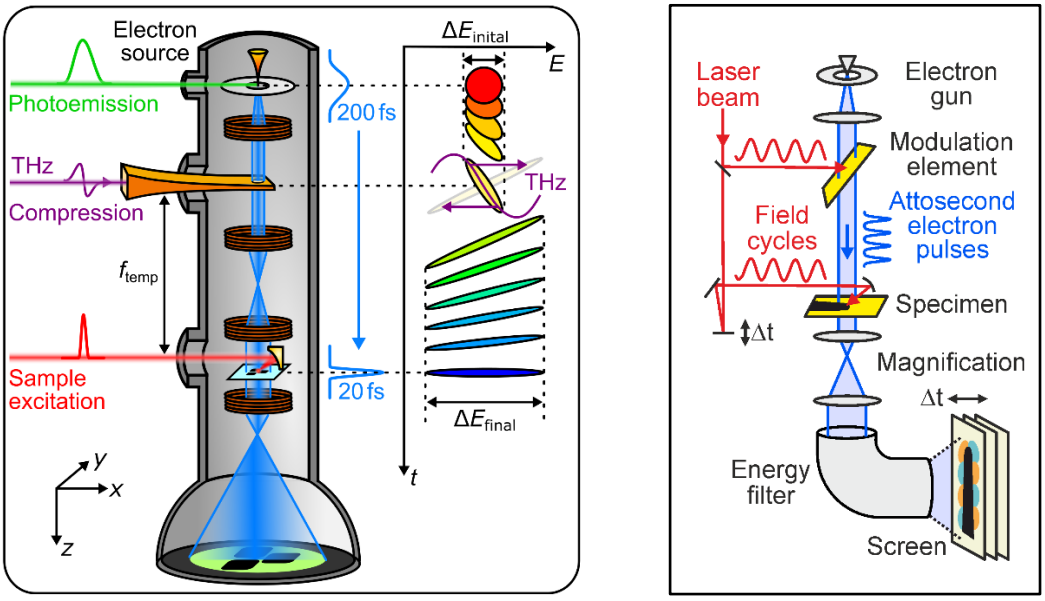

Figure 1. Selected concepts for controlling a single electron or multiple electrons in time by using the optical cycles of laser light. Left: Transmission electron microscope with femtosecond emitter, terahertz compressor and sample excitation. Right: Attosecond electron microscopy with pulse compression in a continuous laser wave.

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Incoherent and coherent cathodoluminescence spectroscopy –nanoscale light distribution in semiconductors

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The growing demand for precise nanoscale characterization tools has driven advancements in techniques capable of probing optical and thermal phenomena beyond the diffraction limit. In this work, we highlight recent developments in cathodoluminescence (CL) spectroscopy within a scanning electron microscope (SEM), enabling high-resolution studies of light-matter interactions at the nanometer scale.

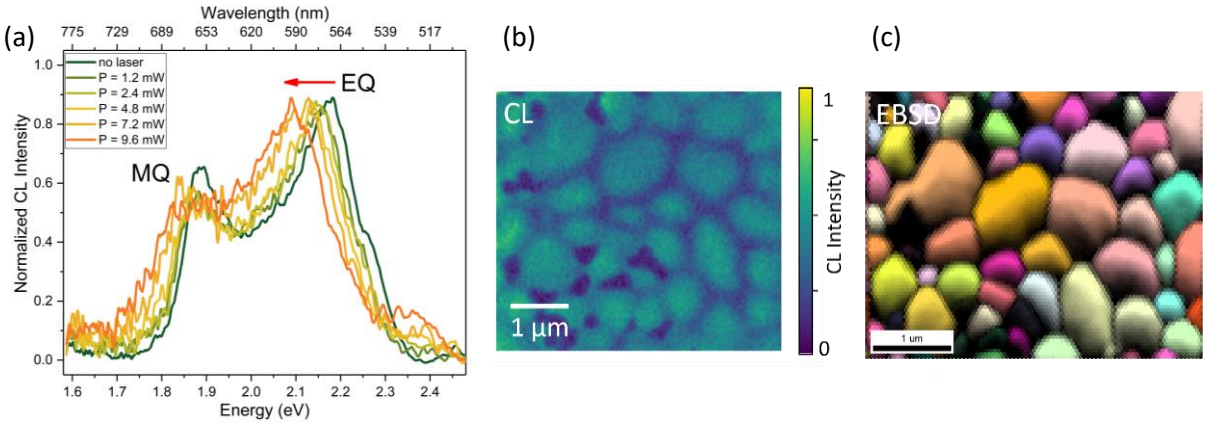

Figure 1. (a) Normalized CL spectra of an individual Si NP (d = 250 ± 2 nm) on a 15 nm-thin Si_3N_4 membrane averaged over the NP's rim (b = 90 - 120 nm), showing laser-induced red-shift of EQ (λ = 442 nm, cw, up to 9.6 mW, 1 µm spot size). (b) CL map (c) and EBSD map of the same area of the CsPbBr3 thin film.

In the first part, we demonstrate a novel approach to nanoscale thermometry using coherent CL from individual crystalline silicon nanospheres (NPs, d ~ 200–240 nm) exhibiting optical Mie resonances. By coupling 442-nm laser light into the SEM, we locally heat individual nanoparticles and use the electron beam as a probe to monitor spectral shifts in the Mie modes (see Fig. 1a). The electric quadrupole (EQ) mode serves as a sensitive temperature indicator, exhibiting red-shifts up to 100 meV corresponding to temperatures as high as 575°C. Thermal simulations reveal that the heat dissipation is highly dependent on the NP's contact area with the 15 nm-thin SiN membrane. We further implement a pump-probe CL technique by synchronizing ns-laser and electron pulses, allowing us to investigate the temporal dynamics of heating and cooling processes in a single NP. This approach opens new pathways for localized, time-resolved thermometry with potential applications in integrated electronics, optoelectronics, and nanoscale heat management.

In the second part, we explore the structural-optical correlations in polycrystalline $CsPbBr_3$ perovskite films. By combining electron backscatter diffraction (EBSD) and CL spectroscopy on the same region, we achieve the first direct mapping of crystal orientation with optical emission at the nanoscale (cf. Fig. 1b, c). While grain orientation does not significantly affect intra-grain CL emission, a strong decrease in CL intensity is observed at grain boundaries. Depth-resolved CL and optical simulations reveal the key roles of carrier diffusion and outcoupling in shaping the spectral response.

Together, these results illustrate the strength of SEM-based CL spectroscopy for in-situ exploration of structural, optical, and thermal properties in functional nanomaterials, with broad relevance to materials science, photonics, and device engineering.

Acknowledgments

ERC Grant Agreement No. 101019932 (QEWS), European Innovation Counsel (EIC) under Grant Agreements No. 101017720 (EBEAM) and No. 101151994 (EXPLEIN).

Controlling and Sensing Spin Systems with Free Space Electrons

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Coherent control and detection of quantum systems lie at the heart of quantum optics and quantum technologies. While optical and microwave control techniques are well established, the use of free-space electron beams as both manipulator and sensors opens an entirely new regime for spatial resolution and spectroscopic access.

In this talk, I will first outline intuitively our theoretical work [1] showing how the non-radiative near-fields of temporally modulated free-space electron beams can coherently drive quantum systems. I show that such manipulation can be performed with only classical control over the electron beam itself and that potential challenges like shot noise and decoherence through back action on the electrons are for certain parameter ranges insignificant for our approach. I will discuss possible implementations and show preliminary data on manipulating spin systems in a sample.

I will then present our recent experimental setup on SPINEM (Spin Electron Microscopy) [2], which integrates continuous-wave electron spin resonance (ESR) spectroscopy directly into a transmission electron microscope. By combining a custom microwave resonator with a free space electron beam [3], we achieve in-situ, phase-locked detection of microwave-driven spin precession with *picoradian* deflection sensitivity (~280 prad) and spatial mapping capabilities down to 30 µm. This approach directly senses the magnetic fields generated by precessing spins in the GHz regime, enabling localized spin spectroscopy in a TEM.

I will conclude with perspectives on pushing SPINEM towards atomic resolution and single-spin sensitivity [4]. These advances could open new frontiers in nanoscale spintronics, magnonics, and quantum materials research, and provide an electron-based analogue to magnetic resonance imaging at the level of individual nanostructures.

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Demonstrating Electron-heralded Photon State Preparation & Electron-Photon Entanglement

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Extending inelastic electron-light scattering to the single-electron single-photon regime has generated considerable interest, with theoretical studies promising the generation [1] and probing [2] of quantum states of light as well as entanglement-based quantum-enhanced sensing [3]. The advent of event-based electron detection, enabling electron-photon coincidence measurements [4,5], as well as a boost in the underlying interaction strength through velocity phase matching and resonant field enhancement [6,7], have brought these proposals within reach of state-of-the-art experiments. The preparation of nonclassical states of light and the demonstration of electron-photon entanglement are two critical milestones in the development of free-electron quantum optics.

In this contribution, we present the experimental realisation of these cornerstones: The demonstration of electron-heralded photon number state generation and the observation of free-electron-photon entanglement. For the former, we place integrated optical waveguide structures inside a transmission electron microscope (TEM) to enhance the electron interaction with a single optical mode, creating electron-photon pair states [4]. Using time- and energy-resolved detection of both particles, we trace the resulting correlation between energy-loss electrons and photon generation. Extending the photon detection setup by a second detector, we study the photon statistics heralded by such loss-electrons in a Hanbury Brown and Twiss (HBT) setup and observe an anti-bunching that verifies the generation of non-classical photon number states [8]. The underlying inelastic electron-light scattering is expected to induce entanglement, which can be investigated using a quantum eraser-type scheme involving the electron position and photon polarisation [9,10]. In the experiments, we pass two partial electron beams, generated in a coherent superposition utilizing an amplitude grating, near the two edges of a sub-micron-sized, metal-coated glass prism. At these edges, the electrons can spontaneously emit photons with a polarisation tied to the electron path [11]. Observing the interference of the recombined electron beams in coincidence with measurements of the photon polarisation in different bases, we experimentally implement a state tomography of the complete electron-photon quantum state and find a violation of the Peres-Horodecki entanglement criterion by more than 7 standard deviations. This constitutes a first unequivocal demonstration of free-electron-photon entanglement [11,12].

In summary, we discuss the preparation of electron-heralded photon number states and the verification of electron-photon entanglement. These results have significant implications for the development of free-electron quantum optics, demonstrating the feasibility of electron-driven sources of quantum light and paving the way to quantum-enhanced sensing in electron microscopy based on photon-mediated electron entanglement.

Acknowledgements: We gratefully acknowledge the contributions by our co-authors from the Göttingen UTEM, in particular R. Haindl, J. Kappert, O. Kfir & H. Lourenco-Martins, as well as the group of Tobias Kippenberg at EPFL, who fabricated the integrated photonic structures utilised in Refs. [4,6,8].

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Pushing the limits of detection in STEM EELS: magnon spectroscopy in an electron microscope

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Nearly a decade since first demonstration, vibrational electron-energy-loss spectroscopy has pushed the capabilities of analytical in a scanning transmission electron microscope (STEM) [1]. Phonon eigen modes can now be detected at atomic resolution [2], along with their dispersion in momentum space [3], and related to local atomic structure and chemistry. Magnons are quasiparticles representing the collective excitation of spins in magnetic materials. They, along with hybrid magnon-phonon quasiparticles (magnon polarons), are the basis for the operation of new spin wave transfer logic devices. They occupy the same energy loss windows as phonon modes, ranging from a few to a few hundred meV in solid-state materials, suggesting that STEM-EELS may offer the ability to detect them at the nanoscale.

Here, we show that bulk THz magnons can be excited and detected at the nanoscale using high-energy-resolution STEM EELS [4] with the help of hybrid-pixel electron detectors. Momentum-resolved (ω-q) vibrational EELS measurements on antiferromagnetic and ferromagnetic material systems (NiO and yttrium iron garnet, YIG, respectively) reveal the unambiguous dispersion behaviour of the magnon signal (Fig. 1) in NiO, and magnon-polaron bands in YIG. The experimental findings are shown to be in excellent agreement with theoretical momentum-resolved magnon EELS dispersion curves (Fig. 1), calculated using theoretical methodologies to electron inelastic scattering of magnons and phonon-magnon coupling in an electron microscope [5]. Finally we explore the limits of spatial resolution, by performing atomically resolve measurements and discuss of the contrast formation in atomically resolved magnon maps.

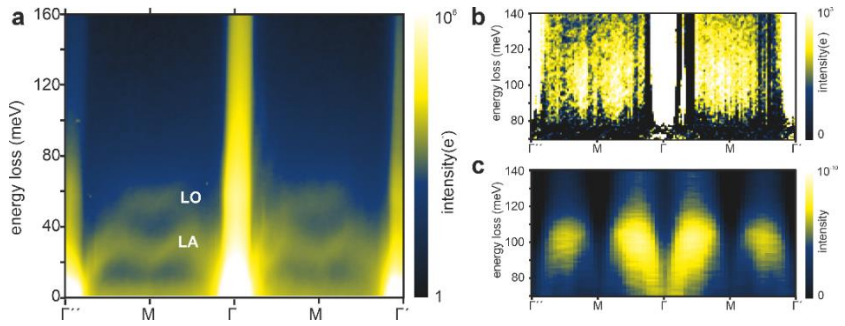

Figure 1. ***a.****ω*-**q** *EELS map along the* Γ → M **q**-path *of NiO, showing the dispersion of the NiO LA / LO phonon branches.* ***b*** *Experimental background-subtracted and* ***c*** *calculated ω-q EELS maps showing the dispersion of the magnon bands.*

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Nanoscale characterization of optical chirality

Malo Bézard[1], Simon Garrigou[2], Heyu Wang[1], Yves Auad[1], Jérémie Béal[3], Xiaoyan Li[1], Florian Castioni[1], Luiz Tizei[1], Odile Stéphan[1], Davy Gérard[1], Hugo Lourenço-Martins[2], Mathieu Kociak[1,4]

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In this talk, we will report recent results on nanometer-scale circular dichroism in canonical 3D chiral systems and discuss their interpretation. We will first present nanoscale, circularly polarized cathodoluminescence (pCL) experiments performed in a scanning transmission electron microscope on plasmonic Born–Kuhn systems (BKS, Fig. 1, a).

Using a novel detection scheme, we observe highly localized and strongly dichroic signals (Fig. 1, b), in excellent agreement with simulations (Fig. 1, c). At first glance, such strong dichroism could be attributed to the well-known large geometrical chirality of BKS. However, a systematic experimental and numerical study as a function of BKS parameters reveals that this contribution is unexpectedly weak. Instead, we show that a continuous pseudo-scalar quantity—the *plasmonic chirality characteristic*—can be rigorously defined to characterize the chirality of individual plasmonic modes [1]. Remarkably, we demonstrate that electron spectroscopy, unlike optical spectroscopy, provides direct experimental access to this quantity.

We will then extend cathodoluminescence excitation spectroscopy (CLE)—i.e., spatially resolved, time-correlated measurements of EELS and CL [2]—to the polarized CL case using experiments on BKS. If time permits, we will discuss how combining such experiments with phase shaping paves the way to probing entanglement between electrons and photons mediated by plasmons.

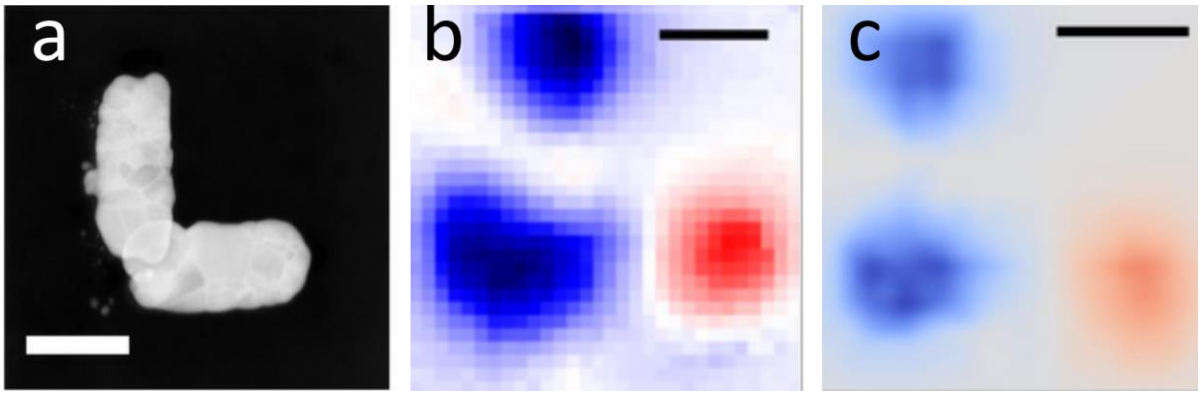

Figure 1. Circular dichroic measurement of a plasmonic BKS structure by spatially resolved cathodoluminescence in a STEM. a: High angle annular dark field image of a BKS with a schematic of the BKS structure in the inset. b: Experimental and c: simulated circular dichroic cathodoluminescence maps filtered at ~ 1.38 eV.

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Acknowledgments

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Electron wave shaping in space and time using structured light

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Stimulated interaction between photons and free electrons allow to coherently modulate the phase of electron wave function and to create photon sidebands in electron energy/momentum spectra. Such manipulation has been utilized for high-resolution imaging and spectroscopy with electron beams. It can be generalized to the case of optical fields structured both in space and time domains.

In this talk I will summarize our recent theoretical and experimental results on free electron wave function shaping using tailored light waves allowing to generate chiral electron beams, to shape the electron pulses, to monochromatize pulsed electron beams or to compensate spherical and chromatic aberrations of electron lens. I will also discuss development and demonstration of a new ultrafast 4D-STEM (scanning transmission electron microscopy) technique allowing to image the transverse component of Lorentz force acting on the electrons during their interaction with optical near-fields of nanostructures or with ponderomotive potential of optical waves in free space.

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Acknowledgments

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Probing phonons and thermal states within nanocavities with electrons

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Heat transfer processes between two surfaces spaced by a nanoscale gap (nanocavity) are assisted by surface phonon polaritons [1]. Determining the spectral distribution of the heat transfer across a nanoscale gap or the thermal state of each coupled phonon modes remain as open scientific questions. Addressing those challenges requires the use of local spectroscopy probes with nanoscale sensitivity. We will present spatially-resolved high energy resolution EELS studies [2] of coupled phonons in dielectric cavities (nanogaps) subjected to different temperatures.

We fabricate cavities with gaps raging between 5 and 150 nm which were subjected to temperatures between 300 – 1000 K. Temperatures gradients across the cavities were also generated to probe them under hear transfer conditions. We detect a large variety of coupled phonon modes within the cavity indicating the existence of several available channels for heat transfer processes (i.e. spectral distribution). We also map out coupled phonons across the cavity gap revealing strong temperature-dependent behavior (Fig. 1), which suggests a complex interplay between thermal and phonon behavior. The EELS scattering signal across the cavity gap exhibits a parabolic distribution, which is strongly dependent on both the gap distance and the temperature profile across the gap. Temperature near the cavity was measured locally with high precision [3].

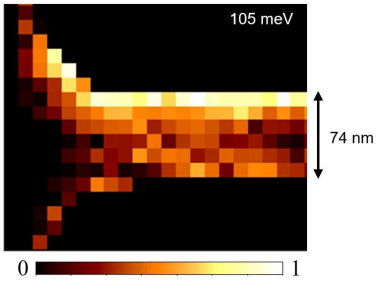

Figure 1. Spatially resolved Phonon EELS map of a nanoscale cavity subjected to a temperature gradient.

This work represents progress towards interpretation of temperature-dependent EELS data and understanding of the interplay between thermal and phonon properties of nanoscale cavities.

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Measuring Symmetry, Coherence, and Phase in STEM-EELS

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Interferometric methods can be applied in electron spectroscopy to measure the phase of excitations. In this method, a pair of nanoscale phase gratings [1] are used as diffractive amplitude beamsplitters placed before and after a specimen to create an electron interferometer in a STEM [2-4]. These beamsplitters coherently divide the electrons into separated probes at the specimen, and then recombine the two paths after the specimen, creating a set of discrete interfering outputs. We demonstrated using this setup to induce optical dipole transitions in nanoparticles, showing that the scattered electrons remain in a superposition, retaining phase coherence between the two paths [3]. In this setup, both electron paths pass near an aluminum nanoparticle, inducing an optical dipole transition. The scattered electron wavefunctions overlap and interfere at the second beamsplitter, recombining to discrete output beams that can be measured in EELS. When the interferometer is initially tuned to have constructive interference at the EELS detector when the paths pass through free space, introduction of the nanoparticle and inducement of a plasmon imprints a π phase difference between the two paths results in destructive interference and a decrease in counts at the detector. Conversely, if the interferometer output is initially tuned to result in destructive interference at the detector before introducing the nanoparticles, the π phase shift induced by the plasmon excitation results in constructive interference at the detector. When the probes are place at 90 degrees from each other relative to the particle, there are initial indications that path entanglement with orthogonal plasmon polarization may reduce coherence at the output. We propose to continue developing this setup to explore electron-photon entanglement as well as applications such as demonstrating electron energy loss sensitivity without the use of a spectrometer [5].

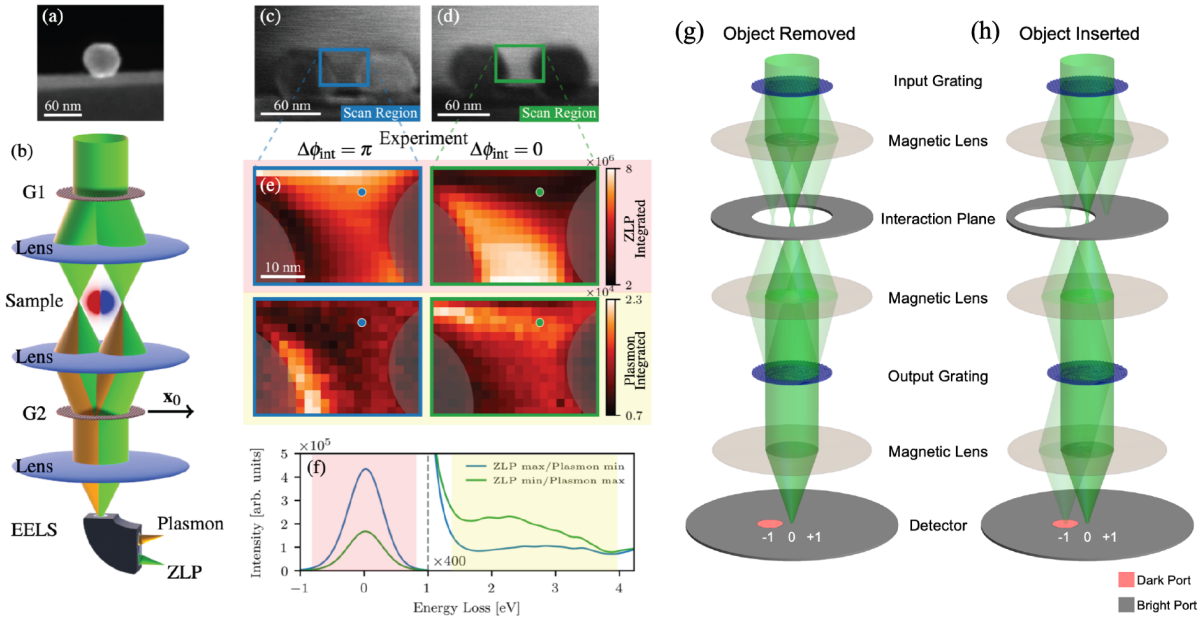

Figure 1.

(a-f) Adapted from [3]. (a) ADF-STEM image of a 60 nm gold nanoparticle (NP) isolated on the edge of a carbon substrate. (b) Sketch of the two-grating electron Mach Zehnder interferometer consisting of a STEM with two gratings used as beamsplitters. The first grating (G1) divides electrons in a superposition of two separate paths, each of them interacting with the NP sample, with some probability of losing energy to an excitation such as a plasmon resonance (orange). (c,d) The electron paths are then recombined using the second grating (G2), which can be positioned for (c) destructive (blue borders) and (d) constructive (green borders) interference, conversely modifying the elastic and inelastic signals. Two NPs are observed in the image because of the two-spot beam

configuration, with the central frame selecting the interference region (i.e., each beam passing by one side of the NP). (e,f) For both alignment schemes, we integrate over the plasmon (yellow-shaded) and ZLP (red-shaded) regions of the energy loss spectra (f) at every scan location to create the spectral images shown in (e). The raw spectra in (f) correspond to the dotted positions in (e). (g) and (h), borrowed from [4], illustrate the interaction free measurement.

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Plasmonics in Oxide-Forming Metals: Opportunities and Challenges

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Mg is the cheapest plasmonic metal by volume, is biocompatible, and can sustain strong LSPRs across the UV-Vis-NIR owing to its lack of interband transitions at those energies, making it an exciting material for nanoplasmonics. However, while Mg alloys have been studied for a century, nanoscale Mg is a recent and growing research field, akin to the state of Au NPs 50 years ago (after millennia of research on Au alloys). For Mg, the challenge of developing a nanoscience toolbox is compounded by its distinction from all other plasmonic metals: it has a different crystal structure (HCP, not FCC) and a high negative reduction potential, such that the synthesis and coating approaches of Au and Ag are not suitable.

Within such challenges, we have found multiple exciting opportunities over the past seven years. Crystallographically, Mg forms different twin planes from FCC metals, leading to fundamentally different shapes that have been a fructuous playground for tomography and Wulff modelling approaches. The bright EELS signal of metallic Mg at ~10.5 eV has also provided plenty of interesting characterization opportunities, where one can map metallic (with EELS) vs all Mg (with EDS). Finally, the plasmonic behavior of Mg has led to many new opportunities in low-loss EELS mapping, as well as optical spectroscopy investigations, including our latest work on twinned Mg cubes.

Yet there remain challenges in need of advanced spectroscopy approaches—mostly revolving around to the coupling of the plasmonic Mg with the defective oxide layer surrounding it, as well as Mg coupling with other engineered surface oxides.

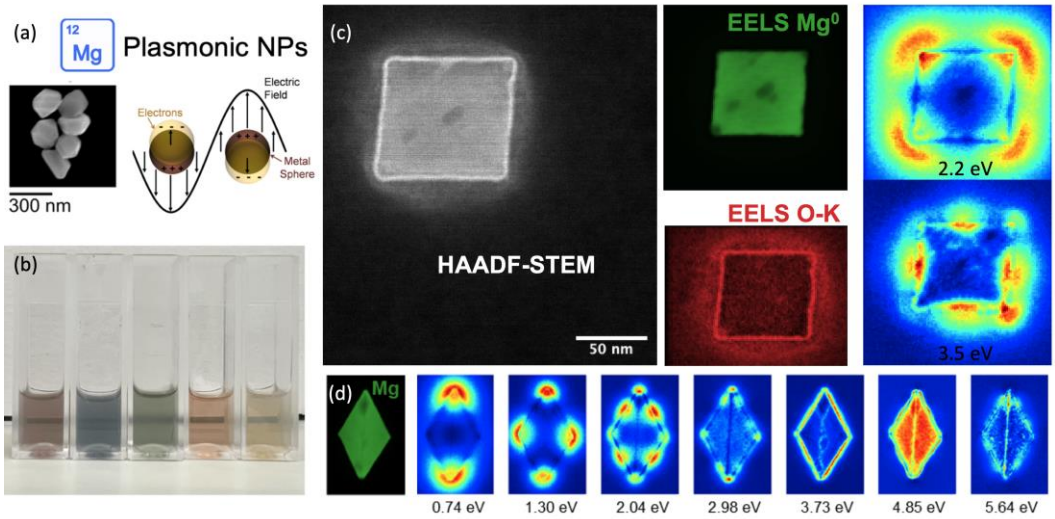

Figure 1. (a) Mg as a plasmonic metal, (b) optical properties of various sizes of Mg nanoparticles, (c) Mg nanocubes, displaying ma metallic core, thin oxide shell, and multiple LSPRs (d) Mg nanokites.

Acknowledgments

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Coherent and incoherent cathodoluminescence photon statistics

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Recent CL studies using Hanbury-Brown=Twiss interferometry (as Fig.1a) uncovered unexpectedly strong photon bunching behavior in the $g^{(2)}(\tau)$ measurement [1], meaning that CL photons excited by CW electron beams show apparent super-Poissonian statistics. This CL photon bunching can be understood through the mixture with zero photon states introduced by random electron excitation with some intervals (Fig.1c). While such bunching behavior is useful to measure the emission lifetime, it blurs the "true" photon statistics by free electron excitation.

We here extract the photon statistics within the single electron excitation event by comparing the "bunching" and the flat background parts in the $g^{(2)}(\tau)$ curves (Fig.1b) [2]. This allows calculating a parameter corresponding to $g^{(2)}(0)$ in the single electron excitation, which we call the *correlation factor* κ_{corr}. In coherent CL processes, where light is generated through a direct electromagnetic interaction (Fig.1d), κ_{corr} is found to be ~1 in various coherent CL systems. This confirms that coherent CL obeys Poisson statistics, in agreement with the coherent states. However, in incoherent CL, which involves multiple mediator particle generations (like bulk plasmons or secondary electrons, Fig.1d), κ_{corr} exceeds 1, meaning that incoherent CL is super-Poissonian ("bunching") even within the single excitation event.

Through a model analysis with multiple (cascade) particle generation steps, we were able to reproduce this super-Poissonian behavior in incoherent CL even when each particle generation process follows Poissonian statistics. This approach not only offers a refined understanding of CL mechanisms but can also be extended to the analysis of internal particle generation processes in general.

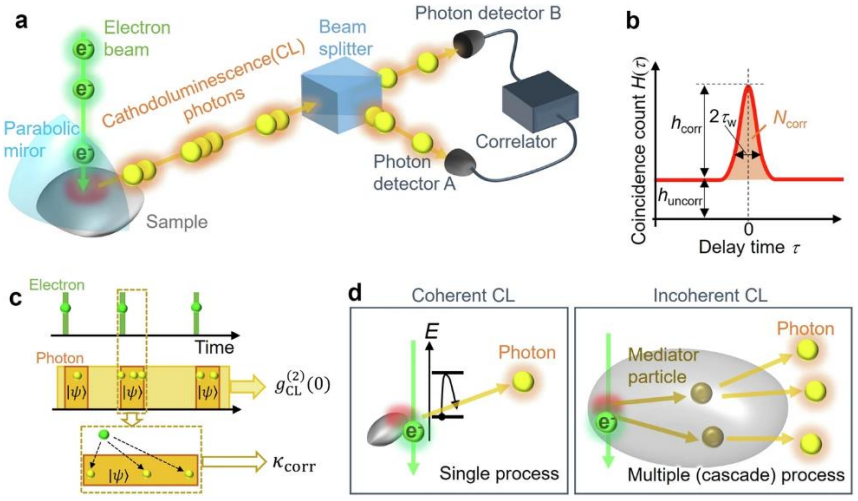

Figure 1. a Schematic illustration of the CL photon statistics measurement and CL processes.[2]

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JSPS KAKENHI (JP21K18195, JP22H01963, JP22H05032, JP24H00400), JST FOREST (JPMJFR213J)

"Novel Electron Imaging Methods Based on Light-Mediated Coherent Electron Wave Function Shaping"

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Abstract:

The interaction between light and electrons can be exploited for generating radiation, or for controlling electron beams for dynamical investigation of materials, enabling new applications in quantum technologies and microscopy. In this contribution, I will describe an innovative method for coherent and versatile longitudinal/transverse manipulation of a free-electron wave function. Using appropriately shaped light fields in space and time, I will demonstrate how to modulate the energy, linear and orbital angular momenta, as well as spatial and temporal distributions of the electron wave function. The experiments have been performed in an ultrafast-TEM, where a pulsed electron beam was made to interact with a shaped optical field generated via a spatial light modulator, and the energy-momentum exchange resulting from such interaction was directly mapped in the electron multidimensional phase space. We will demonstrate how our approach for arbitrary longitudinal/transverse electron modulation at the sub-fs timescale is fundamental for the first time implementation of new imaging techniques, such as Ramsey-like holography and Single Pixel Imaging, as well as for enhanced TEM performance, such as spherical aberration correction. Our results would pave the way for achieving unprecedented insights into non-equilibrium phenomena in advanced quantum materials, playing a decisive role in the rational design and engineering of future photonics and electronics application.

Synthetic gain for electron-beam spectroscopy

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There exist significant challenges in resolving spectral features in electron-beam spectroscopy. These challenges include low signal-to-noise ratios, spectral overlap, and low probability of detecting spontaneous multi-photon events. Recent developments in the synthetic complex-frequency wave (CFW) approach have demonstrated notable improvements in optical microscopy and spectroscopy. Building on this foundation, the present work [1] extends the synthetic CFW methodology to the realm of electron-beam spectroscopy and microscopy, aiming to address analogous challenges in electron-based measurements.

To enhance spectral features in electron microscopy and spectroscopy (Fig. 1a), a mathematical approach involves replacing the real frequency ω with a complex frequency $\tilde{\omega} = \omega - i\frac{\tau}{2}$ to compensate for intrinsic losses. Exploiting causality, one can reconstruct a complex-valued loss probability $\tilde{\Gamma}(\omega)$ using the Kramers-Kronig relations, where the real part is given by the principal value integral $\mathrm{Re}\{\tilde{\Gamma}(\omega)\} = \frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\mathrm{Im}\{\tilde{\Gamma}(\omega')\}}{\omega'-\omega}d\omega'$, and the imaginary part corresponds to the experimentally measured real-frequency loss probability . By applying a Fourier transform, this approach enables the synthesis of the complex-valued loss probability $\tilde{\Gamma}(\tilde{\omega})$ at complex frequencies from a linear combination of experimentally obtained real-frequency probabilities $\tilde{\Gamma}(\omega_n)$, which can be expressed as $\tilde{\Gamma}(\tilde{\omega}) \approx \sum_n \tilde{\Gamma}(\omega_n) e^{-i\omega_n t + i\tilde{\omega}t}\frac{\Delta\omega}{2\pi i(\tilde{\omega}-\omega_n)}$. This synthesis effectively incorporates loss compensation and enhances spectral resolution in electron-beam measurements.

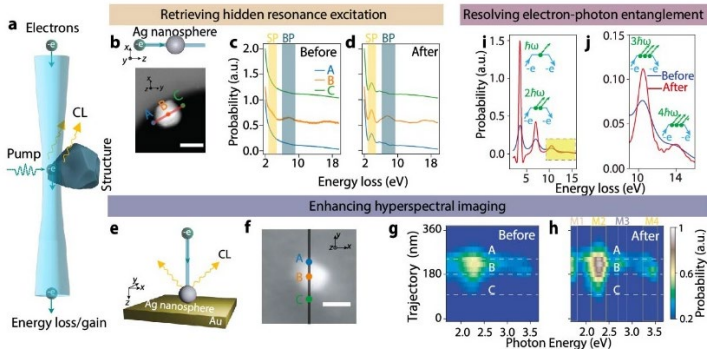

Figure 1. **Synthetic gain for free-electron–light interaction. a.** General schematic. **b–d.** Retrieval of localized surface plasmon resonances obscured by the zero-loss peak: **e–h.** Enhanced CL spectroscopy of a film-coupled nanosphere **i,j.** Resolution of spontaneous multiphoton events in free-electron–photon quantum interactions.

To demonstrate the experimental capabilities of the CFW technique, we present measurements on several representative systems. We first investigate a suspended silver nanoparticle (~20 nm diameter) positioned within a TEM grid hole, where a 200-keV electron beam is scanned across the particle and electron energy-loss spectroscopy (EELS) spectra are acquired at three representative positions (A, B, and C). After CFW processing, the dipolar Mie resonance becomes pronounced at positions A and C, while the bulk plasmon remains absent due to the aloof interaction geometry; notably, CFW enhances both surface and bulk plasmon features at position B, where they are otherwise weak. We further apply the CFW method to cathodoluminescence in a film-coupled nanoantenna system, consisting of silver nanoparticles (~100 nm diameter) placed on a 100-nm-thick gold film and excited by a 10-keV electron beam. Hyperspectral CL imaging along a scan line across a nanoparticle reveals, in the unprocessed data, a dominant mode at 2.3 eV and a faint mode near 3.4 eV; however, following CFW processing, four distinct optical modes (labeled M1–M4 in ascending energy) emerge, with simulations attributing M1 and M2 to gap-plasmon modes at lower energies and M3 and M4 to Mie resonances at higher energies. Finally, in the quantum regime, we simulate an EELS spectrum using a Poisson statistics model at strong quantum coupling ($g_q = 1$), enabling spontaneous multiphoton processes; after CFW processing, not only does the four-photon peak become clearly identifiable, but the lower-order photon peaks are also significantly enhanced. These results collectively demonstrate that the CFW technique substantially improves spectral feature recovery and resolution in both classical and quantum regimes of free-electron–matter interactions.

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Control free-electron—light interaction for nanophotonic particle accelerators and quantum applications

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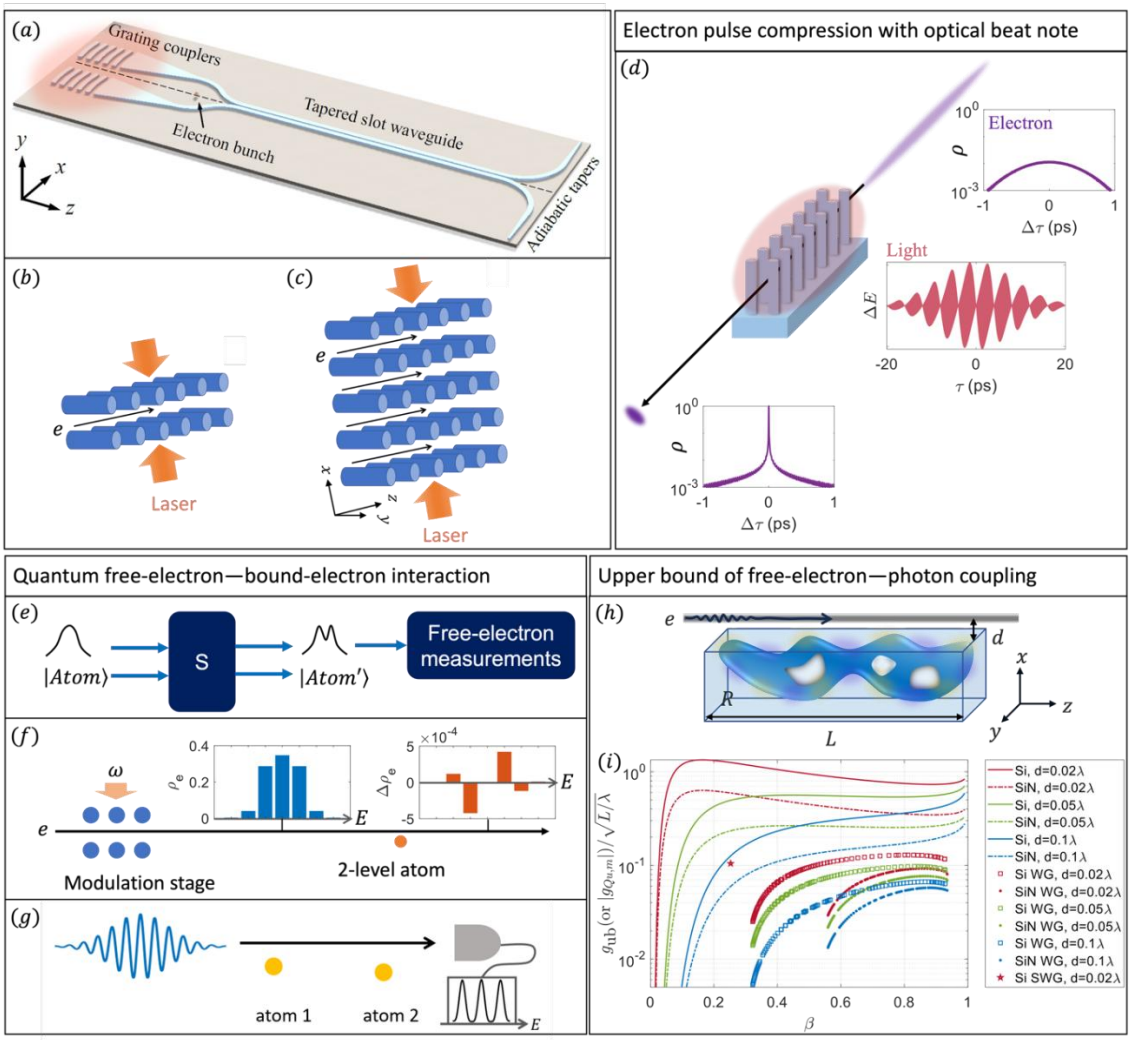
Electron accelerators are important in studying fundamental science, research, medical diagnosis and treatment, and industrial processing. In dielectric laser accelerators (DLAs), laser pulses interact with electron beams in the vicinity of dielectric nanostructures and can accelerate the electrons [1,2]. DLAs have the potential to provide acceleration gradients at least one order of magnitude higher than conventional radio-frequency accelerators, due to the high damage threshold of dielectric materials, with a compact size.

Key challenges of DLAs include the integration of the optical power delivery waveguide with the DLAs, extending the acceleration distance, and increasing the current throughput. In this presentation, we show examples of how photonic design and control address these challenges. In a compact DLA system, the electrons generated by on-chip sources are sub-relativistic. A major challenge of long-distance sub-relativistic acceleration is dephasing, when the velocities of the accelerated particles increase and become mismatched with the phase velocity of the accelerating field. To solve the dephasing, we designed a tapered slot waveguide DLA (Fig. (a)) where the phase velocity of the slot waveguide matches the electron velocity continuously [3]. The slot waveguide supports a guided mode that has a longitudinal electric field in the vacuum slot and co-propagates with the electrons. The waveguide width is changed gradually along the particle trajectory, such that the phase velocity is matched with the velocity of the accelerated electrons. Moreover, light can be coupled into the slot waveguide through grating couplers (Fig. (a)). Another challenge is the low current throughput due to the sub-wavelength narrow channel (Fig. (b)). To increase the DLA current throughput for science and medical applications, we introduced a photonic crystal DLA that has multiple electron channels (Fig. (c)) [4]. Through engineering the band structure of the underlying photonic crystal, we can make the acceleration field in different channels to be almost identical. The photonic crystal DLAs can increase the current throughput by orders of magnitude.

The development of DLAs opens new opportunities in both classical and quantum applications. Modern electron microscopy requires high temporal resolution to probe ultrafast physics. DLAs can generate a train of micro-bunches of sub-femtosecond duration [5, 6]. Here, we propose the compression of picosecond electron pulses using DLAs, where the energy modulation is generated by the optical beat note (Fig. (d)) [7]. Its performance is comparable to terahertz electron compression, while being more efficient and compact. In the quantum region, DLAs can modulate the wavefunction of free electrons, as in photon-induced nearfield electron microscopy [8, 9]. We studied how resonant modulation of the free electron can enhance the interaction between the free electron and a two-level atom and probe the atomic coherence (Figs. (e)-(f)) [10]. We also found that distant identical atoms can be entangled by interacting with the same free electron (Fig. (g)). Furthermore, a large coupling between free electrons and photons is generally desired for many free-electron quantum optics applications. We derived an upper bound for the coupling coefficient describing the free-electron—photo interaction (Figs. (h)-(i)) [11]. The upper bound depends on the interaction length, the optical medium, the free-electron velocity, and the separation between the free electron and the optical medium. It can provide guidance to reach the strong coupling between free electrons and photons.

Looking forward, to make free electron beams and free-electron radiation sources widely accessible, it is important to further integrate the optical system and the free-electron components in a compact platform. To explore the ultimate quantum capabilities of free electron probes, it is crucial to develop systems that can enhance the quantum signals of free-electron—matter interactions.



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Acknowledgments

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CONTRIBUTED TALKS

Coherences in Axially and Laterally Spaced Plasmons Excited by Free Electrons

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Coherent cathodoluminescence (CL) spectroscopy is an established, versatile tool for studying nanophotonic structures at the nanoscale. While the coupling between an electron and a single scatterer is well understood, the interaction of one electron with multiple scatterers has not yet been fully studied. Here, we explore the coherent excitation of multiple scatterers with a single electron, addressing key questions: How can multiple scatterers be coherently excited by the same electron? What is the effect of lateral and temporal coherence of the electrons on emitted CL?

To study this, we introduce CL interferometry – an approach that reveals coherent interactions between free electrons and plasmonic scatterers through interference between different components in the CL emission. Using a configuration consisting of a gold nanotip and a planar gold surface, we demonstrate that a single electron can coherently excite both scatterers in sequence (see Figure 1a). Both interactions create coherent CL components that interfere in the far field. From the rich interference pattern, we retrieve the height of the pillar, $\Delta z = 5.36$ µm, and observe that the two scatterers are coherently excited even though they are vertically spaced by micron distances.

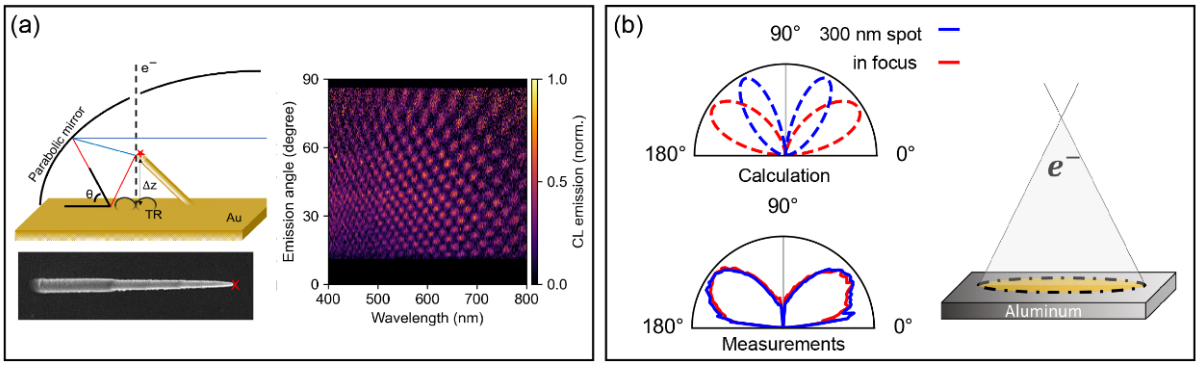

Figure 1. (a) Representation of the sequential excitation of a gold nanopillar above a gold planar surface and their resulting CL interferogram in the far field. (b) First experiments to study the effect of the lateral coherence of the electron on the angular pattern of the emitted TR from an aluminum planar surface.

Next, we investigate the role of lateral coherence of the electron beam on the emitted CL. First, we demonstrate a method to measure the degree of coherence of the electrons using electron diffraction in the SEM. Then, the question arises, considering the spatial coherence of the electron beam, is it possible to coherently excite multiple scatterers that are spaced horizontally? In a first experiment, we vary the beam spot size, from 1 nm (focused) to 300 nm (defocused), and probe transition radiation (TR) from an aluminum planar surface, as illustrated in Figure 1b. Our measurements reveal no significant change in angular emission pattern, suggesting that we do not coherently excite TR within the spot size.

In summary, we introduce and experimentally validate CL interferometry as a method to probe the coherent excitation of multiple scatterers, revealing the role of lateral coherence in electron-induced light emission.

Acknowledgments

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Femtosecond pump-probe experiments in an RF-UTEM at 75 MHz

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We report the first pump–probe experiment performed at 75 MHz repetition rate in the TU/e RF cavity-based ultrafast transmission electron microscope (RF-UTEM) [1].
The system employs a miniaturized 3 GHz TM_{110} dual-mode cavity to chop the continuous beam from the Schottky field emission gun of a conventional TEM into ultrashort pulses, while preserving its low emittance and narrow energy spread [2]. A fully integrated femtosecond laser system enables synchronized pump–probe operation with ~100 fs timing precision and reduced jitter, while supporting stroboscopic measurements at significantly higher average currents than typical UTEMs.

To demonstrate the system's capabilities, we conducted a photon-induced near-field electron microscopy (PINEM) experiment, probing the coherent interaction between ultrashort electron pulses and the evanescent optical field generated at the sharp edge of a platinum aperture. The resulting modulated electron energy loss spectra (EELS) exhibit clear PINEM sidebands and spectral broadening of several eVs. Measurements were performed with laser pulse durations from 100 fs to ~1 ps and pulse energies between 1.34 and 12 nJ. The observed enhancement in light–electron interaction strength with increasing pulse duration and power is consistent with theoretical expectations, and the measured spectra closely match fitted and simulated results. The delay-dependent evolution of the PINEM spectra provides a robust characterization of the interaction envelope, serving as a sensitive diagnostic of the temporal resolution of the RF-UTEM. We demonstrate an achievable effective temporal resolution in pump–probe experiments of at least 334 fs, provided by the convolution of about 260 fs electron pulses, 100 fs laser pulses and contributions from system-level temporal jitter. Additionally, integrated PINEM spectra clearly expose any RF phase jumps or jitter, establishing the technique as a valuable tool for real-time monitoring of synchronization stability. The high reproducibility across delay scans confirms laser–electron synchronization and RF phase stability within a few hundred femtoseconds. These results demonstrates the feasibility of stroboscopic pump–probe operation at 75 MHz in an RF cavity-based UTEM, where high-quality spectra can be acquired within tens of milliseconds under the experimental conditions employed in this study, i.e., nanoamper continuous beam current and hundreds of femtoseconds electron pulses.

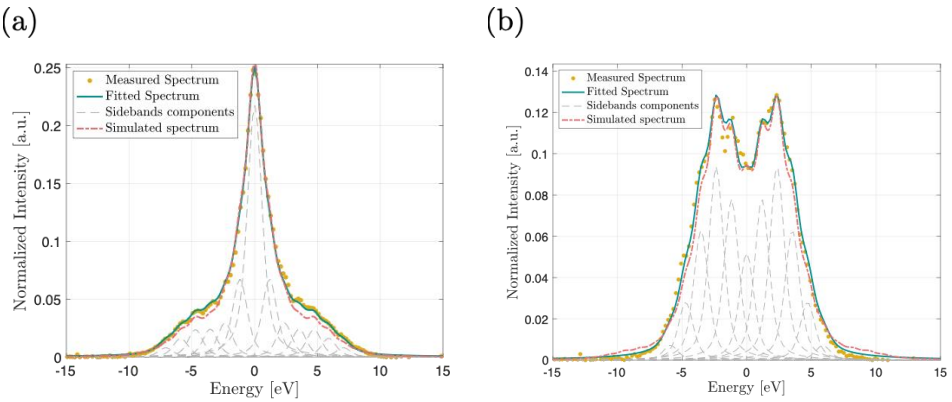

Figure 1. **Electron energy modulation via interaction with the optical near-field at a sharp aperture edge**. EEL-spectra measured at (a) ~100 fs and (b) ~1 ps laser pulse duration and 6 nJ pulse energy. For each laser pulse duration, the figure shows the measured spectrum (yellow dotted line), the theoretically simulated spectrum (dashed red line), the corresponding fitted spectrum (solid teal line), and the individual sideband components (dashed gray lines) located at discrete energies $\pm n\hbar\omega$.

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Time-resolved nanothermometry based on photon-electron pump-probe spectroscopy

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Understanding thermal transport in nanostructures is crucial for technological advancements, particularly as devices shrink to the nanometer scale. Studying out-of-equilibrium behavior requires access to timescales ranging from picoseconds to microseconds. However, existing methods with both high temporal and spatial resolutions remain limited [1]. Recent advances in event-based direct detectors for electron microscopy [2] offer new possibilities.

In this contribution, we present a novel technique for temperature measurement with nanometer and nanosecond resolution in a scanning transmission electron microscope (STEM) (Figure 1). In this pump-probe experiment, a focused (~1 μm) and pulsed (~25 ns) laser excitation is synchronized with electron arrival times, detected using a Timepix3 detector. By analyzing distinct electron energy-loss spectroscopy (EELS) signatures, we determine the temperature of 2D semiconductors, SiNx membranes, and aluminum thin films. Our approach is widely applicable due to the broad spectroscopic excitations spanning from infrared (hundreds of meV) to soft X-rays (tens of eV), providing a universal tool for probing nanosecond thermal dynamics in nanostructures [3]. The observed temperature evolution aligns well with a simple 2D thermal diffusion model. While the current temporal resolution is limited by the Timepix3 detector, further improvements are anticipated.

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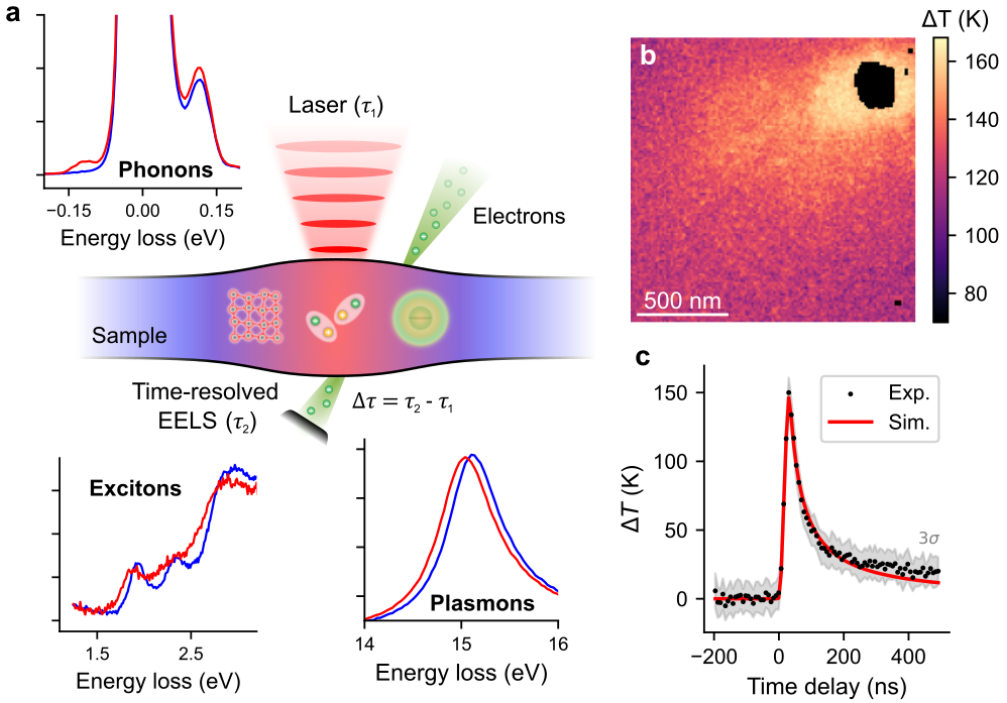

Figure 1. Description of the photon-electron pump-probe experiment for nanosecond-resolved nanothermometry. (a) A 25 ns width pulsed laser in the visible range heats the sample, inducing absorption changes probed by EELS in the IR (phonons) to the soft X-ray (bulk plasmon) energy range in different material systems. (b) Temperature difference spatial mapping of an aluminum thin film following laser exposure. (c) Temperature temporal profile of the aluminum film depending on the time delay Δt following the laser excitation, compared with a heat diffusion simulation in an infinite 2D film.

Probing Optical Nearfields with an SEM

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We present a nearfield imaging approach implemented in a scanning electron microscope (SEM) based on PINEM interaction, enabling nanoscale mapping of optical fields [1-4]. By illuminating nanostructures with ultrafast laser pulses synchronized to the pulsed electron beam, photon-induced energy modulation is imprinted onto the electron spectrum, providing a direct probe of strength of local optical nearfields. The resulting energy-gain patterns are used to reconstruct spatial nearfield distributions with high sensitivity and spatial precision. This approach enables visualization of mode confinement, coupling, and interference effects in nanophotonic systems directly within the SEM.

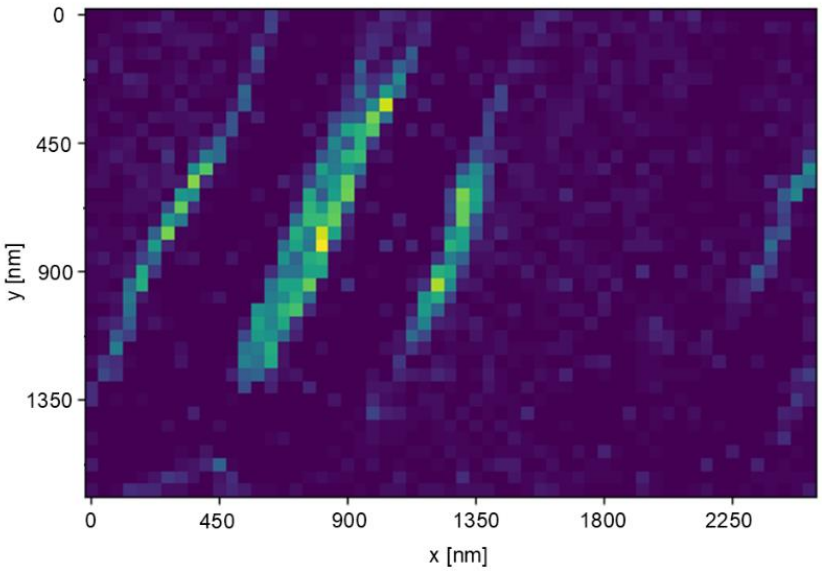

Figure 1. Image of third harmonic acceleration mode of dielectric laser accelerator [5]

In addition we investigate different regimes of pulsed electron beam and how its properties affect imaging and energy resolution performance, we explore multiple regimes of laser-triggered photoemission by tuning extractor voltage, laser wavelength, and pulse energy. These regimes yield distinct trade-offs in terms of beam brightness, pulse duration and energy spread.

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Chirality Control of Free Electrons via Photonic Interaction

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Chirality is a property with broad significance across particle physics, materials science, optics, chemistry, and biology. Chirality is an inherent signature for most of elementary particles with spin, but chiral phenomena can also appear in geometry through a proper composition of multiple non-chiral particles, such as human hands, chiral crystal lattices, rotational phonons, skyrmions, and bacteria. Chiral objects show unique properties and applications compared to their achiral counterparts. So far, it has shown that electrons can be imprinted with chirality by modifying its phase with spiral phase plates, magnetic needle tips, holographic gratings or photon-induced near-field interactions. However, all of them only show a static control over electrons' phase. In our experiment, we create and characterize two intriguing electrons, namely, electron coils [1] and self-torqued electrons [2].

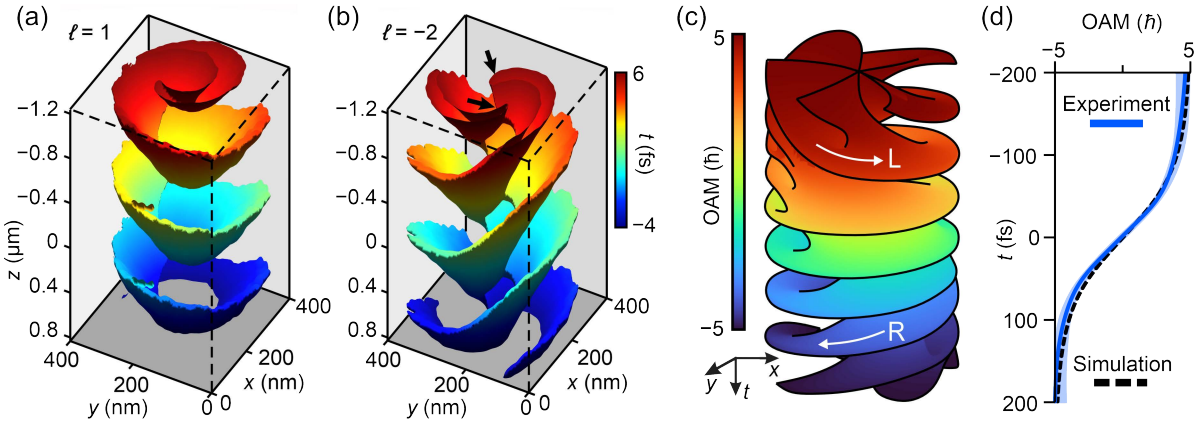

Figure 1. (a) Measured expectation density of an electron coil with a topological charge $\ell = 1$. (b), Measured expectation density of an electron coil with a topological charge $\ell = -2$. (c) Schematic of the phase of self-torqued electrons. (d) Measured and simulated electron orbital angular momentum as a function of arrival time.

We use an ultrafast transmission electron microscope with a Schottky field-emitter source in our experiment. We create femtosecond electron pulses by photoionizing the field-emitter tip and modulate the electron pulses with a beam of femtosecond optical vortex. In their interaction region, we place a silicon nitride membrane as a modulation element. Our experiment reveals that if the photon-modulated electron propagates for a half Talbot distance, the electron is shaped into an attosecond electron coil wave (Figs. 1 (a) and (b)). Such electrons carry no orbital angular momentum (OAM) but they acquire chirality directly from their spatial expectation of charge and mass. The chirality of electron coils is directly controlled with the topological charge ℓ of the optical vortex. More interesting, in a scenario with a high photon-modulation strength, lower electron energy and long propagation distance, the modulated electron manifests as a time-dependent chirality in its phase (Fig. 1(c)). Its chirality evolves in femtoseconds from left handed at the beginning into achiral in the middle towards right-handed in the end. Such ultrafast time-varying orbital angular momentum makes the electron self-torqued (Fig. 1(d)).

The size of the our electron beams is only limited by the de Broglie wavelength and can therefore reach picometer dimensions for applications in atomic spectroscopy, for example, to induce or probe the ring currents in atomic orbitals or magnetic materials.

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Spontaneous and stimulated electron-light scattering: identifying their respective contribution

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Due to recent experimental developments in integrated photonics, the question of the *Photon Induced Near-field Electron Microscopy* (PINEM) in the low occupation regime – i.e. when the cavity is populated by a weak number of photons - has drawn an increasing theoretical and experimental interest [1-3]. Indeed, in this situation, the classical model of the electromagnetic field falls short to describe the electron-cavity interaction, and a full quantum description becomes required. Several works have already pioneered this problem and predicted strong quantum mechanical effects in PINEM [1-3].

The goal of our work is to pursue this effort in order to understand which properties of the electromagnetic field are imprinted on the electron wavefunction during the interaction. In the classical regime, the intensity and the phase of the field are encoded in the electron wavefunction [4,5], which can be used to reconstruct the near-field of an optical cavity, and its dynamic [6,7]. However, in quantum optics, other parameters are needed to describe light, such as number-phase uncertainty, field fluctuations, and quantum coherence [1,8]. In this presentation, we will show how these additional parameters influence the final electron wavefunction.

Our approach is based on the use of the Wigner function – roughly speaking, a probability distribution of the quantum state in the phase space (momentum-space) – which is a powerful visualization tool which was introduced for PINEM in [4]. This function gives visually access to information on the final electron state and its interaction with light, such as the energy exchange, density modulation and quantum coherence. For instance, in figure 1.a, one can observe the electron density modulated by a coherent state. Then in 1.b, one can observe the final electron state, after interaction with a number state: no modulation can be observed, because of the maximum phase uncertainty of the number state. Finally, in figure 1.c, one can observe the electron density modulated by the field fluctuation of a squeezed state.

In order to further explain these observations, we will make use of different quantum mechanical pictures, which are unitary transformation the photonic phase space. More precisely, we used the vacuum picture [9] – i.e. a translation of the phase space – in order to split quantum and classical contribution of the field in the interaction, and the squeezed picture – i.e. a hyperbolic rotation of the phase space – to identify the modulation of the electron due to field fluctuation. In other words, these pictures allow us to go from a complex interaction in a simple space, to a simpler interaction in a more complex space.

In the spirit of [10], by combining these technics, we will identify different types of scattering processes, e.g., spontaneous and stimulated decays, quantum and classical electron density modulations. More generally, for a wide range of initial photonic states, we will quantify the contribution of each of these processes to the total scattering, and connect them to different properties of the electromagnetic field (number of photons, mean field, and fluctuation). For instance, using the vacuum picture, we will show that the interaction with a coherent state is a combination of classical density modulation (due to the mean field) and spontaneous decay (due to the fluctuation of the state, identical to the vacuum). In other words, the PINEM in the quantum regime can be seen as combination of classical PINEM and EELS.

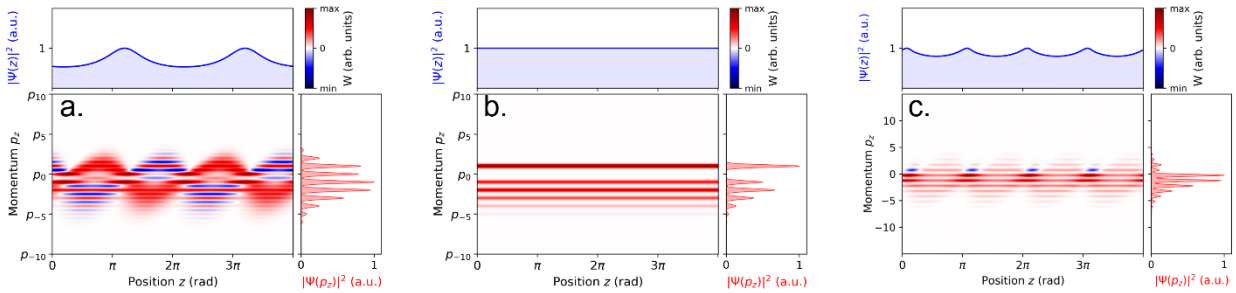

Figure 1. Electronic Wigner function after interaction with (a) a coherent state, (b) a number state and (c) a squeezed state. The Wigner function (central panel) provides a direct visualization of the energy distribution (right panel) and the temporal modulation (top panel) of the electron.

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Probing Strongly Confined Mid-infrared to Terahertz Phonon Polaritons in Few-Unit-Cell Freestanding $SrTiO_3$ Membranes by STEM-EELS

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Surface phonon polaritons (SPhPs) enable deep subwavelength light confinement, offering distinct advantages for applications in infrared sensing, imaging, and optoelectronic devices. However, their application in the terahertz (THz) regime has been limited by the narrow Reststrahlen bands of conventional polar materials. Strontium titanate ($SrTiO_3$), a quantum paraelectric perovskite with large LO-TO splitting, has recently emerged as a promising THz polaritonic platform. Here, we use monochromated scanning transmission electron microscopy-electron energy loss spectroscopy (STEM-EELS) to probe SPhPs in freestanding $SrTiO_3$ membranes down to a few unit cells thick. STEM-EELS offers broadband spectral access, large momentum transfer, and high spatial resolution—key advantages for detecting polaritons in ultrathin films where the dispersion flattens. We directly observed SPhP modes in 30 nm, 8 nm, and 3 nm $SrTiO_3$ membranes, covering both THz and MIR regimes. In 3 nm membranes (~8 unit cells), we extracted confinement factors >500 and group velocities down to ~10^{-5} *c*, rivaling leading 2D polaritonic systems. These findings demonstrate the viability of oxide-based nanophotonics in the THz regime and highlight the power of STEM-EELS for exploring vibrational polaritons in ultrathin materials. [1]

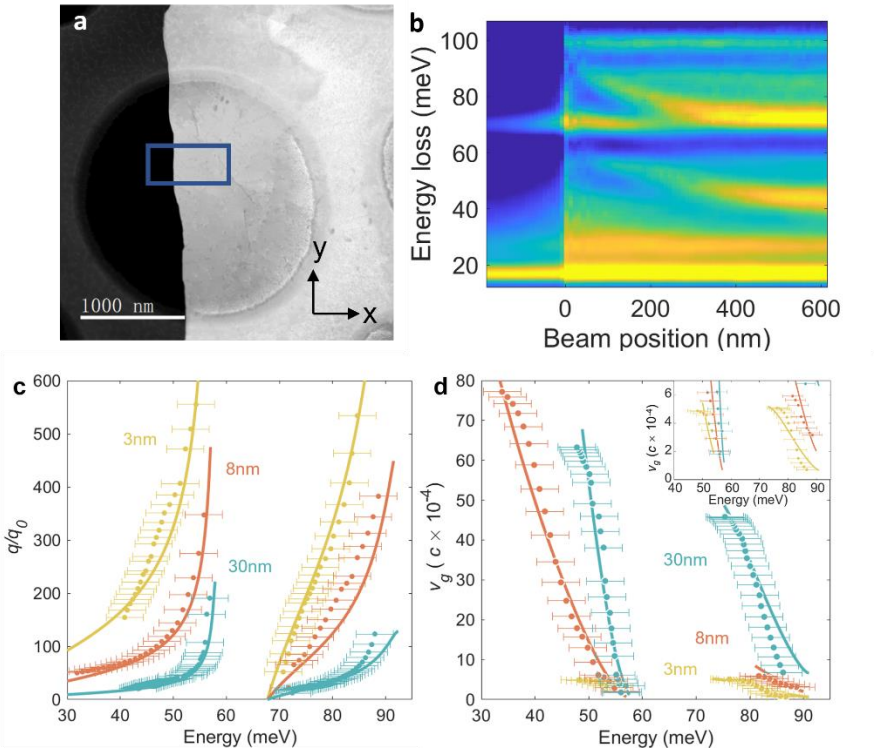

Figure 1. (a) HAADF image of the 30 nm thick freestanding $SrTiO_3$ membranes. (b) EELS line-scan taken in the blue box in (a) by summing data along y-direction. Confinement factor (c) and group velocity (d) of SPhPs for different thicknesses.

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Electron-double pulses reveal longitudinal optical field modulation

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Characterizing the optical properties of nanoscale structures is crucial for advancing nanophotonics with applications in catalysis, quantum and bio-sensing, and light harvesting. Electron beam spectroscopy methods [1], such as inelastic electron-light scattering, allow for the mapping of optical modes and the retrieval of characteristic quantities [2-4]. However, these techniques typically lack access to the field distribution along the electron beam trajectory, except for advanced works that rely on a second electron modulation stage to provide a reference phase [5].

In this work, we propose using electron double pulses of a variable time delay to probe the optical field distribution along the electron trajectory. We demonstrate this scheme by characterizing an optical beat note formed by the superposition of two continuous-wave (CW) lasers inside a Si3N4 microring resonator in a transmission electron microscope (cf. Fig. 1a) [6]. The two electron pulses sample the optical field at different times with a variable temporal delay of τ_i as sketched in Figure 1b. For each detected 2-electron event, with the two electrons randomly distributed over the two pulses [7], we determine the energy differences between the electrons as a function of the pulse delay. By Fourier transforming the measured data (cf. Fig. 1c), we recover the beating frequency of ~600GHz which modulates the field along the electron trajectory.

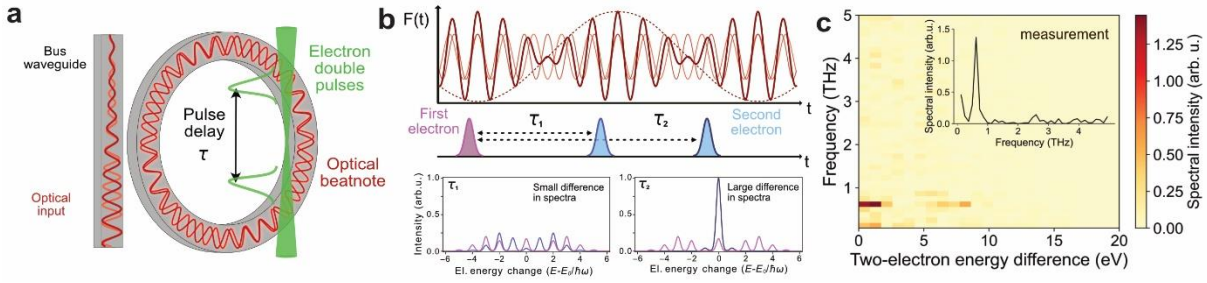

Figure 1. (a) Experimental setup: The ring cavity is pumped by two CW lasers detuned by 600 GHz, resulting in the formation of an optical beat note. The double electron pulses pass the ring resonator in an aloof geometry. (b) Schematic of the variable time delay of the electron double pulses and the different electron energy distributions after interaction. (c) The Fourier transform of the measured data after principal component analysis. The inset shows a line cut at 0 eV two-electron energy difference.

In conclusion, by utilizing electron-double pulses, we introduce a novel technique that enables obtaining information about the longitudinal optical field distribution without the need for additional modulation or synchronization of the electron pulses. This method can be extended to investigate field distributions of functional nanophotonic devices, enhancing the capabilities of electron microscopy.

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Toward Photon Induced Near-Field Electron Tomography

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New techniques for imaging electromagnetic near-fields in nanostructures drive advancements across nanotechnology, optoelectronics, materials science, and biochemistry[1]. Most existing techniques probe near-fields along surfaces, lacking the ability to extract near-fields confined within the structure. Notable exceptions use free electrons to traverse through nanostructures, integrating the field along their trajectories, extracting 2D near-field projections rather than the complete field[2]. However, electron tomography can extract the photonic density of states from the volume of the sample using electron spectrometry[3-4]. Here we introduce photon-induced near-field electron tomography (PINET), a computed-tomography–inspired framework and experimental setup for reconstructing full 3D vectorial time-harmonic near-fields.

Our setup combines two electron–field interaction points in a pump–probe scheme with a custom-made tomography holder that preserves the illumination geometry during a full 360° rotation. A single-mode, polarization-maintaining fiber guiding the beam throw the holder, maintains constant coupling conditions. To overcome the severe phase instability inherent to multi-platform optical paths, we implement a self-referenced phase-correction method: interference between the upper interaction point and the field scattered from the supporting membrane produces a fringe pattern (Fig. 1e1), enabling algorithmic frame-by-frame phase correction. This restores phase stability and allows reconstruction of complex-valued field projections[5-7] at arbitrary orientations (Fig. 1e).

Full tomographic inversion is achievable with a Radon-like algorithm that incorporates the electron wave nature and the time dependence of its interaction with vector fields (Figs. 1a-b). To show the prospects of electron near-field tomography, we propose and analyze its ability to resolve the sub-wavelength zigzag profile of highly confined hyperbolic polaritons[8] and to reconstruct 3D phase singularities in a chiral near-field (Figs. 1a-b) – raising exciting goals for next-generation experiments in ultrafast transmission electron microscopes.

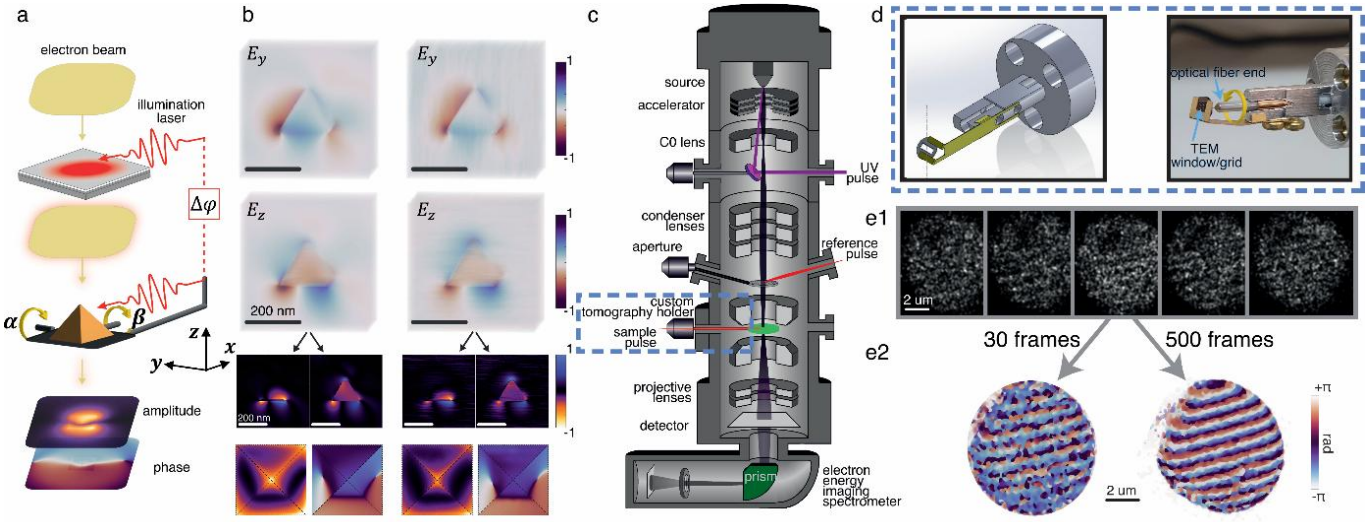

Figure 1. Photon Induced Near-Field Electron Tomography (PINET): simulation and experiment. (a) PINET scheme demonstrated on a gold nano-pyramid, denoting rotation angles α, β. For each orientation, a scan over multiple phase delays Δφ provides the complex-valued field projection. The illumination laser is rotated together with the sample. (b) Comparing the reconstructed field to the simulated confined field, showing the real parts of the y,z components in a 3D view (top), 2D slices view (center), and field perpendicular to the pyramid's faces (bottom). (c) Experimental setup. Ultrafast transmission electron microscope operated at 200 keV with 800 nm sample illumination. (d) Custom-engineered tomography holder supports 360° rotation and a polarization-maintaining fiber guiding the beam passing through it, maintains constant coupling conditions. (e) Algorithmic solution for system instability mitigation. (e1) Raw measurements taken from a single tilt ($\alpha = -12^o, \beta = -45^o$). (e2) Complex-valued projection reconstruction[5-7] after algorithmic analysis providing better signal-to-noise ratio for longer scan.

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Spherical and chromatic aberration of electron microscope compensated by a laser

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Aberration correction is crucial for achieving atomic resolution in electron microscopy. While conventional correctors use multipoles, our work explores a fundamentally different approach tailored specifically for ultrafast electron microscopes operating in pulsed mode. We propose using spatially structured intense laser pulses to manipulate electron wavefronts via free-space electron–photon interactions [1]. Importantly, this concept is not intended as an alternative or comparison to existing static aberration correctors, but rather as a novel tool compatible with the dynamic nature of pulsed electron beams. Simulations show that such laser fields can effectively compensate for spherical and chromatic aberrations of subsequent magnetic lenses, leading to substantial improvements in probe size. These findings open a pathway toward new optical elements designed for ultrafast electron microscopy.

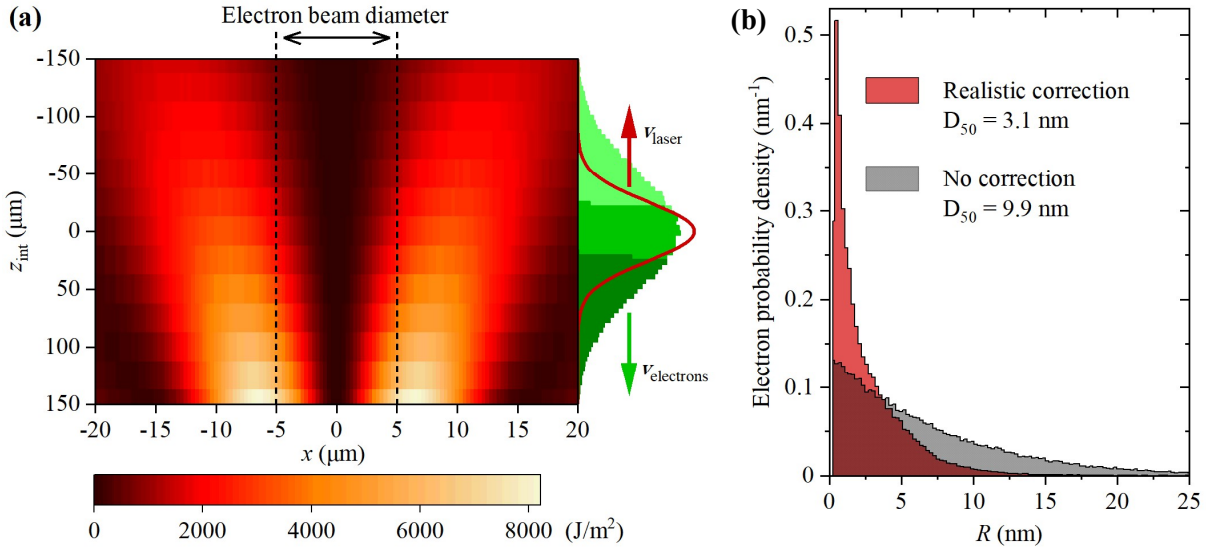

Figure 1. Chromatic aberration correction in an electron microscope operating at low acceleration voltage (500 V). (a) Cross-sections of laser planar energy density in the interaction volume. Laser pulse energy is 3.9 μJ. The histogram shows the energy distribution within the electron pulse in the interaction region: dark/bright green corresponds to the most/least energetic electrons. The red curve shows laser pulse intensity distribution along optical axis. (b) Histogram of electron distribution at z = 0 in the aberrated (gray) and corrected (red) setup.

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Towards Ghost Imaging with Electron-Photon-Pairs

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Since the introduction of time-resolved direct electron detectors to transmission electron microscopy (TEM), there has been a growing interest in studying the correlations between an electron and the cathodoluminescence (CL) photons it produces [1,2,3]. In this contribution, we demonstrate "ghost imaging" [4], a method adapted from photonic quantum optics that leverages position or momentum correlations within electron-photon pairs for imaging.

In a TEM, working at 200 keV, we generate electron-photon pairs via the process of transition radiation using a thin silicon membrane (see Fig. 1). We collect the photons using a custom-made parabolic mirror and let the photons interact with an arbitrary absorptive mask placed in the image plane of the collection system (see grating mask in Fig. 1b), thus imprinting the shape of this mask in the measured photon distribution. Using a single photon counting module and time tagger, we can match each detected photon to its emitting electron and retrieve the mask shape in the distribution of the corresponding electrons (Fig. 1e). This is remarkable, as the electron did not interact with the object; the information about the mask shape is mediated purely through the correlations between the two particles.

In a recent publication [5], we employed ghost imaging in order to demonstrate electron-photon entanglement. We will present ghost imaging with considerably better spatial resolution and explain how this might allow us to show the Einstein-Podolsky-Rosen paradox using electron-photon pairs.

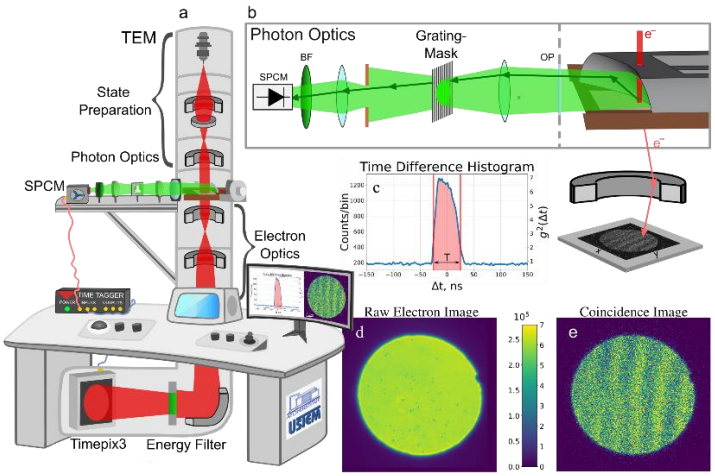

Figure 1. (a) Overview of the experimental setup, (b) photon collection system with transmission mask as imaging object, (c) temporal cross-correlation histogram showing the coincidence interval, (d) raw electron distribution, (e) coincidence-filtered electron distribution showing the imaging object.

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Acknowledgments

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Nanolaser Optical and Structural Properties at the Nanometre Scale

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Since their first demonstration by Huang et al in 2001 [1], semiconductor nanolasers have attracted a lot of interest especially for their application in optoelectronic devices. They have the advantages to be cost-effective, easy to fabricate and of a micron size. Since 2001, various semiconductors and geometries demonstrated lasing properties, for example ZnO [1] or GaN [2] nanowires.

Lasing can be induced by optical pumping, usually it is characterized by a drastic reduction of the emission spectrum width and an increase of the emitted light coherence [3]. The key lasing properties of nanolasers include the emission wavelength, the value of the lasing threshold and the carrier lifetime. The optical modes of the nanolasers observed above the lasing threshold are directly related to the dimensions of the nanolaser, since the cavity is formed by the nanowire itself. Thus, the shape of the nanowire has a direct impact on all laser characteristics. Given that the structural parameters of the nanowires vary at a nanometric scale, electron microscopy is well-suited for their analysis. Moreover, the lasing properties can be directly linked with the nanolaser shape provided that the electron microscope is equipped with a photoluminescence setup.

We aim to probe the near field of optically pumped nanolasers within their lasing regime with a focused electron beam in Ultrafast - STEM. When the electrons interact with the field created by the laser light, electrons absorb or emit quanta of the field energy. By mapping the interactions of the field with the electron beam, Photon-Induced Near-field Electron Microscopy (PINEM) enables us to measure the transversal field intensity [4] and the lasing regime dynamics with a sub-picosecond resolution.

In this presentation, we will discuss how we can study the lasing characteristics of GaN nanolasers at the nanoscale scale within a UTEM using PINEM and photoluminescence.

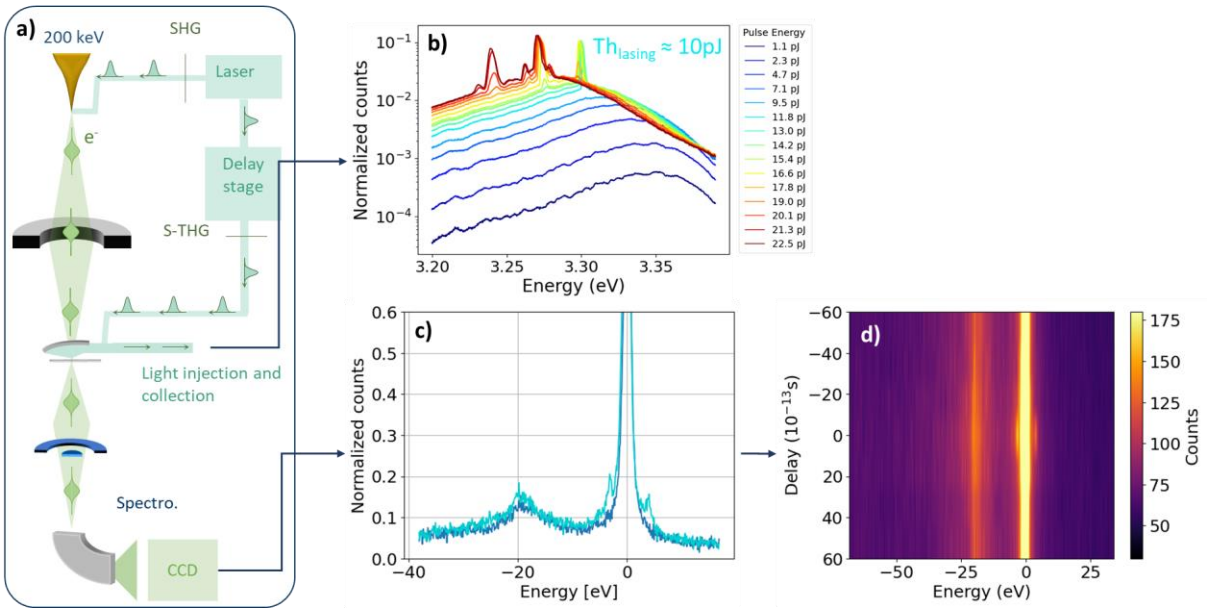

Figure 1. Use the format (a), (b), etc. for panel labels. (a) UTEM set-up, (b) Nanolaser photoluminescence spectrum depending on the pumping power, (c) PINEM spectrum on the GaN direct scattering, (d) PINEM delay scan

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Quantum Sensing and Metrology with Free Electrons

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The non-classical properties of quantum light play a key role in quantum science and technology, enabling high-precision measurements. For example, squeezed states are crucial for the detection of gravitational waves, observed as space modulations several orders of magnitude below the wavelength of the employed lasers [1]. Similarly, the so-called NOON states, a maximally entangled superposition of number states, have been proposed as an alternative way to perform measurements displaying super-sensitivity and super-resolution, with potential applications in quantum metrology, computing, and lithography [2]. The increase in sensitivity and resolution gained when employing NOON states scales with the number of photons N in the entangled state. However, generating photonic NOON states with high N at a high rates remains a challenge [3]. In recent years, free electrons have emerged as a promising tool for generating quantum states of light, where their nature as sources of broadband evanescent fields allows them to couple to strongly confined modes inside cavities and waveguides [4]. In this work, we will theoretically demonstrate that free electrons can be used to generate and detect high-number photon states far beyond the reach of exclusively photonic platforms, and that an unprecedented level of sensitivity and resolution can be achieved by measuring the free-electron currents after suitably designed electron–light interaction events. The key enabling mechanism is the strong electron–light coupling achieved by aloof electron reflection on an optical waveguide, leading to the emission or absorption of a high number of guided photons by each individual electron. We theoretically show that combining electron-beam splitters with two electron–waveguide interactions enables a tenfold enhancement in sensitivity to optical phase shifts using currently available technology. Moreover, we demonstrate that waveguided NOON states containing tens of photons can be generated at megahertz rates via post-selection of the electron energy after interaction. Our work thus presents a novel way to exploit the quantum nature of free electrons and light to perform high-precision measurements and generate exotic states of light.

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State-preserving counting of free electrons

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The emergence of free electron beams with quantum statistical properties, such as heralding [1-3], has a potential for low dose and sub-shot-noise imaging and spectroscopy in next generation electron microscopes. Relying on a co-incident detection of a generated photon to infer the counting of one electron is accompanied by an electron energy loss, making it distinguishable from its initial state.

Here, we employ the concept of *quantum non-destructive measurement* (QND) [4] for electron counting, provide a theoretical framework for the underlying effect, and offer concrete numerical examples. The non-demolition counting emerges from an elastic term in the electron-photon scattering operator. For classical fields, the phase-term governs effects such as Kapitza-Dirac scattering [5] and laser-field electron wave retarder [6]. The phase shift can be represented as $\Delta\phi$ or an equivalent time shift τ. However, in the case of a quantum field governed by the unperturbed Hamiltonian $\hat{H}_{\mathrm{ph}}$, we find that a phase kick-back effect emerges, imprinting the electron number N_e on the optical state in a form $\exp(-\frac{\mathrm{i}}{\hbar}\tau\hat{H}_{\mathrm{ph}}\hat{N}_{\mathrm{e}})$. Fig. 1 illustrates an exemplary system to observe the effect. The electron traversing the optical cavity imprints a phase shift $\Delta\phi = -2g_\phi$, on the confined photonic mode. Provided the electron velocity is mismatched from the phase velocity of the photonic mode, the electron's energy and momentum remain unchanged, preserving its quantum state. The induced phase shift scales linearly with the number of free electrons, which can be quantified as an effective refractive index. This principle would allow the counting of electrons by monitoring the phase shift in a high-finesse optical resonator. The optical phase shift induced by consecutive electrons arriving within the lifetime of the cavity will be indistinguishable, thereby providing a good quantum number estimator for their number.

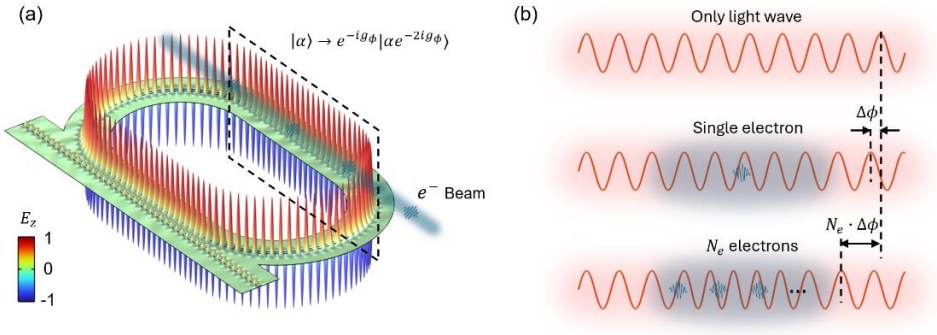

Figure 1. (a) A schematic shows the interaction of free electrons with a confined photonic mode on racetrack microresonator. (b) The amount of phase shift of light is linear proportional to electron number.

To conclude, the phase kick-back effect may act as a QND measurement for counting free electrons, combining a new aspect of electron-photon interaction with concrete technological prospect of sub-shot-noise number-resolved quantum electron microscopy.

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POSTERS

Cathodoluminescence Enhancement Mechanisms in Silica Microspheres

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Cathodoluminescence (CL) analysis of micro-resonators is a powerful analytic tool, probing the properties of photonic excitations with deep sub-wavelength spatial resolution [1,2,3]. In this work, we decompose the various enhancement mechanisms of CL emission from an archetype spherical resonator using its spectral, angular and spatially resolved features [4]. We investigate radiation of optical-whispering-gallery modes [5] in regimes of transition radiation and spontaneous emission.

The role of the excitation position in CL emission is investigated by scanning the impact parameter of the electron beam on the 2.1 μm microsphere (Fig. 1a). The azimuthal emission angle ϕ_{CL} follows the beam position angle ψ with a shift of 180°, as expected from the sphere's positive curvature (Fig. 1b). For small impact radii, the emission angles become poorly defined, evident from the spread and standard deviation of ϕ_{CL} (Fig. 1c). The angular CL maps (Fig. 1d-e) compare excitation at the sphere's edge, yielding collimated emission, to central excitation, resulting in broad angular distribution.

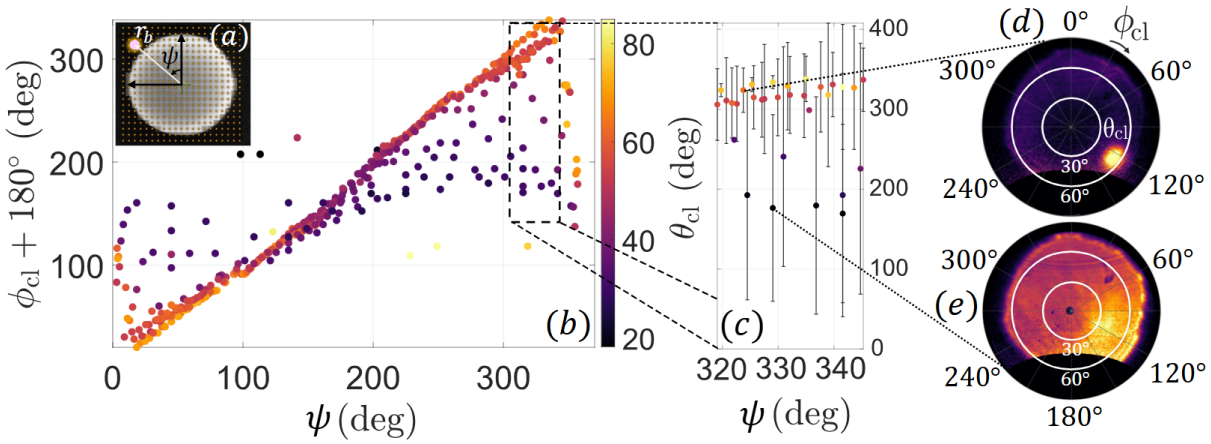

Figure 1. Angular correlation of CL emission with electron position on a 2.1 μm sphere. (a) Excitation grid with angular ψ and radial r_b coordinates. (b) Emission peak angle ϕ_{CL} vs. ψ, showing a strong correlation. (c) Standard deviation of θ_{CL} in the marked region of (b). (d)-(e) CL angular maps for excitation at $\psi = 320°$, with $r_b = R_{sphere}$ (d) and $r_b = 0.2\, R_{sphere}$ (e).

We conclude that CL can isolate the effects of electron-photon coupling form the emissivity, and hence support precision material spectroscopy. In addition, the spheres serve as a high-NA collimating lens for enhancing CL, and we suggest their application for low-signal samples or quantum systems, where the mode profile is important.

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Advancing time-resolved spectroscopies: exploring new frontiers with custom scanning units and event-based electron detection

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In recent years, scanning transmission electron microscopy (STEM) has undergone significant transformation, largely driven by advances in instrumentation. Notable developments include the introduction of sub-5 meV electron energy loss spectroscopy (EELS) [1], as well as progress in photon-electron spectroscopies such as cathodoluminescence and electron energy-gain spectroscopy [2]. Continued improvements in STEM, particularly for time-resolved experiments, hold promise for enhanced electron dose control and the development of novel spectroscopic techniques. Timepix3, although well established in various scientific fields, is still an emerging tool for fast electron detection in electron microscopy [3]. In this work, we explore a recently developed acquisition scheme that employs the event-based Timepix3 direct electron detector for EELS. Specifically, we demonstrate real-time, readout-free hyperspectral imaging acquisition [4], currently limited by the scanning engine to time resolutions on the order of tens of nanoseconds, as illustrated in Figure 1. Additionally, we present a custom-designed, Timepix3-compatible scanning unit developed to fully exploit the advantages of event-based detection. We assess its performance in enabling spatially and temporally resolved experiments and demonstrate its seamless integration with other time-resolved tools, such as pulsed lasers and electron beam blankers.

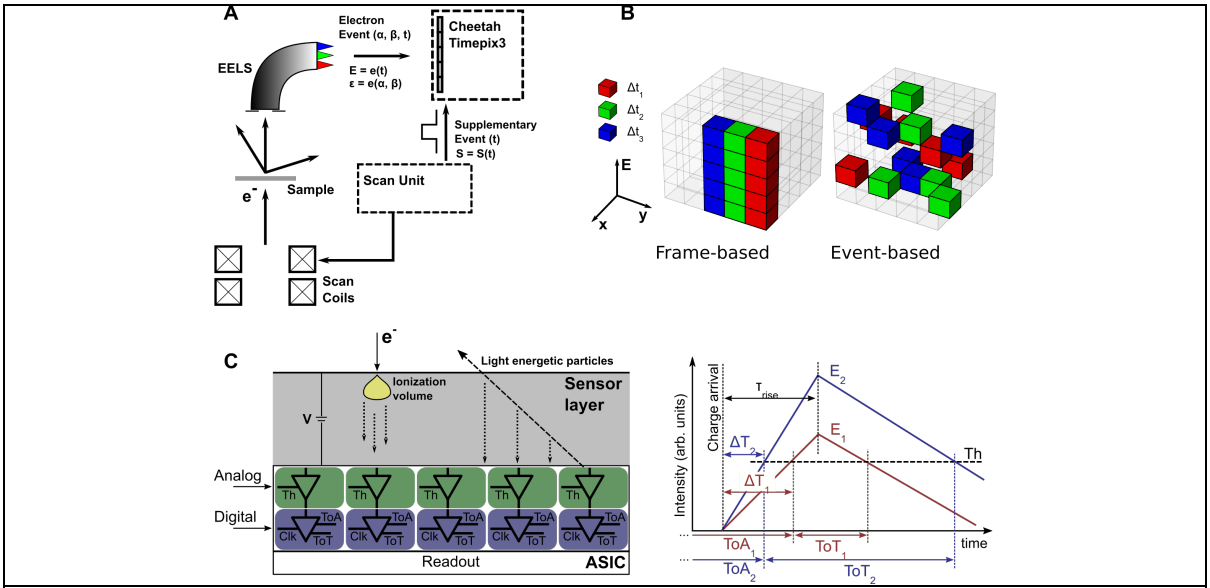

Figure 1: **Usage of event-based direct electron detector in electron microscopy.** (A) Synchronization of the scanning unit with the (B) Comparison between frame-based and event-based hyperspectral imaging. (C) Detailed illustration of hybrid pixel detector architecture.

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High Coherence Beams for Ultrafast Electron Holography

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Recent advancements in ultrafast transmission electron microscopy (UTEM) have expanded its role in investigating nanoscale dynamics with femtosecond temporal resolution. Today, it serves as a versatile tool for time-resolved imaging of structural, electronic, and spin degrees of freedom, while also enabling comprehensive studies of electron-light interactions at the nanoscale. The spatial and temporal resolution achievable in UTEM experiments is ultimately governed by the characteristics of the pulsed electron source. Laser-triggered electron emission from needle-shaped photocathodes, where emission is confined to nanometer-sized areas, results in highly coherent electron beams. Specifically, laser-driven Schottky field emitters showed a normalized beam emittance down to 1.8 nm mrad (r.m.s.), corresponding to a degree of transverse coherence exceeding 10% [1].

Phase-resolved imaging techniques, such as electron beam holography, place even more stringent demands on the coherence of the electron beam. To meet these requirements, a promising new approach involves integrating photoemission in cold field emission guns (CFEGs), which are well-established for achieving excellent brightness and spectral resolution in continuous operation mode. The photoassisted emission process has been experimentally confirmed, and the first linear cold field emission guns are currently being implemented in UTEM [2,3].

Quantitative phase-contrast imaging of optical fields at laser-excited nanostructures has recently been achieved by Lorentz microscopy and free-electron homodyne detection [4,5,6], yet they require an intricate numerical reconstruction or multiple optical interactions. A desirable and more flexible approach involves scanning transmission electron microscopy (STEM) with multiple coherent, focused electron probes [7,8], combined with inelastic electron–light scattering (IELS), which enables tunable electron-light coupling conditions and access to a broader range of local optical responses.

In this contribution, we present the characterization of laser-triggered photoemission employing a recent cold field emission gun design. The CFEG is demonstrated to support sub-nanometer foci and enables photoelectron energy widths of less than 0.3 eV. We systematically study the electron spectral distribution for varying gun parameters and compare it to theoretical models. Furthermore, we introduce multiprobe STEM holography as a phase-resolved measurement technique that enables the direct measurement of elastic and phase-shifts involved in inelastic electron-light scattering. The integration of laser-driven cold field emission is expected to significantly enhance STEM holography, providing high and stable coherent beam currents, smaller focus sizes, and improved energy resolution—all of which are critical for resolving subtle elastic and inelastic phase shifts with high spatial and temporal precision.

Ultimately, the ongoing effort in developing new electron sources will enable the routine application of coherent beam techniques in UTEM, including ultrafast holography and measuring optical phase shifts.

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Correlating optical emission with atomic structure of electron-irradiated defects in hexagonal boron nitride

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Defect engineering in two-dimensional materials offers a powerful route to tailor their electronic and optical properties for quantum technologies, particularly for single-photon emission and sensing applications. Among these materials, hexagonal boron nitride (hBN) is a leading candidate due to its wide bandgap and ability to host a variety of optically active defects. These defects exhibit bright, stable, and antibunched emission at room temperature, making hBN highly attractive for integrated quantum photonic applications [1,2,3]. Focused electron irradiation enables site-selective defect creation [1,3], offering a pathway toward reproducible and scalable on-chip devices. However, despite extensive optical characterization, the atomic-scale origin of many luminescent centers remains unresolved.

In this study, we investigate a controlled approach for generating luminescent centers in hBN via focused electron irradiation in a transmission electron microscope (TEM), enabling site-specific defect creation with nanometer precision. Our primary objective is to understand the atomic structure of in-situ TEM-generated emitters and correlate it with their optical signatures.

Defects are introduced in suspended hBN using scanning transmission electron microscopy (STEM). By tuning the beam energy and exposure time, we aim to engineer vacancy and substitutional defects. Resulting structural modifications are characterized by using high-resolution transmission electron microscopy (HR-TEM), allowing direct imaging of atomic-scale changes.

To probe the optical response, photoluminescence (PL) spectra are recorded under 532 nm excitation on both suspended and supported regions, initially at room temperature and subsequently at low temperature. This enables assessment of the emission of the generated defects.

To bridge the resolution gap between electron microscopy and optical techniques, we employ electron energy loss spectroscopy (EELS), focusing on the fine structure of the B and N K-edges [3]. By analyzing both low-loss and core-loss regions, we aim to identify defect-specific spectral features — such as shifts in excitonic resonances and the emergence of localized electronic states—that could serve as fingerprints for different defect types, provided sufficient signal strength.

Looking ahead, the integration of cathodoluminescence (CL) spectroscopy will offer a powerful complementary approach to spatially correlate structural, electronic, and optical properties at the nanoscale [4]. This work contributes to a broader effort to build a reference database linking atomic structure to optical behavior, supported by experimental imaging and molecular dynamics simulations.

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State-Agnostic Electron-Photon Pair Entanglement Certification in TEM

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The integration of quantum correlations into transmission electron microscopy (TEM) offers new possibilities for advanced imaging and hybrid quantum sensing, with potential benefits such as enhanced contrast mechanisms and reduced radiation dose for sensitive samples [1-4]. Recent experiments have demonstrated electron–photon entanglement for the first time [1, 2]. However, these demonstrations often rely on specific assumptions about the states. In this work, we present an approach for entanglement certification that does not depend on such assumptions. Specifically, we propose and analyze a robust, state-agnostic protocol for certifying electron–photon entanglement using mutually unbiased bases in position and momentum [3, 5]. Our simulations, based on a Cherenkov radiation model, describe the entangling interaction between a fast electron and the electromagnetic field generated by a dielectric medium. Conservation of energy and momentum naturally correlates the electron and the emitted photon [6-8]. We quantify these correlations, establish a lower bound on the entanglement of formation, and demonstrate that certification is feasible under realistic experimental conditions—including uncertainties in both position and momentum measurements [1, 3].

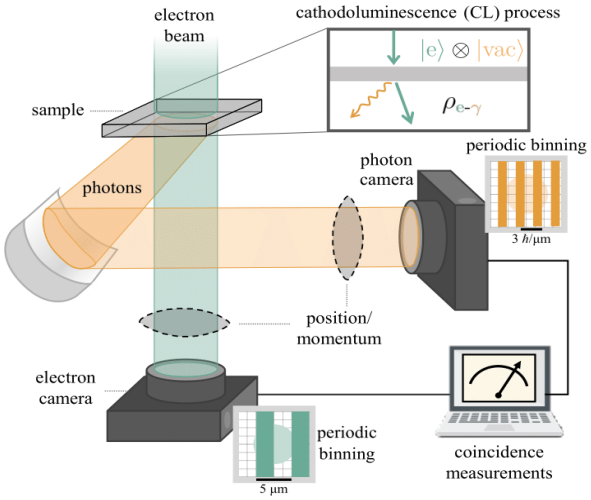

Figure 1. Schematic of the measurement setup for the proposed certification technique. A collimated electron beam (green) passes through a sample, generating electron-photon pairs via coherent cathodoluminescence (CL), with energy and momentum conserved. Emitted photons (orange) are collected and directed to a photon detector via mirror and lens. Lens transformations enable position or momentum measurements. Both particles are detected on pixelated cameras with spatially periodic binning. Coincidences are post-selected based on temporal correlations and used to bound entanglement across mutually unbiased bases.

Our simulation results demonstrate that current TEM systems—with optical specimen access and appropriate measurement implementation—can be adapted to verify this form of entanglement [3]. This work bridges electron microscopy with photonic quantum information techniques and lays the groundwork for entanglement-assisted imaging at the nanoscale. It further establishes a framework for evaluating and comparing quantum resources in electron-photon systems across different platforms.

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Experimental Demonstration of Electron-Photon Entanglement

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Entanglement [1, 2] is a phenomenon which lies at the heart of quantum mechanics and is key to quantum optics experiments and applications. The high resolution and flexibility of transmission electron microscopy (TEM), limited by lens aberrations and radiation damage constraints, motivates the development of free electron quantum optics. However, until recent experimental developments [3, 4, 5], evidence of entanglement in the TEM has remained elusive. Here, we demonstrate the presence of entanglement in position and momentum between an electron and the coherent cathodoluminescence (CL) photon that it emits [3].

We implement coincidence (ghost) imaging, a technique adapted from quantum photonic optics and capable of providing an entanglement witness [6], in an FEI Tecnai G20. Our microscope is adapted with a field emission electron source, a free-space optics collection system with a single photon counting module, and a Timepix3-based camera from Advascope. With this set-up, we perform coincidence matching between electrons and the photons emitted as the electron passes through a thin silicon membrane [7]. We are thus able to form an image of an object placed outside the microscope in the photon path, using the electron camera (Fig. 1). The joint uncertainty product, obtained from a measurement in position space and from a measurement in momentum space, violates the classical uncertainty bound: $\Delta x_{-}^{2}\,\Delta k_{+}^{2} \leq 0.492 \pm 0.047 < 1$, thereby demonstrating the presence of entanglement between the electron-photon pair. This demonstration provides a pathway for the adaption of sophisticated quantum imaging strategies from photonic quantum optics to further improve TEM techniques. We will discuss our experimental entanglement witness and provide outlook on the implementation of quantum imaging techniques in TEM.

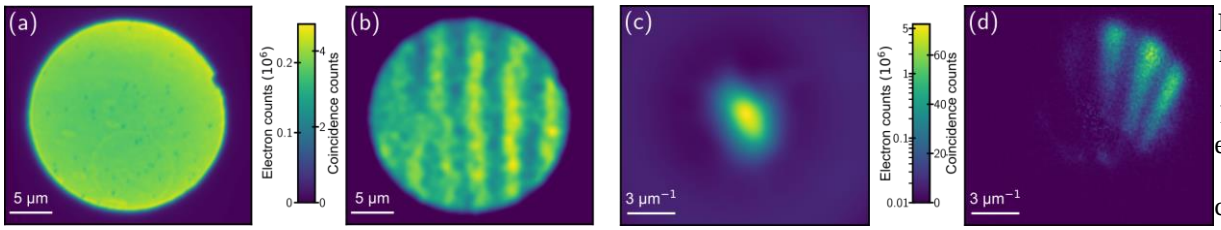

Figure 1. (a) Raw electron counts in image space. (b) Filtering the electrons for electron-photon coincidence counts reveals a ghost image of a periodic grating mask placed in the photon image plane (Δx_{-}=1.4 µm). (c) Low-angle electron diffraction image of the zero beam. (d) Filtering the electron image for coincidence counts reveals a ghost image of a periodic grating mask placed in the photon momentum plane (Δk_{+}=0.49 µm^{-1}).

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Towards using angle-resolved cathodoluminescence interferometry for 3D reconstruction of nanoscale geometries

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As traditional semiconductor scaling approaches the physical limits, a transition is made towards three-dimensional (3D) transistor structures such as FinFETs and gate-all-around devices to enable the continued growth of transistor density. In turn this trend drives the need for high resolution 3D metrology solutions. A well-known technique for imaging 3-dimensional objects is tomography, which is already widely employed in transmission electron microscopy (TEM) to reconstruct the 3D geometry of nanoscale objects. However, TEM tomography is not practical for most semiconductor applications as it involves complex and time-consuming sample preparation and analysis. Therefore alternative, faster methods will need to be considered while preserving the resolution capabilities.

In this work we will explore the use of cathodoluminescence spectroscopy (CL) for 3D geometry analysis at the nanoscale using scanning electron microscopy (SEM). In the SEM a high-energy (1-30 keV) electron beam can excite a material which will emit optical radiation in the UV-IR range. In metals the CL can be generated through coherent processes, such as the emission of transition radiation (TR) when the electron traverses the boundary between two different media. The high lateral imaging resolution of the SEM can then be combined with spectroscopy, polarimetry and angle-resolved detection to yield fast determination of the relevant feature properties in 3D.

In this work we show the angle-resolved (AR) CL measurements on rectangular holes etched into a monocrystalline Au surface using a focused ion beam (FIB) oriented at 52° with respect to the sample surface. Transition radiation can scatter off the hole edges and, due to its coherent nature, interfere with itself in the far-field, which is illustrated in **Error! Reference source not found.** [1]. Applying the Fourier transformation to these interference patterns gives information about distances in the axial dimension on top of the lateral information from the secondary electron image. Boxes of varying lateral dimensions and depths were constructed, and the angle-resolved CL spectra were observed. In **Error! Reference source not found.** we show the results obtained on a 10x10x1 μm box where AR spectra were taken at 5 different locations in the box. A clear evolution of the interference pattern is observed depending on the measurement location within the box, showing that CL interferometry is a promising method for extracting 3D information from a sample.

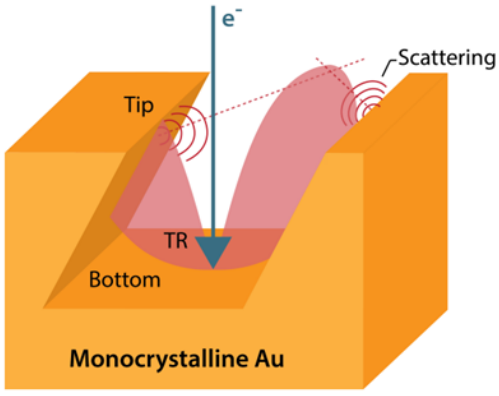

Figure 1. Schematic representation of the rectangular hole fibbed into the Au surface at 52°. The interference fringes found in the angular mission pattern are generated through transition radiation scattering off the hole edges. Image taken from Ref [1].

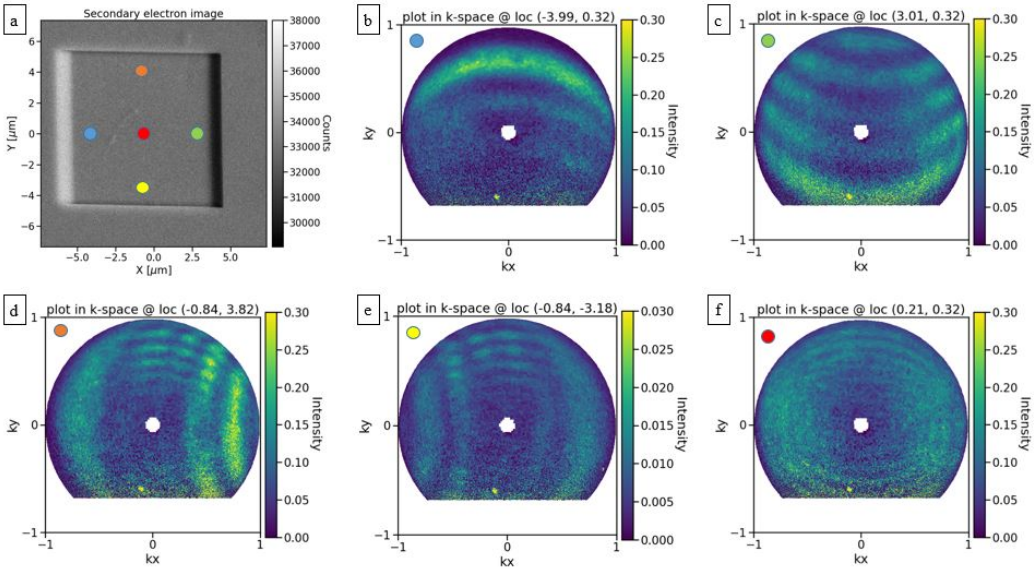

Figure 2. **a)** Secondary electron image of a 10x10x1 μm box with the 52° tip on the right side of the image and 90° walls on the top and bottom. **b-f)** Angle-resolved CL measurements taken at 5 different locations in the box as indicated by the colored markers. Clear differences in the interference fringes are observed depending on the measurement location. The interferograms provide key information on the nanostructure geometry.

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Recoil Enhances Optical Electron Compression

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Shaping the wavefunction of free electrons with light has become a powerful tool with applications in electron microscopy, enabling unprecedented combinations of spatial and temporal resolution for probing electronic and molecular dynamics in materials on the attosecond timescale [1]. Recent advances in light-beam shaping now allow for the creation of tailored optical fields to optimize photon-electron interactions. Since the inception of photon-induced near-field electron microscopy (PINEM), inelastic interactions between free electrons and optical near-fields have been widely used for this purpose, usually spanning over short distances (up to hundreds of nanometers) where they can be adequately modeled in the non-recoil regime [2,3] but recent works have proposed that recoil effects can improve the final pulse compression [4]. In this work, we analyze a scenario where PINEM interaction takes place over a large distance in which recoil effects become critical and strongly affect the evolving electron wave function. Specifically, we consider a simple setup that creates near-fields by total internal reflection of a laser in a prism (Fig.1a). We show that, due to strong recoil effects, the resulting electron energy spectra deviate significantly from those predicted by standard PINEM (Fig.1c and Fig.1d). We exhaustively map different configuration parameters and resulting pulse characteristics, providing a guidelines for achieving pulses with high compression surpassing that of standard PINEM [5]. The parameter optimization was guided by considerations of practical implementability in standard laboratory environments.

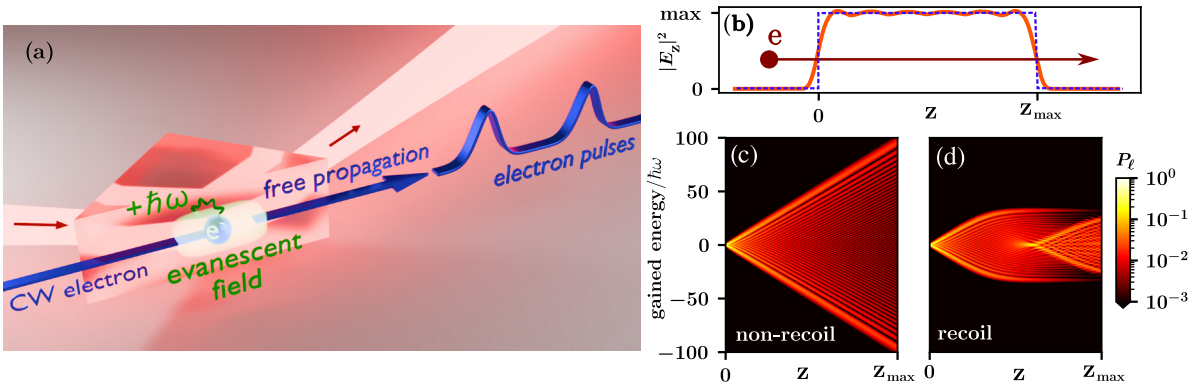

Figure 1. (a) An electron beam interacts with an evanescent field generated by total internal reflection of a laser beam of frequency ω. Subsequent free propagation transforms the electron energy distribution into a spatial train of pulses. (b) Transverse cut of the interaction region, where the electron (arrow indicating the trajectory) transverses the evanescent optical field and interacts with a component longitudinal with respect to the electron velocity. Diffraction-limited fields (solid curve) can be shaped to produce results similar to an ideal box-field marked by a dashed-blue line. (c) Probability of electron energy sideband occupation according to standard non-recoil PINEM theory during the interaction with the external optical field. (d) Same as (c) but with recoil effects included.

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Laser-induced plasma for dynamic correction of spherical aberration in Ultrafast Transmission Electron Microscopy (UTEM)

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Over the past decades, advancements in electron microscopy have been driven by the aim of reaching atomic-scale resolution. Thanks to the development of multipole lens systems, we can currently achieve a resolution on the sub-Ångström scale [1, 2], which necessitates elaborate elements to correct aberrations in the electron optics.
Here, we propose an alternative approach to correct spherical aberrations in ultrafast electron microscopes based on a laser-induced plasma that dynamically shapes the transverse profile of a pulsed electron beam. This approach is extremely versatile and does not require significant hardware modifications to the TEM. When a high-intensity infrared (IR) laser pulse is focused onto a metallic structure, electrons are emitted from the metal, forming a negative charge cloud that evolves in space and time, and interacts with the electron beam [3, 4]. The spatial configuration of the plasma electric field depends on the geometry of the metallic structure and the time delay after plasma formation. By tailoring these parameters, the plasma can act as a lens with a negative spherical aberration coefficient (Cs) that compensates for the intrinsic positive aberration of the microscope lenses.
First, we developed a numerical code aimed at solving Poisson's equation with given boundary conditions, to compute the electric field induced by various plasma configurations and estimate their effect on an electron beam. Then, we performed experiments by generating toroidal-shaped plasma from micrometer-sized holes in a gold disk within the column of the UTEM, before the sample plane. We investigated the effect of different plasma configurations by varying the laser fluence and the delay between the IR irradiation and the electron probe.
From the simulations, we demonstrate that a toroidal charge distribution can act as an effective lens for the electron beam, resulting in a negative spherical aberration. Experimental results confirm that, for specific time delays, the measured Cs assumes negative values. Moreover, our measurements show that plasma-induced spherical aberrations can be tuned by acting on the delay time and the laser power.
The ability to tailor light-induced plasma offers a promising strategy for spherical aberration control in electron microscopy. Future research will focus on optimizing sample geometry and laser power to achieve the experimental conditions needed to exploit this effect for high-resolution imaging applications.

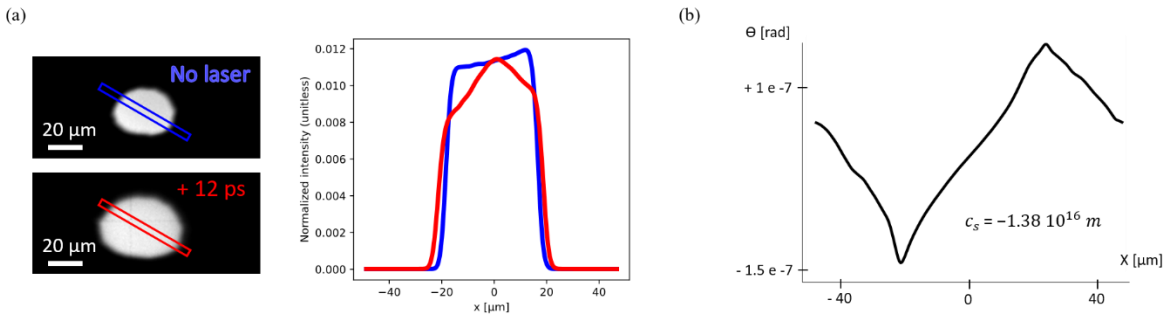

Figure. (a) Distortion of the electron beam image and corresponding intensity profile induced by plasma generated using a 75 mW laser at12 ps delay, (b) Position-dependent angular deflection of the electron beam induced by plasma generated with a 330 mW laser at 18 ps delay.

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Electron Beam Shaping Using Compton Scattering

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The interaction of electron beams with tailored light fields offers promising opportunities for beam shaping. In free space, single-photon processes are forbidden due to energy–momentum conservation, but higher-order processes can be kinematically allowed. Compton scattering is one such process, where an incoming photon interacts with an electron, exchanging both momentum and energy. Here, we study the interaction of electrons with multiple laser fields and explore the use of Compton scattering for shaping of electron beams.

Acknowledgments

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Ponderomotive Control of Aberrations in Electron Optics

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Enhancing the resolution of electron microscopy remains a fundamental challenge, as the field continues to depend primarily on conventional electrostatic and magnetostatic lenses. While multipole correctors have enabled partial mitigation of aberrations, intrinsic limitations, particularly chromatic and spherical aberrations, persist as major barriers to optimal electron optical performance and resolution.

In this work, we present recent advances in light-based aberration correction for electron lenses [1–3]. We demonstrate that spherical aberrations in electron beams can be effectively compensated via interaction with a femtosecond Laguerre-Gauss beam, enabling accurate beam shaping as confirmed by numerical simulations and experimental verification.

Additionally, we show that chromatic aberrations of electron lenses can be mitigated using a pulsed ponderomotive lens, where the energy-dependent focal position of the electron is controlled by synchronizing the temporal chirp of the electron pulse with a time-varying optical potential.

Our findings establish a novel framework for aberration correction in electron optics through the use of structured light fields. Replacing traditional electron lens elements with optically programmable systems opens new possibilities for improving resolution, enhancing contrast, and simplifying the architecture of ultrafast electron microscopes.

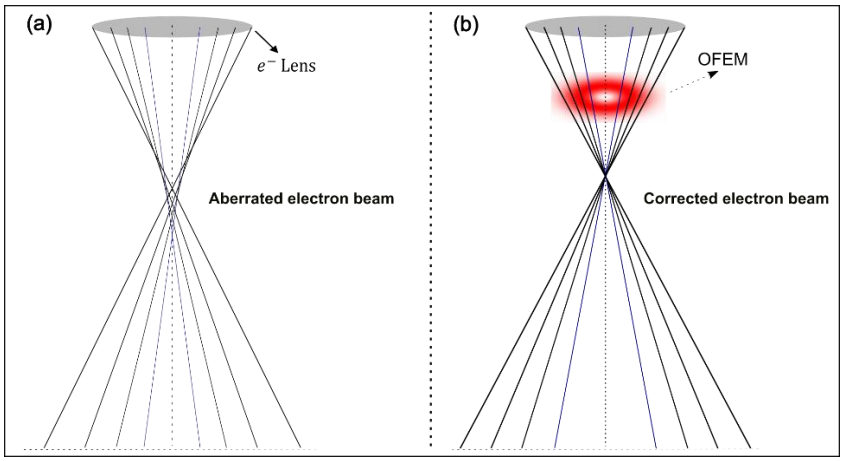

Figure 1. (a) Spherical aberrations in an electron beam caused by an imperfect lens, resulting in a blurred focal spot due to rays focusing at different planes. (b) Interaction with an optical field electron modulator (OFEM) reshapes the electron trajectories, compensating for aberrations and producing a sharp, aberration-free focal point where all rays converge in the same plane.

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Acknowledgments

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Inelastic electron-light interaction probed by holographic scanning transmission electron microscopy

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Ultrafast Transmission Electron Microscopy (UTEM) offers unique capabilities for probing inelastic electron-light scattering (IELS). Harnessing these capabilities, Photon-Induced Near-Field Electron Microscopy (PINEM) can now routinely image the amplitude and shape of optical fields at laser-excited nanostructures. Phase-resolved imaging is enabled by sequential interactions, and dispersive propagation yields attosecond electron pulses, both of which offer a pathway to sub-optical-cycle temporal resolution [1,2,3]. However, these techniques require an elaborate geometry with multiple optical interactions, limiting flexibility in nanostructure imaging.

In this work, we combine IELS with a STEM holography approach. We introduce an amplitude grating into the condenser aperture of our UTEM, which enables the creation of multiple coherent, focused electron probes in the sample plane [4,5]. Two beams are selected with an aperture, which gives access to two inelastic interactions at spatially displaced sample positions. A 2D camera, positioned behind an energy filter, records the far-field interference pattern of these probes in energy-dispersive mode. This provides us with simultaneous, direct access to the amplitude and relative phase of the inelastic interactions as well as elastic phase shifts, which we use to characterize the optical nearfield of a nanostructure quantitatively. Furthermore, this technique enables tailoring the electron-light interaction of each probe through sample design and laser illumination, thereby providing control over the final electron state. The interference pattern reflects the coherent superposition of the individual probe states as a function of their relative phase. We showcase the resulting electron spectra, which display distinct features such as asymmetries, that cannot be achieved using a single interaction.

Our approach enables the direct measurement of phase shifts induced by inelastic electron-light scattering. It may be extended to give access to material dynamics through phase-resolved diffraction. Besides, the superposition of multiple probes undergoing different IELS interactions can be used to synthesize tailored electron states, which could, for example, be optimized for temporal bunching as theoretically predicted [6].

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Active light steering through generalized Smith-Purcell effect

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Smith-Purcell (S-P) effect consists of the constructive interference of cathodoluminescence (CL) arising from the interaction between moving charged particles and periodic structures, and it holds immense promise for various applications, from particle accelerators to broadband radiation sources.

In typical configurations, S-P effect is realized with perfectly periodic arrays where all elements are equivalent and uniformly spaced (with some period a), and the resulting radiation follows a well-known geometric constraint at wavelength λ_0 given by $\cos\theta_n = c/v - n\,\lambda_0/a$ [1], where n is an integer and v is the electron velocity. More recent realizations relied on non-uniformly-spaced arrays designed such that the difference in optical path from the CL emitted by each array element resulted in the focusing of radiation in a controlled position in the near field [2]. In this study, we propose instead a uniformly-spaced array of non-equivalent elements designed to control the far-field emission of S-P radiation.

We start by studying the emission properties of non-uniform arrays with N elements with induced dipole moments $\boldsymbol{p}_j$ $(j = 1, \ldots, N)$. From here, we derive a generalized version of the Smith-Purcell effect with the form $\cos\theta_{n\ell} = c/v - [n - \ell/(N-1)]\,\lambda_0/a$, where $\ell = 0, \ldots, N-1$ denotes the normal modes of the dipole moment distribution according to the Fourier transform $\tilde{\boldsymbol{p}}_\ell = \sum_j \boldsymbol{p}_j e^{2\pi i j \ell/N}$. Regular S-P effect is recovered when all $\boldsymbol{p}_j$ are equal and thus only the $\ell = 0$ mode survives. In Fig. 1, we show different configurations where by adjusting the dipole moment distribution in the array (inset), we achieve highly customized emission profiles.

We envision that such effects can be realized passively by controlling, for example, the size and/or material of each array element, or actively by individually tuning the Fermi level of graphene ribbons disposed in an array. To do so, we are able to calculate the exact doping level of each ribbon necessary to achieve a given value of induced dipole moment, including the ribbon-ribbon interactions, which can be customized actively as desired. Our findings pave the way for the development of advanced S-P radiation sources with tailored and actively-controllable emission properties.

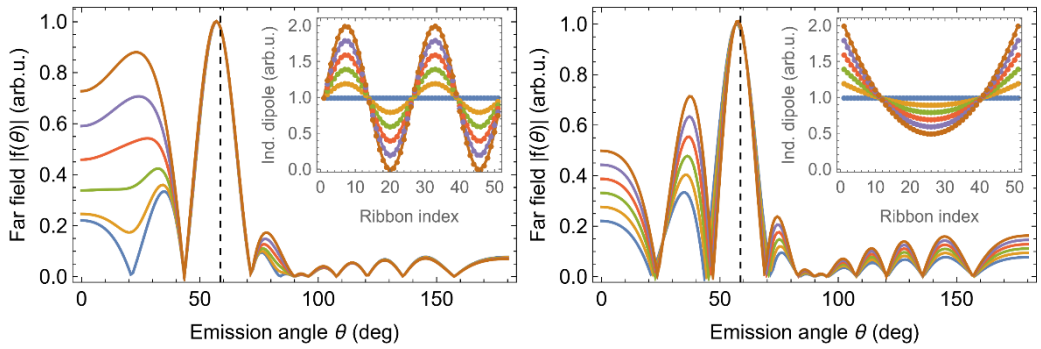

Figure 1. Examples of engineered S-P emission profiles along polar angle θ for the dipole moment distributions shown in the color-coordinated insets. The black dashed line represents the regular S-P emission angle for a uniform array (blue curves). We take $a = 500$ nm, $\lambda_0 = 5.2$ μm, and $v = 0.1c$.

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Substrate-Assisted Cathodoluminescence

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Cathodoluminescence (CL) microscopy is a powerful technique for nanoscale optical characterization [1]. Fundamentally, all CL experiments depend on interactions between electrons and the sample. These interactions may involve direct excitation, where the electron beam penetrates the sample and transfers energy directly. Alternatively, in aloof excitation, electrons do not directly penetrate the sample; instead, they pass very close to its surface, interacting with the evanescent nearfield components of electromagnetic modes confined at the sample surface [2]. A third, less-explored category is indirect excitation of emitters, which has been attributed being mediated by secondary electrons (SEs) [3, 4] or backscattered electrons (BSEs) from the substrate [5].

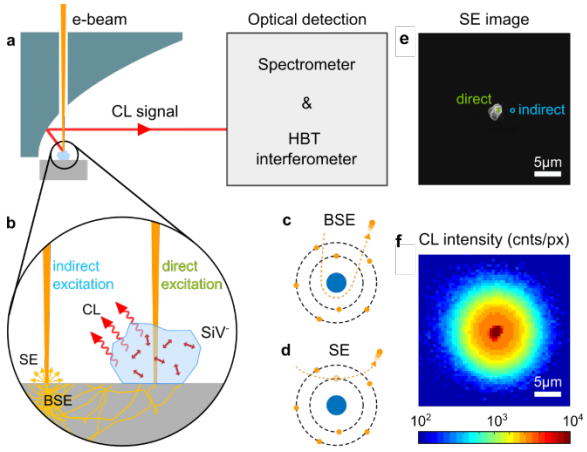

Figure 1. (a) Schematic of the cathodoluminescence (CL) detection setup, utilizing a spectrometer and Hanbury~Brown--Twiss (HBT) interferometer. (b) Close-up illustration comparing indirect excitation (via secondary electrons (SE) and backscattered electrons (BSE) generated in substrate) and direct excitation of SiV^- color centers in diamond. (c,d) Inelastic and elastic collisions between the incoming primary electrons and substrate atoms. (e) SE image of studied diamond. (f) Corresponding to panel (e) log-scaled spatial map of SiV^- CL, where the dark-red core marks the intensity from directly excited centers in the diamond, and the concentric fall-off captures the radially decaying indirect excitation of SiV^- CL.

In this study, we explore the properties of this indirect excitation approach. Using color centers in diamond as sensitive and durable local probes, we investigate the spatial profiles of indirect CL in different geometries and substrates (see Fig. 1). Photon correlation experiments demonstrate increased synchronization of emitters at reduced currents, which we propose utilizing for extracting of effective indirect excitation currents experienced by emitters, achieving remarkably low values down to 0.1 pA, highlighting the potential for minimally invasive probing of sensitive emitters in CL microscopy.

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Smart*Light: an X-band LINAC-based Compton x-ray source with continuous tunability

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At Eindhoven University of Technology, a tabletop x-ray source based on inverse Compton scattering (ICS) has recently been commissioned [1]. In the ICS process monochromatic x-rays are produced by colliding relativistic electron bunches with intense laser pulses. This compact and tunable source holds the promise of a performance in between small-scale x-ray tubes and large-scale synchrotron light sources, making advanced x-ray diagnostics accessible to a wider range of applications. The beamline utilizes a 100 kV DC photo-electron gun in combination with an X-band linear accelerator. The X-band LINAC is based on a design for the CERN Compact Linear Collider (CLIC), but adapted to allow for the injection of subrelativistic electrons. After acceleration the electron bunches collide with femtosecond laser pulses, producing x-ray photons with energies which are continuously tunable between a few and 40 keV. In this presentation we present the design and the successful commissioning of Smart*Light. We discuss the measured characteristics of the x-ray beam, which are in agreement with theoretical expectations, and we present results from the first demonstration experiments.

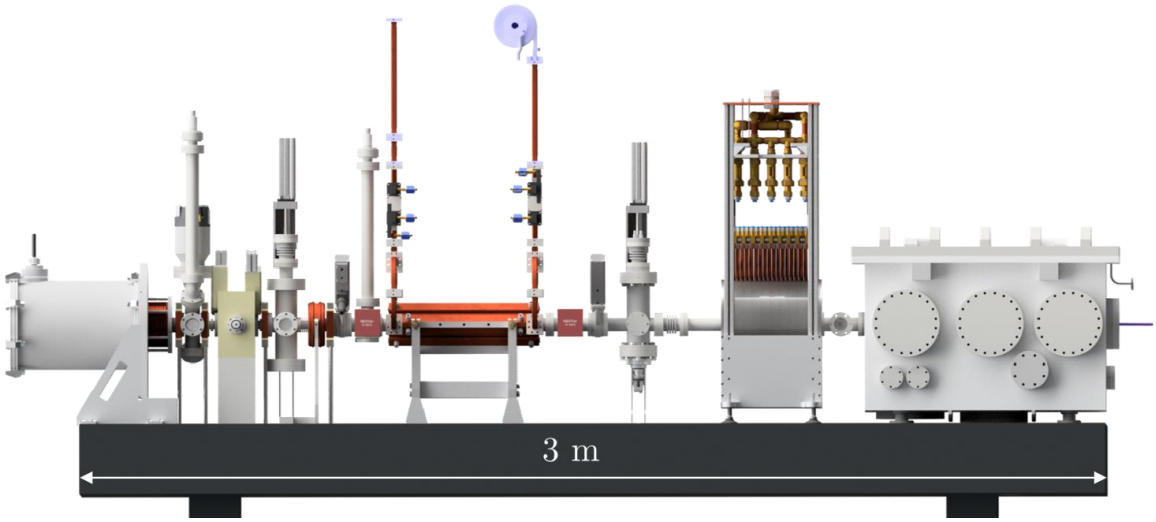

Figure 1. Electron beamline of the Smart*Light setup.

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Cathodoluminescence Excitation Spectroscopy: probing resonant energy absorption and emission in an electron microscope

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Cathodoluminescence excitation spectroscopy (CLE) in Scanning Transmission Electron Microscopy (STEM) correlates electron excitation with subsequent photon emission events. These are measured via electron energy loss spectroscopy (EELS) and cathodoluminescence spectroscopy (CL) respectively and correlated via nano-second resolved coincidence detection using a Timepix3 detector for electrons and photon multiplier tubes (PMTs) for CL. This setup, currently installed on a Nion Hermes STEM at the LPS-Orsay, allows for the tracking of which excitation energy leads to a photon emission, crucial for addressing the broadband nature of the electron-matter interaction. [3] In addition, the selection of electrons leading to photon emission might be an efficient way to measure energy losses in emitters with weak absorption probabilities, such as single point defects. We are currently investigating single point defects in nanomaterials such as hexagonal boron nitride (hBN) flakes and diamond nanoparticles aiming to demonstrate resonant excitation in materials. CLE's high sensitivity also picks up transition radiation (TR), which overlaps spectrally with in-gap excitations of interest [2]. The TR then appears as a detrimental background that can be filtered temporally, spectrally, and via angle and polarization of the emission. Towards this goal, we are investigating angular filtering via an optical system to be integrated in the CL light collection path, enabling Fourier-plane imaging of angular emission profiles. We also developed a python code to simulate the imaging of different emission profiles (dipolar, TR, Lambertian) and their transformations by a parabolic mirror as in [4]. Already by angular filtering we achieved a signal-to-background ratio enhancement of 2. In the future, we plan to explore polarization and temporal filtering to further improve this background subtraction, aiming to observe excitation and emission at the same energy.

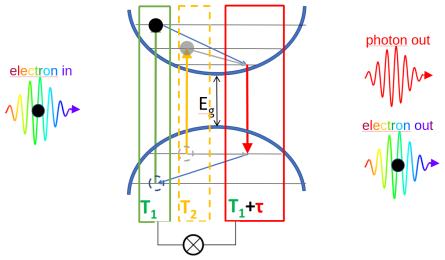

Figure 1. Schematics of electron-photon coincidence detection for CLE.

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Probing Phonon Polaritons in Nanostructures via STEM-EELS

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Phonon polaritons (PhPs), formed by the coupling of optical phonons with electromagnetic waves, enable deep subwavelength confinement and are central to mid- and far-infrared nanophotonics. Scanning transmission electron microscopy combined with electron energy-loss spectroscopy (STEM-EELS) offers distinct advantages over optical techniques, providing sub-nanometer spatial resolution, large momentum transfer, and broadband spectral access. We have employed STEM-EELS to image confined phonon polariton modes in various nanostructures. In SiC nanorods [1] and h-BN nanotubes [2], we observed surface PhPs, and hyperbolic PhPs with ultrasmall mode volumes (~10^{-10}) and Purcell factors exceeding 10^{12}. The technique also enabled direct mapping of phonon polariton dispersion in ZnO nanowires [3] and α-MoO_3 nanoribbons [4], extending well into the far-infrared. The ability of fast electrons to transfer large momentum allows access to the full polariton dispersion, which we demonstrated in multilayer and monolayer h-BN [5]. These measurements revealed PhPs with ultrahigh confinement (>487) and ultraslow group velocities (~10^{-5} c). Similar characteristics were also observed in ultrathin $SrTiO_3$ membranes [6], highlighting their potential as a novel polaritonic platform. These results underscore the growing role of STEM-EELS in advancing vibrational nanophotonics and polaritonic device engineering.

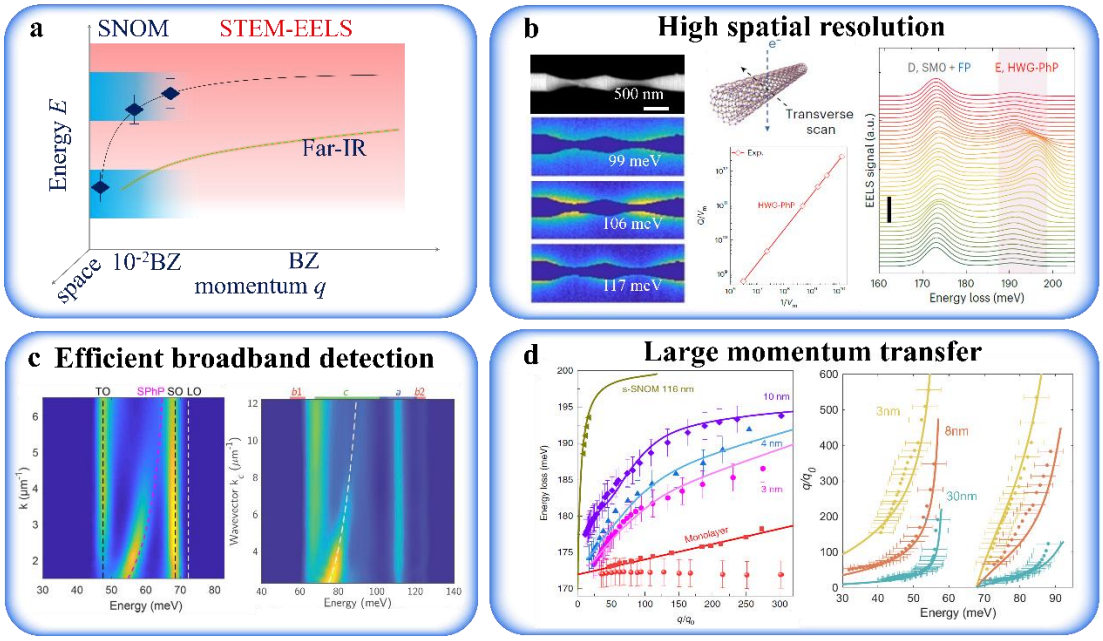

Figure 1. (a) Schematic showing STEM-EELS's advantage for probing phonon polaritons. (b) EELS mapping on SiC nanorod [1] and EELS line-scan on BN nanotube [2]. (c) Dispersion of infrared phonon polariton in ZnO nanowire [3] and α-MoO_3 nanoribbon [4]. (d) Dispersion of phonon polaritons in h-BN flakes [5] and $SrTiO_3$ membranes [6] with different thicknesses.

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Modulated free electrons as quantum sources of light

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Fast electrons in electron microscopes can both excite ground-state samples via spontaneous emission and exchange energy with optically or thermally populated specimens through stimulated processes. Specifically, by synchronizing femtosecond laser illumination with electron pulses at the sample, inelastic electron-light scattering (IELS) multiplexes the electron wave function into a coherent superposition of energy states spaced by multiples of the laser frequency, which can then be recorded using a spectrometer. This effect has enabled studies of ultrafast dynamics in nanostructured materials and on their associated optical near fields.

Recently, due to the strong dependence between the input electron wave function and the output light state, IELS has been used for energy-time tailoring of free electrons, inspiring several works where electrons act not only as probes but as active elements capable of controlling the quantum state of light emitted into photonic structures. When coupling to bosonic light modes, single-electron beams with kinetic energies well above the mode energy behave as semiclassical charges, producing Poissonian

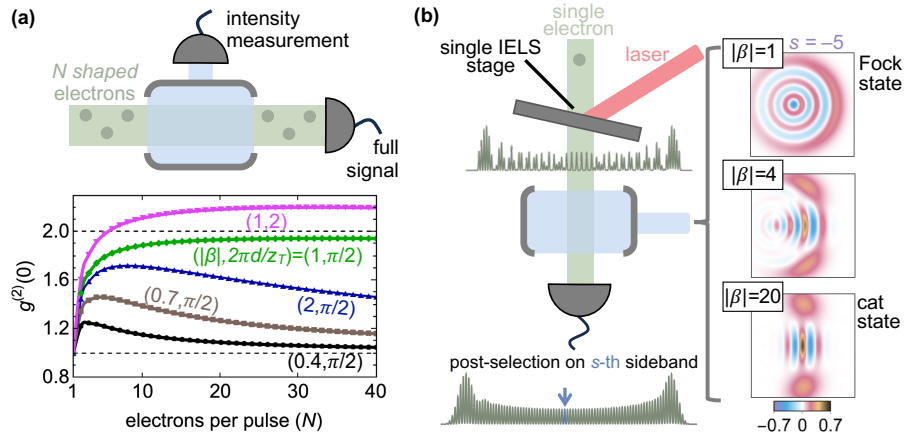

Figure 1. (a) N IELS-modulated electrons with strength $|\beta|$ and free propagation of length d emit light in a photonic structure characterized by a zero-delay autocorrelation function $g^{(2)}(0)$. (b) Quantum light state synthesis by an IELS-shaped electron post-selected on the s-th sideband as a function of $|\beta|$.

statistics with coherences tied to the electron's temporal compression (in the attosecond regime for optical photons). To this end, complex schemes combining IELS interaction stages and free propagation zones have been proposed to create high-contrast attosecond pulse trains, so-called comb electrons. These are fundamental to generating quantum-light states such as cat and GKP states post-selecting electrons in energy after interaction with the cavity [1], though experimental demonstration remains distant due to technical challenges.

In this work [2], we firstly show that N-electron pulses modulated by a single IELS stage gives rise to different types of emission, from thermal to Poissonian light, upon tuning the amplitude of the modulating field and its distance from the second sample (see Fig. 1). Then, we present a scheme employing a single IELS interaction and energy post-selection to create cat states and a multi-interaction IELS stage to produce more complex light statistics with fidelities ~98%. We believe this work marks a significant milestone, bringing electron beams in electron microscopes closer to the first real generation of complex quantum states of light at the nanoscale.

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Probing Strongly Confined Mid-infrared to Terahertz Phonon Polaritons in Few-Unit-Cell Freestanding $SrTiO_3$ Membranes by STEM-EELS

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Surface phonon polaritons (SPhPs) enable deep subwavelength light confinement, offering distinct advantages for applications in infrared sensing, imaging, and optoelectronic devices. However, their application in the terahertz (THz) regime has been limited by the narrow Reststrahlen bands of conventional polar materials. Strontium titanate ($SrTiO_3$), a quantum paraelectric perovskite with large LO-TO splitting, has recently emerged as a promising THz polaritonic platform. Here, we use monochromated scanning transmission electron microscopy-electron energy loss spectroscopy (STEM-EELS) to probe SPhPs in freestanding $SrTiO_3$ membranes down to a few unit cells thick. STEM-EELS offers broadband spectral access, large momentum transfer, and high spatial resolution—key advantages for detecting polaritons in ultrathin films where the dispersion flattens. We directly observed SPhP modes in 30 nm, 8 nm, and 3 nm $SrTiO_3$ membranes, covering both THz and MIR regimes. In 3 nm membranes (~8 unit cells), we extracted confinement factors >500 and group velocities down to ~10^{-5} *c*, rivaling leading 2D polaritonic systems. These findings demonstrate the viability of oxide-based nanophotonics in the THz regime and highlight the power of STEM-EELS for exploring vibrational polaritons in ultrathin materials. [1]

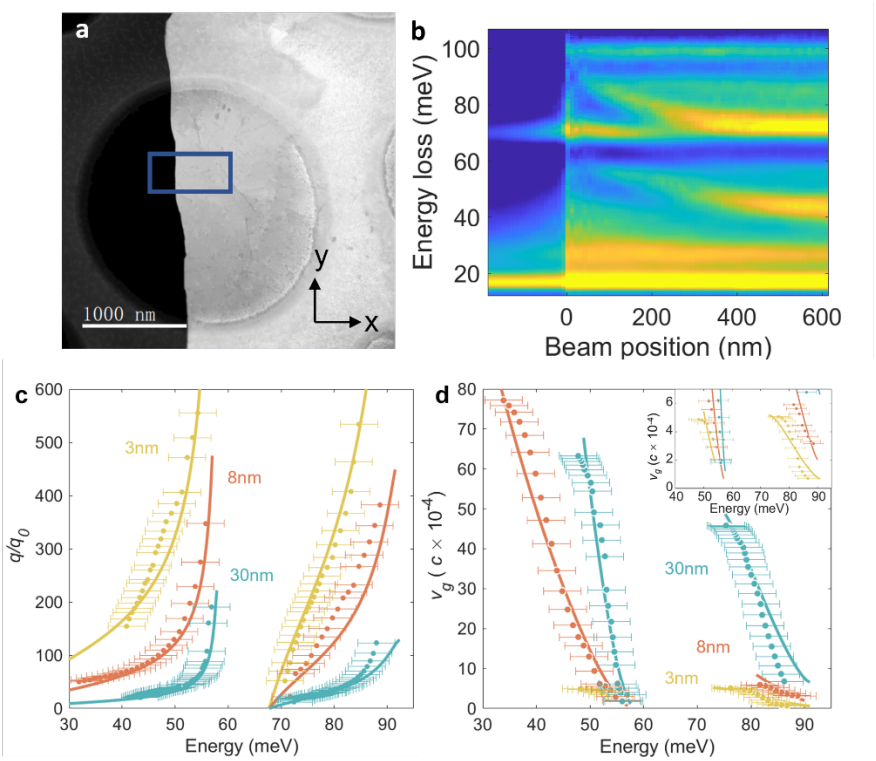

Figure 1. (a) HAADF image of the 30 nm thick freestanding $SrTiO_3$ membranes. (b) EELS line-scan taken in the blue box in (a) by summing data along y-direction. Confinement factor (c) and group velocity (d) of SPhPs for different thicknesses.

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Towards Quantum Computer-Enhanced Measurements in Electron Microscopy

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Electron microscopy inherently causes radiation damage to biologically relevant samples, limiting the number of usable probe particles before the sample deteriorates and thereby the achievable resolution. Recent advances in quantum imaging have shown that more information can be extracted per probe particle, using techniques such as quantum polarized light microscopy [1], interaction-free measurements [2], multi-pass protocols [3] and electron-qubit interactions [4] to surpass classical limits.

In this work, we present first steps towards the integration of a quantum computing platform based on laser-cooled Ca^+ ions into an electron microscope. We address key challenges such as the required ultra-high vacuum and the spatial constraints of fitting a planar Ca^+-Paul trap within the pole pieces while ensuring optical access for laser cooling, state initialisation and readout. Our ongoing efforts focus on the design and construction of this integrated system, aiming to demonstrate its feasibility and potential to minimize radiation damage and enhance low-dose imaging resolution in electron microscopy.

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Detecting Spin System Dynamics with Free Electrons

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In recent work [1,2], we developed SPINEM (Spin Electron Microscopy), which synergically combines microwave (MW) spectroscopy with transmission electron microscopy (TEM) to leverage its high spatial resolution [3], enabling localized *in situ* detection of MW-driven spin transitions via a free-space electron probe.

Spin state polarization is achieved via the $\mathbf{B_0}$ magnetic field (170 mT) of the TEM's polepiece, while a custom-designed microresonator integrated into a TEM sample holder drives spin transitions and modulates the electron beam at a frequency of 5 GHz (see Fig. 1). This modulation enables phase-locked detection, allowing the isolation of spin precession contributions to the electron beam deflection with a sensitivity down to the picoradian scale. By sweeping the polarizing $\mathbf{B_0}$ field across the specimen's resonance, SPINEM spectra can be acquired with a spatial resolution of 30 µm.

These results advance quantum electron microscopy by uniting spin physics with high-resolution electron optics. SPINEM enables sensitive and precise detection of spin excitations and, with future improvements, such as the incorporation of advanced STEM techniques, promises high-resolution mapping of spin excitations through time-resolved 5D STEM measurements.

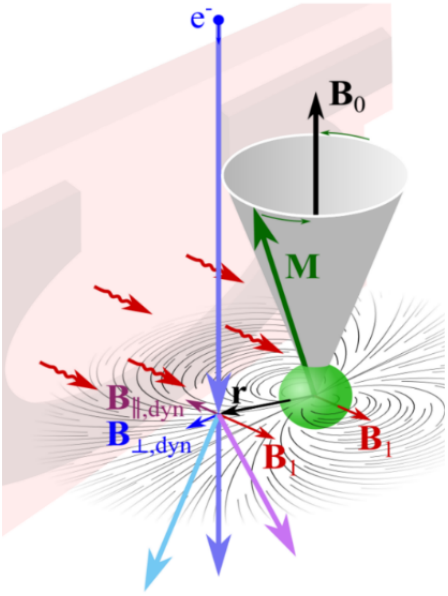

Figure 1: SPINEM spectra are acquired by sweeping the polarizing $\mathbf{B_0}$ field across the resonance, while keeping the driving $\mathbf{B_1}$ magnetic field constant to maintain a stable phase reference. At resonance, the specimen's magnetization **M** begins to precess, inducing dynamic magnetic fields $\mathbf{B_{dyn}}$ that deflect the electron beam.

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Acknowledgments

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Al plasmonic metasurfaces studied with optical spectroscopy, EELS and ultrafast TEM

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We present a study of plasmonic metasurfaces probed using conventional electron energy-loss spectroscopy (EELS), ultrafast TEM (UTEM, imaging and diffraction modes) and transmission optical spectroscopy. By fabricating aluminium plasmonic metasurfaces on ultrathin membranes that are optically and electron transparent, we can transfer the sample between the three platforms and measure on the same structure (see Figure 1). Our conventional EELS measurements enable nanoscale mapping of the different optical modes of the nanostructures and identification of the spectral feature present in the optical data [1-2]. The UTEM measurements carried out with a laser beam of the identified energy reveal information about the dynamics of the optical excitations. Collectively, the results provide a comprehensive insight into the energy absorption mechanisms and allow a detailed spatiotemporal mapping of the optical response of the metasurface nanostructures.

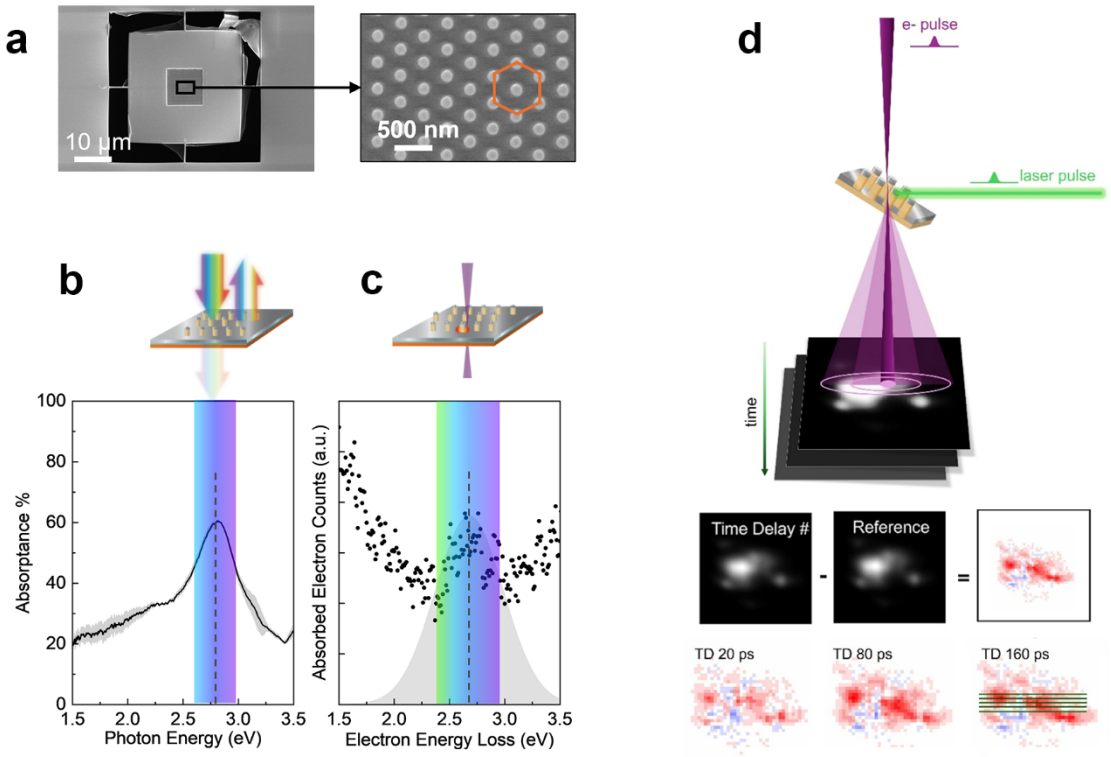

Figure 1. (a) SEM images of the fabricated metasurfaces on 30 nm thick membranes, which were cut out and placed on TEM compatible grids using the focused ion beam (FIB), (b) example of the optical absorption spectrum and (c) EELS measurements from the structures. (d) Configuration of the UTEM diffraction measurements.

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Ultrafast 4D scanning transmission electron microscopy for imaging of localized optical fields

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We report on the development of ultrafast 4D scanning transmission electron microscopy (U4DSTEM). This technique expands the family of methods based on ultrafast electron microscopy, which are utilized to image transient phenomena occurring on nanoscale [1]. Visualization of photon coupling with various nanostructures has been enabled by photon-induced near-field optical microscopy (PINEM) [2], which is based on spectral filtering of electrons inelastically scattered due to the stimulated interaction with the near-field. PINEM is sensitive only to the longitudinal component of Lorentz force in the electron propagation direction and requires spectrally-filtered detection. In contrast, U4DSTEM allows us to image the transverse components of the optical near-field without the need of electron spectral filtering [3]. We demonstrate the applicability of the method by imaging the integrated Lorentz force generated by optical near-fields of a tungsten nanotip and the ponderomotive potential of an optical standing wave with a spatial resolution of 21 nm.

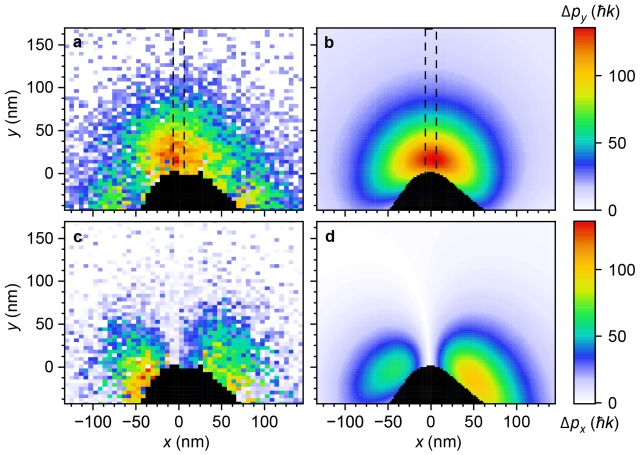

Figure 1. Imaging of the transverse component of Lorentz force of optical near-field generated on the surface of a tungsten nanotip by coherent optical excitation. (a) Maximum of the measured *y*-component of the transverse momentum change of the electrons Δp_y and (c) its *x*-component Δp_x compared to the numerical results shown in (b), (d). All panels use the same color scale.

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Acknowledgments

Czech Science Foundation (project 22-13001K), Charles University (SVV-2023-260720, PRIMUS/19/SCI/05, GAUK 90424), the European Union (ERC, eWaveShaper, 101039339) and TERAFIT project No. CZ.02.01.01/00/22_008/0004594 funded by OP JAK, call Excellent Research.

High-Resolution Cathodoluminescence Imaging of AlGaN Deep-UV Quantum Wells

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Cathodoluminescence (CL), utilised in both steady-state and time-resolved configurations, has significantly advanced exploration of the optical properties of semiconductor materials, playing a crucial role in the development of optoelectronics. Recently, a state-of-the-art time-resolved cathodoluminescence (TRCL) microscope was installed at the Paul Drude Institute, which incorporates an ultra-high-performance scanning electron microscope equipped with a distinctly stable electron source for optimised imaging conditions and the opportunity for ultra-fast beam pulsing and operation at very-low acceleration voltages, down-to 0.35 kV. The latter is critical for achieving high spatial-resolution cathodoluminescence (HR-CL) imaging. By coupling this with specially designed samples, we show that a lateral spatial-resolution of 30 nm can be achieved through measurements performed at 1 kV and 40 K on a 255 nm emitting quantum well (QW) shown in Figure 1. The samples consist of ultra-thin (1 nm) QWs along with thin (5 nm) barriers to limit carrier diffusion and the excitation volume respectively allowing HR-CL imaging to be achieved. We apply HR-CL to investigate the spatial densities and optical behaviours of point defects (PDs) in AlGaN QWs. This work is crucial with regards to resolving the origins behind the poor external quantum efficiencies currently faced by high-Al content deep-UV light-emitting diodes (LEDs). Our work follows that by Weatherly *et al.* who showed that individual PDs in InGaN QWs can be resolved using CL imaging when the sample and measurement conditions are carefully chosen [1]. Through this technique, we estimate PDs on the order of 10^{17} cm^{-3} and aim to further investigate the trend in PD density with varying Al content. Furthermore, the time-resolved characteristics of individually resolved PDs will be studied through TRCL.

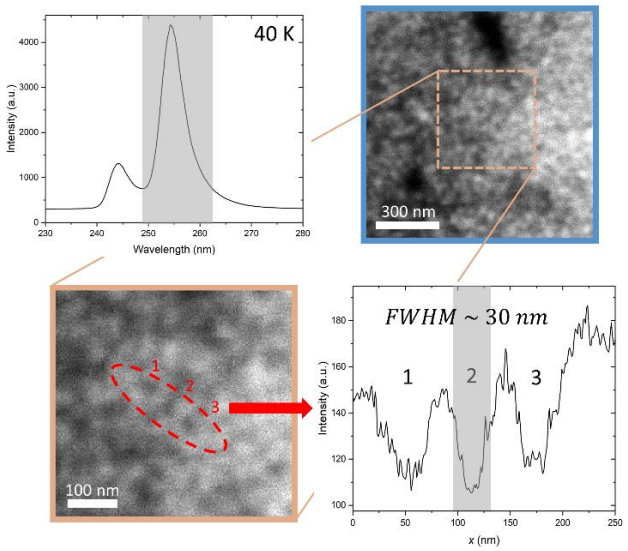

Figure 1. CL spectrum and images recorded from a 255 nm emitting AlGaN QW at 40 K. A line-scan taken across three PDs allows a lateral spatial-resolution of 30 nm to be estimated from the full-width half maximum.

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Photo-Field Emission from GaN Tips: new Virtual Source Cathodes for Ultrafast Electron Microscopy

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Ultrafast and time-resolved electron microscopy techniques rely on pulsed electron beams, typically generated by photoexciting conventional emitters such as Schottky- and cold- field emitters with ultrafast lasers. This approach is limited by cathode poisoning, low efficiency, and reduced brightness [1-2].

In this work, we propose and test a novel class of electron sources based on photo-assisted field emission from semiconductors. Our design is based on field concentration at the apex of a p-doped gallium nitride (GaN) micro-tip. The vacuum barrier reduction combined with the absence of free electrons in the conduction band enables emission only upon photo-excitation (Fig. 1a).

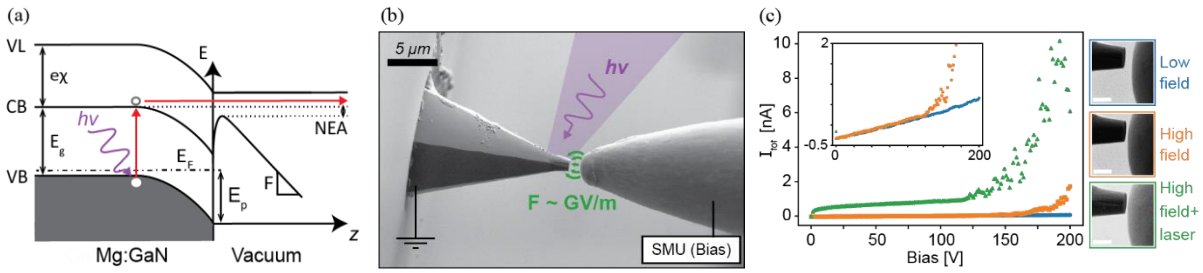

Figure 1. **(a)** Band structure of the device surface. Negative Electron Affinity (NEA) is achieved by exploiting Fermi level (E_F) pinning (E_P) and by the application of the field F. **(b)** The experimental setup reproducing a laser-driven electron gun inside a scanning electron microscope, with the GaN micro-tip on the left and the micromanipulator on the right. **(c)** Current emitted from the micro-tip as a function of distance to the micromanipulator, bias voltage and photoexcitation.

To validate our approach, we fabricate GaN tips with the geometry of Schottky field emitters – 40 µm tall and with micron-sized apex radii. Their performance is tested by recreating a laser-driven electron gun inside a scanning electron microscope (Fig. 1b). A biased micromanipulator is brought into proximity to the tip, inducing electric fields in the GV/m range, sufficient to enable field emission at room temperature (Fig. 1c). Focusing a laser beam into a 4-µm spot at tip apex lowers the threshold bias for current emission and dramatically increases current intensity. Emission current scales linearly with laser power, consistently with a tunneling mechanism from the conduction band to vacuum.

These results point toward a new class of ultrafast electron sources, combining the brightness of virtual sources with enhanced photoemission efficiency, room-temperature operation, and scalability to emitter arrays. This approach could enable stable, coherent, and tunable electron beams, opening new possibilities for time-resolved electron microscopy.

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Plasmonic Smith-Purcell metagratings generate polarization-tunable free-electron radiation

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Merging metasurface design concepts with free-electron radiation phenomena presents a powerful approach to shape the interaction between light, plasmons, and electrons. Grazing excitation geometries offer enhancement of electron-plasmon coupling via the coherent excitation of multiple emitters. One such example is the Smith-Purcell (SP) effect, whereby electrons grazing a periodic structure induce far-field light emission characterized by a discrete scattering pattern [1]. Previous work has experimentally demonstrated the spatial tunability of SP radiation via aperiodic metagrating design [2].

Here we propose a metagrating design for the generation of polarization-tunable SP radiation, shown in Figures 1(a-b). We first perform finite-difference-time-domain simulations, demonstrating control over the polarization state of SP emission via spatially-selective excitation of distinct plasmonic modes within elliptical gold nanocylinders. As shown in Figure 1(c), when the electron beam is placed above the centre of the structure, the electron excites the mode parallel to its trajectory, resulting in p-polarized emission; whereas when placed above the edge of the structure, the electron couples more strongly to the mode perpendicular to its trajectory, evidenced by the dominance of s-polarized emission.

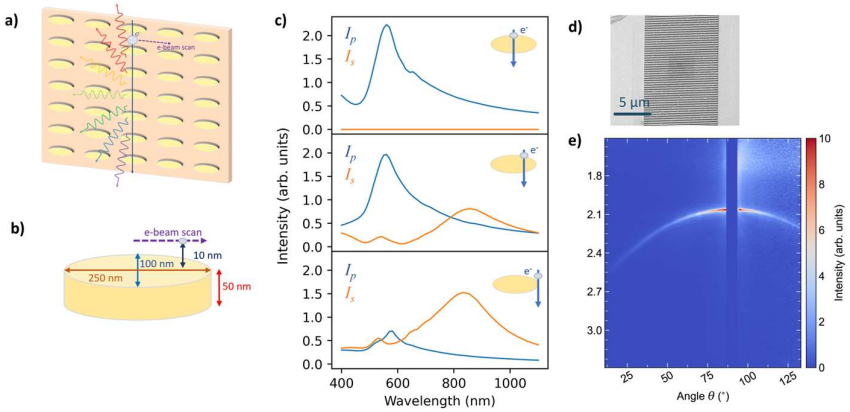

Figure 1. Schematic of (a) metasurface design and (b) nanocylinder geometry; (c) simulated far-field intensity contributions from p- (blue) and s- (orange) polarized light as the electron beam position is scanned above a single nanocylinder; (d) scanning electron microscopy (SEM) image of a gold on silicon SP grating with pitch = *298 nm*; (e) angular and spectrally resolved cathodoluminescence measurements showing the measured SP dispersion for the grating in (d) excited by a 30 keV electron.

We then fabricate these samples using electron beam physical vapour deposition and focused ion beam milling. Finally, we investigate the angular, spectral, and polarization properties of a number of SP metagratings using hyperspectral angle-resolved cathodoluminescence (HSAR-CL) spectroscopy with polarimetry imaging. Figure 1(d) shows the measured SP dispersion from a bar grating excited by a 30 keV electron. We present further HSAR-CL results for nanocylinder and nanohole arrays, demonstrating several opportunities to exploit the localized nature of electron-light-matter interactions to enable highly tunable free-electron-driven photon sources.

These results exemplify the broad potential of combining free-electron phenomena with nanophotonics: we suggest that future work could explore simultaneous control over polarization and functionalities such as lensed emission, and propose an optical-fibre integrated metasurface geometry to further exploit the unique interaction between grazing electrons and metasurfaces. Finally, we explore how the concepts examined here can be applied to the inverse process, whereby the quantum mechanical wavepacket of free-electrons is controlled via interactions with designed optical near-fields.

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Towards Dynamical Lattice Characterization of Native and Induced Defects in Monolayer WS_2 from CVD

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Chemical versatility of monolayers of Transition Metal Dichalcogenides such as WS_2 makes them highly attractive for a wide range of applications, including optoelectronics as well as hydrogen catalysis. However, the role of defects and their impact on the electronic, optical and structural properties of TMDs remains a topic of ongoing debate.
Firstly, in this study we combine High-Resolution Transmission Electron Microscopy (HRTEM) and High-Angle Annular Dark-Field Scanning Transmission Electron Microscopy (HAADF-STEM) techniques, coupled with Global Phase Analysis (GPA) to generate strain maps of WS_2 samples grown on various substrates using Chemical Vapor Deposition (CVD). These local atomic deformation maps are analyzed in relation to the presence, symmetry, and origin of native defects. Different substrates and transfer methods on TEM grids are compared. The results show the presence of both line defects (i.e. dislocations) [1] and point defects (i.e. single or cluster of tungsten vacancies) [2] in the grown crystals of monolayer WS_2. The pristine flakes are then compared to those subjected to ion bombardment, and the defects density and nature are evaluated in the two cases. [3] Secondly, we have performed benchmark ultrafast electron diffraction (UED) experiments on different nanomaterials to prove our ability to evaluate their structural dynamics on the sub-picosecond timescale. We are planning experiments on pristine and bombarded WS_2 flakes to investigate the role of the defects on the material lattice dynamics. Our results will be relevant for the ability to use such engineered defect network in WS_2 for demonstrating a proof-of-concept Quantum Reservoir Computing (QRC) approach. [4]

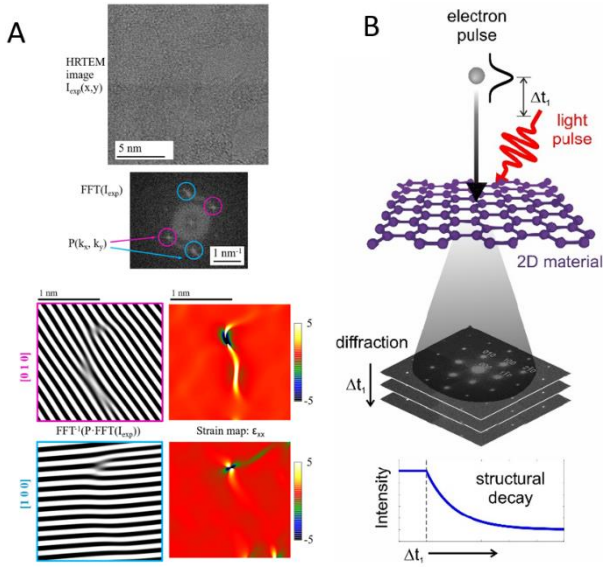

Figure. Investigation of line defects using HRTEM and strain mapping (panel A). Schematic diagram of the UED experiments investigating structural dynamics (panel B).

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An atlas of photonic and plasmonic materials for cathodoluminescence microscopy

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Cathodoluminescence (CL) microscopy has become a key technique for exploring light–matter interactions at the nanoscale. To support the growing use of CL in nanophotonics and materials science, we present an atlas of CL emission from a broad selection of photonic and plasmonic materials [1]. The dataset includes metals, semiconductors, dielectrics, two-dimensional materials, and fabrication-related compounds, studied under varying electron beam energies (Fig.1).

Through systematic CL measurements, we resolve both spectral and angular emission characteristics, highlighting coherent and incoherent light emission processes. In parallel, Monte Carlo simulations are used to model electron trajectories, penetration depth, and energy deposition in each material, offering essential context for interpreting the measured signals.

This atlas serves as a practical reference for researchers designing electron-beam-based experiments and devices. It provides a foundation for material selection, supports the identification of emission mechanisms, and helps avoid common misinterpretations of CL data in complex systems.

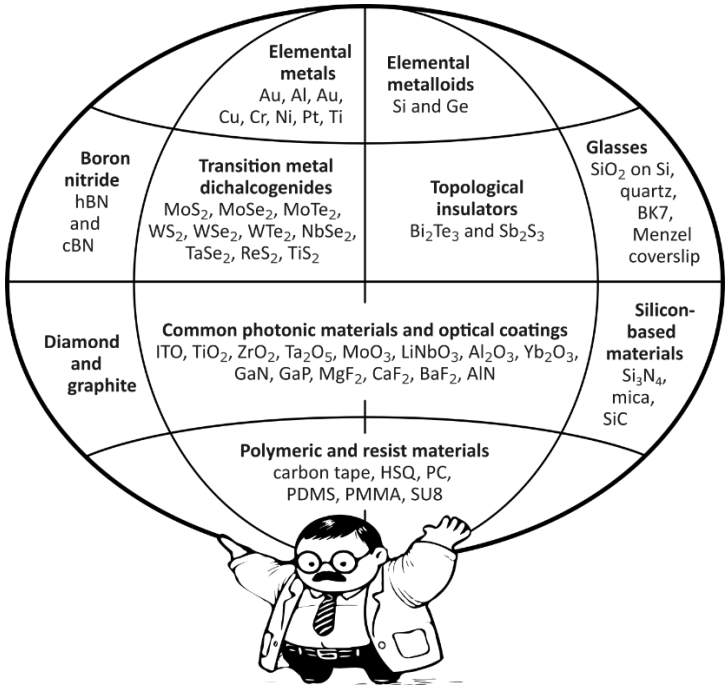

Figure 1. An atlas of investigated photonic and plasmonic materials.

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Relativistic Ultrafast Electron Diffraction and Imaging (RUEDI) National Facility for five scientific themes in the UK

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While ultrafast electron diffraction and imaging has been developed over the past two decades, its advancement has been often limited by the stroboscopic method, the thickness of a specimen, its environment, its external stimuli, and the time resolution. This makes it difficult to study ultrafast dynamic in biology, chemistry, and that of materials under strong excitations, because those dynamics are often irreversible, in liquid phase and spatially not homogenous in addition to μm thick specimens.

To overcome these difficulties, both stroboscopic and single-shot method have been developed using bright pulsed MeV electrons over the past 20 years. Whileit is limited to diffraction, RUEDI aims to integrate these and expand the range of applications by adding unique MeV imaging for the next 20 years [1].

The facility will provide MeV electrons in two different beamlines [2]. The electron diffraction beamline will reach the shortest possible timescales down to 10 femtoseconds resolution, while the imaging line will enable time-resolved single-shot electron microscopy at nanometre-scale resolution. The diffraction line will be based on a radiofrequency photocathode, combined with a magnetic beamline for bunch compression. The imaging line will be based on a DC electron source, combined with a fast chopper. State-of-art sample environments such as gases, liquids and cryogenic temperatures will be made available. RUEDI will be equipped with rich external stimuli such as lasers, high-frequency electromagnetic fields, and ions, to induce a variety of dynamics.

RUEDI is driven by the five scientific themes [3]: (i) Dynamics of chemical change, (ii) Energy generation, conversion & storage, (iii) In vivo bioscience, (iv) Materials in extreme conditions and (v) Quantum materials & Processes with a cross-cutting theme of Artificial Intelligence/Data Science underpinning all activities. RUEDI will provide a unique gateway to new applications and new phenomena. I will present the current status of RUEDI and discuss the new research areas it opens up.

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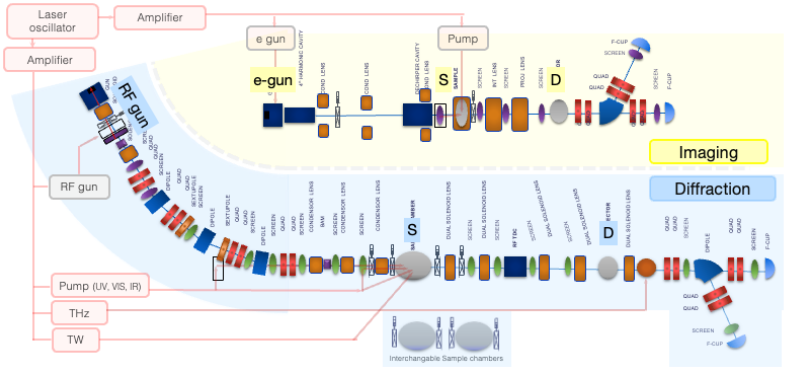

Imaging of Degenerate and Hybridized Plasmonic Modes by Cathodoluminescence Spectroscopy

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Metal nanoparticles host localized plasmon excitations that enable the manipulation of optical fields at the nanoscale. Detailed knowledge of the electromagnetic field distribution associated with surface plasmons (SP) at nanometer spatial resolution is of fundamental importance for a wide range of applications. Here, we present a theoretical and experimental investigation of plasmonic-state symmetries in individual gold nanoprisms and coupled nanoprism pairs arranged in a bow-tie antenna configuration using cathodoluminescence (CL) spectroscopy. Specifically, we use a focused 80 keV electron beam to excite multiple localized SPs over a broad spectral range. The induced optical radiation emitted from the structures is detected and imaged through angle-, polarization-, and space-resolved CL measurements (Fig. 1a). This combined technique grants access to both spectral information (Fig. 1b) and spatially resolved maps (Fig. 1c) of multipolar SP modes localized on the nanoprisms with nanometer spatial resolution. Remarkably, we reveal the spatial distribution of energetically degenerate SP states in a single nanoprism and hybridized SP states in a coupled nanoprism pair [1]. We demonstrate that both degeneracy and hybridization can be unveiled through examination of the symmetry in the s- and p-polarized photon emission maps. Our approach enables a systematic study of plasmonic states in high-symmetry nanostructures. Finally, we analyse our findings by comparing full-wave electromagnetic simulations with experimental results.

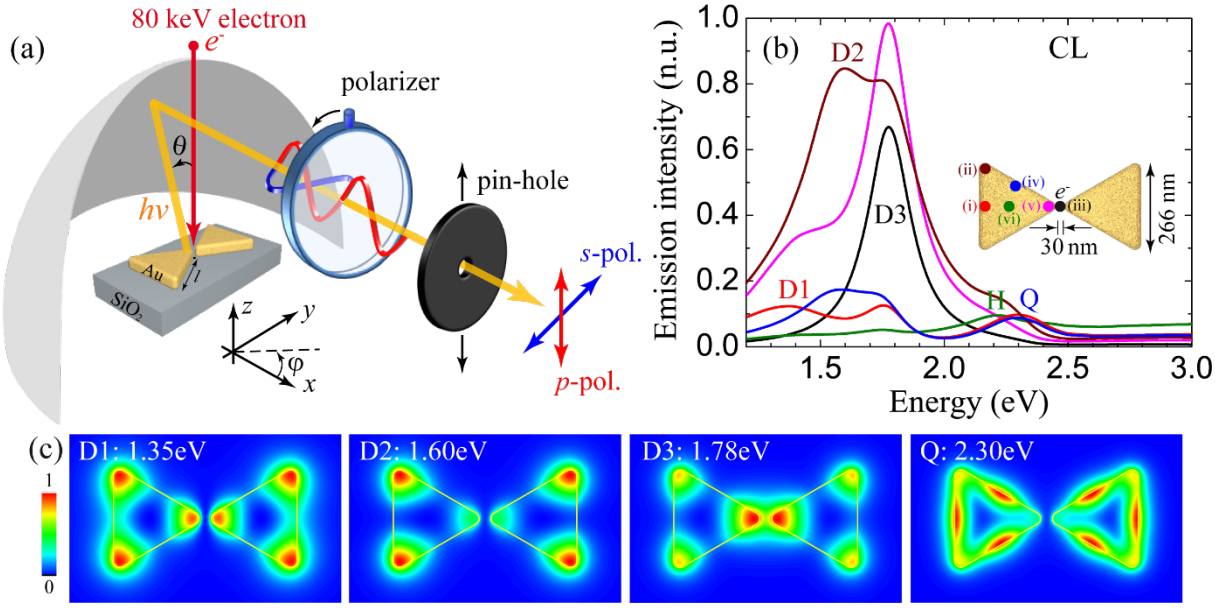

Figure 1. (a) Schematic representation of our angle- and polarization-resolved CL setup. (b) Calculated photon-emission intensity spectra collected at different electron-beam probe positions, as shown in the inset. (c) Photon-emission probability maps of the modes, marked by colored letters in (b), as a function of electron probe position.

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Characterisation of a Cathodoluminescence Setup in an Ultrafast Transmission Electron Microscope

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Advances in nanotechnology have enabled the creation of optical nanostructures with tailored optical properties that can be probed by electron beam spectroscopy [1], facilitating applications in sensing and optoelectronics [2]. The collection of light generated by an electron beam, i.e. cathodoluminescence (CL) radiation, in particular allows for optical mode mapping on the nanoscale with high spectral resolution, enabling the investigation of plasmonic particles and semiconductors [3]. With the advent of temporally resolved CL measurements [4], studies of excitation and emission pathways [5], and correlation-enhanced imaging have become possible [6]. These advanced methods require highly specialised instruments, and thus, a thorough understanding of their capabilities and limits is essential.

Here, we present a detailed study of our CL measurement system implemented in the Göttingen ultrafast transmission electron microscope. We combine a commercial off-axis parabolic mirror with our custom-built detection setup, and employ this to investigate the plasmonic modes of aluminium discs and nanorods of different size in detail. Scanning the highly focused electron beam over an area of interest, the emitted cathodoluminescence is collected by the aligned parabolic mirror. The collected collimated light is coupled to a single-photon avalanche detector to map the intensity for example of a plasmonic mode or coupled into a grating spectrometer to spectrally analyse the generated light. Additionally, the far-field mode profile of the light reflected by the parabolic mirror is monitored. The collection area of the mirror is determined by mapping the beam position-dependent photon count rate on a homogeneous sample. Combining this with an electron current measurement, the conversion and collection efficiencies are evaluated. Furthermore, we characterise polarisation-resolving capabilities by implementing a polarimetry setup including a quarter- and a half-waveplate, a polarizing beam splitter and two single-photon detectors, allowing to measure non-polarized and polarized emission from plasmonic nanostructures.

In summary, we present the implementation and characterisation of a CL setup capable of spectral and polarisation resolved mode mapping by the example of plasmonic nanodiscs and rods. In the future, this setup can be combined with the ultrafast capabilities of our TEM for time-resolved CL-measurements. This includes intensity auto-correlations and electron-photon correlation measurements with applications in enhanced imaging, lifetime analysis and free electron quantum optics. The potential for exploiting electron-photon entanglement for ghost imaging is particularly promising, offering a new paradigm for imaging and sensing applications [7, 8].

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Electrical design of deterministic 2D/1D heterostructures via electron beam induced current mapping

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The integration of 2D semiconductors on nanostructures enables a new generation of dimensionally hybrid systems (Figure 1a) with a great promise for functional and quantum applications [1,2]. At the same time, the community is looking for high-resolution methodologies able to define the functionality of complex nanoscale elements [3].

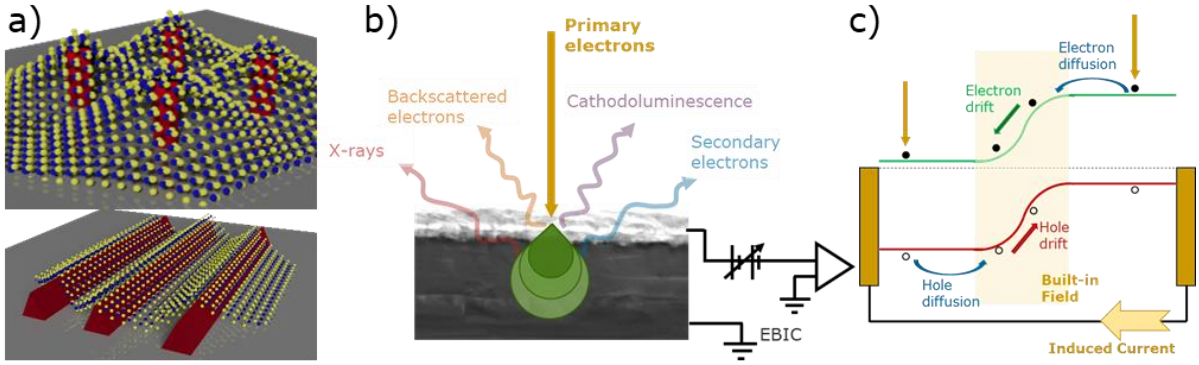

Figure 1. a) Schematic of 2D/1D hybrid heterostructures. b) Interaction volume arising from inelastic scattering of primary electrons and resulting outputs. c) Working principle of electron beam-induced current mapping.

In this work we use electron beam-induced current microscopy (EBIC) to map WSe2/GaAs hybrid heterostructures. GaAs crystals are grown epitaxially on (100) Zn:GaAs substrates in form of planar epi-layers and in-plane 1D nanostructures via metalorganic vapour phase selective area epitaxy. WSe_2 flakes containing terraces with thickness ranging from 2-4L to ~50L are integrated on the epitaxial structures via dry transfer. EBIC maps of the planar heterostructures show a built-in field in selected portions of the heterointerface thus revealing the electron-rich nature of our exfoliated WSe_2. Correlated analyses with atomic force microscopy maps evidence a non-trivial dependence of the carrier collection with the flake thickness. By integrating WSe_2 onto in-plane 1D GaAs nanostructures, we demonstrate lateral confinement of the electric field in deterministically positioned regions as narrow as 200 nm. Modeling clarifies details about carrier drift and diffusion. Epitaxial growth of GaAs nano-networks with complex in-plane geometry allows to achieve a quasi-0D field confinement at the hetero-interface. These results pave the way to engineer nanowire-based heterojunctions and prove the key role of high-resolution mapping in the design of these nanocomponents.

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Excitation and Control of Polaritons Using Periodical Arrays

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Given the growing interest gathered around the control and application of free electrons in the context of high-precision ultrafast electron microscopy, we study the interaction of electron beams with polariton-supporting structures. Due to the wavelength mismatch between photons and polaritons, achieving efficient light coupling to highly confined polaritons through optical methods alone is challenging. This makes the combination of electrons and optical scatterers a promising strategy for generating such polaritons. We analyze the polariton Smith-Purcell emission, which arises from the constructive interference originating from the out-coupling of the evanescent field carried by the electron as it passes near contiguous particles arranged in a linear array aligned with the electron velocity. We investigate the extension of the Smith-Purcell effect to generate surface modes such as plasmons in graphene and thin metal films, as well as phonon polaritons in hexagonal boron nitride (hBN).

We start by addressing a simple system composed of an electron beam passing with a velocity $\boldsymbol{v}$ in the vicinity of a single dipolar point particle, which is placed at a distance z_0 from a polariton-supporting surface. The incoming electron beam passes at a certain distance b from the scatterer, whose optical response is described by its electric polarizability. Under external illumination by the passing electron, a self-consistent dipole moment $\mathbf{p}$ is induced at the point particle, allowing it to radiate and excite the polariton modes of the nearby film. Such field is provided directly by the electron and also contributed by the reflection of the electron and dipolar fields acting back on the scatterer in a self-consistent fashion. We quantify the polariton excitation efficiency by finding an analytical expression for the dipole decay rate of energy into polaritons by describing the surface response in the polariton-pole approximation [1,2]. This quantity is maximized for a specific scatterer-surface optimal separation distance.

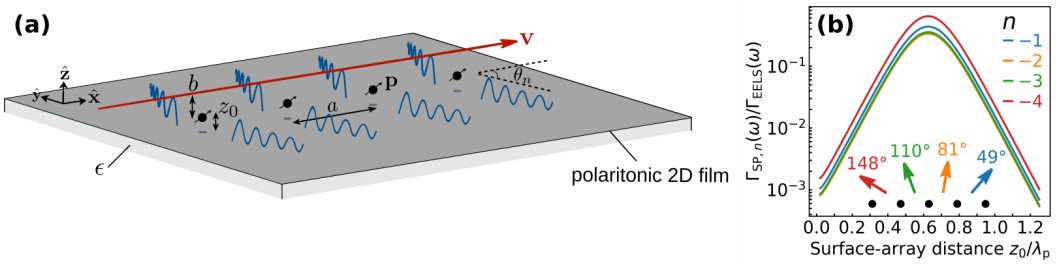

Fig. 1. Polariton Smith-Purcell effect. **(a)** Sketch of an electron moving parallel to a linear array of scatterers placed close to a planar surface. We show several relevant geometrical parameters. **(b)** Probability of surface polariton emission for different Smith-Purcell orders n per particle and divided by the self-standing-particle EELS probability as a function of the scattered-surface distance z_0 normalised to the polariton wavelength λ□, for a fixed frequency ω=0.4 ω_D, where ω_D is the weight of the Drude conductivity of the 2D film, and for a fixed lattice period a=2λ□. The electron velocity is v = 2.2 $v_0 \approx$ 0.2 $c/(\epsilon + 1)$, where c is the speed of light and ϵ is the permittivity of the polaritonic film. The impact parameter b is determined by $k_D\, b$ = 0.1, with $k_D = (\epsilon + 1)\hbar\omega_D/4\pi e^2$. The corresponding emission of the Smith-Purcell angles θ□ are indicated by arrows.

To observe the polariton Smith-Purcell emission, we perform a similar analysis for the alternative case where the single dipole is replaced by an infinite periodic array of well-separated and non-interacting dipoles (see Fig 1**(a)**). This results in a polariton emission pattern that resembles the optical Smith-Purcell effect observed when an electron passes nearby a periodic structure [3]. Depending on the velocity of the electron, the polaritons are preferably emitted along an angle θ□ following the relation $\cos\theta$□ =λ□$(\omega/(2\pi v) + n/a)$, where n represents the diffraction order, a is the lattice period and ω is the polariton frequency. The dependence of the polariton emission probability associated with different orders n on the scatterer-surface separation is illustrated in Fig. 1**(b)**. This result suggests that an adequate array-surface separation can yield polariton emission preferably along a specific angle, revealing an interesting tunability of polaritonic 2D films that has not been observed before.

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Theory of Spin Resonance Spectroscopy in an Electron Microscope

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Coherent spin resonance methods, such as nuclear magnetic resonance and electron spin resonance spectroscopy, have led to spectrally highly sensitive, non-invasive quantum imaging techniques. In this talk, I will present joint theoretical work with my collaborators* on a pump-probe spin resonance spectroscopy approach, designed for electron microscopy, based on microwave pump fields and electron probes [1]. Starting from the description of the coupling of free-electrons and localized sample spins, I will introduce the general method and discuss the size of the effect, in particular, the phase shift on the electron wave function and the corresponding deflection angle. I will also present a framework for the simulation of spin resonance spectroscopy in Transmission Electron Microscopy (TEM) that we have recently developed [2] and discuss our results on the quantum metrological precision limits associated with the technique and the optimal measurement strategy [3]. Notably, state-of-the-art TEM provides the means to detect signals almost as small as that due to a single electron spin in principle. This could enable state-selective observation of spin dynamics on the nanoscale.

*Collaborators: S. Beltrán-Romero (TU Wien), M. Gaida (University of Siegen), P. Haslinger (TU Wien), S. Löffler (TU Wien), S. Nimmrichter (University of Siegen)

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Relativistic electron energy-loss spectroscopy in cylindrical waveguides and holes

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Swift electrons interacting with metallic and dielectric structures provide a powerful probe of confined optical excitations such as plasmons and guided modes. In this work, we present a general analytical framework for modeling electron energy-loss spectroscopy (EELS) in systems with cylindrical symmetry, including nanowires, nanoholes, and optical fibers, and accounting for both parallel and perpendicular electron trajectories (see Figure). While previous EELS theories have primarily focused on electrons moving along the axis of symmetry (a,b), we derive, for the first time, closed-form solutions for perpendicular electron trajectories (c-e), incorporating full electrodynamic retardation effects.

These analytical results are validated against numerical simulations using a frequency-domain boundary-element method (BEM) and a discontinuous-Galerkin time-domain (DGTD) finite-element method. This combined approach allows us to systematically explore both infinite (a-e) and finite-length geometries (f), highlighting termination effects and mode confinement in practical structures. We apply our methods to both metallic (a,b) and dielectric systems (c-e), revealing distinct features such as surface plasmon polaritons and guided photonic modes, respectively.

Our compact analytical expressions not only provide physical insight into the electron–photon interaction in cylindrical systems but also offer a robust tool for interpreting EELS measurements and guiding the design of free-electron–photonic hybrid platforms [1]. Extension of the theory to cathodoluminescence is straightforward, broadening its applicability to a range of spectroscopic and nanophotonic investigations.

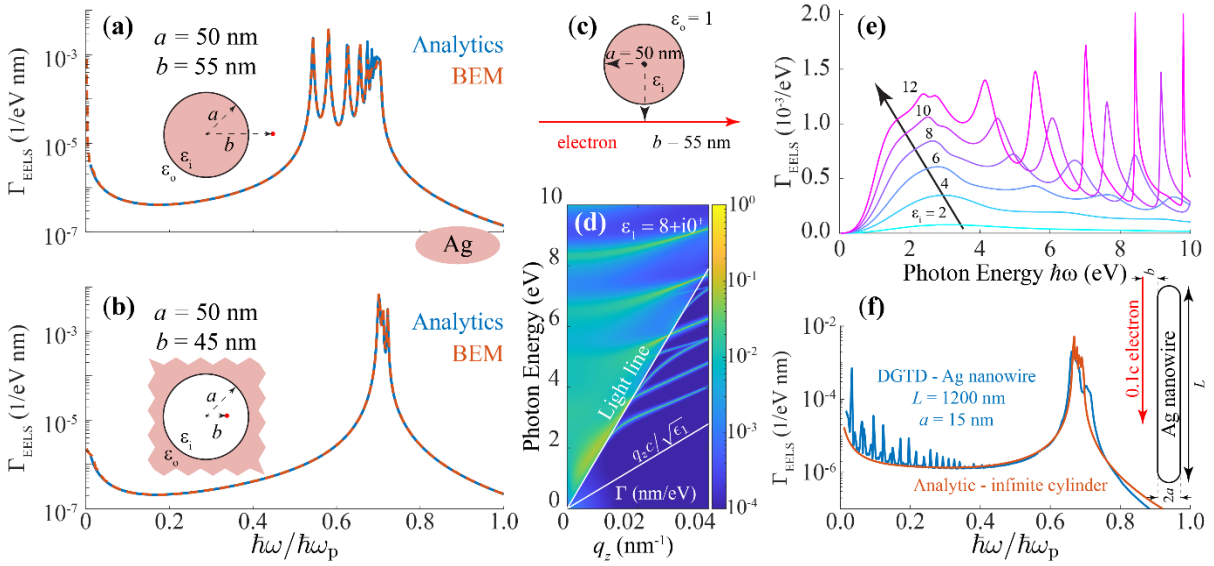

Figure: (a,b) EELS probability for an electron moving parallel to the axis of (a) a silver cylinder and (b) a hole in silver. The electron moves at a distance $|b - a| = 5$ nm from the surface. The probability is calculated by using the corresponding analytical formulas (blue curves) or the numerical BEM (red curves). The metal is modelled by a Drude permittivity with $\hbar\omega_p = 9.17$ eV and $\hbar\gamma = 21$ meV. The electron energy is 100 keV. (c) Illustration of a cylinder with a radius of 50 nm and permittivity ϵ_i, excited by an electron passing 5 nm away from the surface with a kinetic energy of 100 keV. (d) Momentum-resolved EELS probability for $\epsilon_i = 8 + 0.001$i, where a small imaginary part is introduced in the permittivity to help visualize the coupling to guided modes. (e) Momentum-integrated EELS probability under the same conditions as in panel (c) but for several values of the permittivity. (f) EELS probability for a long Drude-like silver nanowire (blue curve) of length L = 1.2 μm and radius a = 15 nm compared to the analytical solution for an infinite cylinder (red curve).

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Coherent and incoherent cathodoluminescence photon statistics

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Recent CL studies using Hanbury-Brown=Twiss interferometry (as Fig.1a) uncovered unexpectedly strong photon bunching behavior in the $g^{(2)}(\tau)$ measurement [1], meaning that CL photons excited by CW electron beams show apparent super-Poissonian statistics. This CL photon bunching can be understood through the mixture with zero photon states introduced by random electron excitation with some intervals (Fig.1c). While such bunching behavior is useful to measure the emission lifetime, it blurs the "true" photon statistics by free electron excitation.

We here extract the photon statistics within the single electron excitation event by comparing the "bunching" and the flat background parts in the $g^{(2)}(\tau)$ curves (Fig.1b) [2]. This allows calculating a parameter corresponding to $g^{(2)}(0)$ in the single electron excitation, which we call the *correlation factor* κ_{corr}. In coherent CL processes, where light is generated through a direct electromagnetic interaction (Fig.1d), κ_{corr} is found to be ~1 in various coherent CL systems. This confirms that coherent CL obeys Poisson statistics, in agreement with the coherent states. However, in incoherent CL, which involves multiple mediator particle generations (like bulk plasmons or secondary electrons, Fig.1d), κ_{corr} exceeds 1, meaning that incoherent CL is super-Poissonian ("bunching") even within the single excitation event.

Through a model analysis with multiple (cascade) particle generation steps, we were able to reproduce this super-Poissonian behavior in incoherent CL even when each particle generation process follows Poissonian statistics. This approach not only offers a refined understanding of CL mechanisms but can also be extended to the analysis of internal particle generation processes in general.

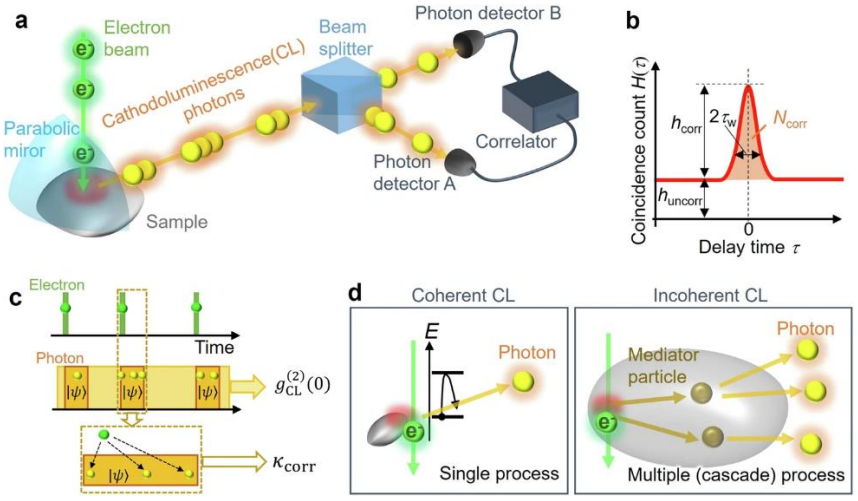

Figure 1. a Schematic illustration of the CL photon statistics measurement and CL processes.[2]

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Nanolaser Optical and Structural Properties at the Nanometre Scale

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Since their first demonstration by Huang et al in 2001 [1], semiconductor nanolasers have attracted a lot of interest especially for their application in optoelectronic devices. They have the advantages to be cost-effective, easy to fabricate and of a micron size. Since 2001, various semiconductors and geometries demonstrated lasing properties, for example ZnO [1] or GaN [2] nanowires.

Lasing can be induced by optical pumping, usually it is characterized by a drastic reduction of the emission spectrum width and an increase of the emitted light coherence [3]. The key lasing properties of nanolasers include the emission wavelength, the value of the lasing threshold and the carrier lifetime. The optical modes of the nanolasers observed above the lasing threshold are directly related to the dimensions of the nanolaser, since the cavity is formed by the nanowire itself. Thus, the shape of the nanowire has a direct impact on all laser characteristics. Given that the structural parameters of the nanowires vary at a nanometric scale, electron microscopy is well-suited for their analysis. Moreover, the lasing properties can be directly linked with the nanolaser shape provided that the electron microscope is equipped with a photoluminescence setup.

We aim to probe the near field of optically pumped nanolasers within their lasing regime with a focused electron beam in Ultrafast - STEM. When the electrons interact with the field created by the laser light, electrons absorb or emit quanta of the field energy. By mapping the interactions of the field with the electron beam, Photon-Induced Near-field Electron Microscopy (PINEM) enables us to measure the transversal field intensity [4] and the lasing regime dynamics with a sub-picosecond resolution.

In this presentation, we will discuss how we can study the lasing characteristics of GaN nanolasers at the nanoscale scale within a UTEM using PINEM and photoluminescence.

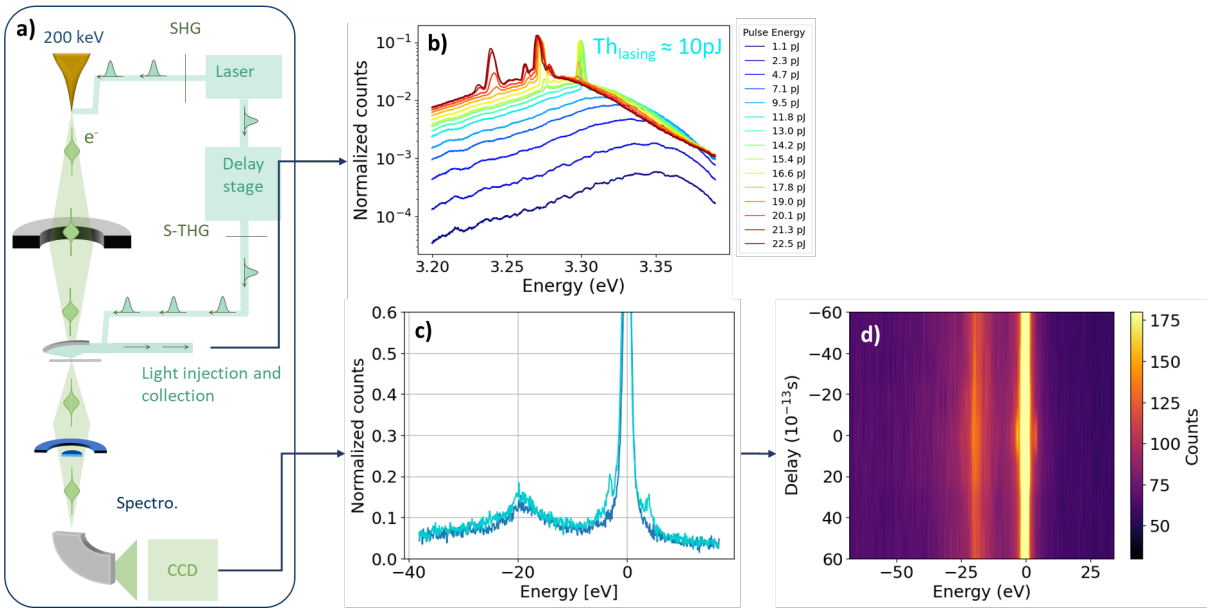

Figure 1. Use the format (a), (b), etc. for panel labels. (a) UTEM set-up, (b) Nanolaser photoluminescence spectrum depending on the pumping power, (c) PINEM spectrum on the GaN direct scattering, (d) PINEM delay scan

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Near-field effects on cathodoluminescence outcoupling in perovskite thin films

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We present spatially-resolved cathodoluminescence (CL) spectroscopy on polycrystalline $CsPbBr_3$ perovskite films to study their optoelectronic properties at subwavelength scale. The CL maps show spectra peaking at the perovskite bandgap of 2.36 eV which is constant across the film. The bandgap CL intensity is strongly reduced for electron beam positions near the polycrystalline grain boundaries. We find a strong correlation of these intensity variations with the surface morphology measured with atomic force microscopy (AFM). Numerical finite-difference time-domain simulations of the light fields generated inside the perovskite film show reduced outcoupling of the subwavelength near fields near the grain boundaries, as a result of enhanced internal reflection. Comparing the measured CL maps with the simulations we conclude that the measured CL intensity variations are largely explained by these near-field outcoupling variations as shown in Figure 1. We also observe lateral CL intensity variations within individual grains. We assign these to variations in the interference of light reflected at the film top surface and substrate interface as the film thickness inside each grain varies. The strong near-field coupling and interference effects observed here are relevant for the analysis of cathodoluminescence and photoluminescence imaging analysis of corrugated films of many different kinds.

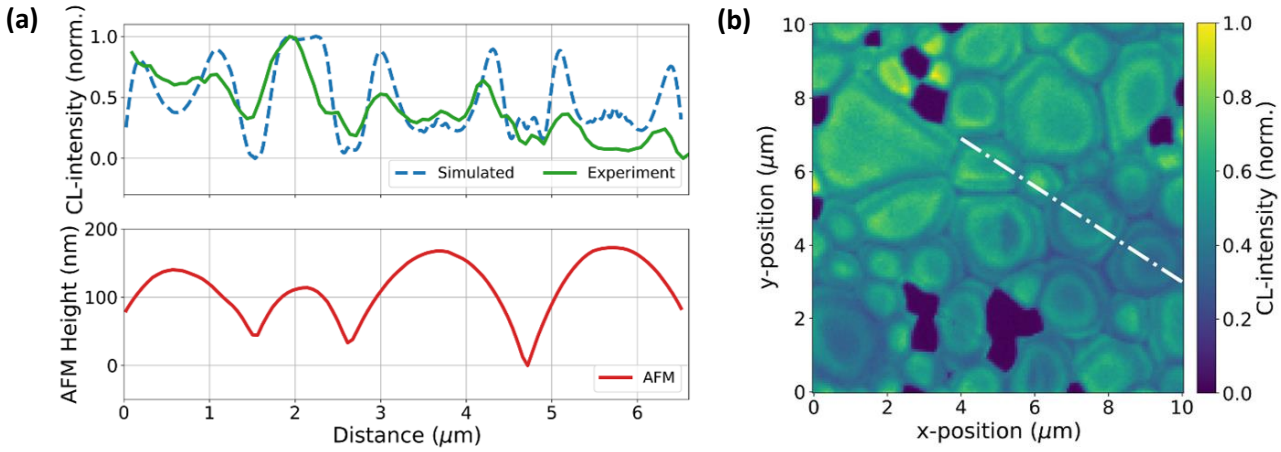

Figure 1. (a) Experimental AFM (red) and CL (green) line traces at λ =525 nm (bandwidth 2 nm; E = 2.36 eV) across the perovskite film at 2 keV. Simulated CL line traces using the AFM line trace as input are shown as blue dashed lines. (b) Corresponding CL map with white dashed line indicating the experimental line scan shown in (a).

Acknowledgments

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Three-Dimensional Imaging of the Modal Structure in Dielectric Cavities

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Recent research aims for designing dielectric optical cavities that confine light with high efficiency at sub-wavelength scales, enabling stronger light-matter interactions. These advances hold great promise for transformative applications in optoelectronics, quantum technologies, and quantum electron microscopy [1]. However, realizing such structures demands characterization techniques capable of high-resolution, three-dimensional imaging – capabilities that remain limited with current technologies.

We demonstrate the first use of electron energy loss spectroscopy (EELS) for tomographic reconstruction of a tailored optical mode with sub-wavelength confinement and a resonance wavelength of 1550 nm in a topology-optimized dielectric photonic structure (see Figure 1) [2]. The experimental results closely match theoretical predictions, confirming the presence of the designed mode. Moreover, we uncover additional modes with distinct profiles that significantly influence the local EEL spectra.

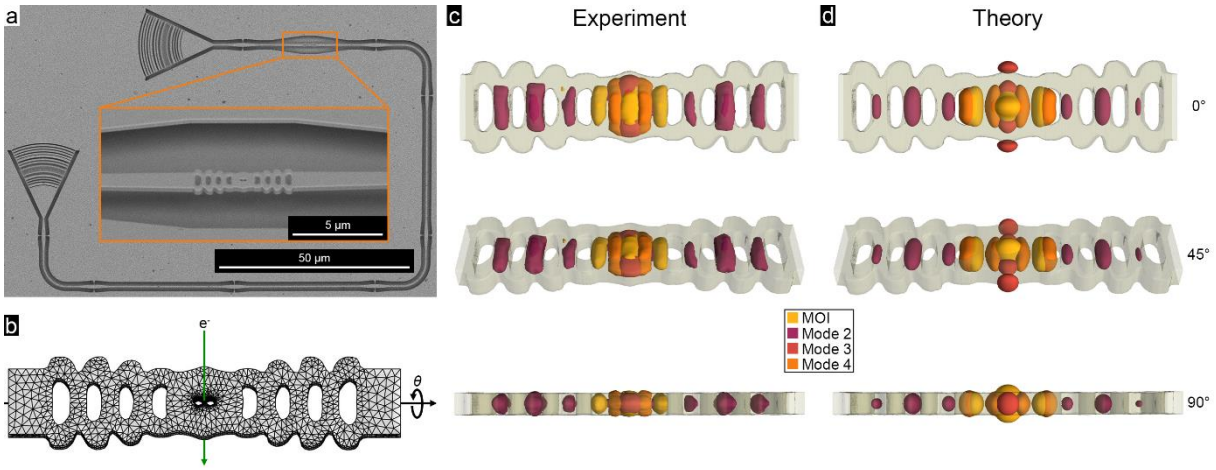

Figure 1. (a) Scanning electron microscopy (SEM) image of the fabricated waveguide-coupled optical cavity. (b) Schematic showing the experimental approach. (c) Isosurface renderings at different sample tilt angles obtained from tomographic reconstructions of the mode of interest (MOI) and additional modes (Modes 2-4) match well with those of the theoretically calculated mode profiles presented in (d).

These findings demonstrate the potential of electron microscopy to probe sub-wavelength field confinement in dielectric systems, offering new insights into light-matter interactions and enabling their precise control. The integrated waveguide structure provides a pathway for coupling light into the system, facilitating electron energy gain spectroscopy, which could enhance signal strength and deepen the understanding of the underlying phenomena [3].

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Acknowledgments

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4D Near-Field Electron Tomography

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New techniques for imaging electromagnetic near-fields have great importance in life science, material science, and nanotechnology [1]. Prominently, ultrafast imaging in electron microscopy has seen a surge of interest with the development of photon-induced near-field electron microscopy (PINEM) [2]. This capability and its generalizations [3,4] provide unprecedented spatio-temporal resolutions, applicable to broad regimes of the electromagnetic spectrum, from THz [3] to optical [2] frequencies. However, all works so far only provided a 2D projection of the near-field, limited by the direction of motion of the electron passing through the sample.

Drawing inspiration from the field of computed tomography (CT), we present a novel technique for 3+1D reconstruction of the near-field profiles, achieved by collecting time-resolved field measurements from multiple angles (Fig. 1a). Unlike reconstructed objects in conventional CT, near-fields oscillate rapidly and change during the electron transmission time (especially in THz frequencies and above). Consequently, each electron interacts with multiple time-frames of the near-field, mixing them together. We develop a mathematical analysis method that disentangles the mixed projections and separates the time frames, denoted as 4D Near-field Electron Tomography (4D-NET). Our technique reconstructs the full space and time (3+1D) information of the near-field, including its vector nature. We apply the technique on a simulated THz field originating from a mix of photo-Dember [5] and optical rectification [6] effects created by a laser pulse impinging an InAs crystal, achieving a good electron tomographic reconstruction, distinguishing the contributions of the mixed nonlinearities (Figs. 1b,c).

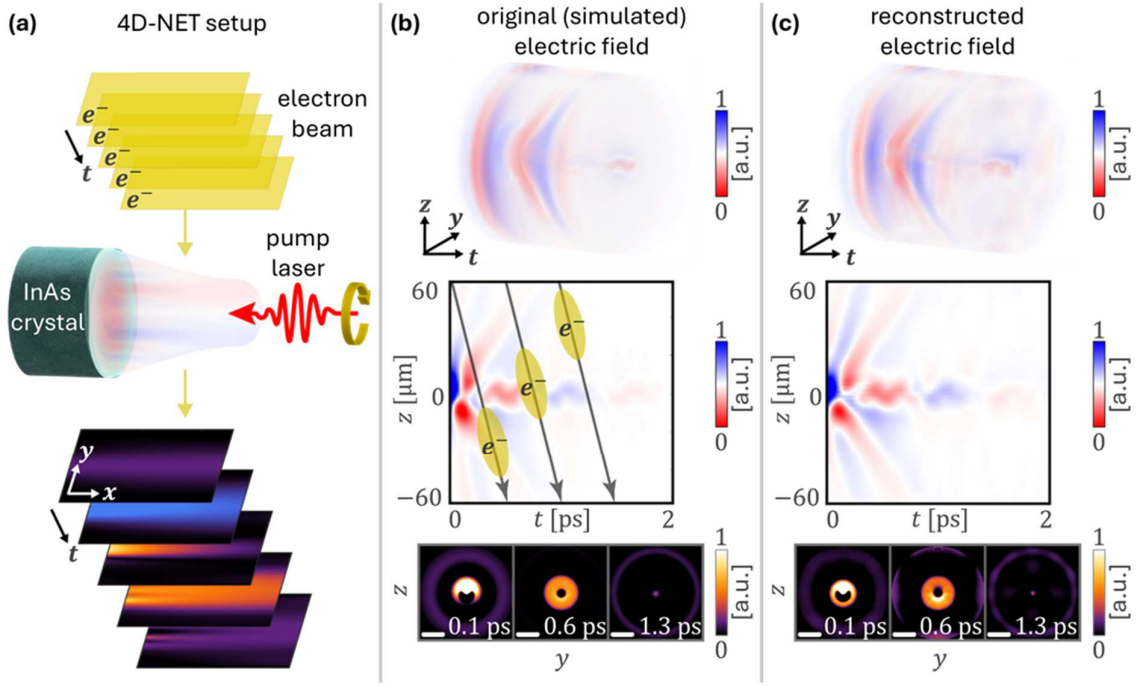

Fig. 1. 4D Near-field electron tomography (4D-NET) of THz near-fields. (a) 4D-NET applied for simulated THz field generated via photo-Dember [5] and optical rectification [6] effects driven by a laser pulse (10 μm diameter, 200 fs duration, 800 nm central wavelength, 10nJ power) impinging an InAs crystal. The measurements are simulated for 200 keV electrons (velocity of 0.7c), as used previously for imaging of THz near-fields [3]. The impinging laser polarization is rotated around its axis, collecting a time series of electron projections of the respective polarizations. **(b, c)** Original (simulated) and reconstructed electric field for distance of x=1 μm from the surface, showing cross-sections of 2D+time view (top, E_z), 1D+time view (center, E_z) and 2D view for different times (bottom, $\sqrt{E_z^2 + E_y^2}$). An illustration of electrons paths is drawn over the 1D+time view of the original field. The reconstruction recreates most of the spatio-temporal features of the electric field. The remaining discrepancies are due to k-ω THz field components that do not interact with the electron. The white bars denote 50 μm.

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Strong Coupling in Coated Silver Nanowires Probed by Fast Electrons

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Composite nanostructures combining components that support confined electromagnetic modes with electronic transitions under certain conditions can enter the strong coupling regime, manifested by the emergence of hybrid light-matter states. The unique optical properties of these polaritonic states are promising for a wide range of applications, including polaritonic chemistry, polaritonic lasing, and Bose-Einstein condensation [1].

Achieving a large coupling strength requires two key ingredients: a nanocavity that sustains tightly confined modes and an electronic transition associated with a strong dipole moment. Plasmonic nanocavities embedded in excitonic materials comprising organic molecules, such as metalorganic frameworks (MOF), are ideal platforms for strong coupling [2]. In that regard, fast electron beams are excellent probes to study strong light-matter interaction, as they can excite tightly confined modes with high quality-factors that are otherwise inaccessible with conventional light-based methods [3].

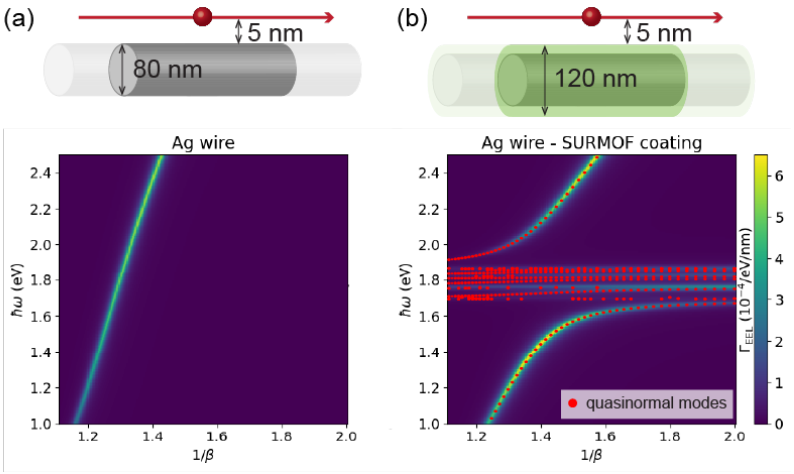

Figure 1. Electron energy loss (EEL) probability versus energy loss and inverse reduced velocity of the electron beam for an electron traveling parallel to the axis of (a) a silver nanowire, and (b) a silver nanowire coated with a concentric layer of SURMOF molecules. The red dots trace the quasinormal modes of the coupled system.

In this work, we study theoretically the strong coupling emerging in an open nanocavity formed by a cylindrical silver (Ag) nanowire coated with a concentric layer of surface MOF (SURMOF) molecules, as illustrated in Figure 1. We probe the coupling with fast electrons in electron energy-loss (EEL) spectroscopy simulations and show that the dispersion relation of the system can be reproduced by varying the velocity of the electron beam, exposing a large anticrossing of the polaritonic branches. The large coupling strength is further confirmed by quasinormal mode analysis.

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Quantum Optical Electron Pulse Shaper

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Ultrafast electron microscopy has been developed with the aim of combining the exceptional spatial resolution of electron microscopes with the time resolution available thanks to pulsed electron sources. While laser-driven photoemission enables few-femtosecond electron pulses, dispersion during acceleration and propagation limits pulse durations to several hundred femtoseconds without additional compression. Techniques involving chirp manipulation have achieved ultrashort electron pulse durations [1–4], but simultaneous control over temporal and spectral resolution remains a challenge.

In this contribution we theoretically describe a technique enabling near arbitrary light-based shaping for electron wave packets. The method is based on the quantum coherent modulation of the electron wave function envelope with optical fields with temporally varying intensity and frequency. The technique can be used to induce a tailored dependence of the electron energy on time, including the generation of one or multiple negatively chirped sub-pulses that compress during free-space propagation. Our method opens new possibilities for precision control in ultrafast electron-based experiments.

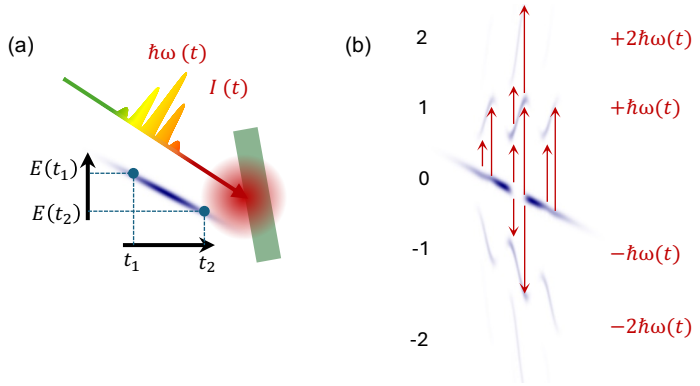

Figure 1.

(a) The electron pulse traverses a dielectric membrane e.g., illuminated by an optical pulse with modulated instantaneous photon energy $\hbar\omega(t)$and intensity envelope $I(t)$. (b) The absorptions and emission of photon quanta result in the generation of tailored-shape energy sidebands, where $\hbar\omega(t)$ determined the instantaneous energy separation between the and $I(t)$ determines the instantaneous band population.

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Acknowledgments

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Ponderomotive microbunching of a relativistic electron beam

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Mircobunched electron beams may improve the brilliance of inverse Compton scattering sources by orders of magnitude via superradiance. However creating the required microbunching using a relativistic electron beam, with energies in the 10s of MeVs, is a feat that has yet to be accomplished.

A new method, termed ponderomotive bunching, has recently been studied [1,2]. Here the electron interacts with two laser pulses, of different wavelength, simultaneously. When the interaction angles are tuned correctly, an optical beatwave is formed that co-propagates with the electron beam. The resulting ponderomotive force imposes an energy modulation which, after a short drift, transforms into a density modulation.

We present progress on a proof of principle experiment to demonstrate ponderomotive bunching using the Smart*Light beamline. A 20 MeV electron beam interacts with infrared and ultraviolet laser pulses under shallow co-propagating angles. After a 20 cm drift microbunching is formed, which is measured via the coherent amplification of transition radiation.

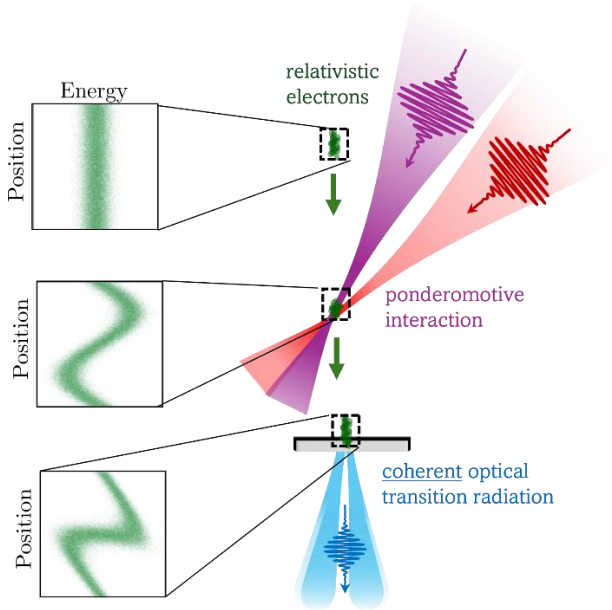

Figure 1 Illustration of the ponderomotive bunching scheme and the subsequent measurement of microbunching. The left panels show how the longitudinal electron phase space throughout the scheme: before the interaction, directly after the interaction, after a drift when microbunching is formed.

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$CsPb(Halide)_3$ alloys nano-optical properties

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Halide perovskite semiconductors are promising materials for applications in photovoltaics, light emitting diodes (LEDs), photodetectors and photocatalysts. They benefit from their variable bandgap as a function of chemical composition. $CsPbI_{3x}Br_{3y}Cl_{3(1-x-y)}$ alloys allow for light emission covering the whole visible range [1]. Despite significant advances in materials quality, it is increasingly clear from the literature that chemical heterogeneity is a limiting factor in $CsPb(Halides)_3$ optical properties, from reduced color purity or phase-separation induced traps. For chemically heterogeneous materials with feature sizes down to the tens of nanometer scale, common in perovskite nano-crystals, macroscopic techniques only allow for average measurements.

In this contribution we will report on the absorption, emission and chemistry of $CsPbBr_{3x}Cl_{3(1-x)}$ individual nanoparticles using electron energy loss spectroscopy (EELS), cathodoluminescence (CL) and energy dispersed X-ray spectroscopy (EDS) in the scanning electron microscope (STEM). EELS and CL for nano-optics were performed on a monochromated Nion Hermes at 100 keV with the sample kept at around 100 K, while EDS was performed at 300 keV on a Thermo Fisher Scientific Titan Themis microscope equipped with a four-detector Super-X detector system. For CL, an Attolight Mönch system was used for light collection. The samples were nominally $CsPbBr_{1.5}Cl_{1.5}$, with nanoparticles stabilized in metal-organic framework glass using a method described in [2]. Typical EELS and CL spectra are shown in Figure 1a. For sufficiently thin nanoparticles, both the excitonic absorption and emission are visible. This allows for a measurement of the Stokes shift on particles also analyzed in terms of chemical composition. These measurements enabled the identification of a correlation between the Stokes shift, the CL full width at half maxium (FWHM), and the chlorine concentration (Figure 1b). The Stokes shift increases while the CL FWHM decreases with chlorine content. Currently, we are exploring phenomenological models and first principle calculations to understand these correlations. The methodology described here provides a path to measure the optical and chemical properties of technologically important semiconductor alloys that cannot be assessed by other techniques.

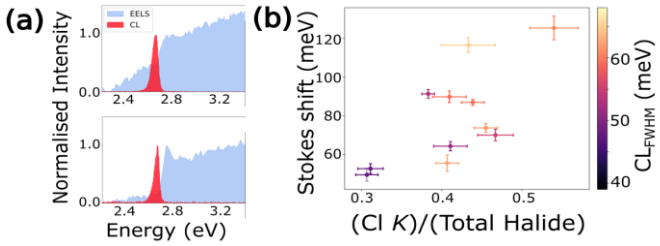

Figure 1. (a) EELS and CL spectra of two $CsPbBr_{1.5}Cl_{1.5}$ nanoparticles in a MOF showing excitonic peaks in absorption and emission. (b) Stokes shift and CL FWHM as a function of chlorine content in the nanoparticles.

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ICS-SAXS: Hard X-ray metrology accelerated by an inverse Compton scattering source

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Small angle x-ray scattering (SAXS) is a well-established, non-destructive technique to determine structural dimensions. It can also be used to determine the critical dimensions (CD) of 3D structures used in semiconductors.

There are two main conditions for CD-SAXS. The first is a trade-off between diffraction efficiency and the transmission through the wafer. The second condition is the need for a source with enough brightness at the right energy to keep the measurement time short. An inverse Compton scattering (ICS) source offers a potential tabletop solution for both these conditions. The continuous tunability of an ICS source allows for operation at the optimal x-ray energy, while also promising a significant increase in brightness compared to other tabletop solutions.

To investigate this potential, we have access to a fully functioning SAXS system and a prototype ICS source that is currently being finalized at the TU/e. The goal of this project is to replace the current source of the SAXS system with the ICS source to do CD-SAXS measurements.

Acknowledgments

ICS-SAXS is funded by Holland High Tech | TKI HSTM via the PPP Innovation Scheme (PPP-I) for public-private partnerships..

Observing the dynamics of a molecular potential through spatiotemporally compressed electron beams

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The interaction of free electrons with near optical fields has emerged as a powerful approach to modulating the former. A suitably engineered light-electron interaction scheme involving parallel interaction at a single lateral plane in distinct separated zones has been proposed to spatiotemporally compress electrons down to Ångström-attosecond scales [1] [Fig. 1(a)]. Here, we investigate the ability of such compressed electrons to perform time-resolved electron microscopy well beyond the capabilities of current instruments. From the scattering distribution of compressed electrons traversing the electromagnetic potential of the probed specimen $V(\boldsymbol{r},t)$ at different times and positions relative to the pump, we infer information on the dynamical evolution of a specimen projected potential [Fig. 1(b)]. We calculate the far-field electron-scattering intensity after interaction with an optically pumped single ozone molecule [Fig. 1(c –d)]. The molecule is excited by a long optical pulse that introduces periodic variations in its valence electron charge density $\rho(\mathbf{r},t)$, resulting in a net potential that evolves on Ångström-attosecond scales and that is probed by the passing electrons. We use the stroboscopic mode of probing, as the optical field that excites the molecule has the same optical period τ_0 as the optical field that compresses the electron. We employ the time-dependent density-functional theory to compute the molecular density and elaborate a formalism to study its interaction with compressed free electrons.

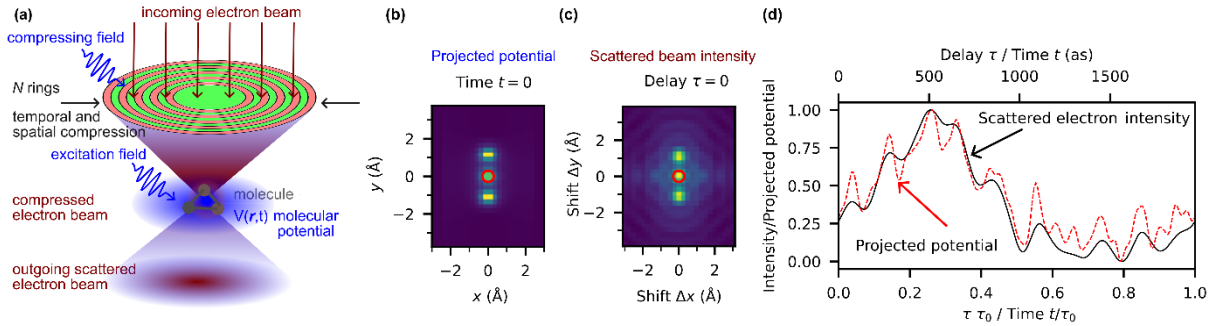

Figure 1. Parallel PINEM compression for probing ultrafast dynamics. (a) Scheme of the proposed experiment. Electrons are focused through parallel interactions with an optical near-field. A sample such as a molecule is excited by synchronized optical field. (b) We aim to reconstruct the dynamics of a projected molecular potential $V_z(x,y,t) = \int V(\boldsymbol{r},t)\,dz$ of the ozone (c) Spatial intensity map for shifts in the transverse directions Δx and Δy, probed by an electron beam with an energy 300 keV. The compression is maintained in the area with a radius 0.48 nm. Notice the artefacts caused by an interaction outside of this area. (d) Comparison of the scattered intensity as a function of delay τ with the temporal profile of the projected potential over an optical period $\tau_0 = 1.97$ fs. Both quantities are scaled to the [0,1] range using min-max normalization. The position corresponds to the beam centred at the atomic core (marked by the red ring in the maps). We are able to distinguish the features with an temporal resolution around $0.1\tau_0$, where $\tau_0 \approx 1970$ as.

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Footprints of atomic-scale features in plasmonic nanoparticles as revealed by electron energy loss spectroscopy

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We present a first-principles theoretical study of the atomic-scale footprints in the valence electron energy loss spectroscopy (EELS) of small metallic nanoparticles (NPs), based on atomistic *ab initio* time-dependent density functional theory (TDDFT) [1,2]. The electron energy loss spectra of small NPs calculated within atomistic TDDFT are particularly sensitive to the atomic-scale features of the surface probed by the electron beam. We trace this behavior with classical local dielectric theories by going beyond spherical shapes and precisely mimicking the atomistic shape of the NPs. Moreover, spectra obtained for penetrating electron beams show the excitation of plasmonic resonances at higher energies than localized surface plasmons (LSPs), which we identify as multipole plasmons (MPs), also known as Bennett modes [3], and confined bulk plasmons (CBPs) [4], which are generally inaccessible with optical spectroscopy techniques. With the aid of a jellium TDDFT model and a linear hydrodynamic model, we characterize their symmetry and excitation rules. In summary, we present a suitable theoretical platform to identify the underlying mechanisms governing the plasmonic response of small metallic NPs as probed by electron beams, which may help clarify the controversy regarding their plasmon energies [5,6].

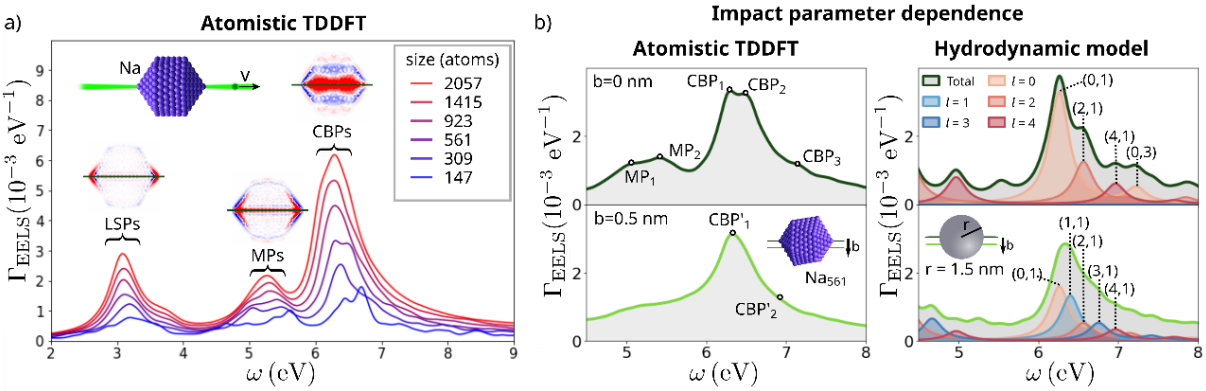

Figure 1. a) EEL spectra of sodium NPs of different sizes calculated using *ab initio* atomistic TDDFT, with different types of plasmon excitations highlighted. The insets display 2D cross-sectional views of the induced charge densities of the corresponding resonances. b) EEL spectra for impact parameters $b = 0$ and 0.5 nm calculated using atomistic TDDFT and a classical hydrodynamic model for a sodium NP of radius $r = 1.5$ nm.

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Spatially resolved EELS and CL coincidence spectroscopy of three-dimensional plasmonic chiral structures

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Cathodoluminescence (CL) and electron energy loss spectroscopy (EELS) are widely used to characterize various materials and nanostructures. Cathodoluminescence excitation spectroscopy (CLE), a temporal combination of CL and EELS [1], has recently been developed to study charge carrier dynamics in semiconductors. In photonic systems, coincident detection of EELS and CL has been proposed to generate heralded photons/electrons [2]. In this presentation, we extend this principle by coincidentally measuring the emission of polarized photons (pCL) and the associated energy loss on chiral plasmonic systems. In this way, we aim to produce heralded polarized photons.

We have designed and fabricated three-dimensional Born-Kuhn system (BKS) chiral structures (Figure 1 a) [3], consisting of staggered silver nano-antennas, suitable for TEM study [4]. Such systems exhibit two low-energy modes resulting from the dipolar coupling of the individual nano-antennas (bonding and anti-bonding mode) with opposite chirality. The experiments are carried out with a NION Hermes200 monochromated microscope equipped with a Timepix3 detector (ASI Cheetah) and a cathodoluminescence detector (Attolight Mönch) modified to measure polarization (Figure 1b). The Cheetah detector enables electrons to be measured as time-resolved events with ns resolution, which can be temporally correlated with the detection of circularly polarized photons performed in parallel.

Figure 1c presents an energy-filtered map extracted from a conventional EELS spectral-imaging, emphasizing the local intensity variations associated with the bonding and anti-bonding mode. Figure 1d shows similar data for the CLE signal polarized along one direction.

We observe a difference in intensity distribution between the two signals, revealing electron-photon correlation dependent on beam position and polarization. In this presentation, we will discuss the implications of this observation for the possibility of producing polarized heralded photons using fast electron beams.

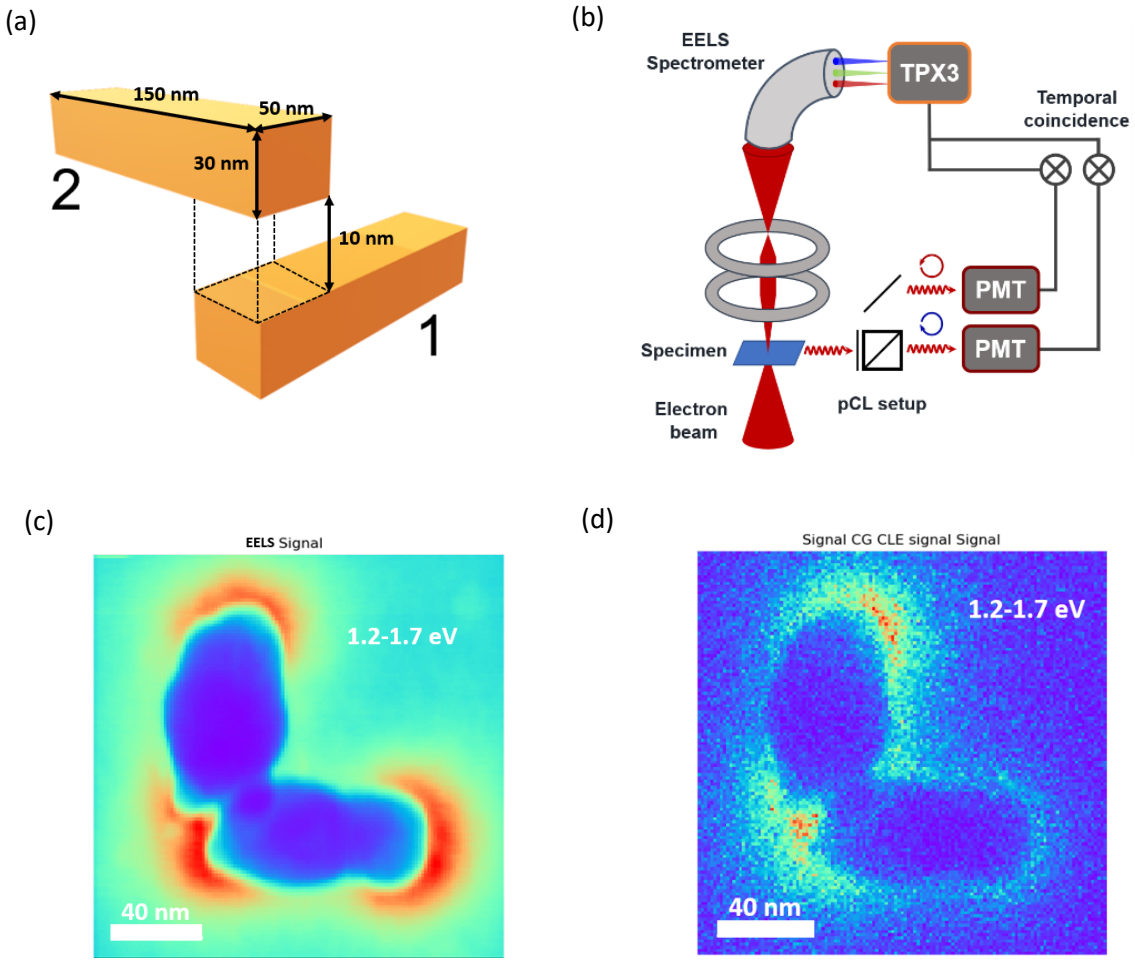

Figure 1. (a) Schematic of a BKS structure with typical dimensions. (b) Schematic of the experimental set-up. (c) Conventional EELS spectrum-imaging (filtered in the 1.2-1.7 eV spectral range) of BKS. (d) Left-polarized CLE spectrum-imaging (filtered in the 1.2-1.7 eV spectral range) of BKS.

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Approaching Free-Electron – Bound-Electron Resonant Intesractions with a Modulated Electron Beam

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Quantum optics with free-electrons is a promising field emerging based on the technology of electron microscopy [1]. One part of the framework of free-electron quantum optics is the resonant and coherent interaction of free-electrons with bound-electron systems [2]. Experimental access to this interaction is particularly challenging, since the bound-electron state has to couple directly to the near-field of the free electrons. With our experimental setup we are aiming to show this interaction for the first time [3]. In this proof of principle experiment (see Fig. 1), we utilize a modulated free-space electron beam in a customized scanning electron microscope, to coherently drive electron spins. The near-field of this spatially modulated beam excites Zeeman levels in a α,γ-Bisdiphenylen-β-phenylally (BDPA) sample placed in a magnetic field. These quantum transitions couple inductively to a micro-coil. The signal in the micro-coil is measured with a lock-in amplifier, sensitive down to the thermal noise floor. A successful implementation of the proposed experiment will lay the foundation for coherently coupling modulated electron beams to bound-electron transitions. Realizing it in a electron microscope, allows to exploit the nano-scopic spatial resolution of electron microscopy. Apart from new spectroscopic methods utilizing the modulated near-field of an electron beam, higher order transitions may also be excited.

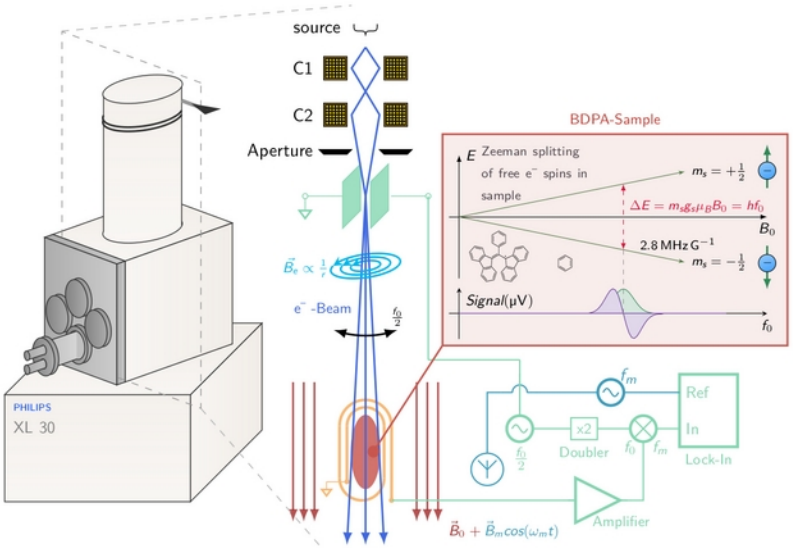

Figure 1. The electron beam in the center (dark blue) of a customized scanning electron microscope (on the left) near field of the beam B_e, couples to the Zeeman levels of the spins in the BDPA sample placed in a bias magnetic field B_0 (red). Sweeping the frequency f_0 and modulating the bias field B_0 at a frequency f_m, enables measuring the spins coupling inductively to a micro-coil (orange), wrapped around the sample, with a homodyne lock-in detection setup.

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Probing Low-Energy Vibrational Excitations in a hBN Nanoparticle via STEM-EELS

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Hexagonal boron nitride (hBN) is a highly promising material for mid-infrared nanophotonics owing to its ability to support strongly confined phonon-polariton modes with long lifetimes and extreme sub-wavelength confinement. These properties enable advancements in applications such as subdiffraction imaging and molecular vibration sensing [1,2]. In this study, high-resolution transmission electron microscopy (TEM) is utilized to investigate low-energy vibrational excitations in hBN semi-ellipsoid nanoparticles, as illustrated in Fig. 1a and b. The observed excitations correspond to two infrared-active in-plane phonon modes within the upper-frequency Reststrahlen band. The maximum intensity is detected when the electron beam passes through the center of the particle, highlighting the strong coupling between the electron beam and the localized vibrational modes.

To interpret these experimental findings, we first model hBN nanoparticles using a dielectric-function-based solution of Maxwell's equation with a finite-element electromagnetic solver (COMSOL Multiphysics). As shown in Fig. 1c, this classical simulation method is based on the local dielectric function of the material, successfully capturing several high-energy features but failing to reproduce low-energy modes. To address this limitation, we formulate an atomistic model, based on density functional theory (DFT) to construct the ground-state atomic configuration and, in turn, calculate the dynamical matrix of the hBN nanostructure. By diagonalizing the dynamical matrix, we obtain the eigenmodes and eigenfrequencies of the system. Using a semiclassical formalism that incorporates long-range dipole-dipole interactions, we describe the coupling between a focused electron beam and lattice vibrations. This approach enables us to predict the spectral features associated with the phonon-induced variations of the EELS spectra of the hBN nanoparticle.

These findings provide new insights into vibrational phenomena in polar materials, advancing our understanding of their fundamental properties. Additionally, they offer valuable guidance for the design of nanophotonic devices based on hBN nanostructures.

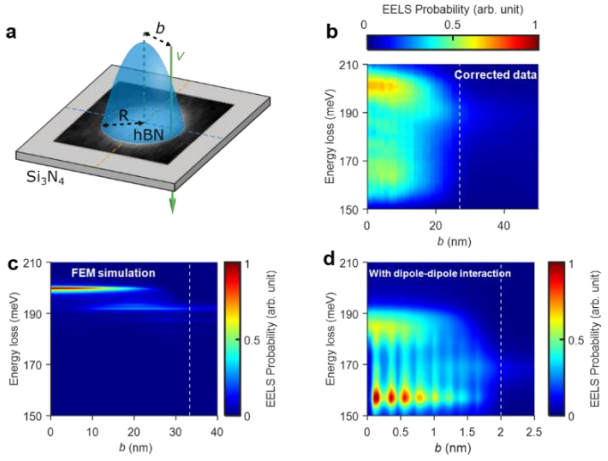

Fig. 1 a Schematic representation of an electron beam interacting with a hBN ellipsoid nanostructure positioned on a silicon nitride membrane. **b** Experimentally measured and normalized EELS probability distribution. **c** Simulated EELS probability using a local dielectric model via COMSOL. **d** DFT-based atomistic simulation of the EELS probability distribution incorporating dipole-dipole interactions.

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Low-energy single-electron detector with micrometric resolution

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Electron detectors are crucial to electron microscopes, electron spectroscopy setups, and quantum electron optics experiments. High spatial and temporal resolution, low noise, and single-electron detection efficiency are among the key features of modern electron detectors.

Due to their high sensitivity and speed, direct electron detectors (DED) [1] are now the gold standard in most electron microscopy and spectroscopy applications [2].

At low electron energies typical for scanning electron microscopy (SEM, lower than 30 keV), DEDs are often hybrid array detectors. While they enable single-electron detection, their spatial resolution is limited by pixel size, typically 55 µm [3] or larger [4]. Monolithic active pixel sensors offer a pixel size down to 5 µm [5-7] and have been employed at energies down to 3 keV. While they are commercially available, they are often prohibitively expensive.

We demonstrate single-electron detection and counting based on a YAG:Ce scintillator. An optical microscope (magnification 11.4x) uses a high numerical aperture objective (NA = 1.4) and a lens (f=50 mm) to image the scintillated light onto a camera chip (pixel size = 4.6 µm). Our detector offers a spatial resolution (FWHM) of 1.1 µm at 18 keV and 1.4 µm at 30 keV. We demonstrate single-electron detection for energies between 16 keV and 30 keV, achieving a false discovery rate (FDR) as low as 1.8% at 30 keV, where the FDR is defined as the ratio of false positives to the total number of detected events: FDR = false positives / (true positives + false positives).

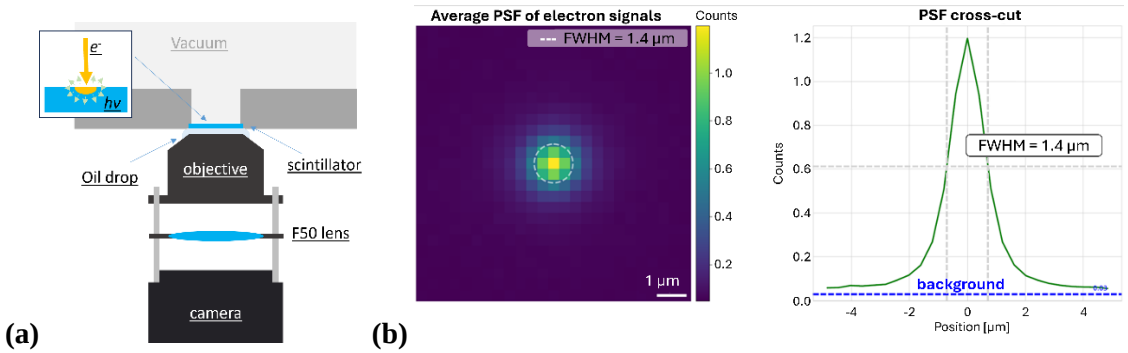

Figure 1. (a) Overview of the detection system: the light emitted by the scintillator due to the impact of an electron is collected by an oil-immersive objective (NA=1.4) and imaged onto the camera chip by a lens (f=50mm). (b) The average point spread function (PSF) of 30 keV electrons (left) and the corresponding crosscut show a FWHM of 1.4 µm.

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Acknowledgments

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Cathodoluminescence study of CdSeTe photovoltaic absorbers using FIBSEM and TEM

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CdTe is the leading second-generation solar technology among thin-film solar cells, offering low cost-solar electricity generation. Extensive research continues to drive CdTe solar cells toward the maximum efficiency predicted by the Shockley-Queisser limit. Typical processes focus on incorporating a gradient of selenium (Se) in the CdTe, the $CdCl_2$ treatment to improve microstructure and passivate grain boundaries, and Group V doping to increase carrier concentration[1]. In this work, we study polycrystalline CdSeTe/CdSe thin film solar cells to correlate the photoactivity directly with grain orientation, defects, and composition from micro- to nanoscale using cathodoluminescence (CL). TEM nanoscale band gap mapping will be shown in Se alloyed CdTe with arsenic doping that demonstrates the effect of Se on the bandgap together with the potential Se interaction with As at grain boundaries.

The instruments employed are (i) FEI xenon Helios G4 plasma FIB attached with Monarc Pro, Gatan and (ii) TFS Talos F200i STEM attached with Mönch, Attolight. The combination of pFIB with STEM allows site-specific sections either parallel (in-plane) or perpendicular (edge) to the film surface to be extracted based on CL for further STEM.

Fig.1a presents a STEM/EDS color-mix map of the Se alloyed CdTe absorber on F doped SnO_2-coated glass substrate. Chlorine (Cl) defines the grain boundaries, and the gradient of Se is evident with high concentration at the front interface. Note Se also segregates along the grain boundary (arrowed). The corresponding band gap derived by the direct band gap energy is given in Fig.1b. The consistent contrast between the Se composition and the band gap map demonstrates the well-known effect of stoichiometry on the band gap of CdSeTe [2]. Furthermore, a halo contrast in the CL map is observed along the grain boundaries associated with Se segregation, as seen in the CL intensity slice at 1.367±0.05eV in Fig.1c. Fig.1d compares spectra of the interiors near the front interface (Point4) and mid grain (Point1), the Se segregated region (Point2), and the opposite of the GB (Point3). Spectra 1(green) and 4(black) shows the direct band gap shift due to the Se concentration. Spectra 1, 2 and 3 suggest Se segregation adjacent to GB affects the defect state at 1.41eV, but no significant effect on the direct band gap at 1.57eV.

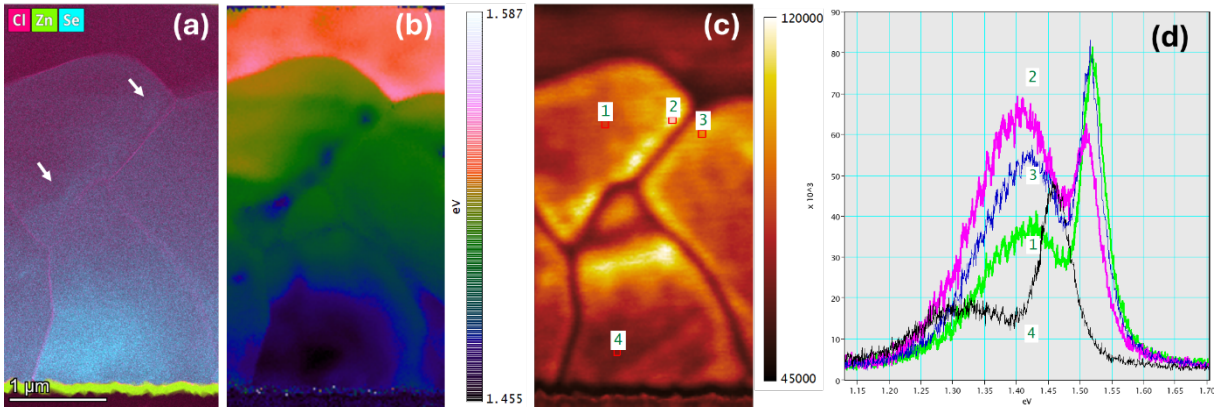

Figure 1.(a) STEM EDS map, (b) CL map of the direct band gap, (c) CL intensity at 1.367±0.05eV and (d) the corresponding spectra in a Se alloyed CdTe thin film doped with As.

References

[1]MA Scarpulla, B McCandless, AB Phillips, et al, Solar Energy Materials and Solar Cells, 2023, 255, 112289.

[2]B Frouin, T Bidaud, S Pirotta et al, APL Mater. 12, 031135 (2024); doi: 10.1063/5.0195398.

Acknowledgments: The project is funded by UK EPSRC strategic equipment grant EP/X030245/1.

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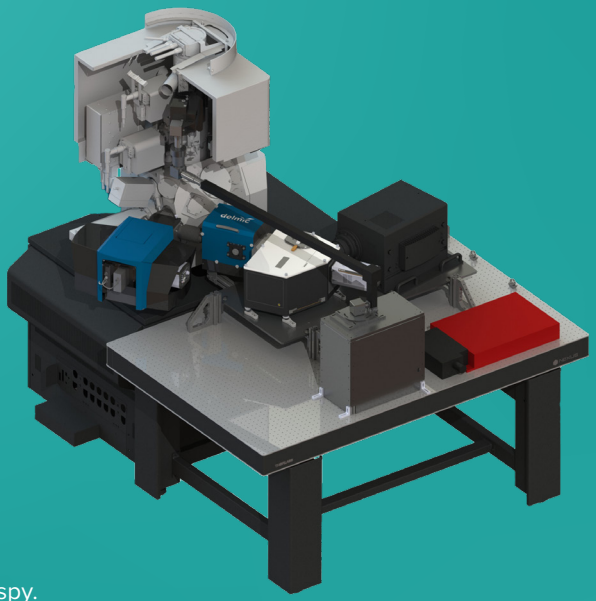

Applications

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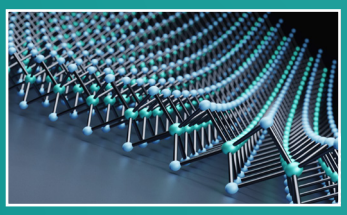
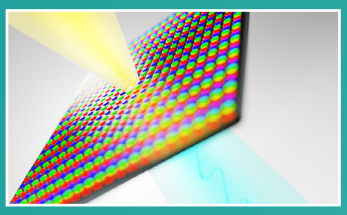
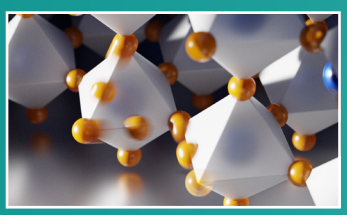


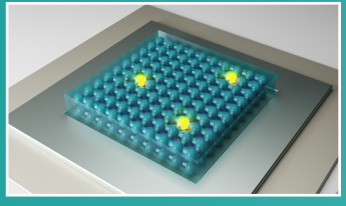
III/V semiconductors



Perovskites



Quantum emitters



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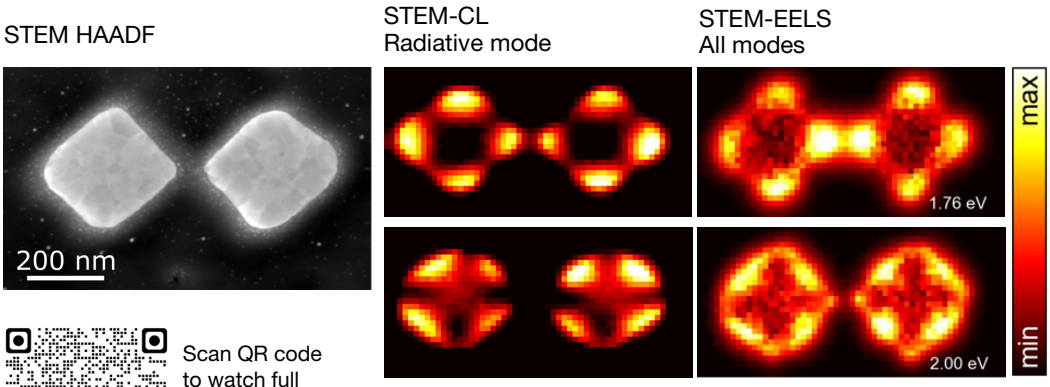
Separation of radiative and non-radiative modes of gold plasmonic antennas using STEM CL and EELS



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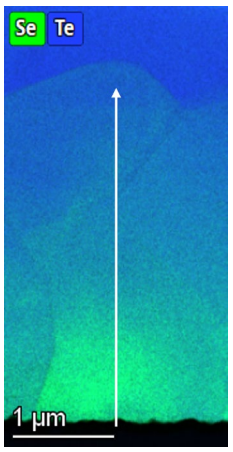
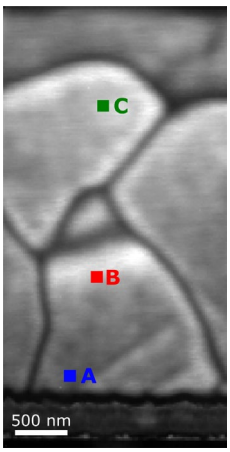
Acknowledgement: Rostislav Repa, Michal Horak, CEITEC BUT

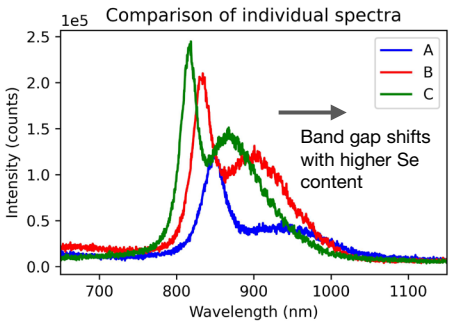
Correlation of composition and bandgap of CdTe-CdSeTe absorber layer using STEM CL and EDS

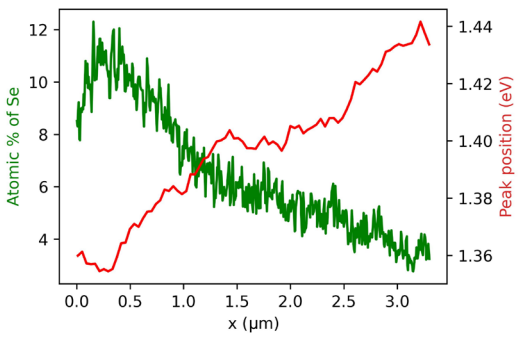


Total CL intensity EDS map of Se and Te



Total CL intensity





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