Personnel remembrance of aberration correction over more than four decades and state of the MC-development

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In early 1980, during an open house at the Institute of Applied Physics at TU Darmstadt, I had the opportunity to gain insight into the theoretical and experimental work in electron optics. At that time, the first Cc and Cs corrector for a TEM was under development in the groups of Scherzer and Rose. The astigmatic beam path necessary for the correction was generated by quadrupole fields with octupole elements. Calculations by Rose had shown that these octupole elements generate a strong intrinsic 5th-order fourfold aberration. As part of my diploma thesis, I was tasked with designing and testing a twelve-pole element to replace the existing eight-pole element [1]. H. Hely, the last member of Scerzer's group, completed his doctorate [2] and moved into industry. Shortly before his departure, I learned from him how to operate this homemade TEM with the corrector. During this handover, the corrected TEM was fully aligned, and subsequently, the best resolution available at that time was achieved with this instrument: 10 Å point resolution at 40 kV. In November 1982, O. Scherzer passed away suddenly. As a consequence, H. Rose, as a theoretician, received no further funding for experimental work. This forced the project to be halted.

Although this marked the end of aberration correction in electron microscopy in Germany, another project was underway in Chicago initiated by A. Crewe, who aimed to correct the spherical aberration of a STEM, equipped with a cold-field emitter, using a hexapole corrector. Since this project was also discontinued after a couple of years, funding for aberration correction in Germany also ceased, leading to the general conclusion that correction of aberrations was impossible.

A new opportunity to continue working on the correction arose from the idea of developing a Cc- and Cs-corrected SEM for biology. A theoretical paper by J. Zach, who was completing his doctorate under H. Rose, formed the basis for this corrector. With this Cc and Cs corrected SEM, an improvement of resolution at 1 keV could be demonstrated for the first time [3].

Meanwhile, a compelling scientific justification for material science applications persuaded the Volkswagen Foundation to fund the experimental development of a high-resolution, Cs-corrected TEM. The resulting joint project between H. Rose, K. Urban, and myself could successfully be completed [4]. For this, the present instability had to be circumvented, and with a realignment together with S. Uhlemann at a night session in 1997, images of an interface showing atomic resolution could be acquired. This happened just one week before the TEM had to be shut down. Although in a textbook in the 1980s it was stated that the correction might work in an experiment, however, it is "*unthinkable as part of a commercial instrument.*" As time went by, the story developed differently. Today, over 1700 corrected electron microscopes have been installed to date.

After this long ongoing work with multipole systems, we continued the development with rather complex aberration correctors like a Cc corrector for TEM low and medium voltage and other advanced EO components like a post column energy filter. In addition, a ground potential monochromator (GP-MC) for TEM and STEM is currently under development [5]. The latter is part of the **MO**mentum and position **RE**solved mapping TEM project (MoRe-TEM) [6] funded by a Synergy Grant of the ERC. The state of this project will be given.

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EELS OF BACKSCATTERED ELECTRONS: WHAT ABERRATION CORRECTION CAN DO FOR AN ULTRA-LOW ENERGY SEM

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The correction of spherical and chromatic aberrations has been very successfully applied in high-end TEM and STEM [1-3], realizing highest spatial resolution as well as unsurpassed analytical capabilities. However, for SEM, conventionally operating at 10 – 30 keV, such correction has been less important so far, as resolution is limited rather by the interaction volume than by the probe size. This has changed with the developments of photoemission and low-energy EM (PEEM/LEEM) [4,5], as those instruments operate at very low electron energies, where the point spread function of the imaging limits resolution. While some designs of aberration correction for such instruments were based on the canonical multipole corrector design [6], electrostatic mirror correction proved superior [7]. Such a design has recently been incorporated into a SEM, commonly known as the DELTA-SEM, presented some years ago by Carl Zeiss [8] adapting the electrostatic mirror design developed by Preikszas and Rose [7] for correcting spherical and chromatic aberration. As an additional device, a retarding electrostatic energy filter was integrated, allowing the DELTA-SEM to record images at different selected energy windows.

We have now evaluated the novel imaging and analytical possibilities provided by the new ‘analytical' DELTA-SEM: In our experiments we have extended the energy range for imaging to ultra-low landing energies, such as 10 – 100 eV. While in a conventional microscope at such energies no meaningful imaging can be realized, we show typical resolution levels at such energies of about 1 – 2 nm for carbon materials. By imaging monolayers of organic dyes, we could also show that beam damage for fluorescent materials induced by electrons is largely reduced for electron energies below 50 eV.

The development and incorporation of an electrostatic energy filter also facilitates electron spectroscopic imaging in the SEM: Images at different electron energy loss can be recorded, providing a direct spectral signal. Such imaging can be used to obtain either spectra of Secondary Electrons (SE) or Backscattered Electrons (BSE).

In a pilot study we used the spectroscopic imaging of BSE (bsEELS – electron energy loss spectroscopy of backscattered electrons) to obtain low-loss spectral signal from bulk materials. bsEELS is a very surface sensitive signal, as is also confirmed by the imaging of DNA origami of only one double-helix thickness. In fact, such images of DNA are formed from only about 40-50 light atoms per pixel as can be deduced from the known structure of the DNA.

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EXELFS analysis of beam-sensitive CeO_2 nanoparticles

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Electron energy-loss spectroscopy (EELS) offers detailed insights into bonding at the nanoscale. Recently, strong interest is rising in using high-energy losses above 3 keV for extended-range fine-structure (EXELFS) studies. Similar to EXAFS – EXELFS's X-ray-based counterpart – EXELFS oscillations beyond the ionization edge reveal the element-specific radial distribution function (RDF), making the method valuable for analyzing amorphous metals and catalytic nanoparticles [1]. However, the weak ionization cross-sections at these energies demand long exposures or high beam currents, which can easily damage beam-sensitive materials. This challenge can be mitigated by operating at substantially lower dose rates [2] requiring highly sensitive, next-generation direct-detection cameras capable of capturing weak high-energy signals without sacrificing spectral fidelity.

In this presentation, high-energy EXELFS analyses of beam sensitive CeO_2 nanoparticles at the Ce L_3 edge (5.7 keV) are demonstrated. This is achieved by using a JEOL F200 which is equipped with a GIF Continuum energy filter and state-of-the-art direct-detection cameras. Spectrum-imaging was carried out with dedicated dose-distribution scan strategies across multiple energy windows while maintaining a dose rate below 3.1×10^5 e/Å²/s to prevent reduction of Ce^{4+}. Optimized lens coupling enabled efficient energy-filter performance [3]. The resulting spectrum data were analyzed with Athena software package to obtain the RDF [4].

The acquired high-loss EELS spectra show clear EXELFS oscillations and the RDF analysis reveals Ce–O and Ce–Ce coordination consistent with the CeO_2 fluorite structure [5]. The preserved Ce oxidation state confirms the effectiveness of low-dose acquisition. These results illustrate how advanced detectors and tailored STEM protocols enable EXELFS on beam-sensitive nanomaterials, providing spatially resolved structural information beyond the reach of conventional synchrotron EXAFS.

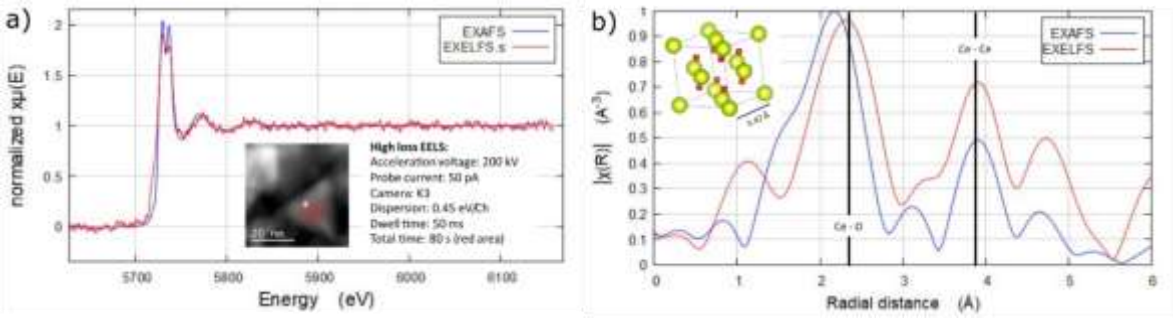

Fig. 1: a) EELS and XAS spectra at Ce L_3 edge. b) RDFs deduced from the spectra in a).

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Toward Attosecond Electron Tomography of Optical Near-Fields

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Imaging electromagnetic near-fields inside complex nanostructures remains a central challenge in ultrafast electron microscopy. While photon-induced near-field electron microscopy (PINEM) enables nanoscale probing of optically driven fields, it typically provides only projected information along the electron trajectory and does not directly recover the full complex-valued three-dimensional field. Prior tomographic approaches in electron microscopy have been highly successful for static quantities, but they are not directly applicable to oscillatory optical near-fields, where the electron integrates over a time-dependent field during transit. In this talk, I will discuss recent developments that build toward **attosecond electron tomography** of optical near-fields. These include phase-resolved electron–light interferometry, controlled electron wavepacket shaping, homodyne-style detection schemes based on multiple interaction points, and experimental methods for stabilizing and retrieving the optical phase with attosecond precision. Together with dedicated sample-holder geometries for tilted illumination and tailored reconstruction approaches inspired by tomography, these tools enable recovery of complex-valued near-field information from multiple viewing angles. Attosecond tomography emerges from the convergence of several emerging directions in ultrafast TEM: PINEM, phase-sensitive free-electron measurements, and computational inversion of electron–field interactions.

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EMERGENT STATES OF MATTER IN FUNCTIONAL 2D MATERIALS ENABLED BY Cc/Cs-CORRECTED LOW-VOLTAGE HRTEM

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Energy storage, nanocatalysis, ultrathin dielectrics, and quantum materials increasingly rely on single-atom control of defects, dopants, interfaces, and phase formation in two-dimensional systems. Achieving this level of precision requires electron microscopy that combines true atomic resolution with carefully tuned interaction energies while preserving beam-sensitive and metastable structures. The Cc/Cs-corrected Sub-Ångström Low-Voltage Electron Microscopy (SALVE) instrument at Ulm University uniquely provides this capability. Applied to transition metal dichalcogenides, transition metal phosphorus trichalcogenides, confined nanoparticles, and ultrathin oxides [3–8], SALVE reveals defect-mediated functionalization pathways and atomic-scale reactivity with unprecedented clarity. Most strikingly, we identify a previously unrecognized atomic state in liquid metals: stationary atoms coexisting with mobile liquid atoms during solidification [9], uncovering fundamentally new atomic-scale physics. In metastable 2D hybrid systems, we demonstrate vapor-phase epitaxy of hexagonal CuI on graphene, formed unexpectedly through reaction of HI vapor with the copper TEM grid and stabilized on monolayer graphene by cooperative mechanical coupling between the soft ionic overlayer and the stiff substrate [10].

Extending atomic-scale control from inorganic to molecular-scale organic systems, we image single-crystalline two-dimensional covalent organic frameworks (COFs) [11] at 120 kV, an operating condition that optimally balances chemical integrity and structural resolution [12–14]. Through in-situ liquid-cell TEM of metal–organic frameworks (MOFs), we further create and directly observe new MOF structures during growth [15]. In addition, we visualize twisted two-dimensional polymer layers and their Moiré-modulated lattice relaxation, thereby expanding twist engineering beyond transition metal dichalcogenides to covalently bonded 2D frameworks. Finally, by correlating atomic structure with optoelectronic functionality, we demonstrate exciton confinement not only in perovskite-WSe_2 heterostructures [16] but also in hybrid 2D polymer–WSe_2 systems, extending quantum localization concepts across fundamentally different bonding classes.

Together, these results establish Cc/Cs-corrected low-voltage TEM as a key enabling platform for discovering and engineering emergent states of matter in functional two-dimensional materials.

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Model for High-Resolution Secondary Electron Imaging

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We introduce a model for secondary electron imaging at high resolution, obtained as an atomic-sized probe is scanned across the specimen in STEM. This model considers the dynamics of the underlying ionization events that are the origin of the signal providing the atomic resolution images. We argue that the short mean free path for inelastic scattering of the secondary electrons inside the specimen largely renders the directional dependence, inherent in the ionization dynamics, unimportant when they finally leave the sample. It therefore suffices to consider appropriately scaled total ionization form factors, which can be calculated for each atomic sub-shell individually [1], allowing us to study their relative contributions to the SE image contrast. Dielectric screening of the ionization, particularly pertinent for loosely bound electrons, is taken into account using a simple parametrization requiring only the input of a characteristic screening energy. Due to the screening, only localized sub-shells remain effective, providing the high-resolution contrast. The model also employs a straight-forward approximate description of the attenuation of the secondary electron signal from different depths in the specimen, requiring only an effective attenuation length as input. The model is illustrated by application to secondary electron imaging of the MCM-22 zeolite [2]. The comparison with experiment shows good agreement at atomic scale resolution. In a second example we show that the model suffices to reproduce the general features of secondary electron images of Ni nanoparticles on a $SrTiO_3$ substrate [3].

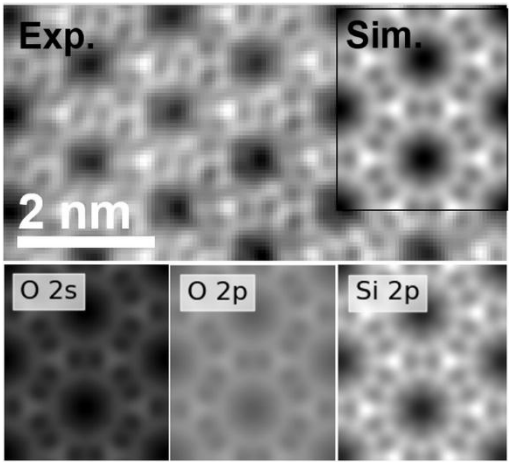

Fig. 1: STEM SE images of the MCM-22 zeolite from experiment and simulation (overlayed) with the main contributions shown at the bottom.

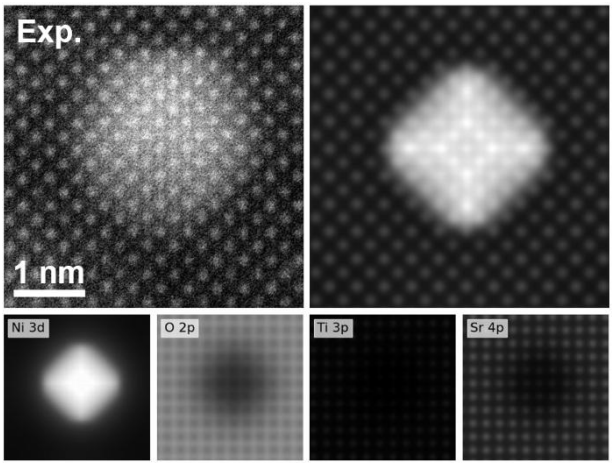

Fig. 2: STEM SE images of a Ni nanoparticle on $SrTiO_3$ [100] substrate from experiment [3] and simulation with the main contributions shown at the bottom.

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