# Oral Presentation

**333** Investigating the ultrafast phase transition of vanadium dioxide using time-resolved three-dimensional electron diffraction
Dr. Arthur Niedermayr[1,2], Prof. Hongyi Xu[3], Michael Yannai[2], Dr. Gaolong Cao[1], Prof. Bertina Fisher[4], Prof. Lior Kornblum[2], Prof. Ido Kaminer[2], Prof. Xiaodong Zou[3], Prof. Jonas Weissenrieder[1]
[1]Materials and Nano Physics, School of Engineering Sciences, KTH Royal Institute of Technology, Stockholm SE-100 44, Sweden, [2]Department of Electrical and Computer Engineering, Technion–Israel Institute of Technology , Haifa 32000, Israel, [3]Department of Materials and Environmental Chemistry, Stockholm University, Stockholm, Sweden, [4]Department of Physics, Technion—Israel Institute of Technology, Haifa 32000, Israel
**379** Probing the chiral behavior of nano-optical near-fields through angular momentum resolved PINEM
Simon Garrigou[1], Mr. Arnaud Arbouet[1], Mr. Hugo Lourenço-Martins[1]
[1]CEMES-CNRS, Université de Toulouse, CNRS, Toulouse, France
**427** Femtosecond electron beam probe of ultrafast electronics
Maximilian Mattes[1], Dr Mikhail Volkov[1], Prof Dr Peter Baum[1]
[1]Universität Konstanz, Konstanz, Germany
**685** Attosecond electron microscopy using free-electron homodyne detection
John Gaida[1,2], Hugo Lourenço-Martins[1,2], Murat Sivis[1,2], Thomas Rittmann[1,2], Armin Feist[1,2], F. Javier García de Abajo[3,4], Claus Ropers[1,2]
[1]Dept. of Ultrafast Dynamics, Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany, [2]4th Physical Institute, University of Göttingen, Göttingen, Germany, [3]ICFO-Institut de Ciencies Fotoniques, Castelldefels (Barcelona), Spain, [4]ICREA-Institució Catalana de Recerca i Estudis Avançats, Barcelona, Spain
**702** Electron-photon quantum interaction enables novel ultrafast electron imaging approaches
Prof. Giovanni Maria Vanacore[1], Cameron J. R. Duncan[1], Beatrice Matilde Ferrari[1], Irene Ostroman[1], Maria Giulia Bravi[1], Tomer Bucher[2], Ron Ruimy[2], Ido Kaminer[2], Javier Garcia de Abajo[3], Thomas LaGrange[4], Fabrizio Carbone[4], Paolo Rosi[5], Enzo Rotunno[5], Vincenzo Grillo[5], Jean-Cristophe Olaya[6], Sang-Tae Park[7], Dan Masiel[7]
[1]Laboratory of Ultrafast Microscopy for Nanoscale Dynamics (LUMiNaD), Department of Materials Science, University of Milano-Bicocca, Milano, Italy, [2]TECHNION, Israel Institute of Technology, Haifa, Israel, [3]ICFO-Institut de Ciencies Fotoniques, Mediterranean Technology Park, Castelldefels (Barcelona), Spain, [4]LUMES, Ecole Polytechnique Federale de Lausanne (EPFL), Lausanne, Switzerland, [5]CNR-Nano S3, Modena, Italy, [6]Holoeye Photonics Ag, Berlin, Germany, [7]JEOL-IDES, Pleasanton, USA
**748** Development and characterization of a laser-driven cold-field emission source
Alexander Schröder[1], Mukai Masaki[2], Kohno Yuji[2], Sascha Schäfer[1,3]
[1]Department of Physics, University of Regensburg, Regensburg , Deutschland, [2]JEOL Ltd., , Japan, [3]Regenburg Center for Ultrafast Nanoscopy (RUN), Regensburg, Germany

# Poster Presentation

**9** Electrochemical lithium intercalation & exfoliation in 2D TMDs and its in-situ studies
Zhiyuan Zeng[1]
[1]Department of Materials Science and Engineering, and State Key Laboratory of Marine Pollution, City University of Hong Kong, Kowloon, Hong Kong
**151** Approaching picosecond temporal resolution in off-axis electron holography
Dr. Tolga Wagner[1], Hüseyin Çelik[2], Simon Gaebel[3], Dirk Berger[4], Christian M. Günther[4], Ines Häusler[1], Sebastian Siewert[2], Michael Lehmann[2], Christoph T. Koch[1]

[1]Department of Physics, Humboldt-Universität zu Berlin, Berlin, Germany, [2]Institute of Optics and Atomic Physics, Technische Universität Berlin, Berlin, Germany, [3]Max-Born-Institut, Berlin, Germany, [4]Center for Electron Microscopy (ZELMI), Technische Universität Berlin, Berlin, Germany

**208** Imaging MEMS motion at nano scale by time-resolved scanning electron microscopy

Mohamed Zaghloul[1,2], Abbas Kosari Mehr[1,2], Riccardo Bertacco[1,3], Simone Cuccurullo, Federico Maspero[1], Giulia Pavese[4], Hao Chen[1,2], Aldo Ghisi[4], Alberto Corigliano[4], Silvia M. Pietralunga[2,5], Alberto Tagliaferri[1,2]

[1]Dipartimento di Fisica, Politecnico di Milano, Milano, Italy, [2]CNST@PoliMi, Istituto Italiano di Tecnologia (IIT), Milano, Italy, [3]Polifab, Politecnico di Milano, Milano, Italy, [4]Dipartimento di Ingegneria Civile e Ambientale, Politecnico di Milano, Milano, Italy, [5]Institute for Photonics and Nanotechnologies (IFN)—National Research Council (CNR), Milano, Italy

**336** Quantum phase modulation and retrieval in an ultrafast transmission electron microscope

Dr. Cameron Duncan[1], Beatrice Ferrari[1], Irene Ostroman[1], Maria Giulia Bravi[1], Sang Tae Park[2], Daniel Masiel[2], Jean Christophe Olaya[3], Eduardo Dias[4], Javier García de Abajo[4], Giovanni Maria Vanacore[1]

[1]Unversità degli Studi di Milano-Bicocca, Milan, Italy, [2]JEOL-IDES, Pleasonton, United States of America, [3]Holoeye Photonics, Berlin, Germany, [4]ICFO-Institude Ciencies Fotoniques, The Barcelona Institute of Science and Technology, Barcelona, Spain

**553** Laser particle acceleration in an ultrafast scanning electron microscope

Leon Brückner[1], Tomas Chlouba[1], Roy Shiloh[1], Stefanie Kraus[1], Julian Litzel[1], Peter Hommelhoff[1]

[1]Physics Department, Friedrich-Alexander-Universität Erlangen-Nürnberg, Erlangen, Germany

**600** Enhanced Free-Electron Wavefunction Modulation via Photon-Induced Near-Field Electron Microscopy (PINEM) with Shaped Light Fields

Beatrice Ferrari[1,7], Cameron J. Duncan[1], Irene Ostroman[1], Maria G. Bravi[1], Paolo Rosi[2], Enzo Rotunno[2], Jean-Cristophe Olaya[3], S. T. Park[4], D. Masiel[4], Vincenzo Grillo[2], Ido Kaminer[5], Francisco J. Garcia de Abajo[6], Thomas Lagrange[7], Fabrizio Carbone[7], Giovanni M. Vanacore[1]

[1]LUMiNaD UniMiB, Milano, Italy, [2]CNR-Nano, Modena, Italy, [3]HOLOEYE Potonics, Berlin, Germany, [4]JEOL IDES, Pleasanton, USA, [5]TECHNION, Haifa, Israel, [6]ICFO, Bacelona, Spain, [7]LUMES EPFL, Lausanne, Switzerland

**620** Electron energy-gain spectroscopy of optical excitations in integrated photonic structures

Jan-Wilke Henke[1,2], Dr. Yujia Yang[3,4], F. Jasmin Kappert[1,2], Dr. Arslan S. Raja[3,4], Germaine Arend[1,2], Guanhao Huang[3,4], Dr. Armin Feist[1,2], Zheru Qiu[3,4], Rui Ning Wang[3,4], Aleksandr Tusnin[3,4], Dr. Alexey Tikan[3,4], Prof. Dr. Tobias J. Kippenberg[3,4], Prof. Dr. Claus Ropers[1,2]

[1]Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany, [2]University of Göttingen, 4th Physical Institute, Göttingen, Germany, [3]Institute of Physics, Swiss Federal Institute of Technolog Lausanne (EPFL), Lausanne, Switzerland, [4]Center for Quantum Science and Engineering, Swiss Federal Institute of Technology Lausanne (EPFL), Lausanne, Switzerland

**696** Coulomb-correlated multi-electron states in a transmission electron microscope beam

Rudolf Haindl[1,2], Dr. Armin Feist[1,2], Till Domröse[1,2], Dr. Marcel Möller[1,2], John H. Gaida[1,2], Dr. Sergey V. Yalunin[1,2], Prof. Dr. Claus Ropers[1,2]

[1]Department of Ultrafast Dynamics, Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany, [2]4th Physical Institute -- Solids and Nanostructures, Göttingen, Germany

**881** Towards electron beam shaping in an Ultrafast Transmission Electron Microscope

Valentin Rollo[1], Sebastien Weber[1], Arnaud Arbouet[1]

[1]CEMES - CNRS, Toulouse, France

**888** Free electron spectroscopy seen through the prism of quantum optics

Tom Fraysse[1], Hugo Lourenço-Martins[1]

[1]CEMES-CNRS, Toulouse, France

**991** Temporal characterization of femtosecond electron pulses inside ultrafast scanning electron microscope

Kamila Moriova[1], Martin Kozak[1]

[1]Faculty of Mathematics and Physics, Charles University, Prague,

**1010** Imaging ultrafast spin dynamics at the nanometer scale
Dr Gaolong Cao[1], Sheng Jiang[2], Yuzhu Fan[1], Johan Åkerman[3], Jonas Weissenrieder[1]
[1]Royal Institute of Technology (KTH), Stockholm, Sweden, [2]South China University of Technology, Guangzhou, China, [3]University of Gothenburg, Gothenburg, Sweden
**1034** Fabrication of electron transparent membranes and nanostructures in fluidic devices by NIL and "Flow-Through"-gas-phase deposition
Jeremy Teuber[1], Manuel Müller[1], Rukan Nasri[1], Dr. Irene Fernandez-Cuesta[1]
[1]University of Hamburg, Hamburg, Germany
**1082** Resolving single-electron and multi-electron distribution functions with event-based electron detectors
Alexander Schröder[1], Andreas Wendeln[1], Prof. Sascha Schäfer[1]
[1]University Regensburg, Regensburg, Germany
**1092** Molecular dynamics simulation of the Brownian motion of biomolecules in liquid phase electron microscopy
Mr Xiaopeng Wu[1], Dr Peng Wang[1]
[1]Department of Physics, University of Warwick, Coventry, United Kingdom
**1163** Spatiotemporal observation of ultrafast magnetization dynamics with 4D Lorentz Transmission Electron Microscope
Phd Yuzhu Fan[1], Researcher Gaolong Cao[1], Professor Jonas Weissenrieder[1]
[1]School of Engineering Sciences, KTH Royal Institute of Technology, Stockholm, Sweden

333

# Investigating the ultrafast phase transition of vanadium dioxide using time-resolved three-dimensional electron diffraction

Dr. Arthur Niedermayr[1,2], Prof. Hongyi Xu[3], Michael Yannai[2], Dr. Gaolong Cao[1], Prof. Bertina Fisher[4], Prof. Lior Kornblum[2], Prof. Ido Kaminer[2], Prof. Xiaodong Zou[3], Prof. Jonas Weissenrieder[1]
[1]Materials and Nano Physics, School of Engineering Sciences, KTH Royal Institute of Technology, Stockholm SE-100 44, Sweden, [2]Department of Electrical and Computer Engineering, Technion–Israel Institute of Technology , Haifa 32000, Israel, [3]Department of Materials and Environmental Chemistry, Stockholm University, Stockholm, Sweden, [4]Department of Physics, Technion—Israel Institute of Technology, Haifa 32000, Israel

IM-08, Lecture Theater 2, august 28, 2024, 10:30 - 12:30

Background incl. aims
Vanadium dioxide ($VO_2$) exhibits a remarkable phase transition at around 68°C, accompanied by significant changes in its electronic and structural properties. This transition holds great promise for the development of advanced electronic switches, optical detectors, logic and memory storage and nonlinear circuit components [1,2]. However, the ultrafast dynamics of this transition are not fully understood, limiting its practical application. The temporal evolution of the structural phase and the question whether thermal or nonthermal effects dominate remains a controversial, debated topic to date [3], in particular, whether $VO_2$ transitions directly from the monoclinic phase (M1 - $P2_1/c$) to the rutile (R - $P4_2/mnm$) phase, or whether an intermediate monoclinic phase (M2 - C2/m) exists under certain conditions in a transient state. This potential intermediate phase which typically occurs under applied strain was proposed by X-ray spectroscopy and later disproved by ultrafast electron diffraction (UED). Our experimental approach which allows for a complete determination of the phase will shine light on this most recent debate.

Methods
Here, to investigate the spatio-temporal evolution of the phase transition we use an ultrafast transmission electron microscope (UTEM) which combines the nanometer spatial resolution of the TEM with the temporal resolution of femtosecond pulsed lasers. Our approach relies on the combination of an optical pump pulse for exciting the sample and a photoemitted electron probe pulse to detect structural dynamics in our sample. The UTEM allows us to probe the atomic structure on a sub-micron area with femtosecond resolution, thus extending the capabilities offered by UED [4] and X-ray techniques. Using this pump-probe technique, a change of the delay of the laser excitation with respect to the electron probe allows us to capture time-resolved diffraction movies in a stroboscopic approach. Given the similar diffraction patterns of the different structural phases of $VO_2$, the phases taken along well-defined zone axes can easily be confused. This is especially true with the conditions relevant for ultrafast electron microscopy, where one is further limited by low electron count rates (SNR), low spatial coherence and resolution. We overcome these difficulties by extending ultrafast electron diffraction, for the very first time, to ultrafast three-dimensional electron diffraction (3D ED [5]), where at each time delay the diffraction pattern of the sample is collected at multiple angles oriented off zone axes. This tilt series of diffraction patterns can then be combined and analyzed to build a complete 3D model of the atomic crystal structure without prior knowledge of the material. This approach allows us to clearly distinguish the different structural phases in $VO_2$ at moderate SNR and without the need of atomic structure simulations for comparison.

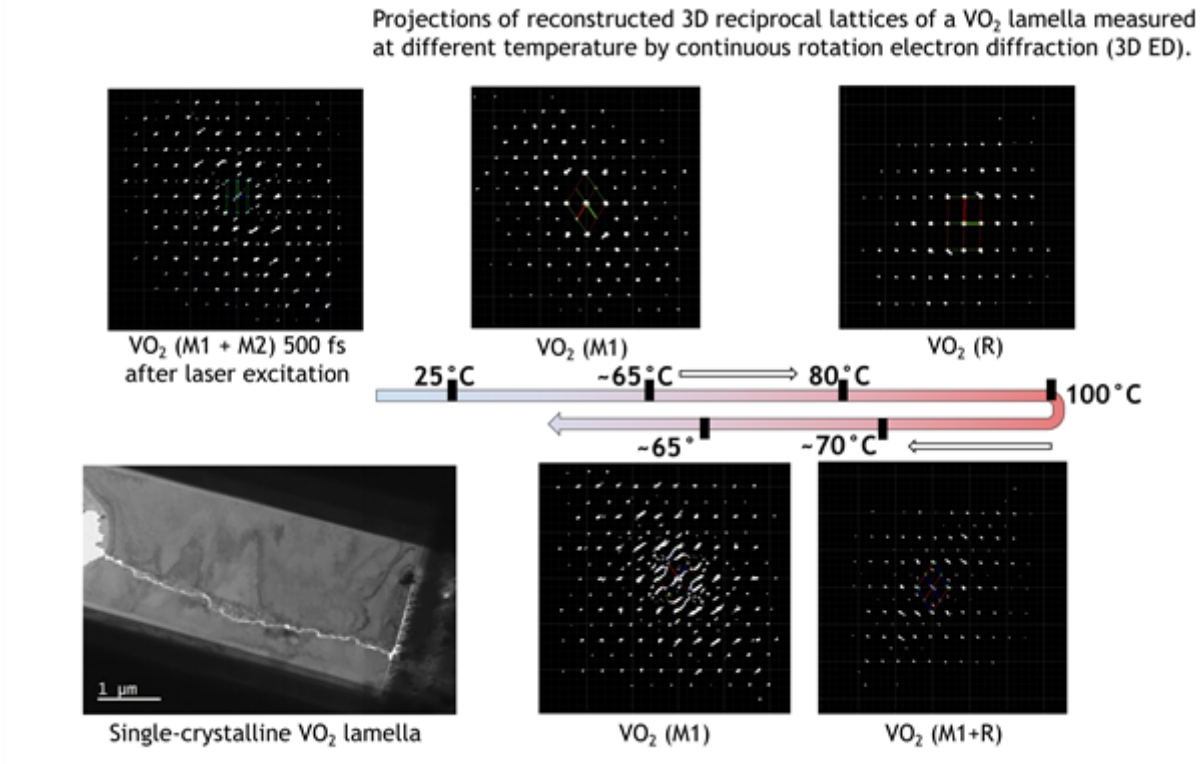
Results
We characterized the ground state of a single-crystalline $VO_2$ lamella made via focused ion-beam milling in a conventional TEM and confirmed its excellent crystalline quality by electron diffraction.

Defocused imaging revealed bending and stress contrast in the lamella (see Graphic). Static 3D ED of $VO_2$ as a function of temperature (between room temperature and 100° C) showed the structural transition from the M1 to the R phase, without any evidence of a stress-induced M2 phase (see Graphic). These results were consistent, regardless of the electron probe size and position on the lamella.

The ultrafast 3D ED results are distinctly different from the static results. We unambiguously observe a transition pathway between the two monoclinic phases (see Graphic), from M1 to M1+M2, on a timescale of 500 fs. This can be indicative of several scenarios: (i) either the nature of the ultrafast photo-induced phase transition is different compared to the thermal phase transition, or (ii) the M2 phase is a metastable, transient state (and thus cannot be resolved in a static measurement), or (iii) the photo-excitation leads to excitation of intense acoustic modes which induce notable stress in the material, placing $VO_2$ on a transition trajectory to a different structure. Our results were reproducible, and the sample did not show any sign of photoinduced damage. Importantly, the laser repetition rate of 12 kHz was kept such that the system could return to its ground state between sequential pump laser pulses.

## Conclusions

In summary, we have for the first time demonstrated the feasibility of 3D ED in the ultrafast regime. The results allowed us to unequivocally determine the crystal structure of $VO_2$ in the time-resolved regime. Surprisingly, and in contradiction to existing literature, we unequivocally detect the presence of a monoclinic M2 transition state in our well-defined, single-crystalline sample. The M2 phase persists over several picoseconds. We plan to extend our study by collecting more data sets at longer temporal delays and with the intention to gain further insights into the underlying mechanisms governing the transition. Of special interest is how photoexcited acoustic modes or different lamella geometries affect the transient stress and ultimately the transition pathway. We believe that ultrafast 3D ED will become indispensable for transient structural determination in the near future. In contrast to three-dimensional X-ray or neutron diffraction, our approach is more readily available, does not require large-scale facilities, and offers better spatial resolution, thus facilitating the exploration of micro/nano-crystals and individual domains.



**Keywords:**

Ultrafast, three-dimensional electron diffraction

**Reference:**

1. Nakano, M. et al. Collective bulk carrier delocalization driven by electrostatic surface charge accumulation. Nature 487, 459–462 (2012).
2. Samizadeh Nikoo, M. et al. Electrical control of glass-like dynamics in vanadium dioxide for data storage and processing. Nat Electron 5, 596–603 (2022).
3. Li, J. et al. Direct Detection of V-V Atom Dimerization and Rotation Dynamic Pathways upon Ultrafast Photoexcitation in $VO_2$. Phys. Rev. X 12, 021032 (2022).
4. Baum, P., Yang, D.-S. & Zewail, A. H. 4D Visualization of Transitional Structures in Phase Transformations by Electron Diffraction. Science 318, 788–792 (2007).
5. Gemmi, M. et al. 3D Electron Diffraction: The Nanocrystallography Revolution. ACS Cent. Sci. 5, 1315–1329 (2019).

379

# Probing the chiral behavior of nano-optical near-fields through angular momentum resolved PINEM

Simon Garrigou[1], Mr. Arnaud Arbouet[1], Mr. Hugo Lourenço-Martins[1]
[1]CEMES-CNRS, Université de Toulouse, CNRS, Toulouse, France

IM-08, Lecture Theater 2, august 28, 2024, 10:30 - 12:30

Background incl. aims
Usually performed in a ultrafast transmission electron microscope (UTEM), photon induced near-field electron microscopy (PINEM) consists in mapping the magnitude of an optical near-field pumped by a laser with a fast electron beam [1]. Benefiting from the sub-nm spatial resolution of TEMs as well as the high fluence and energy resolution of ultrashort lasers, this technique can probe deep-subwavelength structural details of the optical near-field in resonant and non-resonant structures.
Methods
In addition, and contrary to electron energy-loss spectroscopy (EELS), PINEM displays a high degree of selectivity thanks to the laser illumination. For example, by properly tailoring e.g. the wavelength and polarization of the laser, one can selectively excite individual optical modes [2]. This idea can be extended to probe the chirality of nano-optical near-fields. Indeed, as first demonstrated by Harvey and collaborators in 2020 [3], one can perform a so-called “chiral PINEM” experiment – where a right- and left-handed polarized laser beam is sequentially used to excite the structure and measure the differential PINEM signal. In that configuration, spin angular momentum (SAM) of the incoming beam is tuned so that this technique is denoted as SAM-dichroic PINEM (see figure 1).
Nevertheless, by shaping its phase profile, a laser beam can also carry orbital angular momentum (OAM), and a long-standing question remains on the difference between OAM- and SAM-based dichroic experiment [4]. A natural candidate to tackle this issue is thus a combined SAM-dichroic and OAM-dichroic PINEM experiment – an experimental scheme that we will refer to as angular momentum resolved PINEM (AM-PINEM).
Results
In this conference, we will present our latest work on angular momentum resolved PINEM, showing its application to the probing of the coupling between chiral light and optical near-fields. In a first part we will show that the theoretical framework based on the local density of states (LDOS) used to explain EELS and CL experiments [5] can be extended to PINEM, combining in an elegant way these three main TEM spectroscopies. In particular, we will demonstrate that AM-PINEM can be connected to the concept of chiral radiative LDOS.
In a second part we will focus on the numerical simulation of AM-PINEM experiments using pyGDM - an electrodynamics simulation toolbox based on the green dyadic method that we enhanced to simulate polarized electron spectroscopies. The large library of different illuminations allows for simulation of both SAM- and OAM-resolved PINEM experiments. We will focus on practically realizable structures such as the gold trimer (see figure 1) in which OAM- and SAM-dichroism present clear differences.
Conclusions
In a last part, we will show our first experimental results on SAM-dichroic PINEM realized with a new cold field emission UTEM on simple achiral structures, revealing dichroic behaviors at the deep sub-wavelength scale. Finally, we will present our preliminary work towards the realization of the OAM-dichroic PINEM experiments.

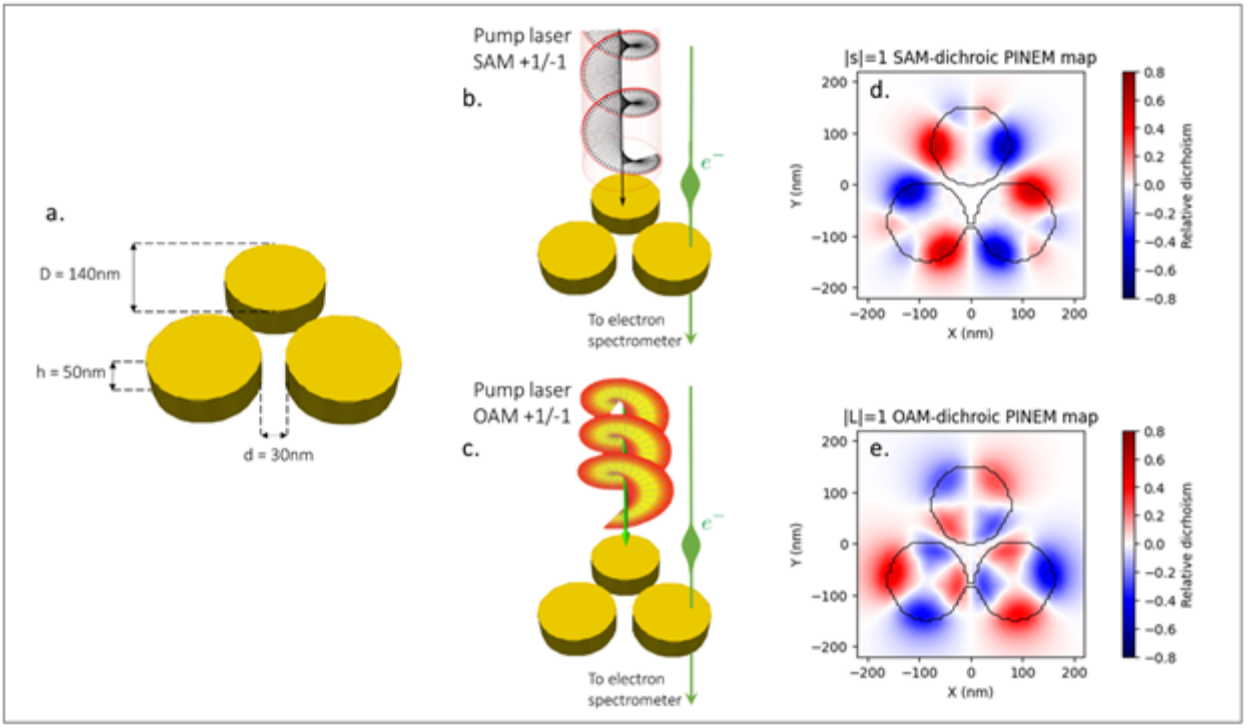



**Figure 1 - (a)** Gold trimere in vacuum with dimensions, **(b,c)** representation of the dichroic PINEM experiment where the sample is pumped with **(b)** SAM +1/-1 beam, **(c)** OAM +1/-1 beam at 650nm wavelength. **(d,e)** Corresponding resulting dichroic maps. Dichroic maps are made by computing the difference of two maps with the same norm of angular momentum but opposed helicity for the illumination beam.

**Keywords:**

PINEM, Angular momentum, Numerical, nano-optics

**Reference:**

[1] A. Feist et al., Nature 521, 200-203 (2015)
[2] M. Liebtrau et al., Light : Science and Application 10, 82 (2021)
[3] T. R. Harvey et al., Nano. Lett. 20 (6), 4377-4383 (2020)
[4] Kerber et al., Communication Physics 1, 87 (2018)
[5] Losquin and Kociak, ACS Photonics 2, 11, 1619-27 (2015)

427

# Femtosecond electron beam probe of ultrafast electronics

Maximilian Mattes[1], Dr Mikhail Volkov[1], Prof Dr Peter Baum[1]
[1]Universität Konstanz, Konstanz, Germany

IM-08, Lecture Theater 2, august 28, 2024, 10:30 - 12:30

## Background and Experiment

The need for ever-faster information processing requires exceptionally small devices that operate at frequencies approaching the terahertz and petahertz regimes. However, electronics itself is too slow to characterize a record-breaking device. Here we show how ultrafast electron beam probe with terahertz-compressed electron pulses can directly sense local electro-magnetic fields in electronic devices with femtosecond, micrometre and millivolt resolution under normal operation conditions [1].

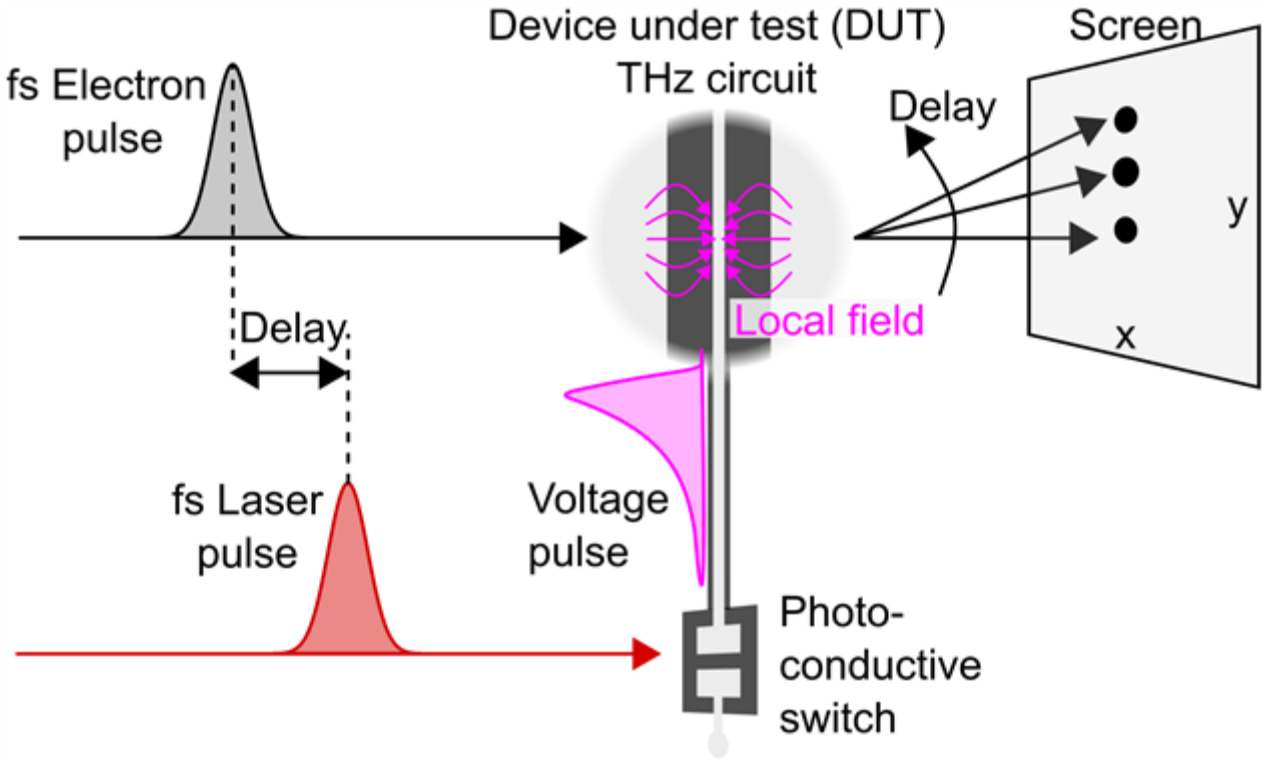
Figure 1 shows the concept of our experiment. We use a femtosecond laser (red) that creates femtosecond electron pulses (grey) with only one electron per pulse. This electron pulse is then further shortened in time to less than 100 fs by using all optical terahertz compression [2]. These ultrashort electron pulses probe the local electromagnetic fields in our device under test (DUT), which is shown in Figure 1. We create a short voltage pulse by exciting a photoconductive switch. The generated femtosecond voltage pulses (magenta) travel through our device under test and the device response deflects the electron beam as a function of space and time. From the resulting electron beam deflections, we then obtain the strength and direction of the local electromagnetic fields [1]. The electron beam does not distort the dynamics under investigation and the measurement is therefore contact-less and impedance-free.

## Results and outlook

In a proof of principle experiment, we investigate a coplanar waveguide which is the standard transmission line in high-frequency electronics. We fabricate the structure by lithography and drill two holes into the substrate for investigation. We observe the impulse response, signal reflections, attenuation and waveguide dispersion directly in the time domain. We find that the experiment has a sensitivity to electric potentials down to tens of millivolt or -20 dBm. With our 100-fs electron pulses, the bandwidth exceeds 10 THz which is well sufficient for almost any modern or near-future circuitry. In order to eventually obtain a full movie in space and time, we also report the design of an ultrafast transmission electron microscope with terahertz compression [3]. The use of an optical cavity with magnetic or electric nodes at the point of electron-terahertz interaction provides an almost aberration-free control. In this way, we can reach nanometer spatial resolution and reach even shorter electron pulse lengths down to 19 fs [3].

Our femtosecond time resolution and the capability to directly integrate our technique into existing electron-beam inspection devices in semiconductor industry make our femtosecond electron beam probe a promising tool for research and development of next-generation electronics at unprecedented speed and size.

Fig. 1. A laser pump pulse (red) creates a voltage pulse (magenta) by closing a photoconductive switch. This pulse then travels into the device under test and triggers its operation. Femtosecond electron pulses (grey) probe the local electric and magnetic field (magenta) [1].




**Keywords:**

Ultrafast electron microscopy, THz electronics

**Reference:**

[1] M. Mattes, M. Volkov, P. Baum, “Femtosecond electron beam probe of ultrafast electronics”, Nat. Commun. 15, 1743 (2024).
[2] C. Kealhofer, W. Schneider, D. Ehberger, A. Ryabov, F. Krausz, P. Baum, “All optical control and metrology of electron pulses”, Science 352, 429-433 (2016).
[3] J. Kuttruf, D. Nabben, A.C. Zimmermann, A. Ryabov and P. Baum, “Terahertz control in a electron microscope”, under review (2024).

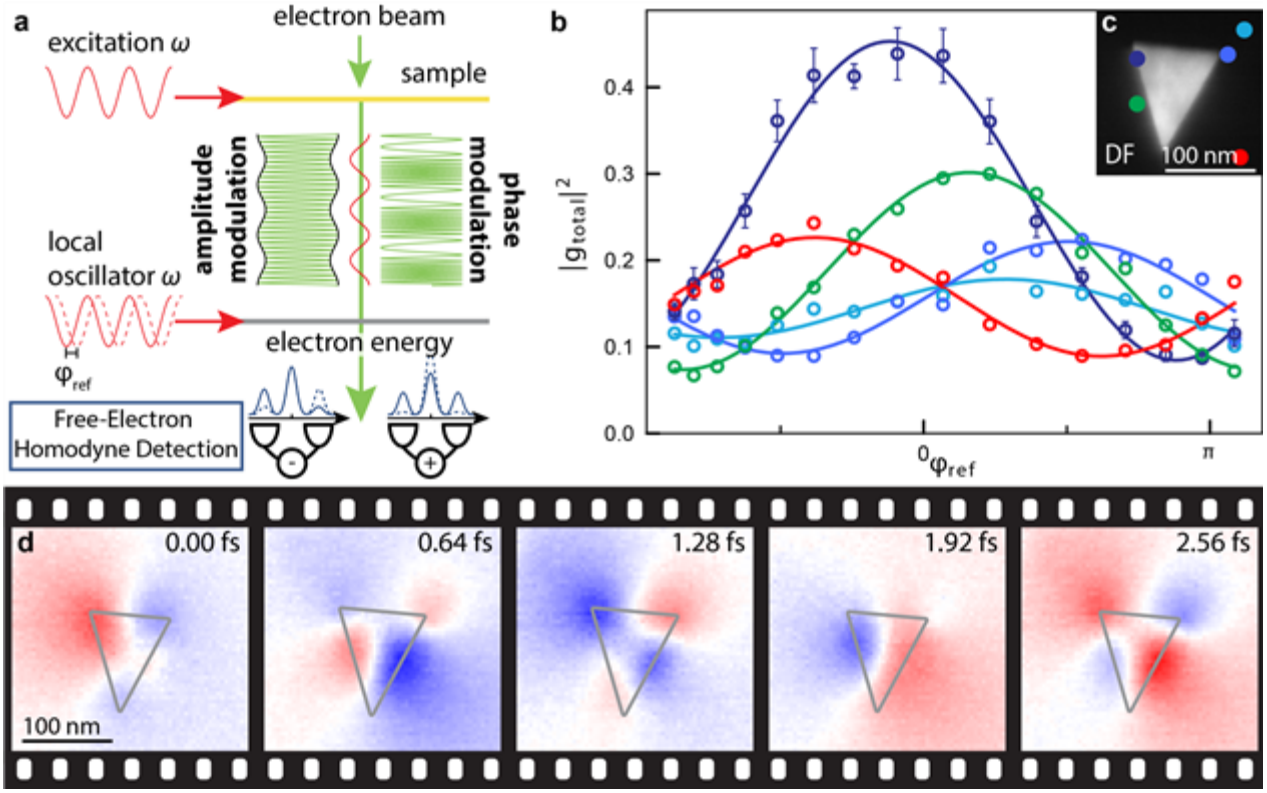
685

# Attosecond electron microscopy using free-electron homodyne detection

John Gaida[1,2], Hugo Lourenço-Martins[1,2], Murat Sivis[1,2], Thomas Rittmann[1,2], Armin Feist[1,2], F. Javier García de Abajo[3,4], Claus Ropers[1,2]
[1]Dept. of Ultrafast Dynamics, Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany, [2]4th Physical Institute, University of Göttingen, Göttingen, Germany, [3]ICFO-Institut de Ciencies Fotoniques, Castelldefels (Barcelona), Spain, [4]ICREA-Institució Catalana de Recerca i Estudis Avançats, Barcelona, Spain

IM-08, Lecture Theater 2, august 28, 2024, 10:30 - 12:30

The desire to map material structures and dynamics at intrinsic scales of ångströms and attoseconds is driving methodological advances in condensed matter science. While X-ray and electron methods provide structural detail, attosecond temporal resolution is being achieved by evolving optical spectroscopy techniques [1]. Photon-induced near-field electron microscopy (PINEM) enables the imaging of near-field intensities with nanometric resolution [2]. However, access to the evolution of nanoscale fields and structures within the light cycle requires sensitivity to the optical phase, e.g. by phase-contrast Lorentz PINEM [3], interferometric detection [4], or electron pulse bunching [5].
Here, we introduce Free-Electron Homodyne Detection (FREHD) as a universally applicable approach to electron microscopy of phase-resolved optical responses at high spatiotemporal resolution [4]. In this scheme, a phase-controlled reference interaction (Fig. 1a) serves as the local oscillator to extract arbitrary sample-induced modulations of a free-electron wavefunction. We demonstrate this principle through the phase-resolved imaging of plasmonic fields of a gold nanoprism (Fig. 1c) with few-nanometer spatial and sub-cycle temporal resolutions. The near field at a sample modulates the phase of the electron wavefunction, and this wavefunction modulation is amplified or attenuated for in-phase and anti-phase reference interactions, respectively, which allows for a coherent read-out of a position-dependent phase (Fig. 1b). The characterization of both the amplitude and phase of the near-field allows to image the electric field at different phases in a time-resolved movie (Fig. 1d).
In the area of phase-resolved near-field imaging, including scanning probe techniques and photoelectron emission, FREHD has distinct strengths. It is non-invasive, exhibits perfect linearity in response, and provides consistently high spatial resolution that is virtually independent of the sample and the optical wavelength. This resolution can be correlated with detailed structural characterization in transmission electron microscopy down to the atomic scale.
The method goes beyond imaging electromagnetic fields, and is also capable of detecting modulations imprinted on an electron beam by electronic or structural material responses. In particular, it encompasses attosecond charge density dynamics, including subtle light-induced changes in the structure factor at both the fundamental frequency and its harmonics.
In conclusion, we introduce Free-Electron Homodyne Detection that generalizes the high-resolution measurement of attosecond materials responses in electron microscopy, without a need for electron density bunching, and offers fascinating new possibilities to image local attosecond and phase-resolved responses on the nanometer scale.




**Keywords:**

Attosecond Lorentz phase-microscopy homodyne-detection holography

**Reference:**

[1] H. N. Chapman et al. “Femtosecond X-ray protein nanocrystallography,” Nature 470, 73–77 (2011).
[2] M.C. Scott et al. “Electron tomography at 2.4-ångström resolution,” Nature 483, 444–447 (2012).
[3] P.B. Corkum, and F. Krausz, “Attosecond science,” Nat. Phys. 3, 381–387 (2007).
[4] F. Calegari et al. “Advances in attosecond science,” J. Phys. B 49, 062001 (2016).
[5] B. Barwick, D. J. Flannigan, and A. H. Zewail, “Photon-induced near-field electron microscopy,” Nature 462, 902–906 (2009).

702

# Electron-photon quantum interaction enables novel ultrafast electron imaging approaches

Prof. Giovanni Maria Vanacore[1], Cameron J. R. Duncan[1], Beatrice Matilde Ferrari[1], Irene Ostroman[1], Maria Giulia Bravi[1], Tomer Bucher[2], Ron Ruimy[2], Ido Kaminer[2], Javier Garcia de Abajo[3], Thomas LaGrange[4], Fabrizio Carbone[4], Paolo Rosi[5], Enzo Rotunno[5], Vincenzo Grillo[5], Jean-Cristophe Olaya[6], Sang-Tae Park[7], Dan Masiel[7]
[1]Laboratory of Ultrafast Microscopy for Nanoscale Dynamics (LUMiNaD), Department of Materials Science, University of Milano-Bicocca, Milano, Italy, [2]TECHNION, Israel Institute of Technology, Haifa, Israel, [3]ICFO-Institut de Ciencies Fotoniques, Mediterranean Technology Park, Castelldefels (Barcelona), Spain, [4]LUMES, Ecole Polytechnique Federale de Lausanne (EPFL), Lausanne, Switzerland, [5]CNR-Nano S3, Modena, Italy, [6]Holoeye Photonics Ag, Berlin, Germany, [7]JEOL-IDES, Pleasanton, USA

IM-08, Lecture Theater 2, august 28, 2024, 10:30 - 12:30

Background incl. aims
Coherently controlling the electronic and structural properties of new generation materials and devices down to the sub-nm level with femtosecond precision is one of the ultimate challenges in quantum technology, and has the potential to engineer novel states of matter with exceptional macroscopic functionalities. Here, we respond to such pressing need by introducing a new paradigm where the interaction between light and free-electrons can be exploited for coherently manipulating electron beams [1,2], enabling dynamical investigation of materials with enhanced sensitivities to specific materials degrees of freedom, unattainable using conventional schemes. We develop innovative ultrafast electron microscopy (UEM) methods based on light-induced electron modulation aiming to observe, understand, and control the non-equilibrium behavior of low-dimensional systems at such ultrasmall and ultrafast scales, which would be otherwise hidden in standard UEM observations.

Methods
The playground for these experiments is a modified Ultrafast Transmission Electron Microscope (UTEM) with multiple optical accesses for: i) electron pulse generation, ii) electron wavefunction modulation, and iii) sample excitation. Arbitrary longitudinal and transverse modulation of a free-electron wave function is obtained using light fields properly modulated in space and time. In particular, a femtosecond electron pulse interacts with the chosen configuration of the optical distribution as generated via a spatial light modulator (SLM) (see schematics in the left panel of the figure), and the energy-momentum quantum exchanges between electron and photons resulting from such coupling are mapped in the electron multidimensional phase space. [3]

Results
In this contribution, we will first demonstrate how to coherently modulate the spatial, momentum, energy, and temporal distributions of the electron wave function with high versatility on different modulation basis. This derives from the technological development of a new device: a photonic-based modulator for dynamic multidimensional control of electrons. Then, we will show how our approach for versatile transverse and longitudinal electron modulation at the fs-timescale is crucial for the first time implementation of new imaging techniques (see schematics in the right panel of the figure), such as Ramsey-like holography [4] – for phase- and time-resolved imaging of hybridized low-energy modes in strongly-correlated systems – and Single Pixel Imaging [5] – that has been only recently proposed for optical detection and now is possible also in electron microscopy permitting

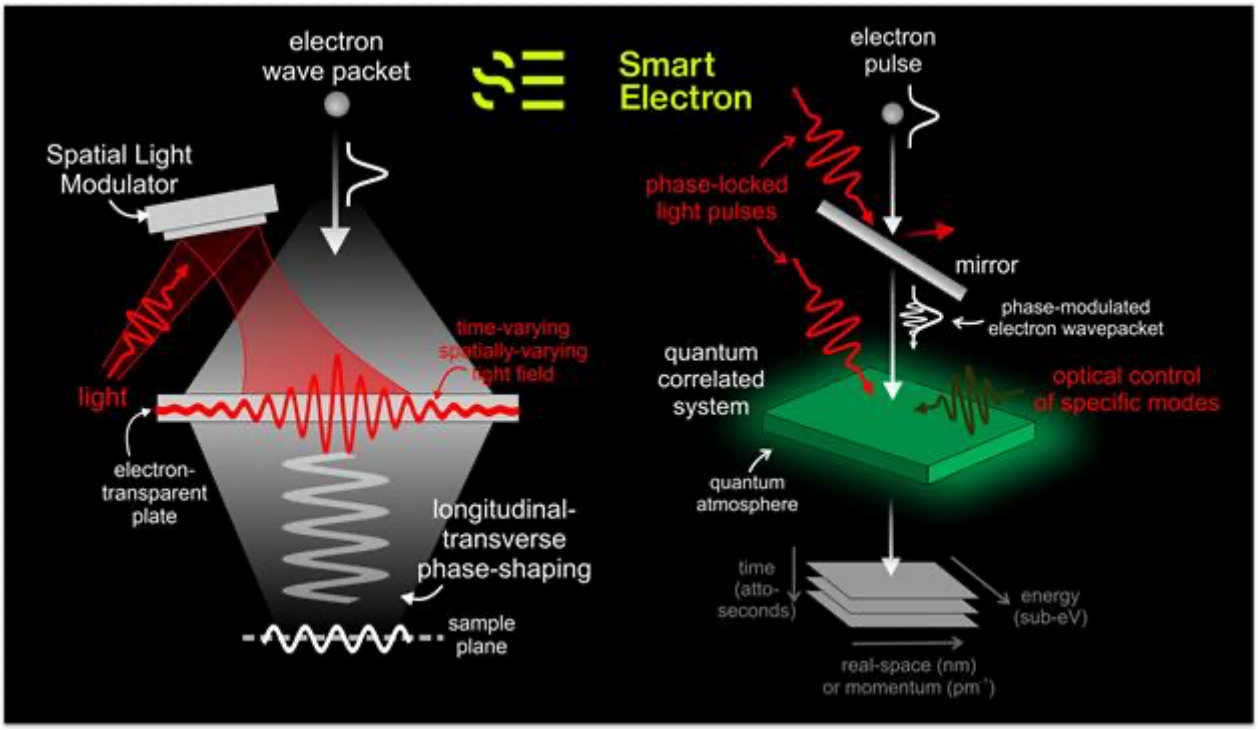
lower electron doses, faster acquisition rates for individual nanoscale objects in their natural environment.

Conclusion

In conclusion, we report on the realization of an all-optical rapidly-programmable phase mask for electrons, which allows us to implement new electron imaging approaches with enhanced sensitivities and performance, thus radically changing how matter is investigated in electron microscopy. This is based on a specific sensitivity to materials' inner coherence when light-modulated structured electron waves are adopted. Our results pave the way for achieving unprecedented insights into new generation materials and their dynamic behavior, impacting also future photonics and electronics applications. More generally, unlocking this potential will play a decisive role in our ability to address the grand challenges that the world is facing nowadays, especially regarding 'energy', 'information' and 'health'.

Funding:

This work is part of the SMART-electron project that has received funding from the EU Horizon 2020 Programme under GA No 964591.




**Keywords:**

UTEM; electron-wavefunction modulation; low-dimensional materials.

**Reference:**

[1] G. M. Vanacore, G. Berruto, I. Madan, E. Pomarico, P. Biagioni, R. J. Lamb, D. McGrouther, O. Reinhardt, I. Kaminer, B. Barwick, H. Larocque, V. Grillo, E. Karimi, F. J. García de Abajo, F. Carbone, Nature Materials 18, 573-579 (2019).
[2] G. M. Vanacore, I. Madan, F. Carbone, La Rivista del Nuovo Cimento 43, 567–597(2020).
[3] I. Madan, V. Leccese, A. Mazur, F. Barantani, T. LaGrange, A. Sapozhnik, P. M. Tengdin, S. Gargiulo, E. Rotunno, J.-C. Olaya, I. Kaminer, V. Grillo, F. J. García de Abajo, F. Carbone, and G. M. Vanacore, ACS Photonics 9, 3215 - 3224 (2022).
[4] T. Bucher, R. Ruimy, S. Tsesses, R. Dahan, G. Bartal, G. M. Vanacore, I. Kaminer, https://doi.org/10.48550/arXiv.2305.02727
[5] A. Konečná, E. Rotunno, V. Grillo, F. J. Garcia de Abajo, and G. M. Vanacore, ACS Photonics 10, 1463 - 1472 (2023)

748

# Development and characterization of a laser-driven cold-field emission source

Alexander Schröder[1], Mukai Masaki[2], Kohno Yuji[2], Sascha Schäfer[1,3]
[1]Department of Physics, University of Regensburg, Regensburg , Deutschland, [2]JEOL Ltd., , Japan, [3]Regenburg Center for Ultrafast Nanoscopy (RUN), Regensburg, Germany

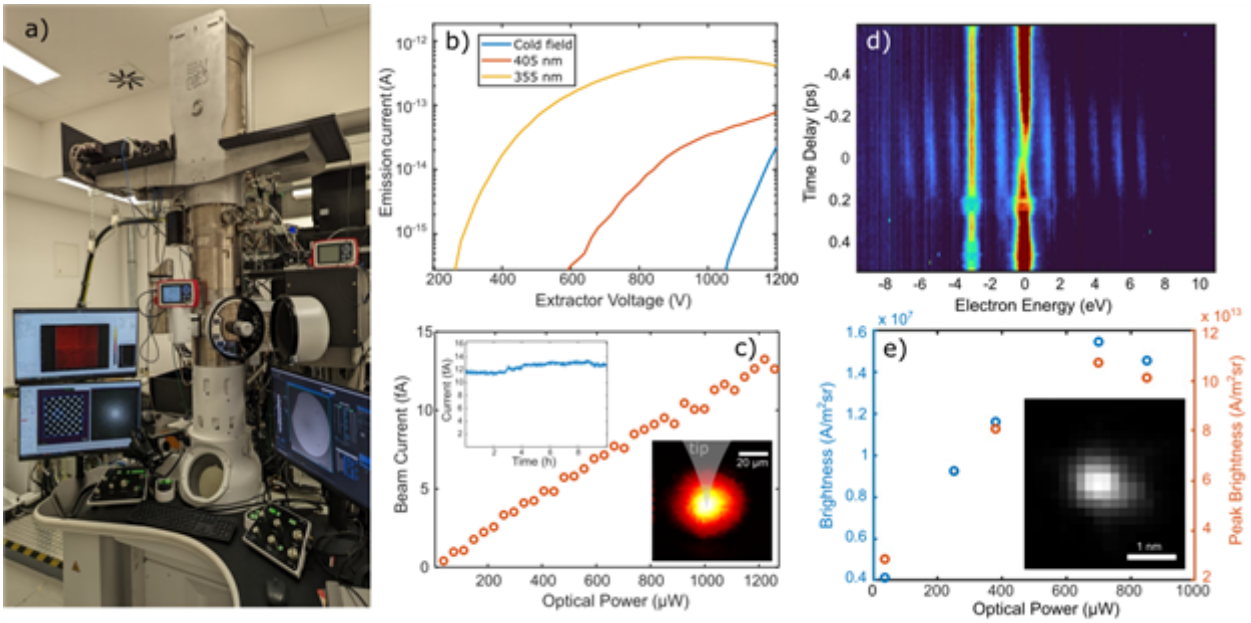
IM-08, Lecture Theater 2, august 28, 2024, 10:30 - 12:30

Background
Ultrafast transmission electron microscopy (UTEM) utilizing pulsed electron sources, has emerged as a crucial tool for probing ultrafast dynamics at the nanoscale. Recent breakthroughs in laser-driven Schottky electron emitters have significantly enhanced the spatial and temporal coherence of electron pulses, achieving temporal pulse widths below 200 fs and electron spot sizes as small as 1 nm [1]. The potential for further enhancement in pulse coherence is anticipated through the application of laser-driven cold-field emitters, especially when operated in a linear photoemission regime [2].
Methods
Here, we present the development and characterization of a long-term-stable laser-driven cold-field emitter source, designed for advanced ultrafast transmission electron microscopes and operating at 200 keV electron energy [3]. Based on a JEOL cold-field emitter gun (CFEG), the source utilizes a sharp single-crystalline (310) tungsten tip within a vacuum level of 10^-12 to 10^-11 mbar. For pulsed operation, the source is modified to allow for a direct optical line-of-sight onto the emitter through opposing optical entrance windows.
Results
In the initial implementations, we conducted a comprehensive characterization of the photoemission yield from the tip using a continuous-wave optical illumination at 3.59 and 3.06 eV photon energy focused to a 20 µm scale spot on the emitter apex (Fig. 1c, right inset). Notably, the employed photon energy is below the work function of tungsten so that linear photoemission is only feasible at the apex of the tip which exhibits a Schottky-reduced effective workfunction. Continuous field emission, following a Fowler-Nordheim description, is observed at large electric extraction fields without illumination (blue curve in Fig. 1b). Illumination at 1 mW optical power reveals an additional photoelectron contribution at low extraction field values, for which continuous field emission is absent (yellow and red curves in Fig. 1b). The onset of photocurrents is dependent on the photon energy of the illuminating light field. Above this threshold the current scales linearly with light intensity (Fig. 1c). Notably, under extreme high vacuum (XHV) conditions near the emitter, no significant drop in photoemission yield is observed over a 10-hour period (Fig. 1c, left inset).
In a subsequent step, the emitter is operated as part of the Regensburg JEOL F200 UTEM (Fig. 1a), using ultrashort laser pulses with a duration of 200 fs at a central wavelength of 345 nm. We systematically carried out an extensive pulse characterization, including the pulsed electron energy distribution, pulse duration, and the spatial attributes of electron brightness across varied excitation fields.
When operated at a state in which both, field- and photoemission can occur, the final kinetic energy of electron generate by the two emission processes are needly separated (Fig. 1d). Performing an electron-photon cross correlation we obtain 300 fs-electron pulse duration and energy width of about 300 meV.
Remarkably, our findings reveal that focal sizes of below 0.74 nm and a normalized peak brightness of 10^14 A/m²sr (Fig. 1e) can be achieved, surpassing the brightness of the laser-driven Schottky field emitter constructed in our lab by an order of magnitude. Currently, small electron focal spot sizes are limited to a low current-regime, due to the onset of spherical aberration at larger convergence

angles. It should be possible to further improve the transversal beam properties at higher pulse charges utilizing a probe aberration corrector.

Conclusion

In conclusion, we demonstrate the successful development of a stable, laser-driven cold-field emitter source for ultrafast transmission electron microscopy applications. Our findings illustrate the potential to significantly enhance electron beam brightness and reduce energy spread of electron pulses, paving the way for further advancements in probing ultrafast dynamics at the nanoscale.




**Keywords:**

UTEM
Electron-pulse
Laser-driven
Cold-field-Emitter

**Reference:**

[1] A. Feist et al., Ultramicroscopy 176, 63 (2017).
[2] F. Houdellier et al., Ultramicroscopy 186, 128 (2018).
[3] A. Schröder et al., to be submitted.

9

# Electrochemical lithium intercalation & exfoliation in 2D TMDs and its in-situ studies

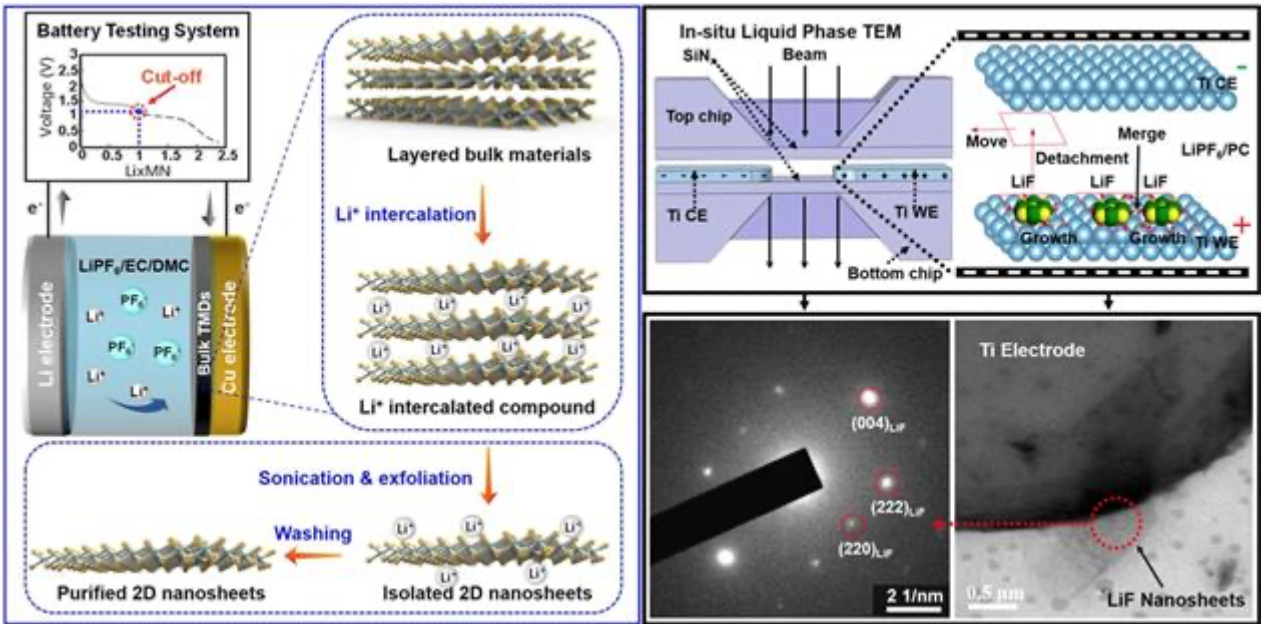
Zhiyuan Zeng[1]
[1]Department of Materials Science and Engineering, and State Key Laboratory of Marine Pollution, City University of Hong Kong, Kowloon, Hong Kong

Poster Group 2

We developed a lithium ion battery intercalation & exfoliation method with detailed experimental procedures for the mass production of 11 two dimensional TMDs and inorganic nanosheets, such as MoS2, WS2, TiS2, TaS2, ZrS2, graphene, h-BN, NbSe2, WSe2, Sb2Se3 and Bi2Te3, among them 3 TMDs achieved mono- or double layer yield > 90%. This method involves the electrochemical intercalation of lithium ions into layered inorganic materials and a mild sonication process. The Li insertion can be monitored and finely controlled in the battery testing system, so that the galvanostatic discharge process is stopped at a proper Li content to avoid decomposition of the intercalated compounds. The intercalation strategy can also be used to tune 2D TMDs' physical and chemical properties for various applications. For example, we developed an one-step covalent functionalization method on MoS2 nanosheets for membrane fabrication, which exhibited excellent water desalination performance. For lithium intercalation mechanism, the state-of-the-art In-Situ Liquid Phase TEM is an ideal technique for identifying the phase changes during intercalation process. With self-designed electrochemical liquid cell utilized, we can directly vapture the dynamic electrochemical lithiation and delithiation of electrode in a commercial LiPF6/EC/DEC electrolyte, such as LiF nanocrystal formation, lithium metal dendritic growth, electrolyte decomposition, and solid-electrolyte interface (SEI) formation. Combining with other in-situ techniques, such as in-situ XAS, XRD and Raman, etc, the underlying lithium intercalation mechanism in TMDs were further investigated, which render us a comprehensive understanding of the intrinsic correlation between the intercalation process and TMDs layered structures.

References

[1] R. Yang, L. Mei, Q. Zhang, Y. Fan, H. S. Shin*, D. Voiry*, Z.Y. Zeng*, High-yield production of mono- or few-layer transition metal dichalcogenides nanosheets by electrochemical lithium ion-intercalation and exfoliation method, Nature Protocols, 2022, 17, 358-377.
[2] R. Yang, Y. Fan, L. Mei, H. S. Shin, D. Voiry, Q. Lu, J. Li*, Z. Y. Zeng*, Synthesis of atomically thin sheets by the intercalation-based exfoliation of layered materials, Nature Synthesis, 2023, 2, 101-118.
[2] R. Yang, L. Mei, Y. Fan, Q. Zhang, H. G. Liao, J. Yang, J. Li*, Z. Y. Zeng*, Fabrication of Liquid Cell for In-Situ Transmission Electron Microscopy of Electrochemical Processes, Nature Protocols, 2023, 18, 555-578.
[3] Q. Zhang, J. Ma, L. Mei, J. Liu, Z. Li*, J. Li*, Z. Y. Zeng*, In-Situ TEM Visualization of LiF Nanosheets Formation on the Cathode-Electrolyte Interphase (CEI) in Liquid-Electrolyte Lithium-Ion Batteries, Q. Zhang, Matter, 2022, 5, 1235-1250.
[4] L. Mei, Z. Cao, T. Ying, R. Yang, H. Peng, G. Wang, L. Zheng, Y. Chen, C. Y. Tang, D. Voiry, H. Wang*, A. B. Farimani*, Z. Y. Zeng*, Simultaneous electrochemical exfoliation and covalent functionalization of MoS2 membrane for ion sieving, Advanced Materials, 2022, 34, 2201416.
[5] H. Peng, R. Wang*, L. Mei, Q. Zhang, T. Ying, Z. Qian*, A. B. Farimani, D. Voiry*, Z. Y. Zeng*, Transition Metal Dichalcogenide-Based Functional Membrane: Synthesis, Modification and Water Purification Applications, Matter, 2023, 6, 59-96.




**Keywords:**

In-situ TEM, 2D TMDs

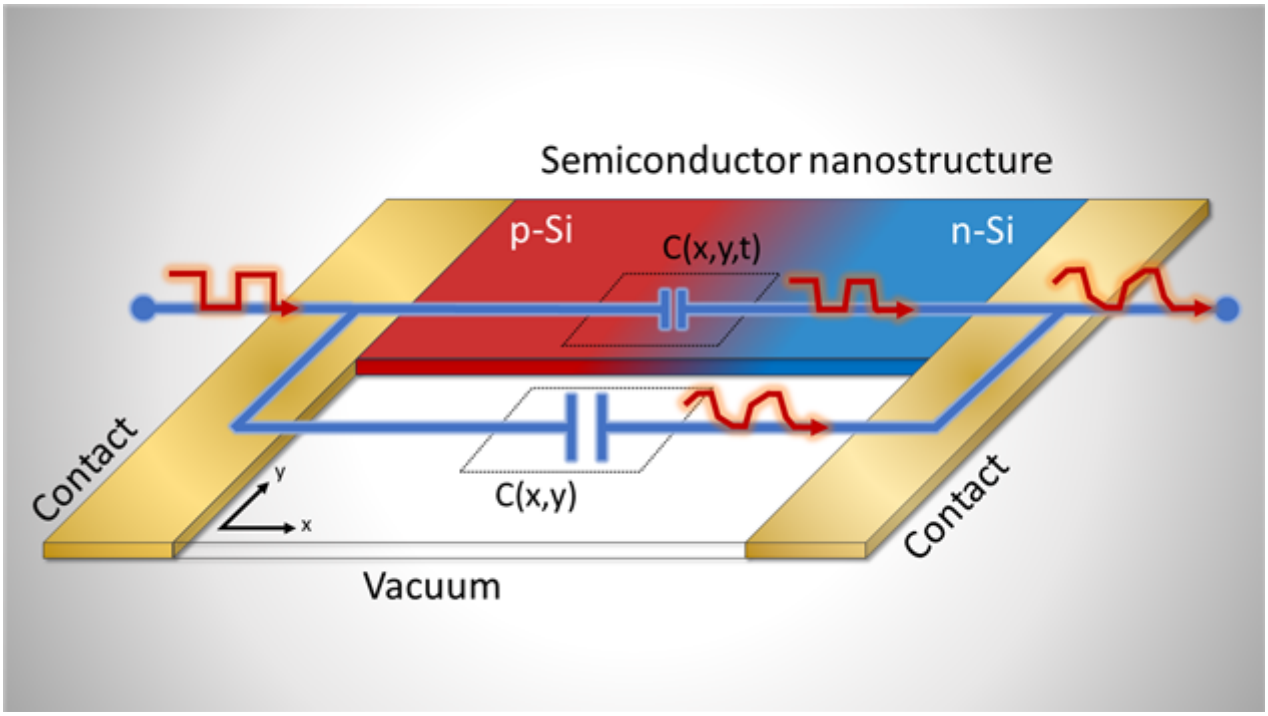
151

# Approaching picosecond temporal resolution in off-axis electron holography

Dr. Tolga Wagner[1], Hüseyin Çelik[2], Simon Gaebel[3], Dirk Berger[4], Christian M. Günther[4], Ines Häusler[1], Sebastian Siewert[2], Michael Lehmann[2], Christoph T. Koch[1]
[1]Department of Physics, Humboldt-Universität zu Berlin, Berlin, Germany, [2]Institute of Optics and Atomic Physics, Technische Universität Berlin, Berlin, Germany, [3]Max-Born-Institut, Berlin, Germany, [4]Center for Electron Microscopy (ZELMI), Technische Universität Berlin, Berlin, Germany

Poster Group 2

In the course of new technological and instrumental possibilities, time-resolved investigations in transmission electron microscopy are currently gaining particular relevance [1]. For phase-resolving methods, however, there are hardly any realizations for the investigation of dynamic processes on ultrashort time scales, although these are of great importance, especially for in-operando investigations of dynamic potential distributions in nanostructures (e.g. switching semiconductor or magnetic topological nanostructures).
Recently, interference gating (or iGate) has emerged as a simple, but robust method to address this circumstance [2]. It is based on the intentional destruction of the interference pattern for a large part of a measurement, utilizing short undisturbed time periods (called gates), which are synchronized to a periodic process under investigation, to generate time-resolved interferometric information. In particular, the simple technical realization of the method (only a two-channel signal generator, two coaxial cables, an dedicated aperture holder and an electrical biasing sample holder are required) enables low-barrier access to the investigation of ultrashort phenomena on existing instruments [3]. Through continuous improvements in sample preparation, automated measurement and reconstruction routines, control signal design, and the necessary electron-optical components, the time resolution achievable with iGate has recently broken through the picosecond barrier. By an application to silicon diodes, switching at repetition rates in the MHz range, variations of the local capacitance C(x,y,t) could be observed and the cause of the anomalous (MOS-cap-like) switching behavior of a defective semiconductor nanostructure was revealed [4]. A schematic representation of the experiment is illustrated in the accompanying figure. The nanostructured diode sits in between two extended metallic contacts, forming an overarching electrical system (e.g. parallel connected capacitances in vacuum C(x,y) and the diode C(x,y,t)).
In addition, the method itself could already be implemented in transmission electron microscopes in further laboratories in very short time frames (less than a work day), whereby the FPGA-based conversion of a JEOL JEM-2200FS into a versatile phase-resolving instrument with sub-nanosecond temporal resolution is discussed here in detail. The presented results will highlight the advantages of image-based investigations of projected dynamic potential distributions with nanometer and sub-nanosecond resolutions and their application to materials science problems, and pave a new path for decoding dynamic processes and their physical causes directly at the point of their occurrence.




**Keywords:**

electron-holography, time-resolved, ultrafast, semiconductor-nanostructures, interference-gating

**Reference:**

[1] A. Arbouet, et al, Advances in Imaging and Electron Physics (2018), 1-72.
[2] T. Niermann et al., Ultramicroscopy 182 (2017), 54-61.
[3] T. Wagner et al., Ultramicroscopy 206 (2019), 112824.
[4] T. Wagner et al., Phys. Rev. B 109 (2024), 085310.

208

# Imaging MEMS motion at nano scale by time-resolved scanning electron microscopy

Mohamed Zaghloul[1,2], Abbas Kosari Mehr[1,2], Riccardo Bertacco[1,3], Simone Cuccurullo, Federico Maspero[1], Giulia Pavese[4], Hao Chen[1,2], Aldo Ghisi[4], Alberto Corigliano[4], Silvia M. Pietralunga[2,5], Alberto Tagliaferri[1,2]
[1]Dipartimento di Fisica, Politecnico di Milano, Milano, Italy, [2]CNST@PoliMi, Istituto Italiano di Tecnologia (IIT), Milano, Italy, [3]Polifab, Politecnico di Milano, Milano, Italy, [4]Dipartimento di Ingegneria Civile e Ambientale, Politecnico di Milano, Milano, Italy, [5]Institute for Photonics and Nanotechnologies (IFN)—National Research Council (CNR), Milano, Italy

Poster Group 2

Background
Reconfigurable micro-optics and on-chip integrated photonics often rely on micro-opto-electro-mechanical-systems to provide dynamical optical processing such as beam steering and focusing or optical coupling and phase tuning. In the process of design and fabrication of micro-electro-mechanical-systems (MEMS), and to assess their quality, a local and direct measurement of their motion on a point-by-point basis would provide unprecedented supporting information. Such a technique must be fast enough to track the MEMS dynamics with sub-micron resolution.
Methods
We introduce and discuss the implementation of dynamical imaging of MEMS by time-resolved scanning electron microscopy (TR-SEM). MEMS resonators are actuated in-operando close to their resonance frequencies, and a synchronized comb of electron pulses is used to image stroboscopically the device at a controlled time delay with respect to the beginning of its oscillation period.
Results and conclusions
We demonstrate the acquisition of stroboscopic movies by a proper sequential acquisition of secondary electron signal. Unprecedented information about local trajectory is provided, in the microsecond scale and at tens of nanometer lateral scale. In-operando nonlinearities in the response of the system, interpretable as related to system hardening are brought into evidence. We also discuss strategies to reach the ultrafast time scale.

*Research supported by Project PE0000021, Concession Decree No. 1561 of 11.10.2022 adopted by Ministero dell'Università e della Ricerca (MUR), CUP, according to attachment E of Decree No. 1561/2022, Project title "Network 4 Energy Sustainable Transition – NEST".

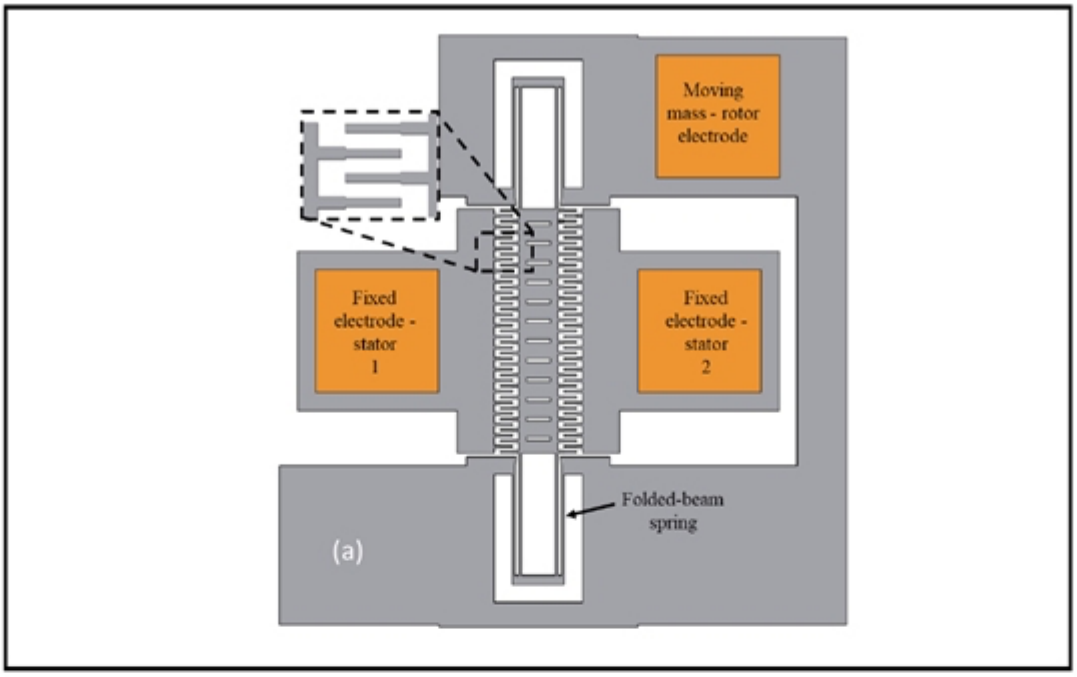



Figure 1. MEMS resonator (*layout*)

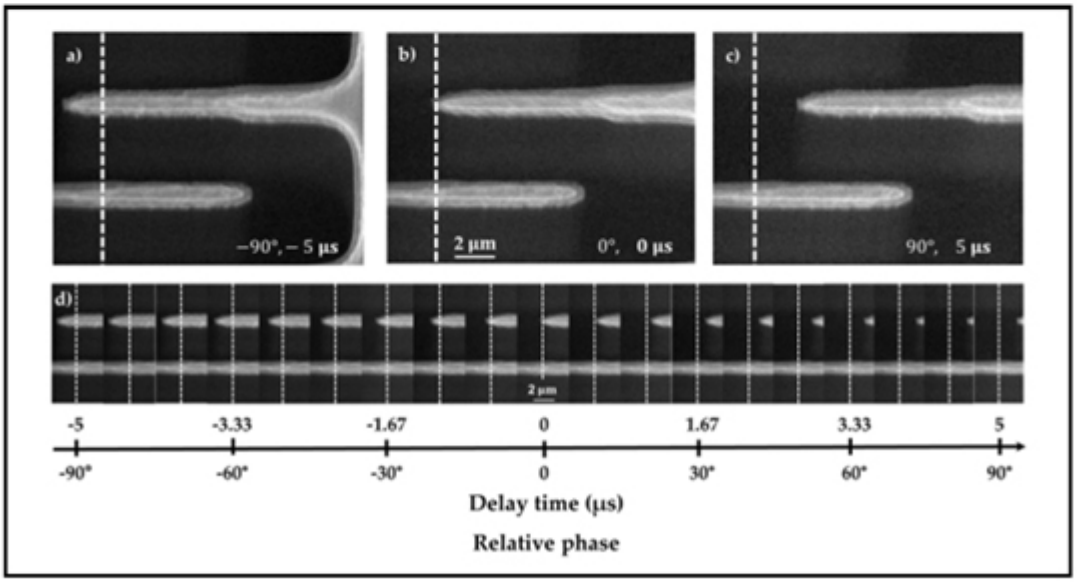



Figure 2. Stroboscopic TR-SEM delay time series, dynamics of the rotor tooth.

**Keywords:**

MEMS, SEM, Dynamics, Nanoscale, Strain

336

# Quantum phase modulation and retrieval in an ultrafast transmission electron microscope

Dr. Cameron Duncan[1], Beatrice Ferrari[1], Irene Ostroman[1], Maria Giulia Bravi[1], Sang Tae Park[2], Daniel Masiel[2], Jean Christophe Olaya[3], Eduardo Dias[4], Javier García de Abajo[4], Giovanni Maria Vanacore[1]
[1]Unversità degli Studi di Milano-Bicocca, Milan, Italy, [2]JEOL-IDES, Pleasonton, United States of America, [3]Holoeye Photonics, Berlin, Germany, [4]ICFO-Institude Ciencies Fotoniques, The Barcelona Institute of Science and Technology, Barcelona, Spain

Poster Group 2

Background incl. aims
Coherent phase modulation of the free-space electron wavefunction has the potential to dramatically extend the applications of electron microscopy by enabling, e.g., enhanced image contrast in Cryo-Electron Microscopy [1], Single Pixel Imaging [2] and quantum information processing [3]. Measuring the phase degree of freedom is challenging because conventional approaches to electron detection are sensitive to deposited energy. Pump-probe ultrafast electron microscopy allows for the implementation of phase retrieval algorithms by comparing pre and post-excitation images, using the pre-excitation image to define a reference phase. Here we demonstrate phase retrieval performed on a probe beam passing through a fast moving, dilute free electron gas.

Methods
Our specimen in pump probe experiments is shown in Fig. 1 a. and consists of a nanometer thin gold film suspended on a standard 3 mm gold TEM grid. We illuminate the specimen with point projection optics (i.e., Lorentz microscopy) and collect the scattered beam at long diffraction camera length, so that the electron intensity on the detector mixes momentum and real-space information, a critical experimental detail for enabling phase reconstruction. We pump the specimen with 1030 nm, 290 fs laser pulses, emitting a low energy cloud of electrons in the path of the probe beam. We record changes in the detected probe caused by the electron cloud as a function of pump-probe delay. The specimen geometry is invariant in one of the two directions transverse to the probe beam trajectory and we improve signal to noise by averaging along the invariant direction, as indicated in Fig. 1 a.

Results
Figure 1. b. shows the modulation of the transmitted beam intensity as a function of position on the sample and delay time. Relying on charge conservation, a deflection angle is computed from the local change in beam intensity as a function of delay as shown in Fig. 2, and from this deflection angle the phase-change of the electron beam is calculated. Figure 3. shows the phase shift as a function of delay time and position using a 2π periodic colormap.

Conclusion
We have demonstrated how the combination of pump-probe specimen modulation with point projection electron optics (Lorentz microscopy) enables phase retrieval. In subsequent experiments, phase retrieval has allowed us to go further and extract key experimental observables from our data, e.g., the charge density of the specimen gas.

This work is part of the SMART-electron Project that has received funding from the European Union's Horizon 2020 Research and Innovation Programme under Grant Agreement No. 964591.

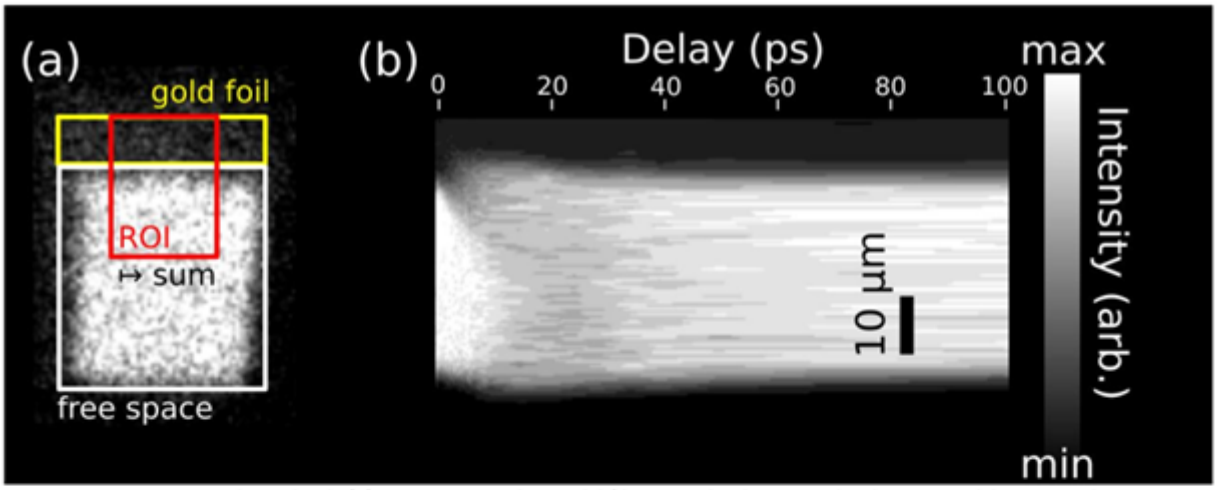



Figure 1. Experimental method. (a) Nanometer thin gold film attached to standard 3 mm gold TEM grid. Intensity is summed along the horizontal direction (b) Pump-probe data series, space on the vertical axis, time on the horizontal axis.

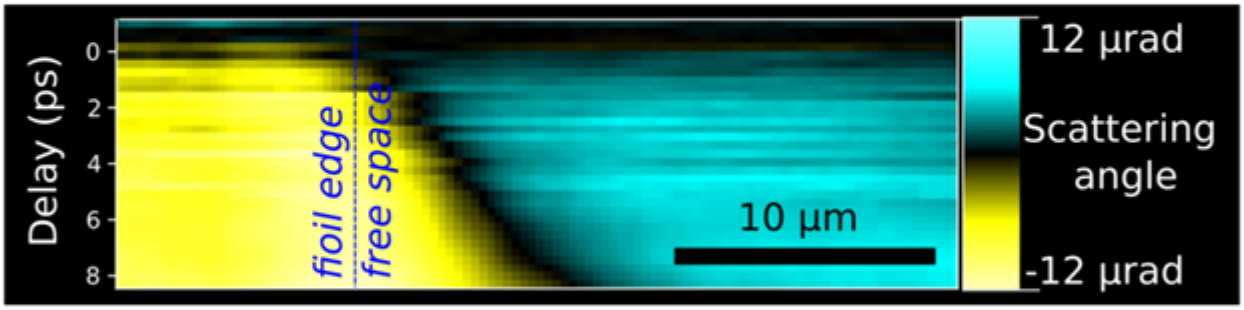



Figure 2. Electron scattering angle as a function of position in the specimen plane, computed from intensity shifts shown in Fig. 1 b. Space shown on the horizontal axis, time the vertical axis.

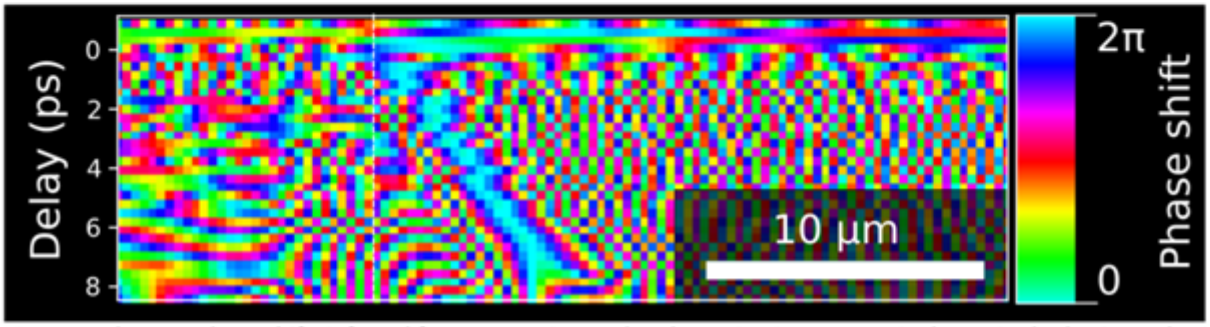



Figure 3. Electron phase shift, inferred from scattering angles shown in Fig. 2. Space is shown in the horizontal axis, time on the vertical axis.

**Keywords:**

Pump-probe electron phase retrieval

**Reference:**

[1] Du, Daniel X., and Anthony WP Fitzpatrick. "Design of an ultrafast pulsed ponderomotive phase plate for cryo-electron tomography," Cell Reports Methods 3.1 (2023).

[2] A. Konečná, et al. "Single-Pixel Imaging in Space and Time with Optically Modulated Free Electrons," ACS Photonics 10, 1463-1472 (2023).

[3] J. O. Reinhardt, et al. "Free-electron qubits," Annalen der Physik 533.2, 2000254 (2021).

553

# Laser particle acceleration in an ultrafast scanning electron microscope

Leon Brückner[1], Tomas Chlouba[1], Roy Shiloh[1], Stefanie Kraus[1], Julian Litzel[1], Peter Hommelhoff[1]
[1]Physics Department, Friedrich-Alexander-Universität Erlangen-Nürnberg, Erlangen, Germany

Poster Group 2

Background incl. aims
Ultrafast electron microscopy is rapidly approaching the limits of electron microscopes in terms of electron pulse length (typically hundreds of femtoseconds) and beam energy (up to 30 keV for a SEM and about 300 keV for a TEM). A miniaturized particle accelerator inside a microscope could overcome these limits by providing a beam of high-energy, ultrashort electron bunches. Dielectric laser acceleration (DLA) aims to achieve this by utilizing pulsed laser light for generating the accelerating field. The high damage threshold of dielectric materials could exceptionally high acceleration gradients of up to several GeV/m [1].
A DLA nanostructure typically consists of two rows of silicon pillars that are illuminated with an infrared laser pulse, which creates an oscillating nearfield mode with a phase velocity given by the period of the structure. Electrons are then injected into the channel in between the pillars with velocity (energy) corresponding to the phase velocity of the nearfield mode which ensures phase-matching and thus efficient energy transfer. By adding a buncher stage, additionally attosecond bunch trains can be generated over very short distances [2].

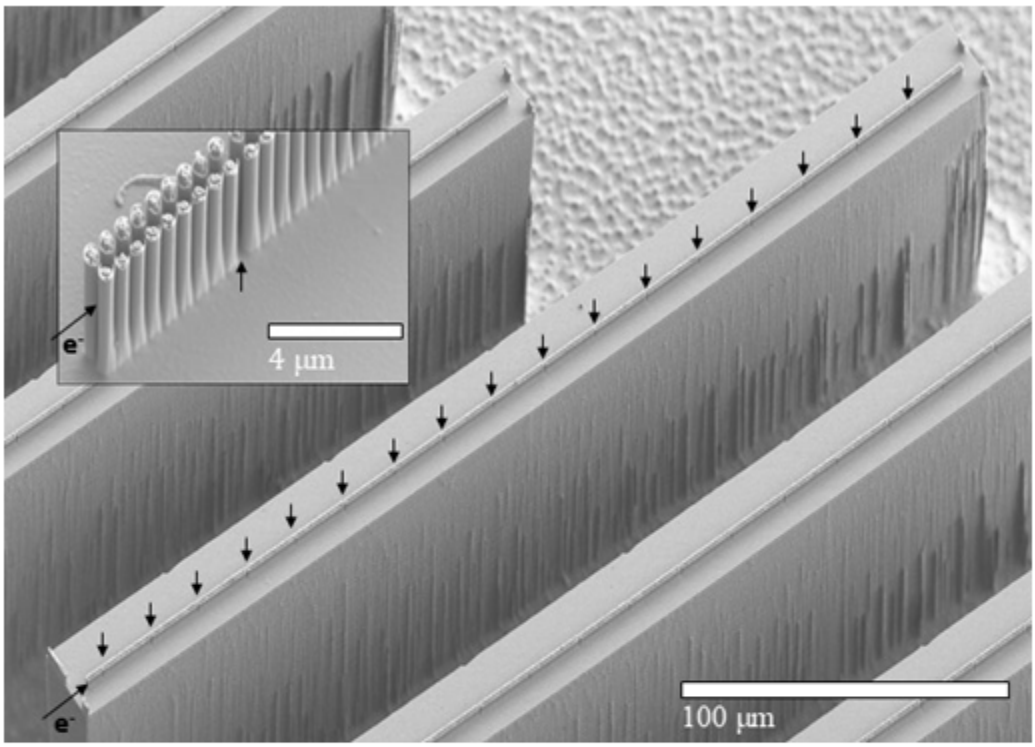
To allow acceleration over long distances, it is necessary to confine the particle beam transversally in order to prevent particle losses. This is realized by exploiting the transverse forces exerted by the nearfield. By incorporating periodic gaps in the structure design, we can instantly change the phase of the propagating electrons and thus alternate at will between transversally focusing and defocusing forces. With this scheme, beam transport over a distance of 78 µm has been demonstrated [3]. Combining guiding with a tapering of the structure period and the periodic gaps, we demonstrated coherent, phase-matched acceleration over long distances [4,5]. This enables significant energy gains, opening the door towards useful applications of these miniaturized accelerators.

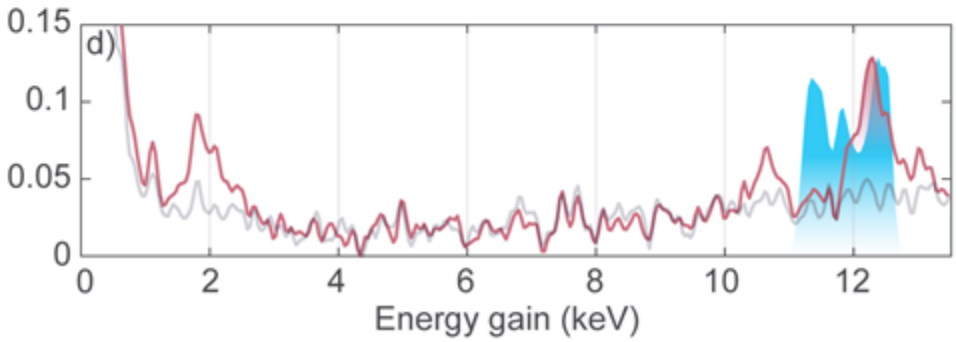
Methods
We have designed and fabricated a 500 µm long acceleration structure with an average gradient of 22.7 MeV/m [4]. In the experiment, an ultrafast SEM serves as the electron source. Ultraviolet laser pulses with a duration of 150 fs are focused onto the SEM tip cathode, emitting electron pulses with a mean energy of 28.4 keV and a pulse length of the order of 700 fs at the sample. The DLA structure is illuminated with a 1.93 µm, 250 fs long laser pulse with a pulse energy of 2.2 µJ that is focused to an elongated spot of 640 µm beam waist along and 13 µm transverse to the structure, creating the design peak field strength of around 600 MV/m. The laser pulse front is tilted by 71° to ensure optimal temporal overlap between electrons and the light field along the propagation. The electron energy is measured with a magnetic spectrometer.

Results
The accelerator demonstrates successful capture, confinement and acceleration of electron bunches. The 500 µm long acceleration structure shows an energy gain of the accelerated bunch of 12.3 keV, meaning a relative gain of 43% and a total energy of 40.7 keV, as predicted by simulation [4].

Conclusion

With careful design and extension of the interaction length, this energy gain could be increased much further, potentially enabling high-energy TEM imaging or electron diffraction in SEMs, for example. Exploring different combinations of dielectric materials with high damage thresholds and excitation wavelengths, particularly in the mid-infrared, will give access to acceleration gradients far in excess of those demonstrated here.




**Keywords:**

ultrafast electron microscopy, light-matter-interaction, lasers

**Reference:**

[1] R.J. England et al., Rev. Mod. Phys. 86, 1337–1389 (2014).
[2] N. Schönenberger, A. Mittelbach, P. Yousefi, J. McNeur, U. Niedermayer, P. Hommelhoff, Phys. Rev. Lett. 123, 264803 (2019)
[3] R. Shiloh, J. Illmer, T. Chlouba, P. Yousefi, N. Schönenberger, U. Niedermayer, A. Mittelbach and P. Hommelhoff, Nature 597, 498-502 (2021)
[4] T. Chlouba, R. Shiloh, S. Kraus, L. Brückner, J. Litzel and P. Hommelhoff, Nature 622, 476-480 (2023)
[5] P. Broaddus, T. Egenolf, D. S. Black, M. Murillo, C. Woodahl, Y. Miao, U. Niedermayer, R. L. Byer, K. J. Leedle, O. Solgaard, Phys. Rev. Lett. 132, 085001 (2024)

600

# Enhanced Free-Electron Wavefunction Modulation via Photon-Induced Near-Field Electron Microscopy (PINEM) with Shaped Light Fields

Beatrice Ferrari[1,7], Cameron J. Duncan[1], Irene Ostroman[1], Maria G. Bravi[1], Paolo Rosi[2], Enzo Rotunno[2], Jean-Cristophe Olaya[3], S. T. Park[4], D. Masiel[4], Vincenzo Grillo[2], Ido Kaminer[5], Francisco J. Garcia de Abajo[6], Thomas Lagrange[7], Fabrizio Carbone[7], Giovanni M. Vanacore[1]
[1]LUMiNaD UniMiB, Milano, Italy, [2]CNR-Nano, Modena, Italy, [3]HOLOEYE Potonics, Berlin, Germany, [4]JEOL IDES, Pleasanton, USA, [5]TECHNION, Haifa, Israel, [6]ICFO, Bacelona, Spain, [7]LUMES EPFL, Lausanne, Switzerland

Poster Group 2

Background and aims
Photon Induced Near-Field Electron Microscopy (PINEM) is a novel imaging technique that relies on the inelastic interaction between a free-electron beam and laser light, mediated by a thin film or a nanostructure [1]. This technique is generally employed to study the ultrafast dynamics of local fields generated by light-induced polaritonic excitations in nanoscale systems. Recently, we further exploited the quantized nature of electron-light interaction, that is intrinsic to PINEM, to achieve coherent control of the electron wavefunction [2-4]. So far, light-induced shaping has relied on Gaussian laser beams illuminating plasmonic nanostructures. In contrast, on-demand electron modulation for dynamic and versatile control has not yet been fully explored. Here, we show that shaped light can be used to introduce an arbitrary modulation on the transverse electron beam profile [5]. In fact, it is already possible to arbitrarily control light fields through spatial light modulators (SLMs) and our goal is to exploit PINEM to transfer this tunability to electron beams, thereby broadening the capabilities of ultrafast TEMs (UTEMs).

Methods
Our experiments are conducted in a UTEM, specifically a JEOL 2100 TEM modified as shown in Fig. 1. A 200 keV electron beam interacts with a femtosecond IR laser beam within a Photonic-based ELectron Modulator (PELM), an additional module of our TEM with an extra sample holder. The laser profile is shaped using a HOLOEYE Phase-Only SLM. We probe the electron-light interaction either by energy filtering imaging (UTEM @ EPFL) or by imaging the momentum distribution of the electron beam in High Dispersion Diffraction (HDD) mode with a 100-m camera length on a direct electron detector (UTEM @ UNIMIB).

Results
We used the SLM to induce a Hermite-Gaussian (HG) shaping of the laser profile and, through energy filtering, we imaged at different focal planes only the electrons that interacted with light. By comparing the obtained data with simulations of an analytically modeled system (Fig. 2), we determined the coherence length δ of the electron beam to be larger than 50 nm. This is a crucial parameter that sets an upper limit to the laser phase profile that can be completely transferred to the electron beam for coherent electron modulation. Additionally, we have directly imaged the momentum distribution of the electron beam and probed the transverse-momentum exchanges at the PELM plane following the electron interactions with quanta of the IR photon beam. In these measurements, we have reached values of transverse coherence on the order of $\delta \sim 1\ \mu m$. By acquiring momentum images in HDD mode with a micrometric aperture at the image plane, we have obtained simultaneous momentum-and-space-resolved information at the PELM plane. Fig. 3 shows the light-modulated electron-beam profile. The inset illustrates the reconstruction of the SLM-modulated laser-beam profile at the PELM plane.

## Conclusions

In conclusion, our findings demonstrate the potential of using shaped light fields in PINEM for enhanced tunability of electron modulation. The ability to control the electron wavefunction with such precision opens up new possibilities for the study of electron-light interactions. This could have far-reaching implications for various scientific domains, including life sciences, materials science, and nanotechnology. The rich dimensionality of light-electron interaction, spanning the transverse, longitudinal, temporal, and energy domains, harbors untapped potential and paves the way for future investigations in this dynamic field.

## Funding

This work is part of the SMART-electron Project that has received funding from the European Union's Horizon 2020 Research and Innovation Programme under Grant Agreement No. 964591.

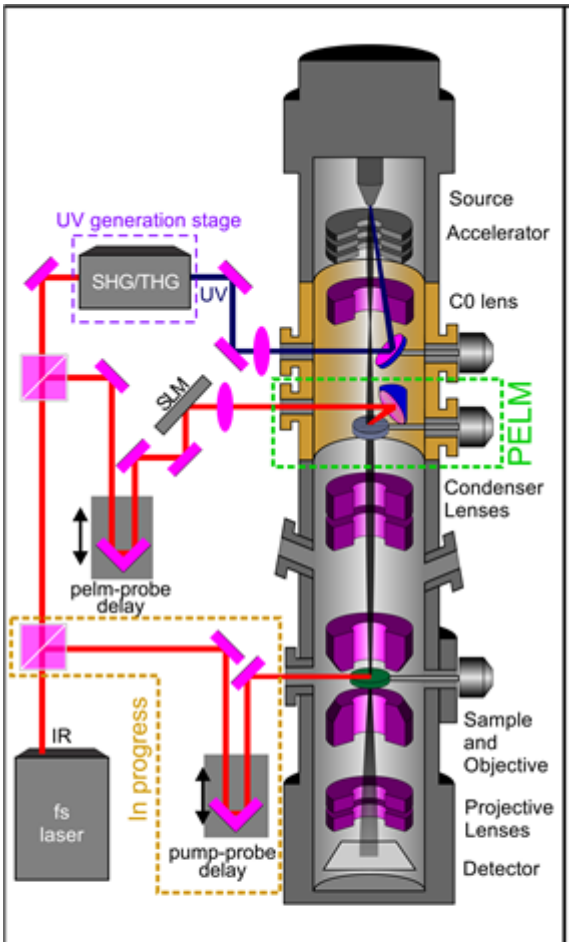



**Fig. 1 | Ultrafast TEM setup at UniMiB.** A pulse produced by a femtosecond IR laser is split in two. One pulse is converted into UV by 4th harmonic generation and it is directed to the TEM cathode to photoemit an electron pulse. The other pulse is syncronized with the electrons such that they reach the Photonic-based ELectron Modulator (PELM) at the same time and interact with each other. This light pulse is previously shaped with a Spatial Light Modulator (SLM) and it is used to dynamically shape the electron pulse. Currently, we are implementing a third laser branch to optically-excite transient states in a sample and to coherently control this excitations.

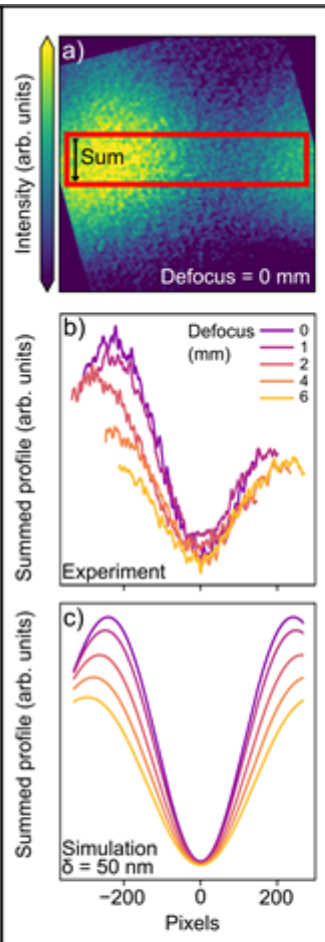



**Fig. 2 | Energy-filtered PINEM with shaped light.** a) Transverse profile of the HG-shaped electron-beam. b) Summed profiles obtained at different defoci. c) Simulations with an electron-beam transverse coherence patch δ of 50 nm. The minimum is preserved at different defoci, reproducing the experimental behaviour observed in panel b.

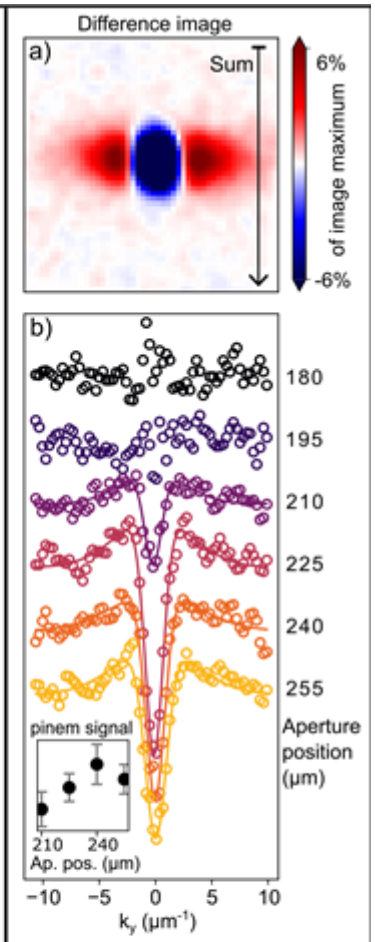



**Fig. 3 | Momentum-resolved PINEM with shaped light.** a) Diffence image of electron-beam momentum distribution at the PELM plane following light interaction. b) Summed profiles obtained with a 20-μm aperture placed at the sample plane at different positions (dots). Solid lines are least-square fits with two summed gaussians. Inset: variance of the positive fitted gaussian.

**Keywords:**

Photon-Induced Near-Field Electron Microscopy PINEM

**Reference:**

[1] Barwick, B., Flannigan, D. J., & Zewail, A. H. (2009). Photon-induced near-field electron microscopy. Nature, 462(7275), 902-906.

[2] Vanacore, G. M., Madan, I., Berruto, G., Wang, K., Pomarico, E., ... & Carbone, F. (2018). Attosecond coherent control of free-electron wave functions using semi-infinite light fields. Nature communications, 9(1), 269
[3] Vanacore, G. M., Berruto, G., Madan, I., Pomarico, E., Biagioni, P., ... & Carbone, F. (2019). Ultrafast generation and control of an electron vortex beam via chiral plasmonic near fields. Nature materials, 18(6), 573-579.
[4] Tsesses, S., Dahan, R., Wang, K., Bucher, T., Cohen, K., Reinhardt, O., ... & Kaminer, I. (2023). Tunable photon-induced spatial modulation of free electrons. Nature Materials, 22(3), 345-352.
[5] Madan, I., Leccese, V., Mazur, A., Barantani, F., LaGrange, T., Sapozhnik, A., ... & Vanacore, G. M. (2022). Ultrafast transverse modulation of free electrons by interaction with shaped optical fields. ACS photonics, 9(10), 3215-3224.

620

# Electron energy-gain spectroscopy of optical excitations in integrated photonic structures

Jan-Wilke Henke[1,2], Dr. Yujia Yang[3,4], F. Jasmin Kappert[1,2], Dr. Arslan S. Raja[3,4], Germaine Arend[1,2], Guanhao Huang[3,4], Dr. Armin Feist[1,2], Zheru Qiu[3,4], Rui Ning Wang[3,4], Aleksandr Tusnin[3,4], Dr. Alexey Tikan[3,4], Prof. Dr. Tobias J. Kippenberg[3,4], Prof. Dr. Claus Ropers[1,2]
[1]Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany, [2]University of Göttingen, 4th Physical Institute, Göttingen, Germany, [3]Institute of Physics, Swiss Federal Institute of Technolog Lausanne (EPFL), Lausanne, Switzerland, [4]Center for Quantum Science and Engineering, Swiss Federal Institute of Technology Lausanne (EPFL), Lausanne, Switzerland

Poster Group 2

Background
Optical shaping of electron beams, e.g. in the form of longitudinal attosecond bunching that promises increased temporal resolution [1], significantly extends the range of experiments possible in transmission electron microscopes (TEM). Based on inelastic scattering with an optical field, the momentum and energy of the electron are modified by absorption or emission of photons [2]. This inelastic interaction can, in turn, be employed to investigate the nano-optical response of samples with high spatial resolution in photon-induced near-field microscopy (PINEM) [3]. However, due to the weak coupling of free electrons and photons, inelastic field probing and beam shaping techniques so far required intense optical pulses and short electron pulses available in ultrafast TEMs.
Methods & Results
Here, we present the efficient modulation of a continuous electron beam by integrated photonics microresonators made from silicon nitride (Si3N4) that are optically pumped with a continuous-wave (CW) laser [4]. The fiber-coupled, chip-based resonator is placed inside a TEM, as illustrated in Figure 1a, such that the continuous electron beam can pass over the chip parallel to its surface before being analysed with an imaging spectrometer. Swift electrons interacting with the resonator's guided optical mode can absorb or emit photons from the laser field coupled to the resonator. This leads to the formation of electron energy sidebands spaced by the photon energy (~0.8eV, corresponding to ~1550nm) in the spectrum as shown in Figure 1b. The inelastic electron-light scattering is facilitated by the velocity matching of the electrons to the optical phase velocity as well as the high-Q resonant field enhancement. We characterise the latter by employing electron energy-gain spectroscopy (EEGS). To this end, the frequency of the CW pump laser is scanned across the cavity resonance at a low input power while electron spectra are recorded in parallel. We retrieve the laser detuning-dependent electron-light coupling strength (Fig. 1c) that exhibits a linewidth of 390 MHz corresponding to a spectral feature of only 3.1µeV width. From this EEGS trace, we infer a cavity quality factor of 7.7*105.
Increasing the optical pump power coupled to the microring resonator, the inherent nonlinearity and anomalous dispersion cause the parametric generation of new optical frequencies via four-wave mixing. We observe the formation of various nonlinear optical intracavity states whose spectral and temporal properties strongly depend on the laser detuning from the cavity resonance frequency. When scanning the laser across the resonance at a power > 100mW, we can thus perform an EEGS measurement on the nonlinear optical states by recording electron energy spectra in parallel [5]. The resulting electron spectral trace, shown in Figure 1d, exhibits prominent changes when entering different nonlinear optical states (marked by dashed white lines). For stable and chaotic intensity modulations (regions 1 and 2), resulting from the superposition of different optical wavelengths, averaging over different instantaneous interaction strengths leads to a smoothening of the electron spectra. However, the interaction of electrons with dissipative Kerr solitons (region 3), self-stable short optical pulses with a broad spectrum, yields a broad, low-intensity plateau and a strong central

peak since only a fraction of electrons interacts with the high-intensity pulse and scatters to high energy changes.

Conclusions

In conclusion, we characterise the inelastic interaction between electrons and the optical mode of an integrated photonics microresonator. By performing EEGS on one of the cavity resonances, we achieve an unprecedented energy resolution that might be transferred to both the study of material excitations as well as the probing of quantum optical excitations with free electrons. The observed strong interaction of a continuous electron beam with a low-power CW laser, moreover, enables efficient longitudinal electron beam modulation with optical fields in a conventional TEM setup. Harnessing the toolbox of optical waveform shaping in integrated photonics, we employ the multicolour fields of optical frequency combs and their impact on the electron energy spectra upon interaction to further extend these beam-shaping capabilities.

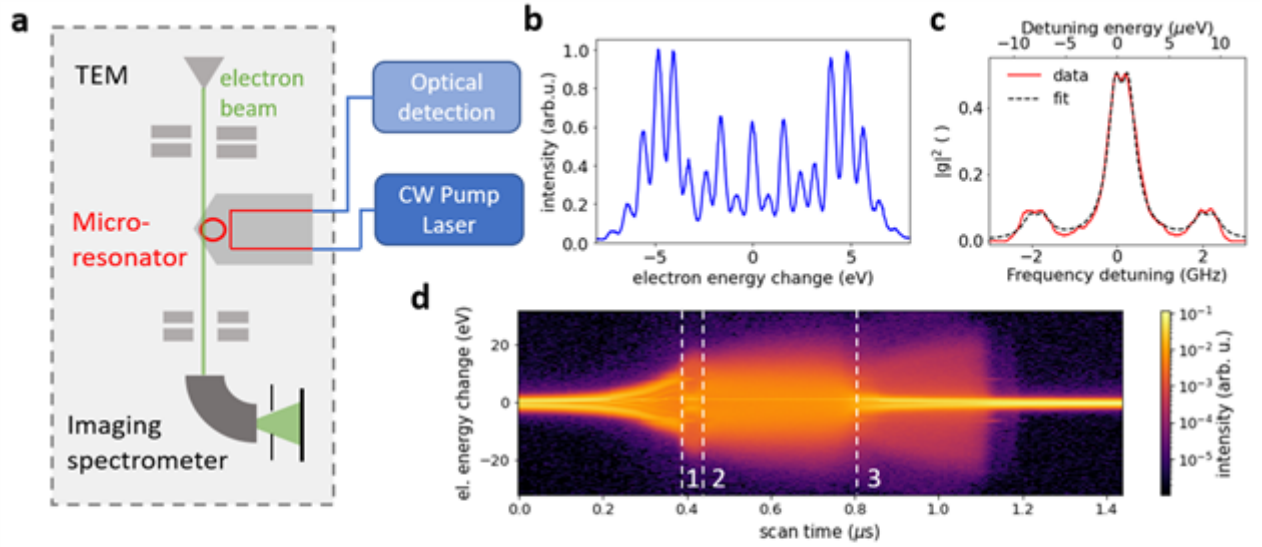



**Figure 1:** a) Schematics of the experimental setup. The fiber-coupled microresonator chip is placed inside the transmission electron microscope (TEM) and pumped with a CW laser. The electrons pass over the resonator surface and interact with the optical mode. Changes to the electron energy distribution are measured using an imaging spectrometer. b) Electron energy spectrum after interaction with a resonator mode pumped with a CW laser. c) EEGS trace of a weakly pumped cavity mode recorded with a frequency-tuneable laser and exhibiting a linewidth of 390 MHz, corresponding to 3.1μeV. d) At higher optical input power, the EEGS trace shows distinct changes to the electron spectrum when entering different nonlinear optical regimes (marked by white dashed lines).

**Keywords:**

EEGS, UTEM, Inelastic Electron-Light Scattering

**Reference:**

[1] K.E. Priebe et al., “Attosecond electron pulse trains and quantum state reconstruction in ultrafast transmission electron microscopy”, Nature Phot. 11, 793-797 (2017)
[2] B. Barwick et al., “Photon-induced near-field electron microscopy”, Nature 462, 902-906 (2009)
[3] A. Polman, M. Kociak & F.J. Garcia de Abajo, “Electron-beam spectroscopies for nanophotonics”, Nature Materials 18, 1158-1171 (2019)
[4] J.-W. Henke et al. “Integrated photonics enables continuous beam electron phase modulation”, Nature 600, 653-658 (2021)
[5] Y. Yang et al., “Free-electron interaction with nonlinear optical states in microresonators”, Science 383, 168-173 (2024)

696

# Coulomb-correlated multi-electron states in a transmission electron microscope beam

Rudolf Haindl[1,2], Dr. Armin Feist[1,2], Till Domröse[1,2], Dr. Marcel Möller[1,2], John H. Gaida[1,2], Dr. Sergey V. Yalunin[1,2], Prof. Dr. Claus Ropers[1,2]
[1]Department of Ultrafast Dynamics, Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany, [2]4th Physical Institute -- Solids and Nanostructures, Göttingen, Germany

Poster Group 2

Background incl. aims
In contrast to possibilities arising from electron correlations in condensed matter systems, stochastic Coulomb interactions in free electron beams are usually considered detrimental. In electron microscopy, electron repulsion leads to stochastic longitudinal and transverse emittance growth and limits the brightness of state-of-the-art electron sources. However, in a regime where few-body interactions dominate, Coulomb interactions can also have beneficial effects based on strong inter-electron repulsion, such as non-Poissonian electron beam statistics [1]. A powerful approach to induce such strong electron-electron interactions is femtosecond-triggered photoemission from nanotips [2, 3].
In this work [4], we characterize the strong Coulomb correlations of multi-electron states from a laser-triggered nanoscale field emitter, with applications in correlated probing applications, shot-noise reduction, and electron heralding schemes.

Methods
The experimental work is carried out in transmission electron microscopes that are modified for photoexcitation of the electron source [4, 5]. A laser pulse train with 2 MHz repetition rate and 160 fs pulse length generates ultrashort electron pulses by close-to-threshold laser-triggered Schottky emission [5]. The photoelectrons are analyzed with an event-based electron detector, which is mounted behind an imaging energy filter and characterizes the spectral and spatial properties of the electrons. The detected electrons are matched to the generating photoemission laser pulse, and the number of electrons n=1,2,3,4 is assigned to each pulse (see Fig. a).

Results
The single electron state spectrum exhibits a single peak at the beam energy of 200 keV (see Fig. b). Strikingly, the electron pulses with n>1 exhibit a distinctive spectral shape with the number of peaks identical to the number of electrons in the pulse (see Fig. c-e), indicating a strong interaction between the electrons in the pulse. The n=2,3,4 -states are plotted with respect to the state-averaged energy. For n=2, a projection of the energies of electrons A and B onto a two-dimensional energy pair histogram (Fig. f) shows a characteristic pair-correlation energy of about 2 eV. The strong inter-particle energy exchange is caused by acceleration-enhanced inter-particle Coulomb interaction, as confirmed by trajectory simulations. State-sorted beam caustics reveal a discrete increase in virtual source size, a longitudinal source shift, and a pronounced angular distribution of the few-electron states compared to single-electron pulses. As the correlation primarily emerges in the initial acceleration stages of the electron gun, controlling the electrostatic configuration enables control over the ratios of transverse and longitudinal Coulomb correlations. Using apertures along the optical axis, which limit the maximum transverse momentum of electrons transmitted into the column, we can decrease the current-current correlation function by lowering the extraction voltage applied to the nanotip cathode.

Conclusion

The high fidelity of few-electron pulses in conjunction with single-particle detection enables the preparation of distinctive electron states for correlated probing, as well as enhanced microscopy and lithography. Specifically, we propose schemes to generate an electron beam with sub- or super-Poissonian two-electron statistics. By means of spatial filtering with an annular aperture or spectral filtering with either a slit or a beam stop, the two-electron state is suppressed or enhanced, resulting in a shot-noise- (annular aperture, slit) or singlet-state-reduced (beam stop) electron beam, respectively. Applications of the two-electron state-enhanced electron beam include heralded single-electron sources and may foster new developments in free-electron quantum optics and quantum-enhanced electron microscopy.

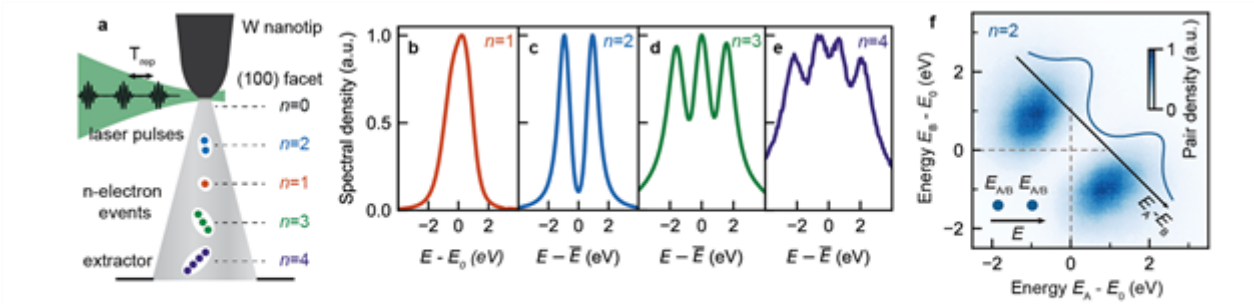



*Figure 1 a) The photoexcitation laser generates multi-electron states via photoemission from a Schottky emitter. b-e) Event-resolved spectra with a distinctive spectral shape for n=1-4. f) The energy pair histogram of n=2-states with electrons A and B shows a spectral correlation of about 2 eV.*

**Keywords:**

UTEM, Electron correlations, Electron sources

**Reference:**

1. H. Kiesel, A. Renz, F. Hasselbach, Nature 418, 392-394 (2002).
2. P. Hommelhoff, Y. Sortais, A. Aghajani-Talesh, and M. A. Kasevich, Phys. Rev. Lett. 96, 077401 (2006).
3. C. Ropers, et al., Phys. Rev. Lett. 98, 043907 (2007).
4. R. Haindl, et al., Nat. Phys. 19, 1410–1417 (2023).
5. A. Feist, et al., Ultramicroscopy 176, 63–73 (2017).

881

# Towards electron beam shaping in an Ultrafast Transmission Electron Microscope

Valentin Rollo[1], Sebastien Weber[1], Arnaud Arbouet[1]
[1]CEMES - CNRS, Toulouse, France

Poster Group 2

Background incl. aims
In 1933, Ernst Ruska developed the first Transmission Electron Microscope (TEM) which became the primary tool for studying matter. The picometer De Broglie wavelength and the numerous signals generated by an electron allow us to probe many properties of a sample at the atomic scale. Indeed, by analyzing the amplitude, phase, deflected angle and the energy of the electron beam after its interaction with the sample we can fully reconstruct its chemical composition or its magnetic/electric properties. However, unlike conventional optics, where the amplitude, phase and polarization of a light beam can be fully controlled using e.g. a Spatial Light Modulator (SLM), the electron beam cannot be easily manipulated in a TEM, making electrons insensitive to certain fundamental properties such as chirality. The development of Ultrafast Transmission Electron Microscopes (UTEM), whose working principle is illustrated on figure 1, has opened new possibilities. In 2020 it has been proposed to shape the electron beam through inelastic electron-light interactions in Photon-Induced Near field Electron Microscopy (PINEM) experiments [1] as sketched in figure 1. The PINEM interaction, based on a pump-probe experiment, relies on energy exchanges between an ultrashort electron pulse and a strong optical near-field induced on a sample by a femtosecond laser pulse. One of the main properties of this interaction is to imprint the laser beam phase on the wavefunction describing the electron after the interaction. Two years later, a first practical demonstration [2] illustrated the capability of this technique to generate Gaussian/Hermite-Gaussian electron beams.
The aim of our work is to develop an experimental set up for electron beam arbitrary shaping on a brand-new UTEM developed at CEMES [3]. This new 300 keV UTEM based on a modified Cold Field-Emission Electron Gun (CFEG) has been equipped with two sample stages that will enable us to use the shaped electron beam to probe a sample of interest.
Methods
This work is divided in three mains parts. First, we will develop the optical set up to spatially shape the laser beam by means of a Spatial Light Modulator (SLM) and fully characterize (amplitude and phase) the laser beam before its injection into the Ultrafast TEM (UTEM). In order to use the full potential of the SLM, we will implement various phase retrieval algorithms depending on the needs i.e. control over the intensity/phase distribution or both. In the second part of the project, we will perform Electron Energy Gain Spectroscopy (EEGS) experiments on our UTEM with a standard spatial shape of a laser beam i.e. a gaussian beam. The last step is to add the laser beam shaper set up in order to use the properties of the inelastic electron-light interaction in PINEM experiments to shape the electron beam.
Results
For the time being, the optical set up already enables us to generate any desired spatial distribution of a laser beam such as vortex, Laguerre-Gaussian beams or more generally any intensity and phase distribution – characterized thanks to a Mach Zehnder interferometer - in excellent agreement with the numerical simulations. Moreover, our new 300 keV UTEM based on a modified CFEG is operational and allows us to perform PINEM experiments. The electron beam shaping experiments are currently under active development. In parallel, we have developed numerical simulations which combine diffraction theory and electrodynamical simulations using the Green Dyadic Method (pyGDM (python package) [4]) to anticipate our future experiments. These calculations allow us to

compute the electron wave function after its interaction with the shaped laser beam and its subsequent propagation in the TEM column.

Conclusion

The double sample stage available on our UTEM will allow us to use the properties of the inelastic electron-light interaction in PINEM experiments to shape the electron beam on the first sample stage in order to probe a sample of interest on the second stage. This poster presentation will mainly be focused on the optical part of the experiment i.e. the generation of shaped laser beams with arbitrary amplitude and phase distributions and also on the numerical simulations of the electron beam shaping experiments that will be realized on our brand new UTEM.

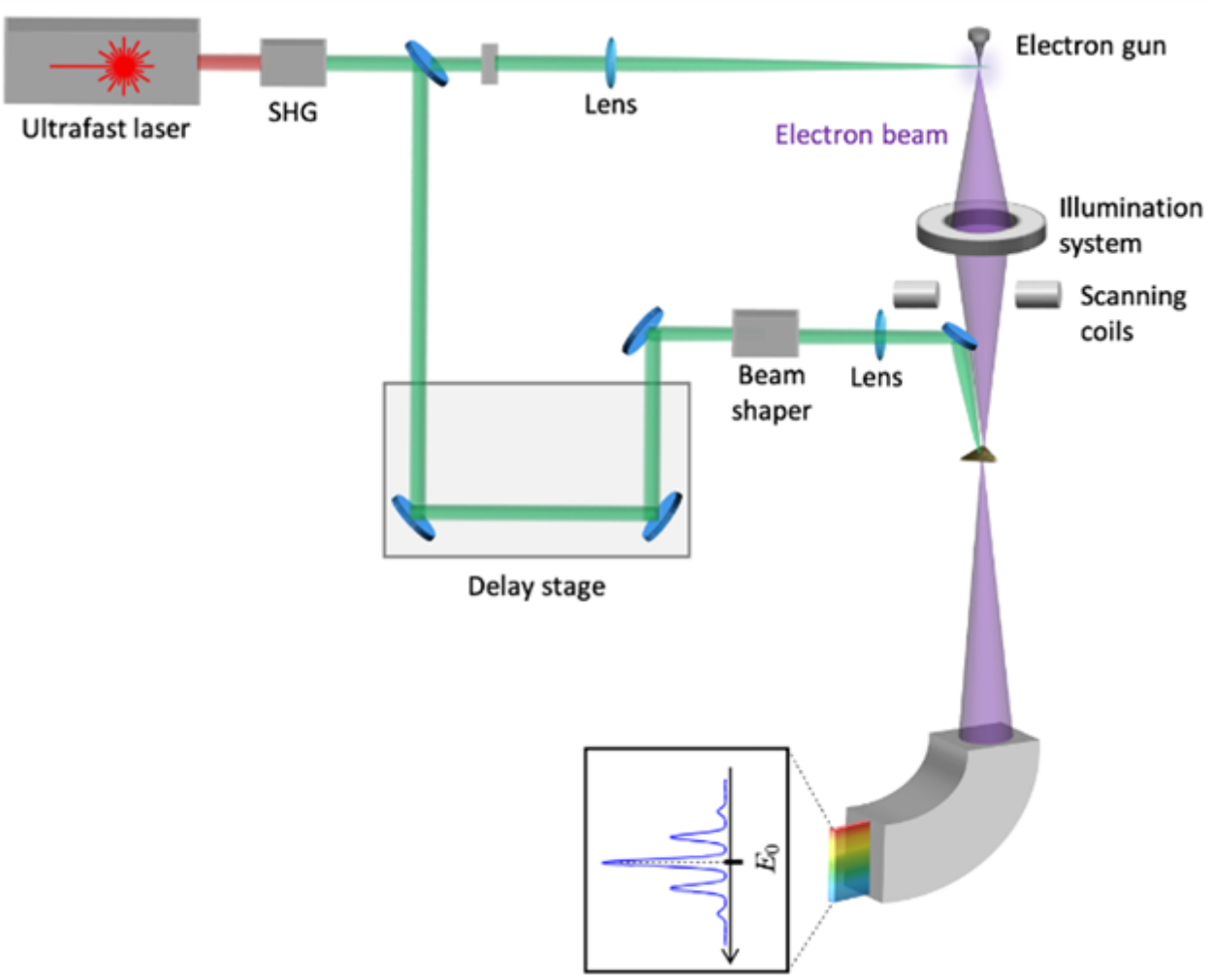



**Fig. 1**: Ultrafast Transmission Electron Microscope and PINEM schematics.

**Keywords:**

UTEM
Electron-beam shaping
PINEM
SLM

**Reference:**

[1] Andrea Konečná and F. Javier García De Abajo. Electron Beam Aberration Correction Using Optical Near Fields. Physical Review Letters, 125(3):030801, July 2020.

[2] Ivan Madan, Veronica Leccese, Adam Mazur, Francesco Barantani, Thomas LaGrange, Alexey Sapozhnik, Phoebe M. Tengdin, Simone Gargiulo, Enzo Rotunno, Jean-Christophe Olaya, Ido Kaminer, Vincenzo Grillo, F.

Javier García De Abajo, Fabrizio Carbone, and Giovanni Maria Vanacore. Ultrafast Transverse Modulation of Free Electrons by Interaction with Shaped Optical Fields. ACS Photonics, 9(10):3215–3224, October 2022.

[3] Houdellier F, Caruso GM, Weber S, Kociak M, Arbouet A. Development of a high brightness ultrafast Transmission Electron Microscope based on a laser-driven cold field emission source. Ultramicroscopy. 2018 Mar;186:128-138. doi: 10.1016/j.ultramic.2017.12.015. Epub 2017 Dec 27. PMID: 29306810.

[4] Peter R. Wiecha. pyGDM – a python toolkit for full-field electro-dynamical simulations and evolutionary optimization of nanostructures. Computer Physics Communications, 233:167–192, 2018

888

# Free electron spectroscopy seen through the prism of quantum optics

Tom Fraysse[1], Hugo Lourenço-Martins[1]
[1]CEMES-CNRS, Toulouse, France

Poster Group 2

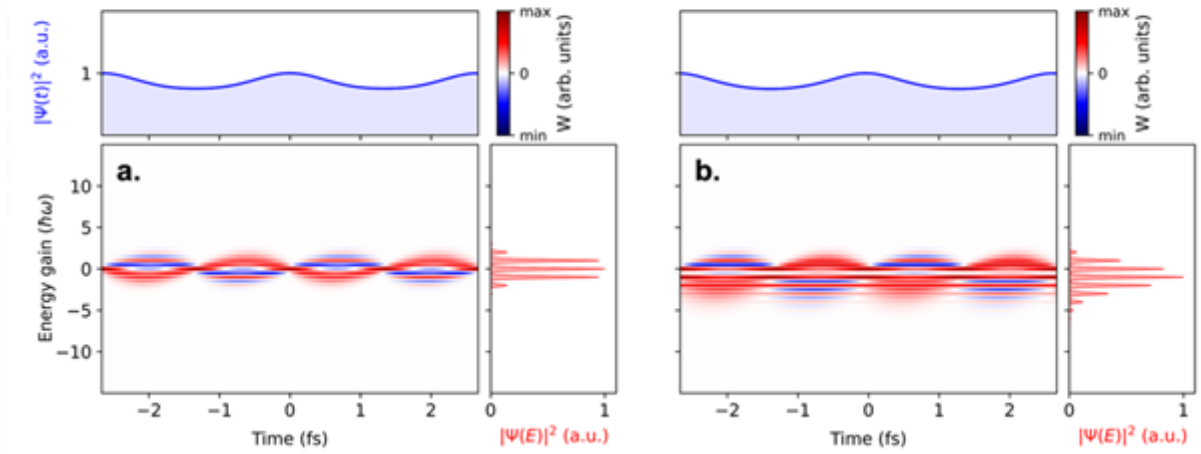
Background incl. aims
Photon Induced Near-field Electron Microscopy (PINEM) denotes the modulations of free electrons by a sample – typically an optical cavity – pumped by a laser source. This effect has been theoretically predicted and experimentally demonstrated in an ultrafast transmission electron microscope (UTEM) more than a decade ago [1]. Following these seminal developments, PINEM has been used to reconstruct the temporal dynamics of nano-optical systems with an attosecond temporal resolution [2].
Due to recent experimental developments in integrated photonics [3], the problem of PINEM in the low occupation regime – i.e. when the cavity is populated by a weak number of photons - has drawn an increasing theoretical interest. Indeed, in this situation, the classical description 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 [4,5] and predicted strong quantum mechanical effects in PINEM. The goal of our work is to pursue this effort and provide an alternative and intuitive approach to this problem by using tools borrowed from the field of quantum optics.
Methods
The goal of our study is thus to determine the electron-cavity state before and after inelastic interaction. To do so, following a standard quantum optics description, we describe the cavity state as a quantum superposition of number (or Fock) states. We then apply perturbation theory to calculate the electron Wigner function after interaction with the cavity state. The latter is the central object of our theory and represents - roughly speaking - a probability distribution of the electron in the phase space (energy-time). It constitutes a powerful visualization tool enabling us to understand the mechanisms of electron-light interaction at the quantum scale, as shown on figure 1.
Results
In this contribution, we will introduce our approach and demonstrate its efficiency on several problems.
In a first part, we will illustrate the influence of the cavity quantum optical state on the energy and temporal structure of the electron beam during the PINEM interaction. For example, after interaction with a number state – i.e. when the initial number of photons is perfectly known – we will show that the electron-cavity coupled state becomes a coherent superposition of entangled (electron-photon) energy-number states. On the other hand, if the electron interacts with a coherent state – i.e. a Poissonian superposition of number states – one would only obtain correlations between the electron energy states and the photon number states.
In a second part, we will show that a PINEM measurement in the quantum regime is deeply connected to the standard quantum limit to the temporal resolution in a UTEM. Indeed, the Heinsberg uncertainty principle governs the inelastic interaction between the electron and the cavity, making impossible to exactly know the number of photons and the temporal dynamics of the field in the cavity at the same time. We will show that this uncertainty relation has a dramatic influence on the quantum path interferences otherwise observed in the classical regime of PINEM. In that regard, the Wigner function appears to be an ideal tool to visualize the effect of the uncertainty principle in phase-space.

In a third part, we will tackle the question of the quantum to classical transition in free electron scattering and show that the Wigner function elegantly highlight the transition between the stimulated and spontaneous scattering regimes in electron energy-loss spectroscopy (EELS).
Finally, in a fourth part, we will introduce to PINEM a central basis of quantum optics: the displaced number state (DNS). This basis will allow us to shade a new light on the interaction between electron and a coherent state of light. In particular, or calculation demonstrate that in this configuration, the final electron-cavity state is a coherent superposition of DNS entangled with similar electron states than the one predicted by the classical model. This creates a bridge between the classical and the quantum description of PINEM.

Conclusion

In this work, we present a complete phases-space description of inelastic fast electron-light interaction, in which the central object – the Wigner function – provides a powerful visualization tool. Using this approach, we tackle important problems in the field of EELS and PINEM, such as the quantum to classical transition, or the distinction between stimulated and spontaneous processes. In that respect, our work demonstrates the central role played by the number-phase uncertainty relation. Beyond these fundamental questions, our approach will find application in the description of future experiments in the field of EELS and PINEM, such as the probing of non-linearity in EELS or the realization of atom-cavity coupling in a TEM.

***Figure 1.*** *Electronic Wigner function after interaction with (a) a classical electromagnetic field and (b) a quantized electromagnetic field.* The Wigner function (central panel) provides a direct visualization of the energy distribution (right panel) and the temporal modulation (top panel) of the electron at the same time.




**Keywords:**

UTEM
Theory
Quantum-Optics
Light-matter interaction

**Reference:**

[1] A. Feist et al., Nature 521, 200–203 (2015)
[2] J. H. Gaida et al., Nature Photonics, in press (2024)
[3] J-W. Henke et al., Nature 600, 653-658 (2021)
[4] O. Kfir, Physical Review Letters 123, 103602 (2019)
[5] V. Di Giulio et al., Optica 6 (12), 1524-1534 (2019)

991

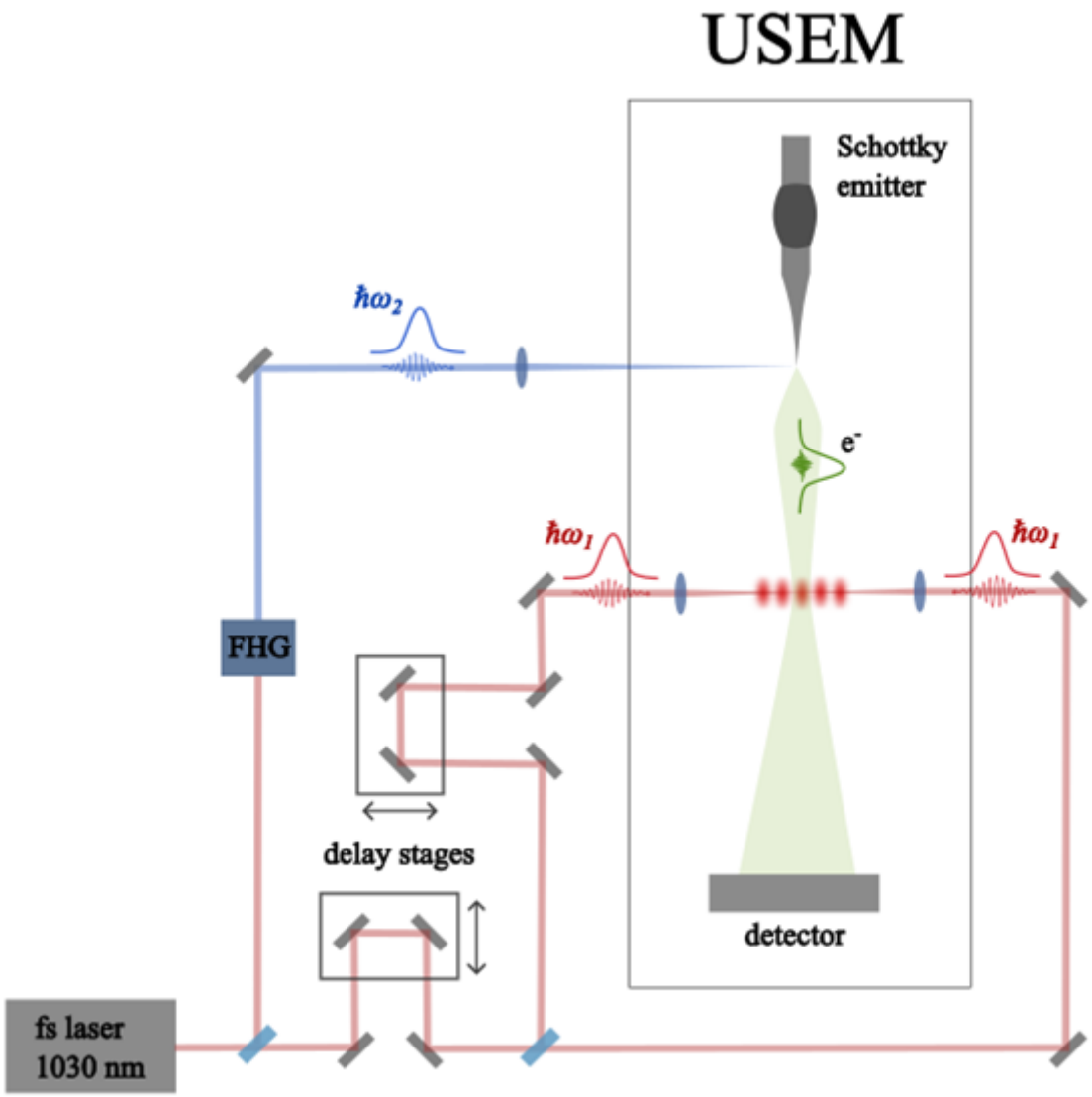
# Temporal characterization of femtosecond electron pulses inside ultrafast scanning electron microscope

Kamila Moriova[1], Martin Kozak[1]
[1]Faculty of Mathematics and Physics, Charles University, Prague,

Poster Group 2

Background incl. aims
In recent years, ultrafast electron microscopy has been proved to be a promising method for examination of the dynamics of the most fundamental processes. The visualization of processes with high temporal and spatial resolution can be achieved with laser pump and electron probe experiments using single electron pulses from a nanoemitter to avoid temporal spread of the pulse by Coulomb forces. In order to perform pump-probe experiments, the exact knowledge of electron pulse duration is required. Only few techniques have been developed to perform the temporal characterization of ultrashort electron pulses with low electron density. One of the approaches, often used in ultrafast transmission electron microscopes (UTEMs), is based on electron-light cross-correlation utilizing the inelastic scattering of electrons induced by the interaction with optical near field around a nanostructure [1]. Such scheme is limited by the fact that the electron microscope has to be equipped with a high-resolution spectrometer, which allows to spectrally filter the inelastically scattered electrons. In this contribution we demonstrate the use of a different method for electron pulse characterization, which is based on scattering of electrons mediated by ponderomotive potential of an optical standing wave. This method is suitable for ultrafast scanning electron microscopes (USEM), for which the experimental determination of the length of the electron pulse is still challenging to this day.
Methods
Free electrons are scattered by an optical standing wave created by two counter-propagating pulsed optical beams of the same frequency in vacuum. This effect is driven by the ponderomotive potential of the optical field and is used as a technique to characterize duration of electron pulses [2,3]. Electrons traveling through the oscillating electromagnetic field of generated standing wave experience ponderomotive force proportional to the gradient of light intensity and are deflected from their original direction of motion. Electron pulse duration is then reconstructed from a time trace, which is given by the measured electron scattered signal taken at different electron pulse-laser pulse time delays, and which is a direct cross-correlation of electron and laser pulse. Although the described method is known, up until now it has not yet been used to characterize electron pulse durations for wide range of electron energies in a commercial scanning electron microscope (SEM).
Results
In our experimental setup, we generate low density electron pulses by a laser-triggered emission from a Schottky emitter in Verios 5 XHR SEM (energy range 1 to 30 keV) by illuminating the apex of the emitter tip with 257.5 nm laser pulses. Inside the SEM chamber, we implement a compact optical setup for the generation of optical grating with two counterpropagating laser pulses with central wavelength of 1030 nm and duration of 220 fs. The scattered electron signal is detected by a hybrid-pixel detector Timepix3. We characterize electron pulse durations for electrons in energy range from 5.5 keV to 30 keV. For the optimal setting with the electron emission regime with less than one electron per pulse, the electron pulse duration at the sample location spans from 0.4 ps full-width-half-maximum (FWHM) for 30 keV electrons to 2.7 ps FWHM for 5.5 keV electrons.
Conclusions
In the past, the development of methods for direct measurement of the temporal characterization of electron pulses has been focused primarily on UTEMs. Electron pulse durations in USEMs were only

estimated on the basis of theoretical calculations. We describe the implementation of all-optical method for direct temporal characterization of femtosecond electron pulses in a commercial SEM.




**Keywords:**

ultrafast electron microscopy, electron pulses

**Reference:**

[1] B. Barwick, D. J. Flannigan, and A. H. Zewail, “Photon-induced near-field electron microscopy,” Nature 462, 902–906 (2009).
[2] C. T. Hebeisen, G. Sciaini, M. Harb et al. “Grating enhanced ponderomotive scattering for visualization and full characterization of femtosecond electron pulses,” Opt. Express 16, 3334-3341 (2008).
[3] M. Gao, H. Jean-Ruel, R. R. Cooney et al. “Full characterization of RF compressed femtosecond electron pulses using ponderomotive scattering,” Opt. Express 20, 12048-12058 (2012).

1010

# Imaging ultrafast spin dynamics at the nanometer scale

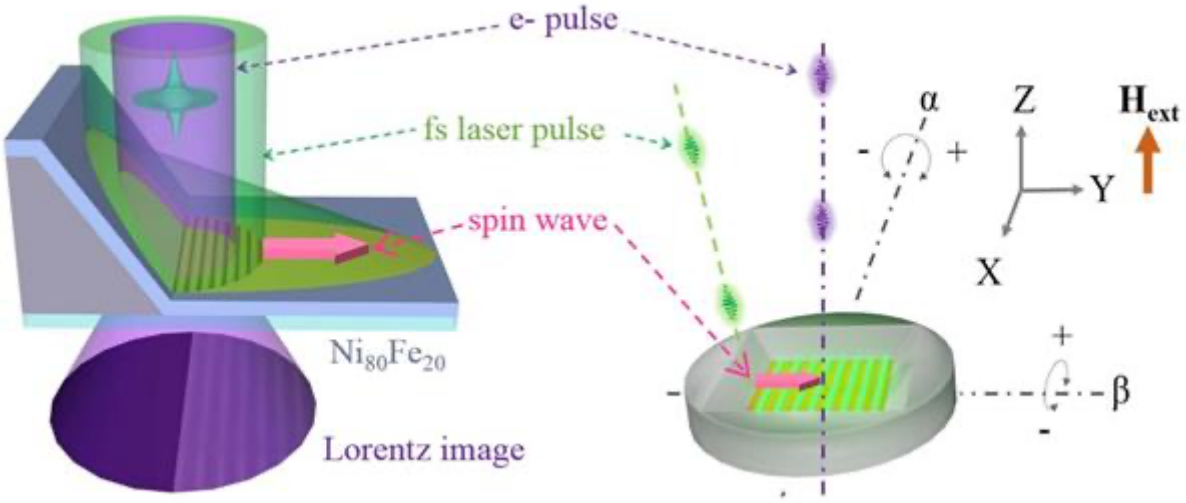
Dr Gaolong Cao[1], Sheng Jiang[2], Yuzhu Fan[1], Johan Åkerman[3], Jonas Weissenrieder[1]
[1]Royal Institute of Technology (KTH), Stockholm, Sweden, [2]South China University of Technology, Guangzhou, China, [3]University of Gothenburg, Gothenburg, Sweden

Poster Group 2

Background incl. aims
Understanding magnetic dynamics is not only important to fundamental physics but also critical to the development of future energy-efficient spintronic devices. Detection of ultrafast magnetic dynamics with high temporal-spatial resolution remains a great challenge. In recent years, the UEM (ultrafast electron microscope) has been rather active in ultrafast magnetism[1-4]. Based on a commercial TEM platform, UEM inherits the capability of high space resolution (nanometer scale) and versatile imaging modes. Moreover, the pulse duration of the photo-emitted electron can reach lower than 1 ps in the premium condition, which is compatible with most of the scenarios for the spin dynamics.
However, it is noteworthy that all these experiments rely on topological magnetic structures. Such limitation originates from the imaging mechanism of the Fresnel mode in the Lorentz TEM, which is the beam-deflecting effect driven by the transverse Lorentz force between the electron beam and local spin state. In short, the Lorentz image contrast in Fresnel mode relies on the in-plane spatial gradient of magnetization other than the magnetization itself. This means the Lorentz UEM could only image the ultrafast perturbations of the topological magnetic structure, and rule out other critical magnetic dynamics, such as ultrafast demagnetization, spin waves etc, which usually do not relate to a static topological structure.

Method and result
Here, we report a novel method of Lorentz ultrafast electron microscope (UEM), which can directly image the spin wave in real space [5]. Unlike traditional methods, our Lorentz contrast does not rely on topological magnetic structure. We introduce a structured-light pulse (transient optical grating (TOG)) onto the ferromagnetic sample, which can induce transient spatial magnetic gradient and then create time-resolved magnetic contrast under Lorentz mode. Then, the propagating spin waves, which are driven by the magnetic-elastic coupling, become ‘visible’ in the UEM.

Conclusion
In summary, we have for the first time directly imaged spin waves in the UEM via structured light, which opens a new door to detect and manipulate ultrafast magnetic dynamics with high temporal-spatial resolution.




**Keywords:**

UEM, Lorentz mode, spin waves

**Reference:**

[1] Hyun Soon Park, J. Spencer Baskin, and Ahmed H. Zewail* 4D Lorentz Electron Microscopy Imaging: Magnetic Domain Wall Nucleation, Reversal, and Wave Velocity Nano Lett. 2010, 10, 9, 3796–3803 (2010)
[2] Ming Zhang, et al. High Spatiotemporal Resolution of Magnetic Dynamics in Mn-Ni-Ga via Four-Dimensional Lorentz Microscopy Phys. Rev. Applied 12, 034037 (2019)
[3] N. Rubiano da Silva, M. Möller, A. Feist, H. Ulrichs, C. Ropers, S. Schäfer, Nanoscale Mapping of Ultrafast Magnetization Dynamics with Femtosecond Lorentz Microscopy Phys. Rev. X 8, 031052 (2018)
[4] G. Berruto, et al Laser-Induced Skyrmion Writing and Erasing in an Ultrafast Cryo-Lorentz Transmission Electron Microscope Phys. Rev. Lett. 120, 117201 (2018)
[5] G. Cao, S. Jiang, J. Åkerman, and J. Weissenrieder, " Femtosecond laser driven precessing magnetic gratings," Nanoscale 13, 3746–3756 (2021)

1034

# Fabrication of electron transparent membranes and nanostructures in fluidic devices by NIL and “Flow-Through”-gas-phase deposition

Jeremy Teuber[1], Manuel Müller[1], Rukan Nasri[1], Dr. Irene Fernandez-Cuesta[1]
[1]University of Hamburg, Hamburg, Germany

Poster Group 2

Background incl. aims
Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) are invaluable tools for visualizing and analyzing samples with nanometer resolution. Integration of liquid cells with these microscopy techniques has expanded their capabilities, allowing dynamic imaging and real-time analysis under controlled liquid conditions [1,2]. However, existing liquid cells still face limitations such as high production cost, restricted geometries, and reliance on slit-like chambers formed by connecting two membranes with spacers.

Methods
We present a novel fabrication method for producing micro-, and nano channels and 3D structures, self-sealed with electron transparent thin Al2O3 membranes, as sketched in Fig. 1a), 1b) and 1c). For this, we make polymeric foils (<10 µm) containing micro-, and nanofluidic structures. Fabrication of the fluidic devices involves direct UV nanoimprint lithography (NIL) using a customizable and versatile stamp design adaptable to specific geometries [3]. Then, the channels are coated selectively from the inside with Al2O3 by using our self-developed gas-phase deposition [4]. These inorganic electron-transparent structures can be suspended by removing the polymer material around them, enabling their use as liquid cells for investigating dynamic molecular behavior within confined spaces, such as in TEM.

Results
Our method allows fabrication of liquid cells with a variety of lateral dimensions, even down to nanometric channels, and complex 3D structures with graded depths and widths. The fluidic device contains two microchannels which connect the inlet and outlet holes. These microchannels are interconnected by several nanochannels. For fluidic devices fabrication, a stamp is placed and aligned onto a polycarbonate plate which has been covered with a UV curable polymer containing pre-patterned holes. The assembly is then cured with UV-light, and the substrate and stamp are separated manually. Next, a polymer coverslip is used to seal the channel system of the fluidic device. The Al2O3 coating is achieved through a specialized gas phase deposition (GPD) reactor operating in a "flow-through" mode (ftGPD), providing conformal coating of the various structures [4]. The reactor's gas and vacuum ports align with the holes in the polycarbonate plate, facilitating connection to the imprinted fluidic system. By precisely controlling precursor gas flow across the microchannel, a controlled pressure gradient is established, enabling conformal deposition of Al2O3 onto various structures, including slits, chambers, and micro- and nanochannels. This method enables to tune the Al2O3 thickness, ranging from a few nanometers to hundreds of nanometers, allowing for adjustment of mechanical stability and electron transparency as needed. This strategy effectively circumvents existing constraints associated with liquid cell geometries, particularly those limited to slit-like chambers. In subsequent fabrication steps, the micro-, nanochannels or membranes can be selectively suspended by masking and reactive ion etching (RIE). Fig 1a) shows hollow Al2O3 microchannel which are used for flow tests. Fig. 1b) shows an example of a suspended, hollow Al2O3 nanochannel with a cross section of 500 x 500 nm and a suspended length of 20 µm. To validate the electron transparency of the Al2O3 membrane, we created a TEM grid sample coated with an Al2O3 membrane fabricated in our GPD reactor. Subsequently, polystyrene beads were

deposited both above and below the Al2O3 membrane, and imaged using SEM and TEM techniques (Fig. 1 d) e) and f), respectively). SEM images (Fig. 1 (d)) captured the same area with two different detectors, where polystyrene beads beneath the membrane were detectable solely by the secondary electron secondary ion (SESI) detector. In contrast, TEM imaging (Fig. 1 (e)) of the sample in transmission mode at a separate location demonstrated the feasibility of imaging beads through the Al2O3 membrane. We also made a sandwich, by placing another Al2O3 membrane on top, and imaged the beads in TEM at 200 keV, as shown in Fig 1 f). Furthermore, we also imaged gold nanoparticles placed below the Al2O3 membranes deposited and suspended from the top in one of our microchannels. The particles were visible in the SEM at an acceleration voltage of 8kV (Fig. 1 c)). All these results confirming the electron transparency of the membranes deposited using our method. To deploy this system as liquid cells in a TEM specimen holder, detachment of the structured and coated polymer foil from the substrate and subsequent cutting to a 3 mm total diameter are necessary.

Conclusion

This fabrication method offers a versatile approach for creating liquid cells with complex geometries, overcoming limitations of existing slit-like chamber designs. The electron-transparent Al2O3 membranes enable dynamic imaging of samples within confined spaces using SEM and TEM, with potential future applications in nanoscale research and analysis. Further investigations using these liquid cells inside SEM and TEM will be presented at the conference.

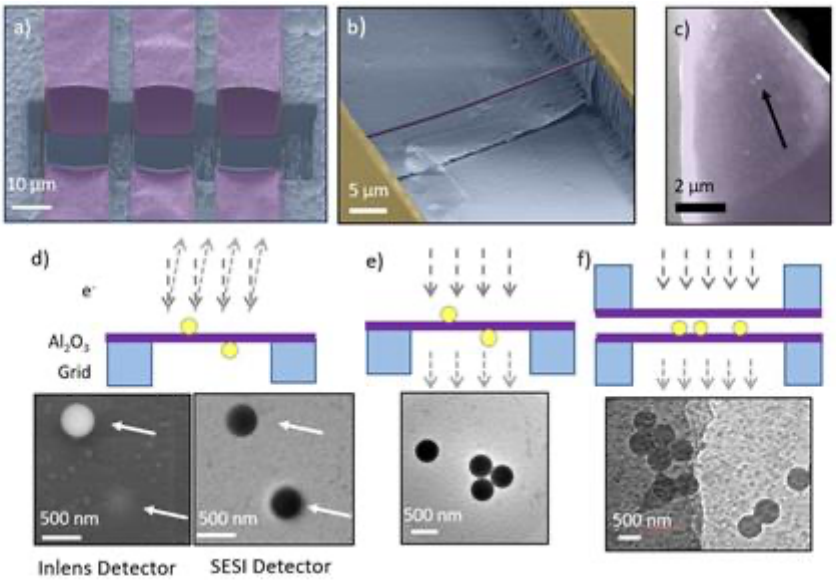



Figure 1: a) shows hollowed $Al_2O_3$ coated microchannel. b) displays a suspended nanochannel with a cross section of 500 x 500 nm and a suspended length of 20 µm. c) shows an SEM image of gold nanoparticles underneath the $Al_2O_3$ membrane. d) Sketch of the experiment to demonstrate the electron transparency of the aluminum oxide coating. A TEM Grid is coated with an Al2O3 layer and polystyrene beads are placed above and underneath. With an SEM, images of the beads could be taken at 5 kV e) This TEM image reveals the beads through the $Al_2O_3$ layer. The image was captured at 200 kV. f) Two TEM grids, coated with $Al_2O_3$, were stacked on top of each other with polystyrene beads placed in between. In order to image the beads using a TEM, the layer must be thin enough to be electron-transparent.

**Keywords:**

electron-transparent membrane, liquid flow cell

**Reference:**

[1] Yoon, A. et al. (2001). Dynamic Imaging of Nanostructures in an Electrolyte with a Scanning Electron Microscope. Microscopy and Microanalysis, 27(1), 121-128. Doi 10.1017/S1431927620024769

[2] Meirong Li and Lan Ling., ACS Nano 2022, 16,15503-15511

[3] I. Fernandez-Cuesta et al., J. Vac. Sci. Technol. B (2011) 29 06F801.

[4] M. Müller et al., DE 102020102076A1 (2020) Hamburg, Germany, Deutsches Patent- und Markenamt.

[5] M. Müller et al., "Suspended Nanochannel Resonators Made by Nanoimprint and Gas Phase Deposition," 2023 IEEE 36th International Conference on Micro Electro Mechanical Systems (MEMS), Munich, Germany, 2023, pp. 1041-1044, doi: 10.1109/MEMS49605.2023.10052412

1082

# Resolving single-electron and multi-electron distribution functions with event-based electron detectors

Alexander Schröder[1], Andreas Wendeln[1], Prof. Sascha Schäfer[1]
[1]University Regensburg, Regensburg, Germany

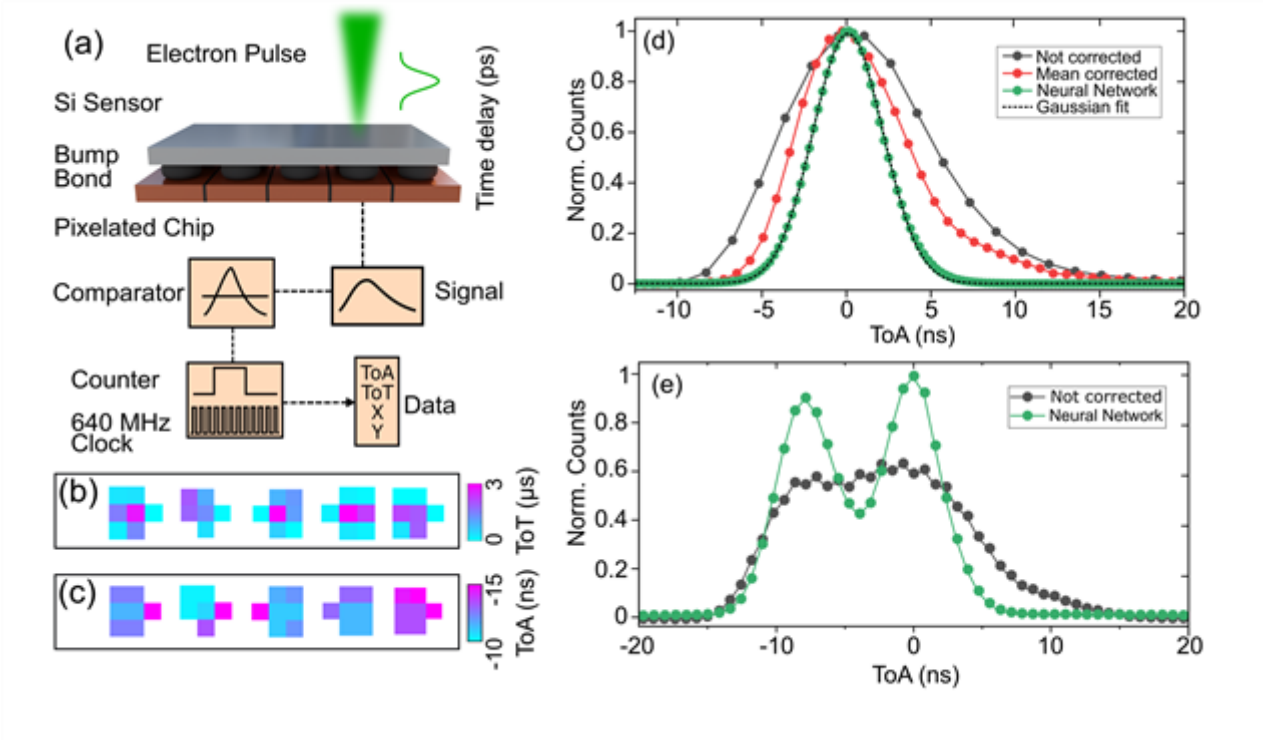
Poster Group 2

Background
The combination of electron probe pulses with a synchronized optical sample excitation as applied in ultrafast transmission electron microscopy (UTEM) has enabled the investigation of ultrafast dynamics on the nanoscale [1]. Besides the traditional optical-pump/electron-probe approach, sufficiently fast event-based electron detectors, based for example on the TimePix3 chip architecture [2], can detect electron events with a temporal bin width of about 1.6 ns. Due to the stochastic electron trajectories in the sensor chip the spatial and temporal resolution is often limited, in particular at higher electron energies [3,4], and requires further data processing to overcome these limitations. In addition to the nanosecond temporal resolution of event-based detectors, such a detection scheme gives access to spatio-temporal electron-electron correlations and has been recently applied to the investigation of Coulomb-correlation within a photoelectron bunch generated by femtosecond photoemission from a Schottky emitter [5]. In general, these new kinds of detectors in principal provide a measurement of multi-electron distribution functions both in electron beams and electron pulses containing additional information with respect to averaged micrographs traditionally recorded.
Methods
To gauge the potential of event-based detectors for UTEM, the temporal response of a TimePix3 detector (Cheetah T3, Amsterdam Scientific Instruments) was characterized utilizing 200-fs electron pulses (400-kHz repetition rate, 200-keV electron energy, 1.3 electron/pulse) as generated in the Regensburg UTEM. As a photoelectron source, both a laser-driven Schottky and cold-field emitter were employed. In a first experiment, we experimentally collected a dataset of electron detection events for up to 4 x 10^6 electron pulses (Fig. 1 (a)). For each event, the TimePix3 collects relative detector position, time of arrival (ToA) and time over threshold (ToT). The events are clustered depending on their ToA and position, resulting in an average of 6.5 events per cluster (Fig. 1 (b) and (c)). The data is synchronized by assigning a timestamp to each photoemission laser pulse. A fully connected deep neural network model is trained using event data from 1.6 x 10^5 time-scrambled event cluster, so that it becomes possible to predict the ToA of the primary electron.
In a second experiment, electrons are emitted by a cold-field emitter gun (CFEG) and around 5 x 10^7 electrons are detected with the TimePix3. To find Coulomb-correlated electron pairs, only clusters with a small mean ToA-difference (< 50 ns) from the dataset are selected. The electron pairs are statistically analysed and the influence of optical excitation, extraction fields and spatial distribution are examined.
Results
Using the non-clustered ToA events leads to a temporal spread of about 10 ns (Fig. 1 (d), grey curve). If a cluster algorithm is used and the mean ToA for each cluster is calculated, then the temporal spread can be reduced to 7 ns (Fig. 1 (d), red curve). The neural network reduces the predicted temporal distribution (for a part of the data not involved in the training) to 4.7 ns (FWHM) (Fig. 1 (d), green curve). In the case of the neural network the distribution is almost Gaussian while the ToA distribution in the previous cases shows a tail for larger ToA.
Applying closely spaced electron double-pulses with a temporal separation of 8 ns to TimePix3 detector (shorter than the intrinsic temporal resolution) shows a broadened flat top distribution (Fig.

1 (e), grey curve). With the implementation of the neural network, the two peaks can be distinctly separated (Fig. 1 (e), green curve), demonstrating the more general applicability of the approach.

Conclusion

In conclusion, we demonstrate, that the accurate prediction of the ToA by a neural network trained by femtosecond electron pulse data, resulting in an increased temporal resolution for the TimePix3 detector. This improved temporal resolution will also be helpful for the mapping of temporal correlations in the imaging of non-static nano-objects and non-trivial multi-particle correlations within electron pulses.




**Keywords:**

ultrafast-TEM, femtosecond-electron-pulses, laser-driven-cold-field-emitter, TimePix3, electron-electron-correlations

**Reference:**

[1] A. Feist et al., Ultramicroscopy 176, 63 (2017).
[2] T. Poikela et al., JINST 9, C05013 (2014).
[3] F. Pitters et al., JINST 14, P05022 (2019).
[4] A. Schröder et al., Ultramicroscopy 256, 113881 (2024).
[5] R. Haindl et al., Nature Physics 19, 1470 (2023).

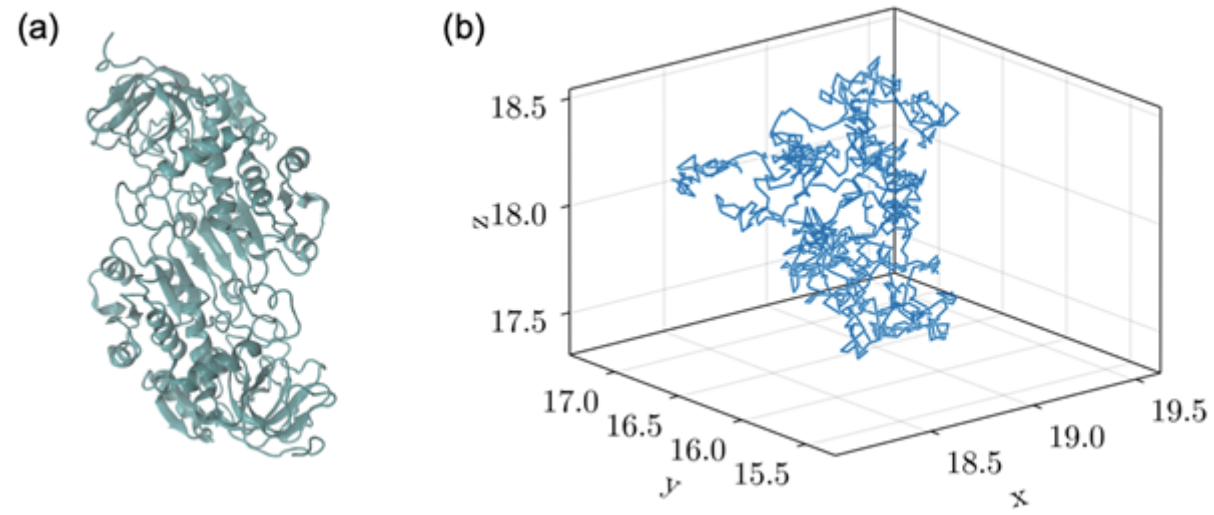
1092

# Molecular dynamics simulation of the Brownian motion of biomolecules in liquid phase electron microscopy

Mr Xiaopeng Wu[1], Dr Peng Wang[1]
[1]Department of Physics, University of Warwick, Coventry, United Kingdom

Poster Group 2

Background incl. aims
Liquid phase electron microscopy (LP-EM) revolutionizes our ability to observe dynamic processes within liquids at nanoscale resolution. It allows for the direct observation of biological, chemical and physical phenomena in their native liquid environments which was not possible with conventional or cryogenic electron microscopy (Cryo-EM). While previous successes of the technique have predominantly been demonstrated with rigid materials such as gold nanoparticles [1], it is anticipated that the application of the technique on soft materials such as biological macromolecules may yield novel insights into the functioning mechanisms from the observed dynamics [2].
In this work, we aim to investigates the behaviour of biomolecular samples in a liquid environment and consider the effect of the Brownian motion.
Methods
We employed molecular dynamics simulations to study the behaviour of the biomolecules in liquid at the nanosecond scale. The structure of the horse liver alcohol dehydrogenase (ADH) enzyme was used for this study [3], as shown in figure (a). We simulated multiple trajectories of the ADH enzyme using the OpenMM library [4], and the ff14SB force field along with the TIP3P water model was used for all simulations [5].
Results
As expected, the diffusivity of the protein is found to be increasing with temperature from the simulations. The protein exhibits significant deviations due to Brownian motion, which can be characterised by a random walk model, in figure (b), we have shown a snippet of the trajectory of the centre of mass of the ADH at 300 K to confirm that the global motion of the protein is indeed random. When the thermal energy of the water molecules is low but still above the freezing point, the diffusive motion of the protein tends to favour the direction with the smallest solvent accessible surface area, this was due to the irregular shape of the ADH protein. Upon further increase of temperature to 300 K, this anisotropy diminishes, as the protein diffuses more isotopically potentially due to more frequent interactions between the solvent and itself.
The protein exhibits thermal vibrations across all temperatures, which collectively influence the overall deviation of its structure. These vibrations are temperature-dependent and exhibit a relatively stable magnitude throughout the period of observation. Additionally, higher temperatures can induce further localised deviations within the protein structure, particularly in regions with higher flexibility, such as the sequence termini, where deviations can be notably pronounced compared to the rest of the protein.
Conclusion
Overall, this study enhances our understanding of the effect of Brownian motion inherent to biomolecules in liquid and may inform the development of more effective experimental designs for LP-EM. Future studies could extend this analysis to samples of different configurations, as well as longer simulation times for more robust estimations over larger timescales. The authors would like to acknowledge funding from the BBSRC International Institutional Partnership Fund.




**Keywords:**

Liquid-phase, electron microscopy, molecular dynamics

**Reference:**

[1] Wu, H., Friedrich, H., Patterson, J. P., Sommerdijk, N. A. J. M. & Jonge, N. de. Liquid-Phase Electron Microscopy for Soft Matter Science and Biology. Advanced Materials 32, 2001582– 2001583 (2020)
[2] Zheng, H., Claridge, S. A., Minor, A. M., Alivisatos, A. P. & Dahmen, U. Nanocrystal Diffusion in a Liquid Thin Film Observed by in Situ Transmission Electron Microscopy. Nano Lett. 9, 2460–2465 (2009)
[3] Herzik, M. A., Wu, M. & Lander, G. C. High-resolution structure determination of sub-100 kDa complexes using conventional cryo-EM. Nat Commun 10, 1032–1033 (2019)
[4] Eastman, P. et al. OpenMM 7: Rapid development of high performance algorithms for molecular dynamics. PLOS Computational Biology 13, e1005659 (2017)
[5] Maier, J. A. et al. ff14SB: Improving the Accuracy of Protein Side Chain and Backbone Parameters from ff99SB. J. Chem. Theory Comput. 11, 3696–3713 (2015)

1163

# Spatiotemporal observation of ultrafast magnetization dynamics with 4D Lorentz Transmission Electron Microscope

Phd Yuzhu Fan[1], Researcher Gaolong Cao[1], Professor Jonas Weissenrieder[1]
[1]School of Engineering Sciences, KTH Royal Institute of Technology, Stockholm, Sweden

Poster Group 2

Background incl. aims
Since the discovery of the sub-picosecond-timescale ultrafast demagnetization induced by femtosecond laser pulse by Beaurepaire et al. in 1996, utilizing ultrafast light to manipulate the magnetization has opened up a new field compared with the conventional ways to control the spins by magnetic fields, charge, or spin currents. Known as 'femtomagnetism', ultrafast optical control of magnetization at femtosecond or sub-picosecond timescales is the currently fastest route known to demagnetize or switch a magnetic material, which conforms to the ever-increasing demand for energy-efficient high-speed magnetic storage devices or random-access memories. To promote further understanding of light-spin interaction and potential technological applications, however, exploiting experimental tools to characterize and understand the light-spin dynamics at the sub-picosecond and nanometer scale plays a decisive role.
Methods
We combine Lorentz Transmission Electron Microscope with transient optical gratings to control laser-induced gradients of the in-plane magnetization. The light absorption distribution is characterized by Photo-induced Near-Field Electron Microscopy (PINEM) technique.
Results
We have spatiotemporally analyzed patterned ultrafast demagnetization dynamics in Ni80Fe20 near the sample discontinuity excited by a transient optical grating. Our findings demonstrate that the propagating surface plasmon polariton (SPP) modulates the ultrafast demagnetization process in space, time, and amplitude. Furthermore, we demonstrate the sub-picosecond all-optical switching dynamics in GdFeCo at a local scale by utilizing the Lorentz contrast from the Bloch domain wall induced by local switching in constructive interference areas. Last but not least, we observe the laser-induced coherent precession in GdFeCo with both FMR mode and exchange mode.
Conclusion
Our findings demonstrate 4D Lorentz Transmission Electron Microscope as an effective tool for investigating laser-induced magnetization dynamics with sub-picosecond and nanometer spatiotemporal resolution.

**Keywords:**

Ultrafast demagnetization, all optical switching,

**Reference:**

1 Cao, G., Jiang, S., Akerman, J. & Weissenrieder, J. Femtosecond laser driven precessing magnetic gratings. Nanoscale 13, 3746-3756 (2021).
2 Fan, Y., Cao, G., Jiang, S., Akerman, J. & Weissenrieder, Spatiotemporal observation of surface plasmon polariton mediated ultrafast demagnetization, submitted.
3 Fan, Y., Cao, G., Jiang, S., Akerman, J. & Weissenrieder, Spatiotemporal observation of all optical switching in GdFeCo with 4D Lorentz Transmission Electron Microscope, to be submitted.