Frontiers in Correlative Material Characterization: Samples, Techniques, Instrumentation and Data Management

Italian-German WE-Heraeus-Seminar

02 Apr - 05 Apr 2024

at the Physikzentrum Bad Honnef, Germany

WILHELM UND ELSE HERAEUS-STIFTUNG

Introduction

The Wilhelm und Else Heraeus-Stiftung is a private foundation that supports research and education in science with an emphasis on physics. It is recognized as Germany's most important private institution funding physics. Some of the activities of the foundation are carried out in close cooperation with the German Physical Society (Deutsche Physikalische Gesellschaft). For detailed information see https://www.we-heraeus-stiftung.de

Aims and scope of the Italian - German WE-Heraeus-Seminar:

Material characterization is essential for providing quantitative information about structure-function and structure-activity relationships in materials. This binational (German-Italian) seminar aims to bring together researchers working on correlative, multiscale and multimodal material characterization from a wide range of scientific disciplines, encompassing experimental technique development, modeling of materials and the development of hardware and software standards for combining data recorded using complemetary analytical techniques. Topics that will be addressed include approaches for sample preparation, sample transfer, the optimization of experimental workflows, data acquisition, data management, image registration, the use of artificial intelligence for image analysis and instrument control, and the creation of digital twins of materials and experiments. In addition to invited talks by experts in the field, scientists and students will be encouraged to contribute actively to the seminar through discussions through poster resentations.

Scientific Organizers:

PD Dr. Axel Neffe   Helmholtz-Zentrum, Hereon, Germany
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Prof. Dr. Rafal E. Dunin-Borkowski   Forschungszentrum, Jülich, Germany
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Dr. Regina Ciancio   Area Science Park, Trieste, Italy
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Introduction

Administrative Organization:

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Registration:

Martina Albert (WE-Heraeus Foundation)
at the Physikzentrum, reception office
Tuesday (10:00 h – 14:00 h)
and Wednesday morning

Program

Program (CET)

10:00 – 14:00	**Registration**	
12:35	*LUNCH*	
14:00 – 14:45	Scientific Organizers	**Welcome and Opening**

Chair: Rafal E. Dunin-Borkowski; Regina Ciancio and Axel Neffe

14:45 – 15:30	Mathieu Kociak	**Light in the electron microscope: from correlated to coincident measurements**
15:30 – 16:00	*COFFEE BREAK*	

Chair: Silke Christiansen

16:00 – 16:45	Rosella Aversa	**Metadata Management in correlative characterization: tales from the Metadata WG**
16:45 – 17:30	Dieter Weber	**Tools and skills to survive the frontier**
17:30 – 18:15	Vincenzo Grillo	**Intelligent experiments in sample analysis and beam control for electron microscopy**
18:15 – 18:30	Stefan Jorda	**About the Wilhelm and Else Heraeus Foundation**
18:30 – 20:00	*DINNER*	
20:00 – 22:00	**Poster Session I**	

Program (CET)

Wednesday, 03 April 2024

07:30	*BREAKFAST*	

Chair: Dorota Koziej

08:30 – 09:15	Regine Willumeit-Römer	**Correlative characterization of degradable Mg-based implants**
09:15 – 10:00	Alberta Bonanni	**Correlative characterization: a perspective for quantum materials**
10:00 – 10:20	*COFFEE BREAK*	

Chair: Fabio Giavazzi

10:20 – 11:05	Pasquale Orgiani	**Crafting the structure of materials for tailoring their quantum properties: the case of TiO2 thin films**
11:05 – 11:50	Jacob Hoogenboom	**From millimeters to molecules: Integrated microscopy with coincident electron, photon and ion beams**
11:50 – 12:35	Subin Lee	**Pushing Boundaries of SEM for Richer Information**
12:35	*LUNCH*	

Chair: Marc-Georg Willinger

14:00 – 14:45	Robin E. Schäublin	**Correlating microscopy methods: the case of precipitates in lean, bioabsorbable Mg alloys**
14:45 – 15:30	Andrew Stewart	**Leveraging Electron Microscopy for Enhanced Process Analytical Technology in Nanomaterials Production**
15:30 – 16:00	*COFFEE BREAK*	

Wednesday, 03 April 2024

Chair: Rafal E. Dunin-Borkowski

16:00 – 17:20   *Short presentations*
(4 selected talks)

	Alexey Boubnov	**Correlative Characterization: Bridging 2D/3D Imaging with Micro-Structuring**
	Berit Zeller-Plumhoff	**Deep learning for digital volume correlation**
	Sandeep Kumar Chaluvadi	**Unveiling Room-Temperature Ferromagnetism in 2D Van der Waals Material: Cr4Te5**
	Malika Khelfallah	**Characterization of self-assembly in ferrofluids**

18:30 – 20:00   *DINNER*

20:00 – 22:00   **Poster Session II**

Program (CET)

07:30	*BREAKFAST*	

Chair: Mathieu Kociak

08:30 – 09:15	Marc-Georg Willinger	**The beauty and complexity of non-equilibrium dynamics in simple reaction systems**
09:15 – 10:00	Fabio Giavazzi	**Probing multi-scale dynamics of soft and bio-soft matter with differential dynamic microscopy**
10:00 – 10:20	*COFFEE BREAK*	

Chair: Axel Neffe

10:20 – 11:05	Francesca Toma	**Correlative Characterization of Light Absorbing and Catalytic Materials for Solar Fuel Generation**
11:05 – 11:50	Lorenzo Albertazzi	**A correlative view on nanomaterials**
11:50 – 12:35	Dorota Koziej	**Probing Hybrid Structures Across the Length Scales with Synchrotron-based X-rays Methods**
12:35	*LUNCH*	
14:00 – 18:30	**Conference photo and Excursion to Adenauer Haus**	
18:30	*HERAEUS DINNER*	
	(social event with cold & warm buffet and complimentary drinks)	

Program (CET)

Friday, 05 April 2024

07:30   *BREAKFAST*

Chair: Regine Willumeit-Römer

08:30 – 09:15   Maria Harkiolaki   **The development of a correlative cryo-imaging platform for the investigation of ultrastructure at the cellular level at near-physiological states to nanometer resolution**

09:15 – 10:00   Alex Aubert   **Correlative probing of macroscopic and microscopic functional properties of magnetic materials**

10:00 – 10:20   *COFFEE BREAK*

Chair: Regina Ciancio

10:20 – 11:05   Markus Kühbach   **On Implementing Comprehensive Documentation for Correlative Microscopy Workflows for Materials Science Examples: Lessons of Fun, Pain, and Gain Learned**

11:05 – 11:50   Silke Christiansen   **Context Microscopy**

11:50 – 12:30   Scientific organizers **Poster Prize Awards & Closing Remarks**

12:35   *LUNCH*

End of the seminar and departure

Posters

Posters I

Arno Annys

Fully automated materials characterization from core-loss electron energy loss spectroscopy by deep learning

Alexey Boubnov

Correlative Characterization: Bridging 2D/3D Imaging with Micro-Structuring

Maria Brollo

ReMade@ARI - Advanced materials characterization for a circular economy

Sandeep Kumar Chaluvadi

Unveiling Room-Temperature Ferromagnetism in 2D Van der Waals Material: Cr4Te5

Tjark Gröne

Towards In-situ X-ray Scattering studies of polynuclear cluster

Maximilian Kabbe

Enhancing correlative characterizations Measurement quality with spatial sample alignment

Andras Kovács

Direct observation of tensile-strain-induced magnetic hardening in a ferromagnet

Qianqian Lan

Quantification of Interface Magnetism of La0.7Sr0.3MnO3 thin film by Off-axis Electron Holography

Yue Liu

Tortoise beetle-inspired sustainable films with tunable properties based on on-demand embedding and release of functional microspheres

Karola Luetzow

Stereochemical Heterogeneity Analysis of Polylactides

Jan Reimers

Correlative in situ synchrotron radiation-based nano CT and (S)TEM imaging of biodegradable Mg-based alloys

Posters II

Mallika Chaukar

Correlative Light Electron Microscopy for Polymeric Material Characterization

Michael Faley

Nanofabrication of superconducting and ferromagnetic structures for operation in TEM

Sumea Klokic

Deducing structural dynamics in responsive crystalline film systems: a multi-technique approach

Thi Thu Le

In-situ Characterization Techniques to the Study of Hydrogen Storage Materials

Josef Armando Mandlule

Correlative characterization of stereocomplex formation in aliphatic polyester P(PCL-b-PLLA) block-copolymers

Ousssama Oulhakem

Effect of water intercalation into tungsten trioxide structure (WO3.xH2O) (x=0,1,2): Correlation between structure and photocatalytic performance

Marc Raventós

XRD steel enhanced-characterization through computational methods

Aurys Šilinga

Advanced transmission electron microscopy of the three-dimensional magnetization distribution in a domain wall-pinning Sm-Co-based permanent magnet

Richard Thelen

Implementation of Correlated Characterization in Surface Metrology

Govind Ummethala

Following Organic Crystal Growth Dynamics During Liquid Reagent Mixing using Liquid Cell Transmission Electron Microscopy

Berit Zeller-Plumhoff

Correlative µCT and histological imaging of bone implants

Abstracts of Lectures

(in alphabetical order)

A correlative view on nanomaterials

L. Albertazzi[1]

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While most of the applications and developments of correlative imaging are in biology, there are many other fields where these techniques can make a big impact. One clear example is the fields of nano- and bio-materials. Chemists and materials scientists are producing materials with complex features for a variety of relevant applications in medicine, and the characterization of such materials is crucial. Understanding materials structure, function and interactions with target cells is important to guide the design of successful materials. Here we will show how correlative imaging can play a pivotal role in the field of nanomaterials. Starting from our expertise on super-resolution microscopy we developed new workflows for nanomaterials imaging. Combination of single-molecule localization microscopy techniques such as STORM and PAINT with electron microscopy proved very useful to study nanomaterials size, shape, functionality and trafficking inside cells. Examples based on polymeric nanoparticles will be provided and the perspective of correlative imaging for nanomaterials science discussed.

Correlative probing of macroscopic and microscopic functional properties of magnetic materials

A. Aubert[1], K. Skokov[1], A. Rogalev[2], F. Wilhelm[2], G. Gomez[3], H. Wende[3], K. Ollefs[3], O. Gutfleisch[1]

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[3]*Faculty of Physics, University of Duisburg-Essen, 47057 Duisburg, Germany*

Materials undergoing first-order phase transitions are crucial for new solid-state refrigeration methods. External stimuli, like altering magnetization with a magnetic field, induce substantial transformations in other subsystems such as crystal lattice, electrical resistivity, sample temperature etc. Understanding the nature of these interactions is vital for optimizing the hysteresis of this material [1,2].

In this talk, we will introduce new instruments which have been implemented at our home laboratory in Darmstadt [2] and at the beamline ID12 of the European Synchrotron Radiation Facility (ESRF) [3]. The "ULMAG—Ultimate MAGnetic characterization" instrument offer the ESRF users a unique possibility to measure under strictly the same experimental conditions the element-specific X-ray absorption spectroscopy (XAS)/ X-ray magnetic circular dichroism (XMCD), high-resolution XRD simultaneously with the measurement of various macroscopic properties (magnetization, volume changes, magnetocaloric properties, resistivity etc.), all as a function of magnetic field (up to 7 T) and temperature (5–325 K) [3].
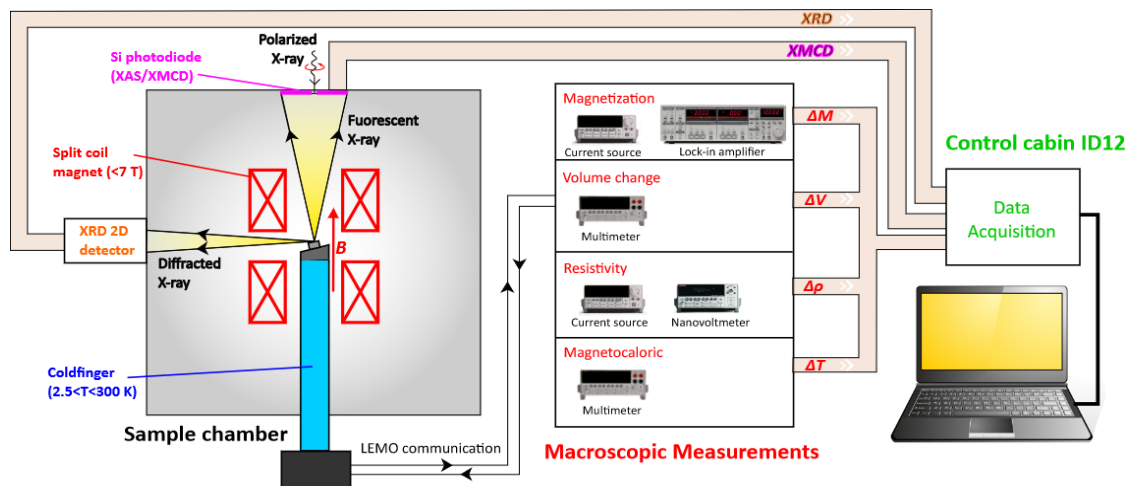

Fig. 1 Schematic of the ULMAG prototype implemented at the beamline ID12 of the ESRF

References

[1] K. P. Skokov, et al. Appl. Phys. Rev. 10 (3): 031408 (2023)
[2] D. Yu. Karpenkov et al. Phys. Rev. Applied 13, 034014 (2020)
[3] A. Aubert et al., IEEE Trans. Instrum. and Meas, 71, 1-9, 6002409 (2022)

Metadata Management in correlative characterization: tales from the Metadata WG

R. Aversa[1]

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Correlative characterization is a highly multi-disciplinary research field where scientists often need to combine different types of information from co-referenced (in time or space) multi-modal data obtained using different techniques. Each measurement or analysis method results in datasets which have to be combined in order to obtain complementary insights on a region of interest and to relate features and properties of different samples across multiple length scales over time.

Metadata management plays an important role in correlative characterization: in particular, the combination of information and the data exchange can be facilitated if the FAIR (Findable, Accessible, Interoperable, Reusable) guiding principles [1] are followed. In addition, effective metadata management enhances discoverability, accessibility, and reproducibility of scientific research data.

For this reason, we founded the Metadata Working Group (WG) [2], including members of the three Helmholtz centers involved in the Joint Lab "Model and Data driven Materials Characterization" (JL MDMC) and of the Nanoscience Foundries and Fine Analysis Europe Pilot (NEP). The activities of the group are focused on the development of communal descriptions and interoperable solutions, driven by the communication with scientists to collect information and requirements.

This talk provides an overview of the current results achieved by the Metadata WG, the ongoing collaborations with other projects, and the planned future steps.

References

[1] M. D. Wilkinson et al., Scientific Data **3**, 160018 (2016)
[2] https://jl-mdmc-helmholtz.de/mdmc-activities/metadata-working-group/

Correlative characterization:
a perspective for quantum materials

Alberta Bonanni

Institute for Semiconductor and Solid State Physics, Johannes Kepler University, Linz - Austria

The field of quantum materials has witnessed a phenomenal success in the prediction and discovery of new symmetry protected topological phases with strong spin-orbit coupling, of ultra-tunable materials platforms based on beyond-graphene van der Waals material systems and of emergent quantum phases in conventional and unconventional superconductors [1,2]. The identification of pathways for the experimental realization of quantum phases of matter and for manipulating and controlling the quantum properties of these materials is a central task of modern condensed matter physics and holds the key to the success of quantum technology.

Correlative characterization, complemented with the applications of machine learning and artificial intelligence is expected to guide the research on quantum materials and quantum technology. A multiscale approach [3] for the collection of multiple datasets from complementary characterization techniques and the spatial registration of multidimensional data library, is in fact needed for a comprehensive databank categorizing large families of quantum material systems based on symmetry, topology and electronic properties.

In this perspective, an outline of a correlative characterization approach used to unravel the magnetic, optical, structural and electronic properties of a variety of relevant quantum materials including the Rashba semiconductor n-GaN:Si [4], the Weyl semimetal T_d-WTe$_2$ [5], Fe doped NbN [6], the magnetic topological insulator SnMnTe [7] and Mn(Bi,Sb)$_2$Te$_4$ [8], the van der Waals antiferromagnet FePS$_3$ [9] and altermagnet MnTe [10] will be presented. Further, an overview of the prospective use of these material systems in quantum sensorics, quantum metrology and of their application as detectors for high energy physics and dark matter physics will be provided.

References:

1. D. N. Basov *et al*. Nat. Mater. **16**, 1077 (2017).
2. Y. Tokura *et al*. Nat. Phys. **13**, 1056 (2017).
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10. D. Bossini *et al*. Phys. Rev. B **104**, 224424 (2021).

Probing multi-scale dynamics of soft and bio-soft matter with differential dynamic microscopy

F. Giavazzi[1]

[1]*Dipartimento di Biotecnologie Mediche e Medicina Traslazionale, Università degli Studi di Milano, Segrate (MI) 20054 , Italy*

Imaging is, in its many forms, a cornerstone of modern science. From astronomy to cell biology, from medicine to material science, obtaining space-resolved maps - images – of the system under study is considered a key step in the process of "understanding" it. Nonetheless, releasing the full informative potential of an image or an image sequence can be a challenging task, in particular when the objects of interest are small, or the system is crowded, or the environment is complex, or the detection chain is noisy, or all the above together.

In my presentation, I will introduce the basics of differential dynamic microscopy - an image analysis scheme based on the study of time correlations in the spatial Fourier domain - enabling the extraction of statistically robust information on the dynamics of a sample from a sequence of images, even in conditions where a direct-space approach, based for example on image segmentation or particle tracking, would be difficult or impossible [1,2].

I will discuss potential and limitations of the method, presenting selected applications out of a broad spectrum of soft and bio-soft matter problems, ranging from thermal fluctuations in liquid crystals and critical fluids to intracellular transport processes, from microrheology of complex fluids to sizing of proteins in diluted solutions.

I will then focus on recent results in the investigation of the microstructural changes induced in soft materials by this imposition of an external mechanical load exceeding the material yield-stress, obtained via a combination of quantitative optical microscopy and oscillatory shear rheology.

References

[1] R. Cerbino, V. Trappe, *Phys.Rev.Lett.* 2008, **100**, 188102.

[2] F. Giavazzi, D. Brogioli, V. Trappe, T. Bellini, R. Cerbino, *Phys. Rev. E: Stat., Nonlinear, Soft Matter Phys.* 2009, **80**, 031403.

Intelligent experiments in sample analysis and beam control for electron microscopy

E. Rotunno[1], P. Rosi[1] L. Viani[1] P. Habibzadeh Kafkani[1,2], M.Beleggia[2], A.Tavabi[3], R.E. Dunin-Borkowski[3] and <u>V. Grillo[1]</u>

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The electron microscope is a very powerful mean not only to observe a sample but also to measure its properties and do spectroscopy. It is also a very powerful optical bench for electrons ideal to explore quantum mechanics.

Having an optimized and automatic control of the experiment, a streamlined workflow of the analysis and a real time alignment of the instrument are all things that increase the relevance of the electron microscopy in many contexts.

At the same time a real time control of the electron beam could allow the creation of innovative experiments that further enhance the resolution and possibilities of electron microscopy.

I will therefore show recent results and suggest a few examples of "intelligent experiments" , the current lines of research and futuristic ideas.
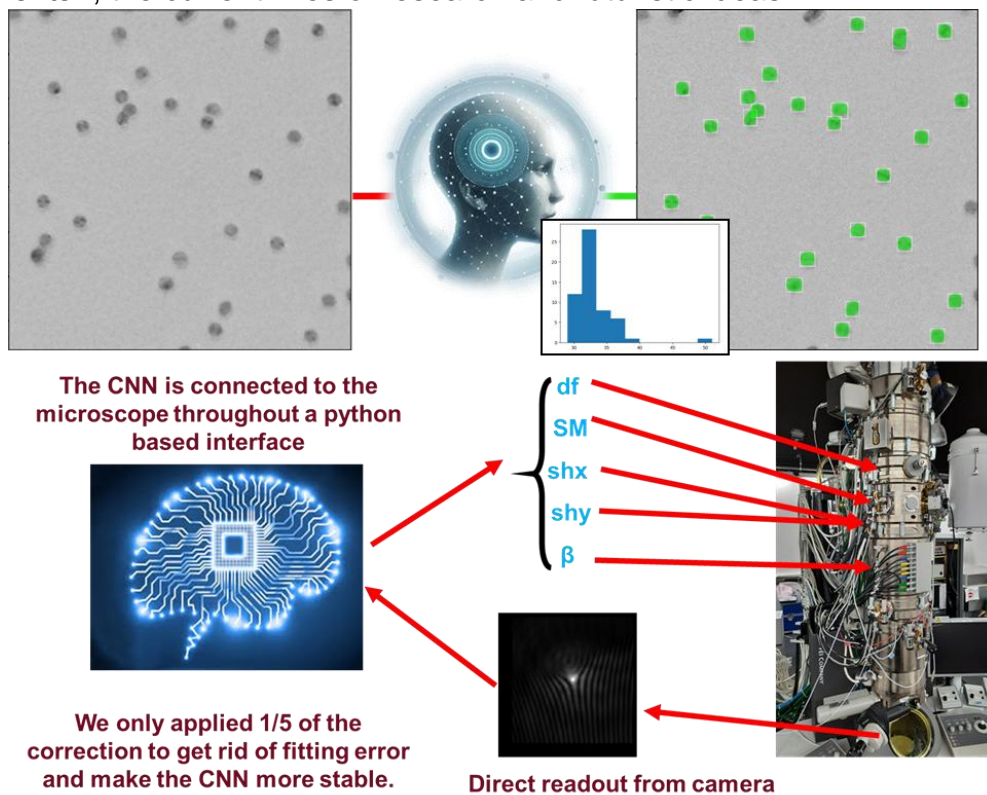

Fig 1 example of use of AI in microscopy for image analysis and for direct microscope control

The development of a correlative cryo-imaging platform for the investigation of ultrastructure at the cellular level at near-physiological states to nanometer resolution.

M Harkiolaki,
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Biological imaging has developed rapidly in recent years across scale and resolution ranges but to capture the intricate interplay of structures and processes within soft-condensed living matter a multitude of imaging methods need to work together. At the biological cryo-imaging beamline B24, at the UK synchrotron, Diamond Light Source, two high end 3D imaging systems have been developed side-by-side to enable the in-depth examination of biological systems at near physiological states[1,2]. The two imaging methods are: soft X-ray tomography (SXT) which uses the natural absorption contrast of hydrated biological material, such as cells and tissues, to deliver nanometer resolution 3D data (Figure 1) and fluorescence structured illumination microscopy (SIM) which highlights localisation of tagged molecules, organelles and other structures within the cellular maps captured through X-ray imaging. The fully commissioned workflow starts with a sample first mapped using conventional imaging techniques at room temperature and cryogenic temperatures which lead to cryo-imaging using SIM to identify and characterise areas of interest before the samples is loaded to the transmission X-ray microscope for SXT data collection on these exact areas of interest. This way, data recorded at different microscopes can be directly correlated enabling the unambiguous interpretation of data. The beamline workflow has been developed with accessibility and ease-of-use in mind and it will be presented with examples of recent data collected along with highlights and lessons-learned of the correlative scheme employed.
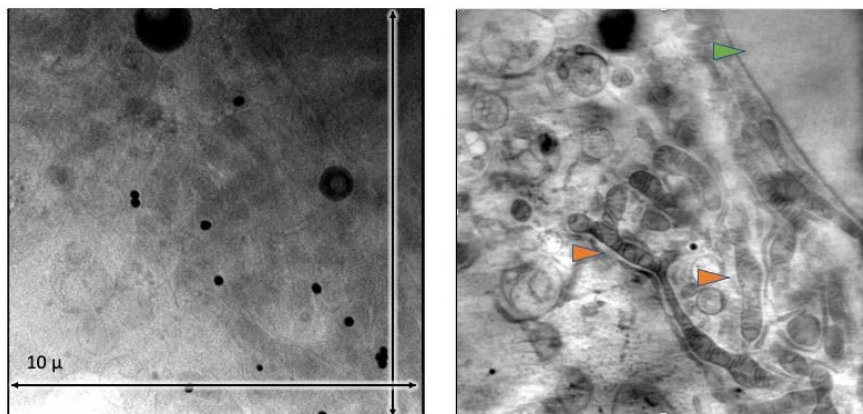
Figure 1. SXT data from a human osteosarcoma cell collected at beamline B24.

Raw data single projection (left panel) and the corresponding reconstructed data instance (panel on the right) with representative mitochondria (orange arrows) and the nucleus (green arrow) labelled.

References
[1] I Kounatidis et al. *Cell*, **182**(0), 1–16 (2020). https://doi.org/10.1016/j.cell.2020.05.051
[2] C Okolo. *Biochem Soc Trans* **50**(2): 649-663 (2022). https://doi.org/10.1042/BST20210886

From millimeters to molecules: Integrated microscopy with coincident electron, photon and ion beams

Jacob Hoogenboom

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Light in the electron microscope: from correlated to coincident measurements

M. Kociak[1], **Y. Auad**[1], **N. Varkentina**[1], **M. Bézard**[1], **J. Baaboura**[1], **F. Castioni**[1], **X. Li**[1], **J-D. Blazit**[1], **O. Stéphan**[1], **L. Tizei**[1]

[1]*LPS, CNRS, Université Paris-Saclay, Orsay, France*

There are many reasons why electron microscopy and optical microscopy should be combined. For example, CLEM is widely used in bio-imaging to combine the (ultra)structural information obtained from electrons in a TEM with the functional information obtained from photons in luminescence. In physics and materials science, the emission of light from an irradiated sample, otherwise known as cathodoluminescence (CL), has long been correlated with the structural information available in a SEM. More recently, with the advent of fast electron nanooptics, it has become clear that EELS spectroscopy also provides access to the optical properties of nanomaterials [1].

Recent advances, both conceptual and technical (optimized light collection/injection systems in STEMs [1], monochromators [2], etc...) have made it obvious that different types of optical spectroscopy need to be correlated in an electron microscope.

In this talk, I will present different strategies for correlating luminescence (CL) and extinction or absorption (EELS) signals in a TEM for physics and material science applications, including plasmonic [3], photonic [4] or semiconductor [5] systems. Beyond correlations, I will show how and why to measure CL and EELS signals *in coincidence* [6] using ns-resolved direct electron detectors [7]. If time permits, I'll finish by describing another form of experiment involving coincidence between electrons and photons [8]. This time, the photons come from a pulsed laser synchronized with the EELS detector, providing the spectral resolution of a laser combined with the spatial resolution of a STEM.

References

[1] M. Kociak and L. F. Zagonel, Ultramicroscopy **174**, 50 (2017).
[2] O. L. Krivanek et al., Nature **514**, 209 (2014).
[3] A. Losquin et al. Nano Lett. **15**, 1229 (2015).
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Probing Hybrid Structures Across the Length Scales with Synchrotron-based X-rays Methods

Prof. Dr. Dorota Koziej

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The key to fabricate complex, hierarchical materials is the control of chemical reactions at various length scales. To this end, the classical model of nucleation and growth fails to provide sufficient information. We will illustrate how a combination of in situ X-ray spectroscopic, scattering, and microscopic studies bridge the molecular- and macro- length scales. [1-3] Moreover, we will present how synchrotron methods, far from merely providing new tools, are extending the ways we study, understand, and design such complex structures. It gives complementary information about chemical reaction in solution and nucleation, growth and crystal phase transition of nanoparticles and their functionality in devices.

On example, of 3D printed Au/TiO_2 aerogels we will show how hierarchical design, inspired by light scattering in clouds during photochemical processes and from mass transfer in plants during photosynthesis booster hydrogen production in the gas phase. [4-5]

Finally, to demonstrate the advantages of the operando X-ray scattering (PDF & SAXS) measurements we will discuss the photodegradation of $CuBi_2O_4$ photoanodes. [6]

References:

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On Implementing Comprehensive Documentation for Correlative Microscopy Workflows for Materials Science Examples:
Lessons of Fun, Pain, and Gain Learned

M. Kühbach[1]

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Workflows for correlative microscopy combine e.g. spatiotemporal or multi-method spectroscopic data that have been collected with different methods or instruments to obtain a more multifaceted characterization of the physical mechanisms and defect structures which control structure-function correlations of materials. Workflows encode specific assumptions about the frames of references, analysis conditions, and the parameterization used for the underlying physical models and algorithms. Different types of representations, conventions, and formats are available for serializing these pieces of information to e.g. files or database entries. Consequently, when multiple of these need to be mastered, such as at the junctions of data analysis workflows, format conversions are a frequently required task. If not documented carefully, if defined ad hoc, or if communicated incompletely, such workflows lack contextualization rendering attempts to repeat the described research difficult. These challenges apply also to correlative microscopy with electrons and atom probe.

In my talk I will provide two examples from materials science how the documentation of workflows for correlating pieces of information collected in experiment and simulation can be improved. The first example will be taken from the field of developing software for analyzing the colocation of microstructural features. For this example, methods from computational geometry were applied and made usable for datasets of domain scientists who wish to characterize how parameterization affects the characterization of microstructural features such as precipitates using atom probe tomography. The second example documents recent work from the FAIRmat consortium within the German National Research Data Infrastructure. I will report how we have formulated com-prehensive data schemes that are vendor- and research data management-system-agnostic including electron microscopy and atom probe. I will exemplify how these can be used for documenting data collected with electron microscopy. I would also like to share some of the practical experiences we had with implementing these examples.

Pushing Boundaries of SEM for Richer Information

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This presentation demonstrates two innovative characterization techniques in scanning electron microscope (SEM) which can provide much more rich information in microstructure analysis by pushing the boundaries of microstructural characterization in SEM.

ECCI emerges as a powerful tool for defects analysis, offering insights into dislocations and crystallographic defects. Despite its potential to replace TEM dislocation analysis, ECCI faces challenges in quantitative analysis. Addressing this limitation, we present a semi-automated tool for dislocation Burgers vector analysis using SEM, enabling comprehensive dislocation characterization in a large area. Our new approach is demonstrated in various materials including ferritic steel, Ni-based superalloys and even ceramic materials such as STO.

In parallel, 4DSTEM (Scanning Transmission Electron Microscopy)-in-SEM extends the capabilities of SEM by capturing information from reciprocal space. By using a direct electron detector, we establish a protocol for fast electron diffraction pattern acquisition within the SEM. Operating in event-driven mode, this method enhances data acquisition rates, facilitating in situ measurements and dynamic observations.

Together, these advancements provide new aspects in microstructural characterization in SEM, promising richer insights and broader applications across materials science and engineering.

Crafting the structure of materials for tailoring their quantum properties: the case of TiO$_2$ thin films

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Quantum materials continuously attract huge interest due to the widespread technologically important phenomena which can display. In this respect, the thin film technology is currently able to go beyond the single-crystals technology by allowing the investigation of the physical properties of quantum materials in the limit of single atomic layers, through the controlled deformation of the structural parameters by substrate induced strain and growth of multi-layers, thus ultimately providing key information for the development of real layered device applications. The full control of the electronic properties of quantum materials can be therefore achieved through the crafting of their structural properties.

As key-example, I here show a cross-correlative investigation of surface-vs-bulk electronic/chemical/structural properties of anatase titanium dioxide TiO2 thin films grown by Pulsed Laser Deposition [1,2] by a combination of synchrotron-based photo-emission, absorption and optical spectroscopies, x-ray/electron diffraction and electron microscopies. In more detail, localized in-gap states located around 1.5-2.0 eV of binding energy have been correlated to the presence of Ti-ions into a 3+ valence state and can be controlled by the overall oxygen content. In addition, a 2-dimensional electron gas (2DEG) has been also observed at its surface and correlated to Ti4+ electronic configuration. Differently from other Ti-based system, the 2DEG appears to be decoupled by the localized in-gap states and generated by a very local chemical doping in the near-surface region. Its occurrence has been also investigated as a function of the film thickness down to a single-unit-cell of anatase TiO2 thus ruling out the request of a minimum critical thickness as shown in other oxide systems. Moreover, it can be modulated in the k-space by different surface reconstruction of the films as well as by substrate-induced epitaxial strain mechanism. This last, in particular, can be also used to tune the optical absorption of the TiO2.

The case of anatase TiO2 thin films can fully show how to refine the understanding of the surface-vs-bulk electronic properties of quantum materials by taking advantage of a cross-correlative approach. The capability to shed light on the evolutions of the electronic properties allows the grasping of important concepts in fundamental problems related to the interface physics and ultimately contributes to their practical use ad building bricks of an emerging new technology.

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Correlating microscopy methods: the case of precipitates in lean, bioabsorbable Mg alloys

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Many of the properties of metallic systems are driven by nanoscale precipitation. We are deploying a correlative approach, utilizing mainly atom probe tomography (APT) and transmission electron microscopy (TEM), to cover a wide range of spatial length scales and obtain quantitative information. Despite its power, the correlative approach presents technical hurdles: sample preparation requires meticulous methods, including focused ion beam. Ideally, the same region of interest should be analyzed across all instruments, which can be challenging considering the vastly different size scales involved. Minimizing damage or corrosion during transfer between instruments, which usually occurs in air, is also crucial. Finally, to fully leverage the harvested data, one must apply data-treatment techniques, including AI and simulations to aid data interpretation.

We present our workflow in the correlative study of "ZX" lean Mg alloys, which contain zinc and calcium, as a test case. Such alloys are promising candidates for bioabsorbable implants due to their excellent biocompatibility and degradation rate matching tissue regeneration. Because the nanoscale intermetallic precipitates within the alloys drive the mechanical properties and degradation behavior, it is critical to understand their type and role to tailor the alloys for specific applications. Starting from the solid solution, we were able to elucidate the precipitation sequence via APT at the smallest scales, and, as precipitates matured and grew in size, with STEM imaging and EDS chemical mapping supported by simulations. We revealed that the debated equilibrium phase of the ternary precipitates is based on the $Ca_2Mg_5Zn_5$ crystal [1]. However, its composition derived from EDS and APT presented a broader range, which we also noted in the correlative nanoscale analysis of Fe–Cr alloys [2, 3]. This showed us that the correlative evaluation of APT- and EDS-derived results is beneficial when assessing a composition. We conclude with an outlook on further correlative approaches, including X-ray nanotomography.

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Leveraging Electron Microscopy for Enhanced Process Analytical Technology in Nanomaterials Production

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The integration of Process Analytical Technologies (PAT) into the manufacturing processes of advanced materials is pivotal for optimizing production efficiency and ensuring compliance with Good Manufacturing Practice (GMP) [1] and regulatory standards. As nanoscale materials transition from research to industrial applications [2], the limitations of traditional PAT tools at the nanometer scale become evident. Traditional methods, including laser scattering, dynamic light scattering (DLS), Raman spectroscopy, and turbidity measurements, while effective at the micron scale, offer averaged data that may not meet the stringent quality requirements for nanomaterials. These requirements demand precise control over the size and morphology of nanomaterials, beyond what averaged metrics can provide.

Electron microscopy (EM) emerges as a promising alternative, offering unparalleled resolution and analytical capabilities at the nanoscale. However, its integration into PAT frameworks faces challenges, such as the manual sample handling and the slow, qualitative data interpretation. We propose a pathway for incorporating electron microscopy into a multi-PAT environments for nanomaterials analysis. By addressing the current bottlenecks in sample handling and data processing, electron microscopy can significantly enhance the quality control of nanomaterials in a range of applications, from pharmaceuticals to additive manufacturing and beyond.

Furthermore, this approach aligns with the evolving landscape of automated research laboratories across various scientific disciplines, particularly where the push for rapid discovery and development of new materials and medications is critical. As the demand for advanced materials grows, integrating sophisticated analytical tools like electron microscopy into PAT systems will be key to meeting the challenges of the 21st century. We will outline strategies for overcoming the limitations of current EM-PAT integration and discuss the broader implications for material science, engineering, and the biosciences.

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Correlative Characterization of Light Absorbing and Catalytic Materials for Solar Fuel Generation

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Carbon neutral energy sources that are scalable, deployable, and cost effective will be required at an unprecedented scale to halt irreversible climate change. To design novel materials for solar fuel generation that can efficiently produce energy with minimal impact on the environment, correlative characterization of the material components and integrated systems under operating conditions can provide complete understanding of their properties and inform how to tune their behavior. Here, I will focus on different light absorbing materials and catalysts and their correlative characterization, spanning microscopy, spectroscopy, and scattering to shed light on their working mechanism. We will show the synthesis and the advanced characterization of integrated semiconductors and catalysts for (photo)electrocatalytic systems as they can be used under realistic operating conditions for solar fuel production.

Tools and skills to survive the frontier

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Life at the frontier (of Correlative Material Characterization): A pathless wilderness that draws in fearless explorers. Adventure and opportunities await, but also hardship. Here we discuss which tools and skills can help to survive in such an unstructured and untamed environment off the beaten paths.

Just like in the wilderness, we cannot expect that others build our infrastructure for us before we arrive: *WE* are the frontier, meaning we are tasked with getting the basics up and running quickly for our own survival and the people arriving after us. Following selected examples, we explore how composition of simple tools combined with suitable development methodologies can deliver quick results that are suitable for this dynamic environment.

The beauty and complexity of non-equilibrium dynamics in simple reaction systems

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As stated in the conference description, material characterization is essential for providing quantitative information about structure-function and structure-activity relationships in materials. In many cases, this relationship can only be understood if the material is characterized while it is in a relevant functional or working state. Processes and dynamics that dominate at different length and time scales should be considered. This is certainly the case in catalysis, where we try to understand processes that lead to the emergence of catalytic function. Taking the example of simple redox reactions catalyzed by non-noble metals, we find that the active catalyst operates near a phase boundary where metallic and oxidized phases coexist [1]. Real-time imaging reveals fascinating oscillatory redox dynamics that increase in complexity as the chemical potential of the gas-phase increases [2]. Moving from simple model catalysts to industrially relevant metal nanoparticles supported on reducible oxide supports, we use in-situ transmission electron microscopy to study effects related to a strong metal-support interaction (SMSI) under reactive conditions [3,4]. Correlative operando electron microscopy clearly shows that catalysis cannot be understood based on the study of static atomic arrangements of a system in thermodynamic equilibrium. Instead, we must consider complex non-equilibrium dynamics in which oscillatory behavior near phase boundaries provides conditions for continuous re-generation of active sites and catalytic turn-over.

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Correlative characterization of degradable Mg-based implants

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Mg and its alloys are a new class of load bearing metal implants because they degrade under physiological conditions. This amazing property offers significant advantages over classical, non-degrading implants which have to be removed upon the completion of the healing process. However, it is not easy to deal with Mg-based implant materials because the degradation in a biological environment is extremely complicated. In addition, we have not yet fully understand how the surrounding tissue and the immune system deals with the degradation products. Furthermore we need structural and chemical characterization across various length and time scales - for the material as well as for the biology. We therefore utilize several techniques for correlative and multimodal characterization, in some cases also in situ or operando approaches. This presentation will give an overview of the results for degradation phenomena ranging from nm to μm scale, load bearing degradation and correlated multimodal imaging, and will point out where there is need for further developments.

Abstracts of Posters

(in alphabetical order)

Fully automated materials characterization from core-loss electron energy loss spectroscopy by deep learning

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Electron energy loss spectroscopy (EELS) when combined with scanning transmission electron microscopy (STEM) forms a powerful tool for obtaining spatially resolved chemical information. Although core-loss EELS reveals chemical content in a very direct manner, conventional analysis methods such as peak finding algorithms struggle with the low signal-to-noise ratio and dominant background that are common in EELS. Identification of core-loss edges in EELS spectra thus usually remains a manual task for a human operator. This is slow, inconvenient, and inevitably leads to experimenter's bias and reproducibility issues. Deep learning is especially suited to tackle this problem because the analysis can be learned from training data instead of having to be algorithmically predefined. In this work, a simulated dataset is introduced with 736,000 labeled EELS spectra that represents 107 K, L, M or N core-loss edges through all 80 elements from Be to Bi. This dataset is used to train and evaluate a series of neural network (NN) architectures including convolutional NNs and attention-based transformer networks. The use of an ensemble of NNs allows for a further increase in performance and is used to demonstrate fully automated elemental mapping in a spectrum image. The element identification network is useful as a stand-alone tool, but an even more complete analysis is obtained by combining the NN with more flexible existing physical model-based tools like EELSMODEL [1]. This combination provides an entirely unsupervised workflow for quantification of the large datasets generated in modern STEM-EELS experiments. [2]
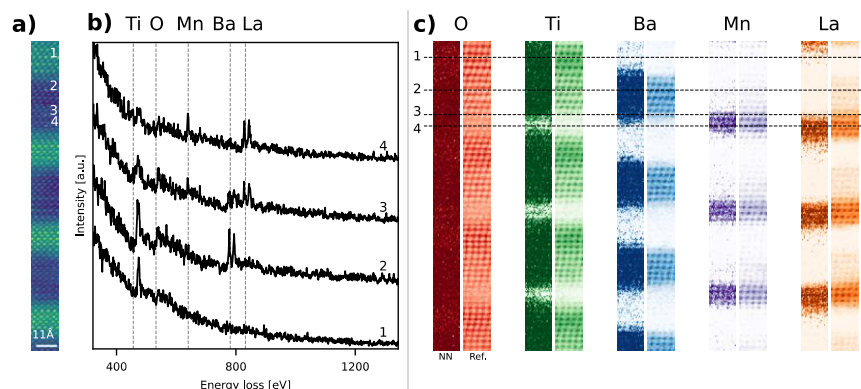
Figure 1: Fully automated elemental mapping of a $LaMnO_3/BaTiO_3/SrTiO_3$ superlattice.

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Correlative Characterization:
Bridging 2D/3D Imaging with Micro-Structuring

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Correlative characterization is the spatial registration of several imaging modalities with the aim to combine complementary information on the same region of interest [1]. At the same length scale, analyzing identical regions of interest using several modalities is well established in electron microscopy, e.g. various STEM imaging modalities (BF, DF, HAADF) with EELS and EDX spectroscopy or also the combination of APT and TEM. Across length-scales, this requires combination of different instruments. Without a correlative approach, the characterization of the full length-scale hierarchy relies on statistically random sample extracts, which only provides a good representation for very homogeneous samples. However, for macroscopically inhomogeneous samples, such as catalyst pellets, a correlative approach is indispensable to characterize the individual components of the sample. Therefore, for linking the local micro-structure to the macroscopic level of the sample, a full correlation of all applied techniques on the entire sample is desired.

We present a case study on an iron molybdate catalyst for selective oxidation of methanol to formaldehyde [2], possessing a complex mixture of single and mixed oxides, exhibiting different porosity and density, easily distinguished by color, each potentially influencing catalytic performance. Our workflow allows the combination of 3D analysis across several length scales from the bulk sample with defined laser pre-structuring for micro-CT at the sub-mm level, identification of regions of interest on the 10-100 μm scale in a FIB for 2D/3D surface and tomography visualization by SEM/EDX/EBSD and light microscopy followed by defined sample extraction and transfer to nano-CT and TEM.

Finally, concepts for 2D and 3D reference markers at different length scale (global markers and local markers prepared by FIB on the sample), their tracking, as well as their accuracy, will be presented, aimed towards automated marker recognition in next-generation microscopy software.

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ReMade@ARI - Advanced materials characterization for a circular economy

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ReMade@ARI (REcyclable MAterials DEvelopment at Analytical Research Infrastructures) project provides scientists in academia and industry with analytical tools that enable them to explore the properties of their recyclable materials. The project offers free access to over 50 analytical research infrastructures across Europe comprising synchrotrons, free-electron lasers, neutron sources, high magnetic field laboratories and ion or positron beam facilities. The Smart Science Cluster is composed of 18 early career scientists from which teaming partners are assigned to user support, in a one-to-one format. From the design of the experiments to the data collection and evaluation, the Junior Scientists share their expertise matching the experimental techniques offered within ReMade@ARI. Ideal for first time users of Analytical Research Infrastructures (ARIs) and an opportunity to discover complementary techniques. The project is divided in three different calls: ReMade-TNA for coordinated, easy and supported access for academic and industrial users; ReMade-SME for tailored access for SMEs, confidential, includes data analysis; ReMade-IND for joint access for industry and knowledge providers such as RTOs, service providers and academics.
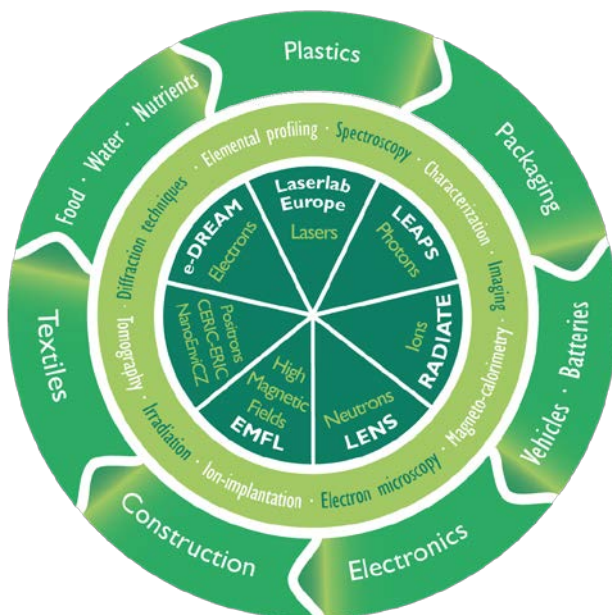
Unveiling Room-Temperature Ferromagnetism in 2D Van der Waals Material: Cr4Te5

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Two-dimensional (2D) Van der Waals (VdW) materials have garnered significant attention across diverse research avenues due to their wide-ranging properties. The recent identification of magnetism in 2D materials has further attracted material scientists, particularly for potential advancements in magnetic spin valves and VdW heterostructures. Achieving nanoscale precision in the downsizing of 2D transition metal dichalcogenides (TMDs) is crucial for their integration into technology, with a particular emphasis on reaching the ultrathin film limit.

In this work, we demonstrate the successful fabrication of epitaxial Cr_4Te_5 thin films on Al_2O_3 substrates with layer-by-layer precision control of thickness through pulsed laser deposition (PLD) using 1st harmonic Nd: YAG pulsed laser source [1], [2]. X-ray diffraction (XRD) and scanning tunneling microscopy (STM) reveal high crystalline quality as well as atomically flat surfaces. Our investigation reveals the thickness threshold beyond which room temperature ferromagnetic order persists. We have explored the electronic and magnetic structure of Cr_4Te_5, employing both experimental techniques and theoretical analyses. Our findings indicate a predominant in-plane ferromagnetic character, with spin-orbit coupling playing a significant role in promoting ferromagnetic order. Our study enables Cr_4Te_5 TMDs as a promising material for the development of room-temperature magnetic devices and sensor applications.

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Correlative Light Electron Microscopy for Polymeric Material Characterization

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Correlative Light Electron Microscopy (CLEM) is an advanced imaging approach which integrates the strengths of fluorescent microscopy (FM) and electron microscopy (EM). FM allows the labeling of the molecules of interest with fluorescent dyes to study their interaction with the surroundings and function in their native state while EM can give the structural information and morphology of the sample.[1] Therefore, their combination holds a great promise to get "the best of both worlds" and to enable the holistic analysis of materials. CLEM also holds a great potential to answer open questions in polymer science.[2] The goal of the project is to exploit the strength of FM and EM techniques and get a comprehensive 5D picture (in three spatial dimensions (x, y, z), in time, and function) of polymeric materials.

As traditional FM lacks the resolution required to resolve individual nanoparticles, to study functional information about polymeric nanomaterials (eg. ligand valency, their interaction with surroundings, etc) super-resolution microscopy (SRM) technic is used in this project which surpasses the diffraction limit and can give resolution up to 20-40 nm. For seeking morphological and structural information of polymeric materials transmission electron microscopy (TEM) would be used. Correlating the data from these two techniques will enable the structural as well as functional characterization of polymeric nanomaterials.

To establish CLEM workflow, polymer-coated quantum dots (QD) are used as model nanomaterial. QD are commonly used in many biomedical applications in biosensing, and imaging studies. QDs exhibit photoluminescence intermittency (blinking). This blinking of QD poses challenges in many applications. CLEM can give insights on the relationship of QD morphology and blinking behavior.

Further studies are planned on polymeric materials such as self-assembled nanoparticles, fibers, drug delivery systems, etc.

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# Nanofabrication of superconducting and ferromagnetic structures for operation in TEM

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Nanoscale superconducting (Nb, TiN, NbN and BSCCO) and ferromagnetic (Permalloy) structures were prepared on SiN, SiO_2 and SiC membranes for experiments in a TEM. Exfoliated e-beam transparent BSCCO flakes were fixed on 4-contact chips to study correlations between temperature, electron transport properties and behavior of Abrikosov vortices. Metals Ti and Nb or nitrides TiN and NbN were combined into 3-layer heterostructures for adjusting superconducting parameters through the proximity effect. An on-chip thermometer was used also as a heater, allowing the sample temperature to be changed in the range from 400 K to 5 K and back in a few seconds. NanoSQUIDs with nanobridge Josephson junctions (nJJs) were prepared at a distance of below 200 nm from the tip of a cantilever by using bulk nanosculpturing of the substrate with a focused ion beam. The nanoSQUIDs had a sub-micrometer loop size, which limited the dimensions of the nJJs to below ~100 nm. Electron beam lithography and high selectivity reactive ion etching with pure SF_6 gas were used to pattern nJJs with a width down to 10 nm that is comparable to the coherence length in thin films of Nb and NbN and provides better reproducibility in the case of a Nb functional layer and better long term stability due to enhanced corrosion resistance in the case of NbN layer. A naturally created undercut in the Si substrate was used to prepare nanoSQUIDs on a 10-nm-thick SiO_2 membrane within 500 nm from the edge of the substrate. High-resolution TEM revealed that NbN films on SiN have a columnar structure while they observe cube-on-cube epitaxial growth on SiC membranes. Towards future realization of hybrid superconductor-ferromagnetic nanostructures for spintronics experiments in TEM, Permalloy (Py) nanodisks and triangles with dimensions down to ~100 nm were prepared on SiN membranes and studied by Lorentz microscopy (LTEM) and electron holography TEM methods.
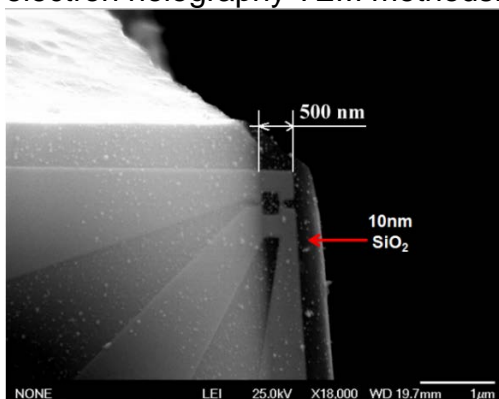
SEM image of nanoSQUID on SiO_2 membrane.
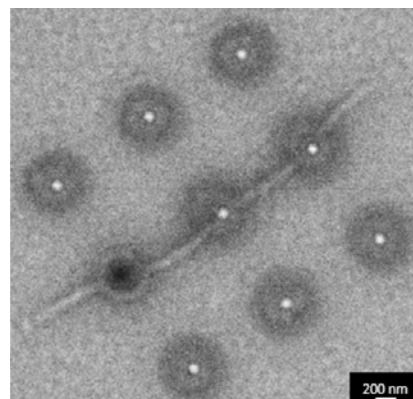
LTEM image of array of 400 nm Py disks

Towards *In-situ* X-ray Scattering studies of polynuclear cluster

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Polynuclear metal nanoclusters form various, catalytically active coordination compounds ranging from smaller metal complexes to larger structures such as metal-organic frameworks, nanosheets or nanoparticles (NPs) [1,2]. Over the years, plenty of cluster syntheses have been developed, however, common characterizations with single crystal diffraction and mass spectroscopy can only be performed *ex-situ* and demand tedious sample preparation. Thus, there is little insight into the growth mechanisms of small metal clusters [3]. To this end, we developed a strategy to adopt X-ray techniques to *in-situ* study cluster synthesis, which has already been proven very successful for NPs in the past [4,5]. In particular, total scattering combined with pair distribution function analysis and X-ray spectroscopy are the ideal techniques to characterize the geometric and electronic properties independent from long-range order. In this regard, we perform in-situ experiments on highly interesting discrete planar metal nanoclusters prepared from Fe and Mn [5,6]. The intermediate stages towards metallic monolayers have a unique symmetry with outstanding catalytic properties [1]. Gaining a detailed insight into these clusters will enable tailoring their properties, scaling with their size ranging from single atoms to full nanoparticles.

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Enhancing correlative characterizations Measurement quality with spatial sample alignment

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Manufactured micro- and nano structures in various forms became indispensable in our daily life. A prominent example might be MEMS (micro electromechanical systems). To improve these structures, it is crucial to gain more information about their surfaces.

Correlative metrological surface characterization can provide more information about manufactured micro- and nano structures. This information can be used to make predictions to new materials and structures and will reduce development time and expenses.

Within a correlative surface characterization, the sample will undergo different measurements by different measurement devices with different characterization methods. Finally, all measurement results will be correlated and interpreted.

Currently a correlative characterization is time consuming, and it is prone to errors. In a first measurement system, a region of interest (ROI) has to be defined. The sample has then to be transferred from one system to another as well as the spatial adjustments needed are, in most cases, done manually.

Apart from the difficulty to find the region of Interest in the second device again there are additional challenges associated with the process of a correlative characterization. One of these challenges is ensuring the correct spatial orientation of the sample across different measurement devices.

A misalignment of the sample's spatial orientation, for example a rotation about one coordinate axle, can lead to a reduction of the usable sensor field. Since the samples surface will not shrink, there is more surface that must be measured with a reduced sensor field. With this reduction the image quality will decrease. Samples rotation about more than one coordinate axle can even cause a distortion of the measurement areas compared to the previous measurements in previous measurement devices.

Deducing structural dynamics in responsive crystalline film systems: a multi-technique approach

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The design of responsive crystalline structures capable to react to external stimuli, such as temperature or light, while maintaining a controllable structural response, holds significant promise for advancing applications like mechanical energy storage, drug delivery or gas sequestration.[1] One key challenge, particularly in film systems, lies in accurately characterizing both the magnitude and duration of the induced structural response. These aspects are vital for tailoring structures with adjustable response kinetics that can be triggered remotely.

Using epitaxially grown metal-organic framework model film systems as a case study,[2-4] we reveal that the dynamic behaviour is intricately linked to crystallite geometry, morphology, and chemical composition. Consequently, employing a multi-technique approach becomes essential to unravel these interdependencies and infer the timescale of initiated dynamics. In this context, we showcase the efficacy of time-resolved grazing incidence X-ray scattering and infrared spectroscopic techniques in precisely quantifying the structural response triggered by light, temperature, or upon gas uptake and release (such as CO_2). Our findings pave the way for a deeper understanding of timescales and responsive behaviours in similar or more advanced soft-matter film systems, offering a new avenue for research in this field.

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Direct observation of tensile-strain-induced magnetic hardening in a ferromagnet

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Magnetoelasticity is the bond between magnetism and mechanics, but the intricate mechanisms via which magnetic states change due to mechanical strain remain poorly understood. Here, we provide direct nanoscale observations of how tensile strain modifies magnetic domains in a ferromagnetic Ni thin plate using in situ Fresnel defocus imaging, off-axis electron holography and a bimetallic deformation device [1]. We present quantitative measurements of magnetic domain wall structure and its transformations as a function of strain. We observe the formation and dissociation of strain-induced periodic 180° magnetic domain walls perpendicular to the strain axis. The magnetization transformation exhibits stress-determined directional sensitivity and is reversible and tunable through the size of the nanostructure. In this work, we provide direct evidence for expressive and deterministic magnetic hardening in ferromagnetic nanostructures, while our experimental approach allows quantifiable local measurements of strain-induced changes in the magnetic states of nanomaterials [2].

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Quantification of Interface Magnetism in $La_{0.7}Sr_{0.3}MnO_3$ Thin Film by Off-axis Electron Holography

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Material interfaces encompass discontinuity in structure and electronic band structure that leads to an enormous number of effects at the interfaces. It is particularly important in magnetic oxide thin films, which systems offer various types of manipulatable magnetic states for information storage and computation. However, a quantitative determination of the interfacial magnetic properties at high spatial resolution is challenging. Here, we use off-axis electron holography to quantitatively determine the interface magnetization, Curie temperature TC in $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) film with a subtle Mn composition change. We reveal an electrostatically-induced long-range magnetic proximity effect that extends over a distance of 40 nm at a ferromagnetic/paramagnetic interface in LSMO. [1] We report a magnetic transition region with a gradual change in the magnitude of magnetization, but no rotation, stable at all temperatures below TC. [2] We show that the behaviour results from carrier diffusion and drift across the interface, which changes the Mn^{3+}/Mn^{4+} ratio and hence the local Curie temperature and density of magnetic moments. The unravelled quantitative relationship between electrostatic properties (i.e., local hole concentration and Mn valence) and local magnetization, as well as Curie temperature, provides a fundamental understanding of the electrostatic-shaping of nanoscale magnetism.

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In-situ Characterization Techniques to the Study of Hydrogen Storage Materials

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Abstract: Finding a solution to efficiently utilize renewable energy sources is a key step in achieving complete independence from fossil fuel energy sources. Hydrogen is considered by many to be a suitable energy vector to efficiently utilize intermittent and unevenly distributed renewable energy sources. However, while the production of hydrogen from renewable energy sources is technically feasible, the storage of large quantities of hydrogen is challenging. Compared to conventional compressed and cryogenic hydrogen storage, solid-state storage of hydrogen has many advantages in terms of safety and volumetric energy density. In general, studies of the hydrogen storage properties of materials consist of their phase information, microstructures, chemical compositions, thermal/structural stability, and chemical bonding, which can be studied using *ex- situ* experimental techniques. However, the experimental results obtained from *ex-situ* measurements provide only a partial description of the phenomena studied, and some important information are lost. In this context, *in-situ* studies offer a number of unprecedented advantages. This work presents the *in-situ* instrumentation (*e.g. in-situ* synchrotron powder X-ray diffraction, *in-situ* XRD cell) and its applications for characterizing the properties of hydrogen storage materials.

Tortoise beetle-inspired sustainable films with tunable properties based on on-demand embedding and release of functional microspheres

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Inspired by tortoise beetle, a novel method for preparing sustainable films, leveraging the on-demand embedding or release behavior of the functional fillers to impart or eliminate functions to the film is reported. The microscopic shape-memory behavior of the film surface was characterized using microscopy techniques and finite-element analysis to investigate the embedding and release of single micro-/nanospheres on the film surface, which were affected by sphere diameters and programming temperatures. [1] By utilizing different functional fillers, the transformation of film functions was demonstrated, such as optical reflection and fluorescence. Additionally, the recyclability and reusability of the substrate materials were verified through the collection and recycling of released microspheres and the film. This approach offers a promising avenue for developing sustainable films with tunable functions and highlights the potential for recycling functional fillers, contributing to both environmental sustainability and material reusability.

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Stereochemical Heterogeneity Analysis of Polylactides

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Polylactide acids (PLA) are commonly used as degradable medical implants, sutures or for the preparation of drug carriers. Furthermore, PLA is an alternative to petrochemically derived polymers. PLA is commercially produced by ring-opening polymerization (ROP) of lactide. Lactide monomer has two chemical stereocenters, thus three stereoisomers (LL, DD, meso) exist. ROP of an enantiopure monomer leads to isotactic polymer chains, however defects caused by e.g. racemization may occur. The specific microstructure of the polymer significantly affects its physical properties, and thus it is essential to determine the stereochemical composition accurately.

A new HPLC method has been developed, which analyzes PLA according to its stereochemical composition. An online hyphenation with size exclusion chromatography allowed the determination of the molecular weight of the analytes by multidimensional chromatography. This analysis was complemented by preparative HPLC to correlate the findings from liquid chromatographic with homonuclear decoupled ^{1}H-NMR and MALDI-TOF-MS analysis.

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Correlative characterization of stereocomplex formation in aliphatic polyester P(PCL-*b*-PLLA) block-copolymers.

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Quantifying the intricate relationship between molecular structure and macroscopic properties in phase-separating semicrystalline block copolymers is vital for their practical applications. Poly[(ε-caprolactone)-block-(L-lactide)] (P(PCL-b-PLLA) blended with Poly(D-lactide) (PDLA) holds a particular interest due to their mechanical properties. Polymers with defined block lengths and molar masses were prepared and showed minimum block lengths for crystallization and stereocomplexation (SC). WAXS showed SC formation in high molar mass blends in the 1:1, 2:1 and 1:3 blends (PLLA:PDLA), with PLA homocrystallite (HC) formation only occurring when no SC is observed. Moreover, SC formation acts as nucleation agent for the crystallization of PCL. Crystallite sizes vary between 1-8 nm (SC), 1-20 nm (PCL) and 1-50 nm (HC). Crystallinities of 6-70 % (SC), 2-50 % (PCL) and 3-80 % (HC) were calculated. We observed a diverse crystal morphology and superstructure, characterized by well-defined Maltese-cross spherulites of PDLA/PLLA and the formation of short and long shish-kebab-like SC crystals in racemic mixtures of PDLA/PLLA. The correlative characterization of Atomic Force Microscopy and Polarized Optical Microscopy was employed to complement Transmission Electron Microscopy results, providing a comprehensive understanding of the polymer matrix's structural features and enhancing the overall opportunities for the development of versatile materials with tunable mechanical and thermal properties.

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Keywords: Structure-Property Relations, Soft Matter, Correlative Characterization, Polymer Composites

Effect of water intercalation into tungsten trioxide structure ($WO_3.xH_2O$) (x=0,1,2): Correlation between structure and photocatalytic performance

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Tungsten oxide (WO_3) is a potential photoanode material for water oxidation, however, its employment is still restricted by its limited light absorption (Eg ~ 2.75 eV). A particular interest was devoted to tuning its electronic structure and optical bandgap in order to enhance its photocatalytic performance in hydrogen production as well as in advanced oxidation process. One of the promising alternatives for the enhancement of these photocatalytic performances, is the introduction of guest molecules into the tungsten oxide structure. In this paper, the photocatalytic performances of tungsten trioxide were highly improved by the intercalation of water into its structure. The intercalated compounds of chemical formula $WO_3 \cdot xH_2O$ were prepared and characterized by TGA-DSC, PXRD, FT-IR, Raman spectroscopy, SEM-EDS and XPS. The structures of intercalated materials were refined using Rietveld method and the electronic proprieties including bandgap energy, CB and VB edges were estimated using UV–visible reflectance spectroscopy. The obtained results show that the intercalation of water molecules into tungsten oxide structure reduces the gap energy of the WO_3 (2.4 eV) to 2.16 eV for $WO_3 \cdot H_2O$ and to 2.31 eV for $WO_3 \cdot 2H_2O$. The photocatalytic efficiency of the obtained materials was studied through oxygen evolution reaction tests in the presence of an electron acceptor (Ag^+) and the oxidative photodegradation of the orange G dye pollutant. We found that intercalated samples exhibit the highest efficiencies compared with anhydrous samples. These findings are correlated with the increase in the average W-O-W tilt angle in $WO_3 \cdot 2H_2O$ and $WO_3 \cdot H_2O$ compared to that of WO_3. This improvement in the tilt angle induces the shrinkage of the energy bandgap and increases the photocatalytic performance. Furthermore, the stability of the investigated materials was studied as a function of pH, the results reveal that the intercalation of water molecules is beneficial for the chemical stability of the hydrated tungsten oxides. These findings shed light on new alternatives for band-gap engineering and improvement of materials stability through intercalation with water molecules.

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XRD steel enhanced-characterization through computational methods

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Third-generation steels rely on the precise distribution of different crystallographic phases to provide higher tensile strength and ductility than previous steels[1,2]. However, the relationship between the crystalline meso and microstructure and the emergent superior mechanical properties is not yet completely understood. To better understand this relationship, we are working on an x-ray diffraction forward model[3] to assist on different computational approaches.

On one hand, we aim to generate synthetic powder diffraction patterns with a labeled phase composition to use supervised learning to identify such phases in real data. Traditional 1D-averaging methods such as Rietveld-refinement often fail to distinguish crystalline phases with similar lattice parameters, and we expect Neural Networks (NNs) to be able to estimate these differences successfully. The use of NNs for scientific applications originates from the medical imaging field but has already been applied in the field of metallurgy using electron[4] and optical microscopy[5] data. The end goal is to directly predict phase distribution with single 2D powder diffraction patterns whithout the need of refinement techniques.

On the other hand, we want to use the forward model to simulate raster-beam powder diffraction datasets to retrieve voxelized diffraction patterns within a sample[6]. The forward model is necessary here to test different algorithms with known synthetic samples and verify the accuracy of the method. The end goal is to obtain ~15um³ spatially resolved 3D phase maps using only x-ray diffraction data.

Our long term goal is to develop new user pipelines for synchrotron techniques applied to material science, and we believe this project can have a significant impact on advanced materials in general and the metallurgical industry in particular.

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Correlative *in situ* synchrotron radiation-based nano CT and (S)TEM imaging of biodegradable Mg-based alloys

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Magnesium-based alloys have emerged as promising candidates for implant materials due to their notable attributes of biocompatibility and biodegradability. [1] To comprehensively understand how the material microstructure influences the degradation process, a multiscale approach is necessary. *In situ* synchrotron radiation nano computed tomography (SRnanoCT) has proven to be an invaluable non-destructive 3D imaging technique capable of investigating dynamic degradation phenomena. Complementary insights into the chemical compositions and microstructure down to the atomic scale are facilitated by (scanning) transmission electron microscopy ((S)TEM). However, the correlation of these techniques presents its own set of challenges, such as the identification of regions of interest (ROIs) (e.g., precipitates), sample transfer, registration of images and the correlative preparation.

With my studies, I have successfully established a correlative workflow for unravelling the biodegradation mechanisms of Mg-based alloys across various length scales. Wire samples (80 µm diameter) of Mg-2wt.%Ag and Mg-2wt.%Gd were subjected to degradation in simulated physiological conditions within a flow-cell setup for *in situ* SRnanoCT. The investigation focused on degradation rates, the homogeneity of degradation layer formation, and influence of insoluble precipitates. The resulting tomographic scans acquired at intervals of 13-30 minutes, allowed for the selection of degradation-relevant ROIs for further in-depth characterization. Subsequent steps involved the transfer of samples into a dual-beam FIB FEI Helios NanoLab 400S, where a lamella at the identified location was prepared using a lift-out method. For correlative STEM imaging and EDX analysis of the microstructure, structural defects and the degradation layer, a probe-corrected FEI Titan G2 80-200kV was employed. [2]

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Advanced transmission electron microscopy of the three-dimensional magnetization distribution in a domain wall-pinning Sm-Co-based permanent magnet

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$Sm(CoFeCuZr)_7$ permanent magnets have a high energy product and are commonly used in applications at elevated temperature. Their high coercivity results in part from the pinning of magnetic domain walls at phase boundaries, which are the result of phase decomposition on the nanoscale. Here, we use the advanced transmission electron microscopy technique of electron holographic tomography, in combination with model-based reconstruction, to measure the three-dimensional magnetization distribution of a pinned domain wall in a needle-shaped sample. The results are discussed by considering the shape of the sample, as well as local variations of its microstructure, chemical composition and magnetocrystalline anisotropy.
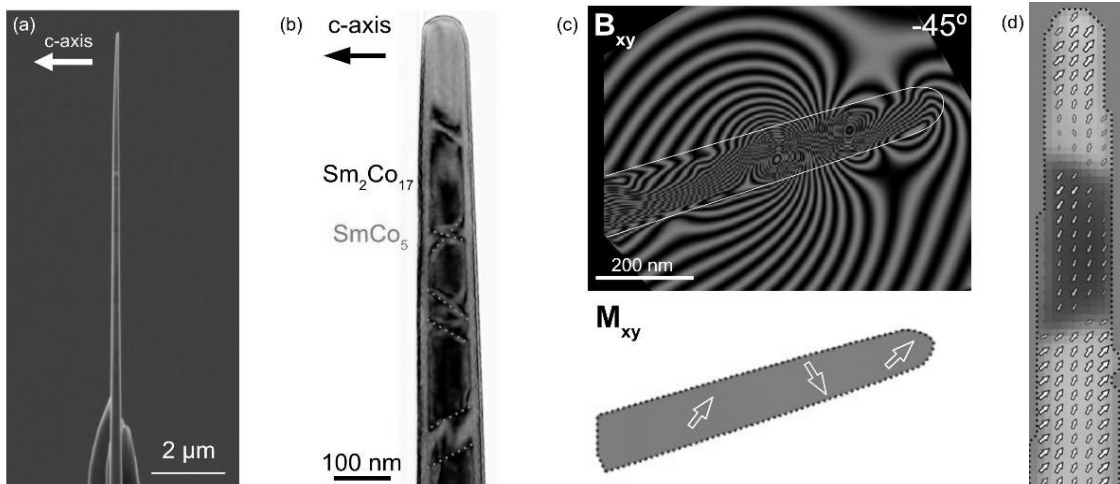
(a) SEM image of the needle and its base. (b) TEM image showing boundaries of different Sm-Co crystal phases. (c) Magnetic contour and diagram showing magnetic domain directions when needle is imaged at -45 degree tilt. (d) Slice of the tomographic reconstruction showing the direction of magnetic moments.

Implementation of Correlated Characterization in Surface Metrology

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Correlated Characterization CC improves information quality in scientific research with higher-level insights into sample properties. While the basic idea is simple, its actual implementation is puzzling. Compared to subsequent measurements, CC needs references to address a Region of Interest RoI on a sample. Size, quality and distance of markers or auxiliary structures determines the ability to address a RoI, regardless of the sample orientation on a carrier inside a device.

Key issues to implement CC include mechanical interfaces, software and data interchangeability, finally information processing and management. All this influences the capability to re-measure one spot exactly, given by its coordinates.

To integrate CC into an Electronic Lab Notebook ELN we even defined a common wording for process description by MDMC [1] to support FAIR data management.

For sample interchange, we use a common mechanical interface with standardized, 3D printed carriers. They include three rotational degrees of freedom: theta rotation, tip and tilt. Improved reference marker design supports easy yet accurate detection. Finally, we transformed available process data into a common data format.

We categorize the metadata from individual processes into three groups: first the identification, with information about task, origin, customer and similar, second the references, needed for CC as it contains everything to readdress RoIs based on references. Finally, settings including all parameters needed to perform a process.

One goal of the ELN integration of devices for CC is to sort important metadata by routines to make the connected process FAIR and easier to apply. We are not yet there, but currently we analyze process structures for commonalities to create a transfer script for metadata for multiple devices with just minor adjustments.

The first CC protocol will still include tasks where users have to add data manually, mainly for identification. A later and more elaborated version, however, will make monotonous data input obsolete and help the operators focus on their jobs.

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Following Organic Crystal Growth Dynamics During Liquid Reagent Mixing using Liquid Cell Transmission Electron Microscopy

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Liquid cell transmission electron microscopy (LCTEM) offers the opportunity to visualise structural, compositional and chemical changes on the nanoscale in real time. However, the study of phenomena in liquids during mixing of two precursor solutions has proven challenging, requiring sophisticated liquid cell designs. Here, we demonstrate that the sequential introduction and withdrawal of solvents makes it possible to maintain optimal imaging conditions while mixing liquids in a commercial liquid cell. We have succeeded in visualising a fast nanoscale crystallisation mechanism, in which methanol was allowed to interact with an organic crystal (R-BINOL-CN) dissolved in chloroform. Scanning TEM (STEM) images were recorded in real time during interaction of the two solvents, revealing the formation of chain-like structures of R-BINOL-CN, which would otherwise have coalesced to form large single particles in the absence of methanol. Our approach of mixing liquids and using STEM techniques to record images with precise electron dose control establishes a powerful new platform for LCTEM studies of electron-beam-sensitive materials such as drug molecules, polymers and molecular amphiphiles during reagent mixing.

Correlative µCT and histological imaging of bone implants

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Multimodal imaging is critical in attaining complementary information on complex materials systems, such as biological tissues and functional materials. If performed in a correlative manner, few samples can be sufficient to provide a comprehensive understanding of underlying mechanisms and interactions. For example, correlative characterization of biodegradable bone implants using synchrotron radiation-based micro computed tomography (SRµCT) and histology provides insight into sample morphologies, including degradation behavior and bone growth in 3D, as well as biological information in terms of cell behavior in 2D [1]. However, the sample preparation for correlative imaging, as well as the image registration later on to extract the desired information from both modalities can present a bottleneck.

We have performed a study in which magnesium-based, titanium and polymer implants have been implanted into rat tibia for healing times of 4, 8 and 12 weeks. After sacrifice of the animals, SRµCT imaging was undertaken at the P05 beamline at the PETRA III storage ring at DESY, Hamburg, followed by slicing and staining of the sample for histology. Quantities of interest, specifically degradation rate and relative bone volume, were determined [1]. We have developed an automated registration tool to determine the image plane in the 3D SRµCT that corresponds to the 2D histology and correlate findings from both modalities. Finally, we are currently using generative adversarial networks, specifically CycleGANs, to generate a 3D virtual histology by artificially staining the SRµCT data based on the registered histologies. The preliminary results obtained for two different histological stainings (H&E and toluidine blue) are promising. In the future, 3D virtual histology may be used to drastically reduce the experimental effort and destructive nature of histological imaging.

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