PICO 2021

Sixth Conference on Frontiers of Aberration Corrected Electron Microscopy

Virtual Conference

2 – 6 May 2021

PROGRAMME PICO 2021, VIRTUAL CONFERENCE, 2 – 6 MAY 2021

Time	Sunday 2 May 2021	Time	Monday 3 May 2021	Time	Tuesday 4 May 2021	Time	Wednesday 5 May 2021	Time	Thursday 6 May 2021
		0800-0900	BREAKFAST	0800-0900	BREAKFAST	0800-0900	BREAKFAST	0800-0900	BREAKFAST
		0900-0935	Xiaodong Han	0900-0935	Ryo Ishikawa	0900-0935	Joanne Etheridge	0900-0935	Holger Stark
		0935-1010	Sang Ho Oh	0935-1010	Miyoung Kim	0935-1010	Philipp Wachsmuth	0935-1010	Wim Hagen
		1010-1045	Qing Chen	1010-1045	Andrew Maiden	1010-1045	Christoph Koch	1010-1045	Lothar Houben
		1045-1115	TEA BREAK	1045-1115	TEA BREAK	1045-1115	TEA BREAK	1045-1115	TEA BREAK
		1115-1150	Eva Olsson	1115-1150	Thomas Walther	1115-1150	Christian Maunders	1115-1150	Bram Koster
		1150-1225	Layla Mehdi	1150-1225	Fabien Massabuau	1150-1225	Ute Kaiser	1150-1225	Friedrich Förster
		1225-1300	Mitra Taheri	1225-1300	Gianluigi Botton	1225-1300	Paolo Longo	1225-1300	Ariane Briegel
								1300-1310	Closing Remarks
		1300-1430	LUNCH BREAK	1300-1430	LUNCH BREAK	1300-1430	LUNCH BREAK	1310-1430	
		1430-1505	Henry Chapman	1430-1505	Randi Holmestad	1430-1505	Kai Grünewald	1430	
1515-1600	WELCOME RECEPTION	1505-1540	Toma Susi	1505-1540	Rachel Oliver	1505-1540	Jürgen Plitzko		
1600-1610	Opening Remarks	1540-1615	Ido Kaminer	1540-1615	Marija Drndić	1540-1615	Ohad Medalia		
1610-1645	Wolfgang Baumeister	1615-1645	TEA BREAK	1615-1645	TEA BREAK	1615-1645	TEA BREAK		
1645-1720	Colin Humphreys	1645-1720	Elizabeth Dickey	1645-1720	Hamish Fraser	1645-1720	Wah Chiu		
1720-1755	Max. Haider	1720-1755	Demie Kepaptsoglou	1720-1755	Robert Sinclair	1720-1755	Peter Rez		
1755-1830	John Spence	1755-1830	Jian-Min Zuo	1755-1830	David Smith	1755-1830	DINNER BREAK		
1830-2000	DINNER BREAK	1830-2000	DINNER BREAK	1830-2000	DINNER BREAK	1830-1915	WINE RECEPTION		
						1915-2000			
2000-2200	Poster Session A	2000-2200	Poster Session B	2000-2200	Poster Session C	2000-2230			

PICO 2021

SIXTH CONFERENCE ON
FRONTIERS OF ABERRATION
CORRECTED ELECTRON MICROSCOPY

and

Colloquia Honouring
Wolfgang Baumeister, Colin Humphreys,
John Spence, and Knut Urban

Virtual Conference
2 – 6 May 2021

Conference Organisers
Rafal Dunin-Borkowski (Jülich)
Joachim Mayer (Aachen)
Carsten Sachse (Jülich)
Karsten Tillmann (Jülich)

DELEGATE INFORMATION
PROGRAMME
POSTER PROGRAMME
ABSTRACTS

The organisers kindly acknowledge support of PICO 2021 by the following enterprises, establishments and organisations:

The cover image displays a high-resolution micrograph of domains and a domain wall in $BiFeO_3$ recorded along the [110] zone axis of the perovskite unit cell under NCSI conditions by Lei Jin. The domain wall area is distinguished by the upward (in the top domain) and downward (in the bottom domain) off-centre displacements of the O- and Fe-columns with respect to the Bi-columns.

PICO 2021

SIXTH CONFERENCE ON FRONTIERS OF ABERRATION CORRECTED ELECTRON MICROSCOPY

Preface

PICO 2021 – the Sixth Conference on Frontiers of Aberration Corrected Electron Microscopy including colloquia honouring the achievements of Colin Humphreys and Knut Urban as well as Wolfgang Baumeister and John Spence on the occasion of their 80th and 75th birthdays in 2021, respectively – will be held as a virtual meeting from 2 to 6 May 2021. The event is hosted by the Ernst Ruska-Centrum für Mikroskopie und Spektroskopie mit Elektronen in Aachen and Jülich and supported by Thermo Fisher Scientific Inc, JEOL (Germany) GmbH, Ametek GmbH Business Unit Gatan, Attolight AG, CEOS GmbH, and Forschungszentrum Jülich GmbH.

The conference has attracted 181 participants from 23 countries plus 71 staff of RWTH Aachen University and Forschungszentrum Jülich GmbH. Organisers have put together a programme including 45 scientific keynote lectures with further 69 contributions being scheduled for poster presentations.

The organisers trust that you will have an enjoyable time and that you will find the meeting stimulating and enthusing.

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DELEGATE INFORMATION

PICO 2021 organisers are delighted that you have decided to attend the Sixth Conference on Frontiers of Aberration Corrected Electron Microscopy to be held merely as a virtual event this year in view of the continued uncertainty surrounding the present pandemic situation. The organisers look forward to meeting you "virtually" and do hope that you will enjoy your participation in PICO 2021. This delegate information is intended to provide information regarding the

overall programme as well as boundary conditions to comply with concerning the preparation and transfer of presentations.

Please read these pages prior to getting ready to the event as they include all of the information you will be in need of for the preparation of your presentations and for participation in PICO 2021. If you require further information, please contact any of the organisers, the conference secretary, or staff at local finance department and at "nice:-) Agentur für Kommunikation GmbH" concerning issues related to invoicing and to the operation of the "virtual" conference area, respectively.

CONTACT

Conference Organisers (programme)	Rafal Dunin-Borkowski e-mail: r.dunin-borkowski@fz-juelich.de phone: +49 2461 61-9297 Joachim Mayer e-mail: mayer@gfe.rwth-aachen.de phone: +49 241 80-24345 Carsten Sachse e-mail: c.sachse@fz-juelich.de phone: +49 241 80-2030 Karsten Tillmann (until 16 April 2021) e-mail: k.tillmann@fz-juelich.de phone: +49 241 80-1438
Conference Secretary (admin. issues)	Gabriele Mertzbach e-mail: g.mertzbach@fz-juelich.de phone: +49 2461 61-4073
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Virtual Venue (technical issues)	Romain Deltour and Christina Schultz nice:-) Agentur für Kommunikation GmbH e-mail: pico2021@team-nice.de phone: +49 2054 87559-0

PAYMENT OF YOUR REGISTRATION FEE

Registered delegates will receive an e-mail invoice by the Jülich Research Centre's finance department (sender: o.sap.fl@fz-juelich.de or alike; subject: PICO 2021 - multi digit number) in between 6 and 16 April 2021. This e-mail will include a link to a personalised webpage to be used to initiate the payment of the conference fee by credit card.

Please bear in mind that you need to initiate electronic payment of the conference fee by Friday 23 April 2021 at the latest as otherwise access to PICO 2021 will not be granted.

PREPARATION OF ORAL AND POSTER PRESENTATIONS

Both, lecturers and presenters of contributed posters will receive instructions on the preparation and upload of their documents by Romain Deltour or Christina Schulz from "nice:-) Agentur für Kommunikation GmbH" by 16 April 2021 the latest. Please check your spam folder in case you should not have received anything by then. You can reach Romain Deltour and Christina Schulz by e-mail (pico2021@team-nice.de) or phone (+49 2054 87559-0) in case of technical queries. Technical requirements for the preparation of oral and poster presentations to comply with are given below.

REQUIREMENTS FOR ORAL PRESENTATIONS

You will receive an email by Romain Deltour or Christina Schulz from "nice:-) Agentur für Kommunikation GmbH" in between 29 March and 11 April 2021 giving you all the information you will be in need of to prepare your presentation. Please check your spam folder in case you should not have received anything by then. All slides will be requested in 16:9 widescreen aspect ratio and you will be given the opportunity for a test run prior to the conference. Romain Deltour and Christina Schulz will assist you if you are in need of any technical support. You can reach them by e-mail (pico2021@team-nice.de) or phone (+49 2054 87559-0).

REQUIREMENTS FOR POSTER PRESENTATIONS

Previously declared and uploaded posters will be discussed on the first to third evenings of the conference. All posters prefixed with number PA will be discussed on Sunday 2 May 2021 from 2000 to 2200 hrs while posters prefixed PB and PC will be presented on Monday 3 May 2021 and Tuesday 4 May 2021, respectively, at the same time. Presenters of posters are asked to make sure that they are available in the in front of their poster in the virtual poster presentation room during scheduled poster sessions.

You will receive an e-mail by Romain Deltour or Christina Schulz from "nice:-) Agentur für Kommunikation GmbH" in between 29 March and 11 April 2021 giving you all the information you will be in need of to prepare your poster. Please check your spam folder in case you should not have received anything by then. All posters will be requested in 16:9 widescreen aspect ratio. Romain Deltour and Christina Schulz will assist you if you are in need of any technical support. You can reach them by e-mail (pico2021@team-nice.de) or phone (+49 2054 87559-0).

COMPANY PRESENTATIONS

A number of oral and poster presentation by leading electron microscopy equipment manufacturers will be given in the regular PICO 2021 oral and poster programme. Delegates are encouraged to take the opportunity to meet with and talk to company representatives during tea, lunch and dinner breaks. You can either do this by having an open discussion or using the private discussion areas of the virtual venue system (see tutorial: https://www.loom.com/share/7f46b049c438454792c325640788fdf3).

LOGGING ON TO THE PICO 2021 VIRTUAL CONFERENCE SYSTEM

Please make sure to have installed and use one of the following web browsers to join PICO 2021 virtually: Mozilla Firefox, Google Chrome, or Opera.


Mozilla Firefox


Google Chrome

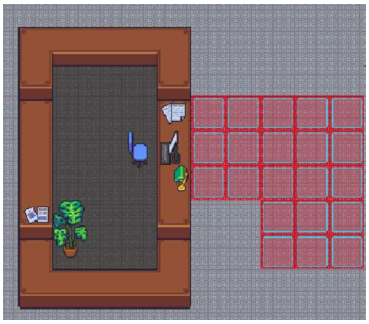

Opera

All registered participants will receive an e-mail by Romain Deltour or Christina Schulz from "nice:-) Agentur für Kommunikation GmbH" shortly before the event giving details on access to the conference platform. Please check your spam folder and contact Romain Deltour or Christina Schulz if you should not have received further information by the end of April 2021. You can reach them by e-mail (pico2021@team-nice.de) or phone (+49 2054 87559-0).

TECHNICAL BRIEFING ON THE OPERATION OF THE AUDIO-VISUAL PRESENTATION SYSTEM AND ANSWERS TO QUERIES BY PARTICIPANTS

A basic overview on the capabilities and operations of the PICO 2021 virtual venue conference system (classical and video tutorials as well as a test run site) is available at https://erc.org/pico2021/virtualvenue.htm .

Participants giving oral presentations will additionally be contacted by "nice:-) Agentur für Kommunikation GmbH" giving them the opportunity to receive an in-depth briefing prior to the event.



Furthermore, a virtual reception desk can be found inside the platform, where staff from "nice:-) Agentur für Kommunikation GmbH" can be directly contacted. Staff will be available throughout the event to assist with technical issues.

CONFERENCE PROCEEDINGS

Manuscripts of the better part of oral presentations given during the conference will be published in the PICO 2021 Special Issue of Ultramicroscopy to be released in summer 2021. Or-

ganisers will not publish any further proceedings nor make electronic versions of the presentations available for download on web pages.

TEA, LUNCH AND DINNER BREAKS

The PICO 2021 conference programme allows for a total of four tea, lunch and dinner breaks in between 30 and 90 minutes and per day. Delegates are encouraged to make use of these breaks for talking to other participants in the various virtual discussion areas.

WINE RECEPTIONS

The conference programme includes a wine reception to be held on Sunday 2 May 2021 from 1515 to 1600 hrs before the opening of the lecture programme and another one on Wednesday 5 May 2021 from 1830 to 2000 hrs. Delegates are invited to attend the former to welcome other delegates and the letter to listen to tributes to the PICO 2021 guest of honour. Delegates are encouraged to help themselves with a drink and to enjoy the programme.

SMOKING AND DRINKING POLICY

Smoking or other use of tobacco products (including, but not limited to, cigarettes, pipes, cigars, snuff, or chewing tobacco) as well as moderate drinking is allowed in any part of the virtual PICO 2021 conference venue. Delegates may smoke and drink during presentations as well as during breaks but are commended to comply with their employer's respective policies.

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PROGRAMME

PROGRAMME

SUNDAY 2 MAY 2021

1515-1600		VIRTUAL WELCOME RECEPTION
SESSION A		**INTRODUCTION**
1600-1610		Opening Remarks by the Organisers
1610-1645	**A1**	Structural biology *in situ*: the promise and challenges of cryo-electron tomography Wolfgang Baumeister, Max-Planck-Institute of Biochemistry – Martinsried (Germany)
1645-1720	**A2**	From electron microscopy to gallium nitride to graphene electronic devices Colin Humphreys, Queen Mary University of London (UK)
1720-1755	**A3**	Instrumentation for advanced electron microscopy techniques Max. Haider, CEOS GmbH – Heidelberg (Germany)
1755-1830	**A4**	Inversion of dynamical scattering from Bragg intensities John Spence, Arizona State University – Tempe (US)
1830-2000		DINNER BREAK
2000-2200	**PA**	**POSTER SESSION A**

MONDAY 3 MAY 2021

SESSION B — **DYNAMIC PHENOMENA AND IN SITU TECHNIQUES**

0900-0935	**B1**	Timely and atomic-resolved high-temperature mechanical testing system Xiaodong Han, Beijing University of Technology (PRC)
0935-1010	**B2**	Stabilisation mechanisms of polar oxide surfaces studied by in-situ electron microscopy at high temperatures Sang-Ho Oh, Sungkyunkwan University - Suwon (Korea)
1010-1045	**B3**	In situ liquid cell TEM study on the wet chemical etching of semiconducting nanowires and nanobelts Quing Chen, Peking University - Beijing (PRC)
1045-1115		TEA BREAK
1115-1150	**B4**	High resolution in situ electron microscopy studies to correlate atomic structure to strain induced effects Eva Olsson, Chalmers University of Technology - Gothenburg (Kingdom of Sweden)
1150-1225	**B5**	Understanding nanoscale processes in Li-ion batteries by operando scanning transmission electron micrscopy Layla Mehdi, University of Liverpool (UK)
1225-1300	**B6**	Intelligent microscopy: a path toward tailored materials at the atomic scale Mitra Taheri, Johns Hopkins University - Baltimore (US)
1300-1430		LUNCH BREAK

SESSION C — **NOVEL TECHNIQUES AND APPROACHES**

1430-1505	**C1**	Convergent beam diffraction at X-ray free-electron laser Henry Chapman, Deutsches Elektronen Synchrotron - Hamburg (Germany)
1505-1540	**C2**	Electron-beam manipulation of lattice impurities Toma Susi, University of Vienna (Austria)

1540-1615	**C3**	Extreme light-matter interactions in the ultrafast transmission electron microscope Ido Kaminer, Technion - Israel Institute of Technology - Haifa (Israel)
1615-1645		TEA BREAK
1645-1720	**C4**	Structural disorder characterised by aberration corrected scanning transmission electron microscopy: real-space 2-D partial pair correlation functions Elizabeth Dickey, Carnegie Mellon University - Pittsburgh (US)
1720-1755	**C5**	Nanoscale functional chemistry and opto-electronic response of organic crystals Demie Kepaptsoglou, Sci-Tech Daresbury (UK)
1755-1830	**C6**	Crystallographic information imaging (CII), how fast detectors and modern computer algorithms have created a new form of electron microscope Jian-Min Zuo, University of Illinois at Urbana-Champaign (US)
1830-2000		DINNER BREAK
2000-2200	**PB**	**POSTER SESSION B**

TUESDAY 4 MAY 2021

SESSION C		**NOVEL TECHNIQUES AND APPROACHES (CONTINUED)**
0900-0935	**C7**	Three-dimensional imaging by depth sectioning Ryo Ishikawa, University of Tokyo (Japan)
0935-1010	**C8**	Dimensionality reduction and unsupervised clustering for electron energy-loss spectroscopy-spectrum images Miyoung Kim, Seoul National University (Korea)
1010-1045	**C9**	Multislice ptychography with structured illumination Andrew Maiden, University of Sheffield (UK)
1045-1115		TEA BREAK

1115-1150	**C10**	Self-calibrating method for effective absorption correction of elements with multiple X-ray lines in transmission electron microscopy Thomas Walther, University of Sheffield (UK)
1150-1225	**C11**	Sequential plan-view TEM imaging of porous GaN Fabien Massabuau, University of Strathclyde (UK)
1225-1300	**C12**	Detecting plasmons' interactions and strain in nanostructures Gianluigi Botton, McMaster University - Hamilton (Canada)
1300-1430		LUNCH BREAK
SESSION D		**GROWTH MECHANISMS, INTERFACE PHENOMENA AND LATTIE IMPERFECTIONS IN SOLIDS**
1430-1505	**D1**	Clustering and precipitate nucleation in al alloys studied by SPED and HAADF-STEM Randi Holmestad, Norwegian University of Science and Technology - Trondheim (Kingdom of Norway)
1505-1540	**D2**	Defect structure and properties in cubic nitrides Rachel Oliver, University of Cambridge (UK)
1540-1615	**D3**	Atomic-scale devices made by electrons Marija Drndić, University of Pennsylvania - Philadelphia (US)
1615-1645		TEA BREAK
1645-1720	**D4**	Uncovering solid-state nucleation mechanisms in titanium alloys using materials characterisation at the nano scale Hamish Fraser, The Ohio State University - Columbus (US)
1720-1755	**D5**	Correlative STEM-EELS and Raman spectroscopy studies to optimise gold nanoparticles for early cancer detection Robert Sinclair, Stanford University (US)
1755-1830	**D6**	Atomic-resolution structure imaging of elemental and compound semiconductors: defects, interfaces and intermixing David Smith, Arizona State University - Tempe (US)
1830-2000		DINNER BREAK
2000-2200	**PC**	**POSTER SESSION C**

WEDNESDAY 5 MAY 2021

SESSION E — **ADVANCED METHODS AND INSTRUMENTATION**

0900-0935	**E1**	Dynamical scattering in picometre steps and other adventures in 4D-STEM Joanne Etheridge, Monash University - Victoria (Australia)
0935-1010	**E2**	Development and application of laser illumination system integrated into TEM Philipp Wachsmuth, JEOL (Germany) GmbH - Freising (Germany)
1010-1045	**E3**	Exploring 2D materials with high-resolution, high speed, high efficiency, and low dose Christoph Koch, Humboldt-Universität zu Berlin (Germany)
1045-1115		TEA BREAK
1115-1150	**E4**	Spectra-Ultra, the latest developments Christian Maunders, Thermo Fisher Scientific - Eindhoven (The Netherlands)
1150-1225	**E5**	Unravel low-dimensional material's contrast and properties Ute Kaiser, University of Ulm (Germany)
1225-1300	**E6**	Recent advances for 4D STEM and EELS data acquisition: Stela and K3 Paolo Longo, Gatan Inc. – Pleasanton (US)
1300-1430		LUNCH BREAK

SESSION F — **CRYO MICROSCOPY METHODS AND APPLICATIONS IN BIOSTRUCTURE RESEARCH**

1430-1505	**F1**	Structural nanotopology and "molecular gymnastics" of herpesvirus glycoproteins Kay Grünewald, Universität Hamburg (Germany)
1505-1540	**F2**	Tools and technologies for visual proteomics with cryo-ET – a short retrospective of 20 years with Wolfgang Baumeister Jürgen Plitzko, Max Planck Institute for Biochemistry - Martinsried (Germany)

1540-1615	**F3**	Lamin filaments, mutations and the nuclear lamina Ohad Medalia, University of Zürich (Switzerland)
1615-1645		TEA BREAK
1645-1720	**F4**	Electron cryo-microscopy of biomolecules at atomic resolution Wah Chiu, Stanford University (US)
1720-1755	**F5**	Coherent and incoherent imaging of biological specimens with electrons and X-rays Peter Rez, Arizona State University - Tempe (US)
1755-1830		DINNER BREAK
1830-2000		**VIRTUAL WINE RECEPTION in honour of Wolfgang Baumeister, Colin Humphreys, John Spence, and Knut Urban**

THURSDAY 6 MAY 2021

SESSION F		**CRYO MICROSCOPY METHODS AND APPLICATIONS IN BIOSTRUCTURE RESEARCH (CONTINUED)**
0900-0935	**F6**	Atomic resolution structure determination by cryo-EM - where are the limits? Holger Stark, Max-Planck-Institute for Biophysical Chemistry - Göttingen (Germany)
0935-1010	**F7**	Efficient sample screening and automation-setup in cryo-EM Wim Hagen, EMBL Heidelberg (Germany)
1010-1045	**F8**	Scanning transmission electron microscopy for cryo-preserved samples in life sciences and materials sciences Lothar Houben, Weizmann Institute of Science - Rehovot (Israel)
1045-1115		TEA BREAK
1115-1150	**F9**	Zooming in on cells and molecules with correlative light and electron tomography Bram Koster, Leiden University (The Netherlands)

1150-1225	**F10**	Structural insights into protein biogenesis at the endoplasmic reticulum Friedrich Förster, Universiteit Utrecht (The Netherlands)
1225-1300	**F11**	Exploring the structure and function of microbial motility: chemotaxis and intermicrobial transport Ariane Briegel, Leiden University (The Netherlands)
1300-1310		Closing Remarks by the Organisers
1310-1430		LUNCH BREAK / END OF CONFERENCE

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POSTER PROGRAMME

POSTER PROGRAMME[1]

SUNDAY 2 MAY 2021

POSTER SESSION A

PA01 Atomic structure and Fe-segeragtion in Σ13 [0001] titanium tilt grain boundaries
V Devulapalli, Max-Planck-Institute for Iron Research in Düsseldorf (Germany)

PA02 Interferometric STEM for phase imaging and interaction-free measurements
Amy Turner, University of Oregon in Eugene (USA)

PA03 Defect dynamics and self-healing in lead-free double perovskite nanocrystals
Sasha Khalfin, Technion – Israel Institute of Technology in Haifa (Israel)

PA04 In-situ atomic-resolution TEM study of phase-transition and energy-storage pathway in antiferroelectric $PbZrO_3$
Xian-Kui Wei, Forschungszentrum Jülich GmbH (Germany)

PA05 PtCu octahedral electrocatalyst for proton exchange membrane fuel cell: structure-performance-durability study
Melina Zysler, Bar-Ilan University in Ramat Gan (Israel)

[1] Posters will be presented on Sunday 2 May 2021 (poster session A), Monday 3 May 2021 (poster session B), and Tuesday 4 May 2021 (poster session C) at 2000-2200 following the dinner break.

PA06 Single-shot automatic recovery of dislocation 3D structures in TEM and quantification of the reconstruction
Gulnaz Ganeeva, EPF Lausanne (Switzerland)

PA07 Three-dimensional characterization of nanoparticles with simultaneously acquired bright-field, dark-field and secondary electron signals in STEM
Jaeyeon Jo, Seoul National University (Korea)

PA08 Introducing physical modelling In NMF decompostion of EDXS spectrum images
Adrien Teurtrie, EPF Lausanne (Switzerland)

PA09 Atomic structure and electron magnetic circular dichroism of individual rock salt structure antiphase boundaries in spinel ferrites
Zhuo Li, Tsinghua University in Beijing (China) and City University of Hong Kong (Hong Kong) and City University of Hong Kong (China) and RWTH Aachen University (Germany)

PA10 Schlieren Imaging of magnetic fields with hollow-cone electron beams
Ken Harada, RIKEN (Japan)

PA11 Measuring the mean inner potential of bernal graphite using off-axis electron holography
Avi Auslender
Tel Aviv University (Israel)

PA12 About the atomic structure of different phases within one grain boundary in pure Cu
L Frommeyer, Max-Planck-Institut für Eisenforschung GmbH in Düsseldorf (Germany)

PA13 HAADF STEM simulation: a key to solve a metal compound structure
R E Schäublin, ETH Zürich (Switzerland)

PA14 Multiple polarization orders in individual twinned hafnia colloidal nanocrystals
Hongchu Du, Forschungszentrum Jülich GmbH (Germany)

PA15 Atomically resolved tomographic reconstruction of nanoparticles from single projection: influence of amorphous carbon support
Chiranjit Roy, Indian Institute of Technology in Madras (India)

PA16 Resolution and contrast in a liquid cell scanning electron microscope platform
Aram Yoon, Fritz-Haber-Institut der Max-Planck Gesellschaft in Berlin (Germany)

PA17 Direct electron imaging of dislocation dynamics at nanoscale
Haw-Wen Hsiao, University of Illinois at Urbana-Champaign (USA)

PA18 Revealing billion-year-old secrets of a metallic meteorite
András Kovács, Forschungszentrum Jülich GmbH (Germany)

PA19 Quantitative application of moire sampling in scanning transmission electron microscopy
Alexandre Pofelski, McMaster University in Hamilton (Canada)

PA20 Presentation off-axis electron holography of Néel-type magnetic skyrmions in multilayer samples
Thibault Denneulin, Forschungszentrum Jülich GmbH (Germany)

PA21 A retrofitable laser-free ultrafast pulser for time resolved electron microscopy
Chunguang Jing, Euclid Techlabs in Bolingbrook (USA)

PA22 Quantitative analysis of magnetic states in an artificial spin ice by off-axis electron holograph
Teresa Weßels, Forschungszentrum Jülich GmbH (Germany)

PA23 Shaping electron wave functions with quantum light
Valerio Di Giulio, The Barcelona Institute of Science and Technology (Spain)

MONDAY 3 MAY 2021

POSTER SESSION B

PB01 How much can inelastically scattered electrons contribute to electron cryotomography of biological specimens?
Joshua L Dickerson, MRC Laboratory of Molecular Biology in Cambridge (UK)

PB02 Macromolecular organization of membrane-associated Atg18 oligomers
Daniel Mann, Forschungszentrum Jülich GmbH (Germany)

PB03 PspA adopts an ESCRT-III-like fold and is capable of remodeling bacterial membranes by fusion and fission
Benedikt Junglas, Forschungszentrum Jülich GmbH (Germany) and Johannes Gutenberg University Mainz (Germany)

PB04 Capture of co-translational multi-domain protein folding with cryo-EM
Nuno Bustorff, Forschungszentrum Jülich GmbH (Germany)

PB05 Presentation title to be confirmed
Raimond Ravelli, University of Maastricht (The Netherlands)

PB06 A millikelvin scanning tunneling microscope in ultra-high vacuum with adiabatic demagnetization refrigeration
Taner Esat, Forschungszentrum Jülich GmbH (Germany)

PB07 Early results from the X-ray Perimeter Array Detector (XPAD) on the Argonne PicoProbe
Nestor J Zaluzec, Argonne National Laboratory (USA)

PB08 Nanoscale thermometry of solids and liquids using in-situ STEM
Robert F Klie, University of Illinois at Chicago (USA)

PB09 Efficient particles size distribution characterization with TEM: recent advances in experiments and analysis automation

Daniel G Stroppa, Thermo Fisher Scientific in Eindhoven (The Netherlands)

PB10 Determination of 3D Strain fields by dark-field electron holography utilizing dynamic diffraction
L Niermann, Technische Universität Berlin (Germany)

PB11 Attolight Mönch add-on for (S)TEM
Maité Blank, Attolight SA in Lausanne (Switzerland)

PB12 Continuous-wave photonic chip-based temporal phase plates for TEM
Armin Feist, University of Göttingen and Max Planck Institute for Biophysical Chemistry (Germany)

PB13 Combining quantitative ADF STEM with SiN_x membrane-based MEMS devices: a simulation study with Pt nano-particles
Katherine E MacArthur, Forschungszentrum Jülich GmbH (Germany)

PB14 Quantification of long-range electric fields by combining 4DSTEM and multislice simulations
Damien Heimes, Philipps University Marburg (Germany)

PB15 Identifying support effects in CO oxidation with Au
Zachary R Mansley, Northwestern University in Evanston (USA)

PB16 Measurement of strain- and doping concentration-dependent electrostatic potential change in GaN wire using off-axis electron holography
Yan Lu, Forschungszentrum Jülich GmbH (Germany)

PB17 How to make electron holography quantitative: demonstrated exemplary for nitride semiconductor interfaces
Michael Schnedler, Forschungszentrum Jülich GmbH (Germany)

PB18 Single event detection in transmission electron microscopy for spectroscopic coincidence experiments
Daen Jannis, University of Antwerp (Belgium) and Forschungszentrum Jülich GmbH (Germany)

PB19 Atomically resolved 3D structural reconstruction of small quantum dots
Pritam Banerjee, Indian Institute of Technology in Madras (India)

PB20 Exploration of atomic structures in S3 [111] aluminium tilt grain boundaries
Saba Ahmad, Max-Planck-Institute for Iron Research in Düsseldorf (Germany)

PB21 XRD and TEM study of the quasicrystalline phase in pellets and metal-matrix composites consolidated using spark plasma sintering
Z L Dong, Jiangsu University in Zhenjiang (China)

PB22 Phase relation and morphology of titania polymorphs in extreme hydrothermal conditions
Alisa Gordeeva, Stockholm University (Sweden)

PB23 Modulation of cathodoluminescence emission by interference with external light
Valerio Di Giulio, The Barcelona Institute of Science and Technology (Spain)

PB24 Structural characterisation of HfO_2 thin films for resistive switching memory applications using transmission electron microscopy techniques
Xiou Hou, Forschungszentrum Jülich GmbH (Germany) and RWTH Aachen University (Germany)

TUESDAY 4 MAY 2021

POSTER SESSION C

PC01 Unveiling nanoscale optical properties of tmd monolayers using combined electron spectroscopy techniques
N Bonnet, Université Paris-Saclay in Orsay (France)

PC02 Direct evidence of an Al alloyed SiN_x interlayer within an ammonia predosed AlN/Si interface via STEM-EELS
Simon M Fairclough, University of Cambridge (UK)

PC03 Direct imaging of structural defects in NMC cathode material
Helen Valencia, RWTH Aachen University (Germany) and Forschungszentrum Jülich GmbH (Germany)

PC04 Combining ETEM/ESTEM and electrochemical impedancespectroscopy
Søren Bredmose Simonsen, Technical University of Denmark in Kgs. Lyngby (Denmark)

PC05 Tailoring of shape and size of platinum nanoparticles to enhance their oxygen reduction reaction performance
J Michalička, Brno University of Technology (Czech Republic)

PC06 Automated mapping of the crystallographic sample orientation from diffraction patterns in momentum-resolved STEM
Mauricio Cattaneo, RWTH Aachen University (Germany) and Forschungszentrum Jülich GmbH (Germany)

PC07 Moiré angle dependent excitonic absorption in twisted bilayer WSe_2 by EELS
Steffi Y Woo, Université Paris-Saclay in Orsay (France)

PC08 Understanding transition metal dichalcogenide absorption line widths in electron energy loss spectroscopy
Fuhui Shao, Université Paris-Saclay in Orsay (France)

PC09 Operando TEM study of all-solid-state battery interface
Shibabrata Basak, Forschungszentrum Jülich GmbH (Germany)

PC10 Interfacial defects in the layered structure of a chalcopyrite compound
Guangming Cheng, Princeton University (USA)

PC11 Sculpting plasmonic resonances in nanoparticles by intelligent electron beam control
Kevin M Roccapriore, Oak Ridge National Laboratory (USA)

PC12 Dosimetry in TEM and STEM
Ray F Egerton, University of Alberta in Edmonton (Canada)

PC13 Visualizing plasmon-driven nanoparticle transformations with optically-coupled environmental TEM
Katherine Sytwu, Stanford University (USA)

PC14 Ag segregation induced nanofaceting transition of a Cu tilt grain boundary and its impact on plastic deformation mechanisms
Nicolas J Peter, Max-Planck-Institut für Eisenforschung GmbH in Düsseldorf (Germany)

PC15 Live 4D-STEM processing at 15'000 detector frames per second for series registration
Benedikt Haas, Humboldt-Universität zu Berlin (Germany)

PC16 Machine learning electron diffraction
Renliang Yuan, University of Illinois at Urbana-Champaign (USA)

PC17 Off-axis holography of polarization fields in InGaN/GaN with monolayer resolution
T Niermann, Technische Universität Berlin (Germany)

PC18 Towards measurement of polarisation-induced electric fields in $PbZr_{0.2}Ti_{0.8}O_3$ by momentum-resolved STEM – Impact of systematic errors
Achim Strauch, Forschungszentrum Jülich GmbH (Germany)

PC19 Effect of probe defocus in momentum-resolved STEM experiments

H L Robert, RWTH Aachen University (Germany) and Forschungszentrum Jülich GmbH (Germany)

PC20 Fabrication of three element Boersch phase shifters and their characterization by voltage-controlled interference of coherent high energy electron beam
Soichiro Tsujino, Paul Scherrer Institut in Villigen (Switzerland) and University of Basel (Switzerland)

PC21 Localized surface plasmon resonances in Sierpiński fractal nanoantennas
Isobel C Bicket, McMaster University in Hamilton (Canada)

PC22 Anomolous contrast in HAADF STEM imaging of dysprosium scandanate
Duncan T L Alexander, EPF Lausanne (Switzerland)

PC23 Real-time interactive ptychography from electron event representation data
Philipp M Pelz, Lawrence Berkeley National Laboratory and University of California (USA)

PC24 Real-time measurement of changes in electrostatic potential in individual TiO_2 nanoparticles during resistive switching using off-axis electron holography
Janghyun Jo, Forschungszentrum Jülich GmbH (Germany)

PICO 2021

SIXTH CONFERENCE ON FRONTIERS OF ABERRATION CORRECTED ELECTRON MICROSCOPY

and

Colloquia Honouring Wolfgang Baumeister, Colin Humphreys, John Spence, and Knut Urban

Virtual Conference
2 – 6 May 2021

ABSTRACTS

STRUCTURAL BIOLOGY *IN SITU* : THE PROMISE AND CHALLENGES OF CRYO-ELECTRON TOMOGRAPHY

Wolfgang Baumeister

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Abstract

Traditionally, structural biologists have approached cellular complexity in a reductionist manner by characterizing isolated and purified molecular components. This 'divide and conquer' approach has been highly successful, as evidenced by the impressive number of entries in the PDB.

However, awareness has grown in recent years that only rarely can biological functions be attributed to individual macromolecules. Most cellular functions arise from their acting in concert. Hence there is a need for methods developments enabling studies performed *in situ*, i.e. in unperturbed cellular environments. *Sensu stricto* the term 'structural biology *in situ'* should apply only to a scenario in which the cellular environment is preserved in its entirety.

Cryo electron tomography has unique potential to study the supramolecular architecture or 'molecular sociology' of cells. It combines the power of three-dimensional imaging with the best structural preservation that is physically possible to achieve. We have used this method to study the 26S proteasome in a number of cellular settings revealing their precise location, assembly and activity status as well as their interactions with other molecular players of the cellular protein quality control machinery.

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FROM ELECTRON MICROSCOPY TO GALLIUM NITRIDE TO GRAPHENE ELECTRONIC DEVICES

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Abstract

This talk will give case studies of how electron microscopy has underpinned and stimulated the research of my research groups, culminating in our current work on manufacturable graphene electronic devices.

In 1998, Gianluigi Botton was working with me on EELS and we found that the calculated EELS spectrum for nickel oxide did not agree with the experimental results. I contacted Sergei Dudarev in Oxford and he developed a new theory to better account for electron correlations in the 3d shell of nickel. It turned out that solid-state physicists were having similar problems with understanding the properties of NiO and related materials, and our paper has been of fundamental importance to them and cited over 8000 times.

In 2003, it was universally believed by the GaN community that GaN/InGaN quantum well LEDs were so bright because of In-rich clusters in the InGaN quantum wells. It was thought that they prevented the carriers from diffusing to dislocations, which would have quenched the light emission. My research student, Tim Smeeton, used high resolution electron microscopy to show that such clusters did not exist, and were due to radiation damage. Aberration-corrected electron microscopy was then used to show that there were atomic-height steps at the InGaN/GaN interface, and calculations showed that these steps localised the electrons. This work was of fundamental importance in understanding and optimising light emission from such LEDs.

Humphreys' group pioneered making low-cost GaN LEDs on large-area silicon by MOCVD. Aberration-corrected electron microscopy was essential for understanding and optimising the interface between AlN and Si. This was a key factor in the exploitation of this technology. Two companies were spun out, CamGaN (2010) and Intellec (2011), which were acquired by Plessey in 2012, who then manufactured such LEDs. Plessey are now manufacturing microLEDs based on this technology and have a partnership with Facebook (2020) to use microLEDs in AR, etc.

Humphreys' group grew large-area, device-quality graphene by a new technique for graphene growth, MOCVD, and patented this in 2015. A company was set up to exploit the technology, Paragraf, which moved into premises in 2018. Its first product is a manufacturable graphene Hall-effect sensor, which is the best-performing Hall sensor in the world and will be launched in March 2021. This sensor raises expectations that graphene biosensors and transistors may be manufacturable. Aberration-corrected electron microscopy will be essential in characterising and optimising such devices.

INSTRUMENTATION FOR ADVANCED ELECTRON MICROSCOPY TECHNIQUES

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Abstract

An electron microscope is a tool for the investigation of small objects in various fields of natural sciences like, for example, in Materials Science or Biological Structure research. Hence, any development or modification of an instrument has to follow the requirements of the proposed users and its applications. The precision of experimental results can only be as precise as the available tools allow for. Therefore, one important aspect of any instrumental development is not only the achievable resolution (spatial/angular/energy resolution etc.) one can show with one acquired image, instead it is more important to lower any noise accompanied with the measurements to reduce the error bars of the acquired data. With respect to the needs of advanced TEM users who are asking for larger fields of view which are well transferred, we developed already some years ago the so-called BCOR for the correction of the linear off-axial coma [1]. This BCOR is successfully used in Materials Science, e.g. for Holography, and in Life Sciences recently contributed to a new resolution record in single particle structure research [2]. However, due to the complexity of this BCOR and its length (about 43 cm) we recently developed the Advanced TEM Corrector (ATCOR) which has the same outer shape as the standard CETCOR. Due to several design changes and additional channels it combines the advantages of the well-accepted advanced STEM corrector (DCOR/ASCOR), i.e. minimized A_5 (5^{th} order astigmatism) by using shorter hexapoles with additional quadrupoles to offer full 4^{th} order axial aberration correction, with an increased available field of view due to substantially reduced intrinsic off-axial aberrations S_{3g} and A_{3g}. However, for very large detectors (≥4kx4k) the off-axial coma still remains the limiting aberration which may justify the use of a BCOR. An additional type of aberration corrected TEM is a Cc/Cs corrected TEM for cryo-tomography of thicker biological objects. For this purpose, a CCOR has been adapted to the cryo-lens of a TFS Krios TEM and optimized for the planned applications. This TEM has been shipped to China and currently the installation is ongoing. For analytical purposes like EFTEM and EELS we developed a post-column energy filter with high performance and more freedom to combine various hardware components like novel cameras and detectors without being linked to one EM vendor. The optics design of our CEFID (CEOS Energy Filtering and Imaging Device) [3] consists of an optimized sector magnet with twelve-pole elements at the entrance and exit plane and some focusing elements for which we used pure quadrupoles. In addition, a useful number of deflectors and separate stigmators are installed for appropriate alignment routines. An important part of this development was the long-time stability and ease of use, i.e. reliable switching between various spectrum modes. The general goal of this development was to offer larger flexibility and leave the choice of the combination of hard- and software to the user. The application the user has in mind and its best realisation dictates the decision of the set-up because he/she knows best about the requirements. Due to increasing demands on higher energy resolution and improved performance, for example, for ω-q mapping or phonon spectroscopy this development will be continued.

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[2] K. Yip et al., Nature 587, 157-161, 2020.

[3] F. Kahl et al. AIEP 212 https://doi.org/10.1016/bs.aiep.2019.08.005

INVERSION OF DYNAMICAL SCATTERING FROM BRAGG INTENSITIES

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Abstract

A method for recovering complex structure factors from many simultaneously excited Bragg beam intensities is described. The method is applied to simulated transmission electron diffraction data over a wide range of crystal thickness and beam energies. The method is based on iterated projections between structure and scattering matrices, which are related by a matrix exponential, which we invert. The algorithm removes multiple-scattering perturbations from diffraction data (such as Micro-ED data) and might be extended to other fields, including X-ray and neutron diffraction and cryo-electron microscopy. Because coherent multiple scattering involves interference between Bragg beams, the method also solves the phase problem. Unlike dynamical inversion from electron microscope images or ptychography data, the method, which starts with Bragg beam *intensities*, provides complex structure factors unaffected by focusing errors or resolution limitations imposed by lenses. We provide inversions to the same complex structure factors from simulated data with 441 simultaneously excited Bragg beams over a range of thickness and beam energy. We discuss the retrieval of chirality information from enantiomorphs (such as crystals of drug molecules), the efficient incorporation of symmetry information using the irreducible representation of the group of structure matrices, and the effect of HOLZ lines to provide three-dimensional information [1,2, 3].

If time allows, a summary of our new iterated projection algorithm will be given which merges into 3D the Cryo-EM images collected on a curved Ewald sphere. These are not projections, because the depth of focus is less than the sample thickness, and so cannot be merged in the usual manner. Curved sphere artifacts appear at lower beam energy, larger sample thickness, and high resolution [4].

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TIMELY AND ATOMIC-RESOLVED HIGH-TEMPERATURE MECHANICAL TESTING SYSTEM

Jianfei Zhang[§], Yurong Li[†], Xiaochen Li[§], Yadi Zhai[§], Qing Zhang[§], Dongfeng Ma[§], Shengcheng Mao[§], Qingsong Deng[§], Zhipeng Li[§], Xueqiao Li[§], Xiaodong Wang[‡], Ze Zhang[§,†], and Xiaodong Han[§]

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Abstract

Timely and atomic-resolved room-temperature mechanical testing system has been developed in the past [1-4]. Revealing the atomistic mechanisms of high-temperature mechanical behavior of materials is important for optimizing their properties for service at high-temperatures and their thermomechanical processing. However, due to the dynamic recovery and the absence of available *in situ* techniques, the high-temperature deformation behavior and atomistic mechanisms of materials are difficult to evaluate. Here, we report the development of timely and atomic-resolved high-temperature mechanical testing system that enables mechanical testing at temperatures above 1000K inside a transmission electron microscope [5]. Several metal and alloy examples including W, Cu and Ni-based superalloys are tested with the system. We will introduce tungsten fractures in a ductile manner at high temperature and the dislocation dynamics of Cu tested at high temperatures. Our research provides an approach for timely and atomic-resolved mechanical testing system of materials at high-temperatures.

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[2] Nature Communications, 3, 609, DOI: 10.1038/ncomms1619, 2012
[3] Nature Communications, 4, 2413, DOI: 10.1038/ncomms3413, 2013
[4] Nature Communications, 5, 4402, DOI: 10.1038/ncomms5402, 2014
[5] Nature Communications, 2021.

STABILISATION MECHANISMS OF POLAR OXIDE SURFACES STUDIED BY IN-SITU ELECTRON MICROSCOPY AT HIGH TEMPERATURES

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Abstract

Polar surfaces of ionic oxides consisted of alternating stacking of oppositely charged layers are unstable if their atomic structure remains the same as in the bulk-truncated form and/or their surface charges are uncompensated. When a polar surface is prepared by cleavage or annealing of ionic oxide in vacuum, it exhibits various structural and chemical modifications to compensate the surface charges, for example, via ionic reconstruction, removal of surface atoms (vacancy formation), the formation of step edges with opposite charge, or massive faceting toward vicinal surface orientation [1]. During crystal growth or evaporation a polar surface may exhibit dynamic structural and chemical evolution, which can critically affect the thermodynamics and kinetics of surface reactions or diffusion. However, it remains largely unknown due to difficulties of in-situ, real-time, and atomic-scale observation of high temperature evaporation or growth processes of polar oxide surfaces.

Recently, we observed unique surface evolution of ZnO (0001) and $LaAlO_3$ (001) polar surfaces using a negative Cs HRTEM imaging (NSCI) at elevated temperatures. We found that the Zn-terminated Zn (0001) and the LaO-terminated $LaAlO_3$ (001) surfaces, both are positively charged, tend to form a regular array of surface steps into vicinal surface orientations to cancel the normal component of dipole moment. The formation of cation vacancies, e.g., Zn vacancies (V_{Zn}) in Zn-(0001) and La vacancies (V_{La}) in $LaAlO_3$ (001), which are negatively charged, is energetically favored and plays important roles in the step formation, reconstruction and even disordering of crystalline lattice before evaporation. The Zn-deficient layer (similar to ZnO_2 in composition) formed by diffusion of V_{Zn} on the Zn- (0001) ZnO surface becomes easily disordered at around 300 °C due to its structural instability and catalyzes the congruent evaporation of ZnO. In the case of LaO-terminated (001) $LaAlO_3$ surface, while the (013) or (015) vicinal surface orientation is preferred to cancel the normal component of dipole moment, the step reconstruction involving the formation of V_{La} and La_{Al} anti-site defect along the step edges is driven to compensate the in-plane dipole moment. At temperatures higher than 800 °C, the step edges containing V_{La} + La_{Al} become disordered and highly mobile on the LaO-terminated $LaAlO_3$ (001) surface.

The present in-situ (S)TEM study reveals that the stabilization mechanism of polar oxide surfaces is driven primarily by the electrostatics that favors the step formation into vicinal orientation and the vacancy formation at low temperatures and the vacancy induced disordering that is driven by the increase of entropy favored at high temperatures.

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IN SITU LIQUID CELL TEM STUDY ON THE WET CHEMICAL ETCHING OF SEMICONDUCTING NANOWIRES AND NANOBELTS

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Abstract

Wet chemical etching is a widely used process to fabricate fascinating nanomaterials. Understanding the etching mechanism and kinetic evolution process is crucial for controlling wet chemical etching. The development of in situ liquid cell transmission electron microscopy (LCTEM) enables the study on wet chemical etching with high temporal and spatial resolutions. Semiconducting nanowires and nanobelts have broad applications, especially in semiconductor technology and nanotechnology. Wet chemical etching is an important method to fabricate semiconducting nanomaterials with designed size and morphology.

In this talk, I will introduce our studies on wet chemical etching InAs nanowires[1], ZnO nanobelts[2], and other one-dimensional (1D) nanowires by LCTEM. The etching rate of InAs nanowires along radial direction in radiolytic water is found to remain constant with the shrinkage of nanowires, even when the diameter reduces to 5 nm. Hillocks are observed on the (000-1) O-terminated surface of ZnO nano/micro belts during in situ etching. Nanoparticles on the apex of hillocks are observed to be essential for the formation of the hillocks, providing a direct experimental evidence of micromasking mechanism for the first time. The surfaces of the obtuse triangular hillocks are characterized to be high-index {01-1-3} facets. High-index facets may have important applications in many fields. O_2 plasma treatment is found to be the reason for the appearance of {01-1-3} facets after etching. We thus develop a method to control the surface facets of ZnO to be high-index {01-1-3} facets or low-index {01-1-1} facets. We further confirm that the method is also applicable to bulk ZnO. Our results are significant for understanding the etching of 1D nanomaterials and nanobelts. Our results on ZnO nano/micro belts are also significant to controllably fabricate high-index surface facets.

[1] M. Sun, X. Li, et al., Nanoscale, 10 (2018) 19733.
[2] M. Sun, B. Yu, et al., Small, 16 (2020) 1906435.

HIGH RESOLUTION IN SITU ELECTRON MICROSCOPY STUDIES TO CORRELATE ATOMIC STRUCTURE TO STRAIN INDUCED EFFECTS

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Abstract

This talk will address and illustrate the use of piezo driven *in situ* manipulation in the electron microscopy to stimulate changes and with site specificity directly correlate atomic structure to strain induced changes at high spatial resolution. The manipulation allows us not only to manipulate but also to stimulate and probe with high precision. The effect of electric fields, light, mechanical strain and temperature on both structure and properties can be quantitatively imaged and studied using spectroscopy. The obtained knowledge is used to tune the properties of advanced materials and devices. Catalytic activity of metal nanoparticles [1] and electrical properties of semiconducting nanowires [2-4] are examples where the strain induced effects have a strong influence on the properties and performances. Our situ studies have also revealed that extremely high electric fields can dramatically change the atomic ordering [5]. In addition, we have noted that the thermal handling capabilities of one atomic layer thin surface film can be used to compensate for local heating effects of the electron beam by comparing observations on bare SiN transmission electron microscopy windows to results from windows dressed with single layers of graphene oxide [6-7]. New aspects of material properties and mechanisms, not obvious from measurements on the macro scale, can be revealed using in-situ electron microscopy. The knowledge is crucial for not only the understanding of the mechanisms that are involved but also for the design or materials and devices with tailored properties.

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[2] L. J. Zeng, C. Gammer, B. Ozdol, T. Nordqvist, J. Nygård, P. Krogstrup, A.M. Minor, W. Jäger and E. Olsson, "Correlation between electrical transport and nanoscale strain in InAs/In0.6Ga0.4As core-shell nanowires, Nano Letters 2018, DOI:10.1021/acs.nanolett.8b01782.

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[6] N. Voskanian, E. Olsson and J. Cumings, "Field-dependent heat dissipation of carbon nanotube electric currents", Scientific reports 2019, DOI: 10.1038/s41958-019-46944-9

[7] H.M. Nilsson, L. de Knoop, J. Cuming and E. Olsson, "Localized resistance measurements of wrinkled reduced graphene oxide using in situ transmission electron microscopy", Carbon 2017, DOI: 10.1016/j.carbon.2016.10.086

UNDERSTANDING NANOSCALE PROCESSES IN LI-ION BATTERIES BY *OPERANDO* SCANNING TRANSMISSION ELECTRON MICRSCOPY

B Layla Mehdi[§,†] , Wequin Li[§,†], Ioannis Siachos[§], Juhan Lee[§,†], Daniel Nicholls[§], Felipe Tontini[§], Mounib Bahri[§], and Nigel D Browning[§,†,‡]

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Abstract

The worldwide need for net-zero carbon emission to minimize global temperature rise requires new solutions in the area of clean energy technologies to power all aspects of society's current and future needs in transportation, industrial processes and a wide range of personal and home devices. In this regard, the pressing need for energy storage systems with both higher energy densities and high-power capabilities requires the development and implementation of new battery electrode nanomaterials as well as high stability liquid/solid electrolytes, in a manner that also permits a circular economy to be developed – recycling Li-ion batteries is a major challenge in itself. Current developments in atomic scale imaging methods for *operando* scanning transmission electron microscopy (STEM) now permit routine testing (direct imaging and analysis) of the dynamic structural changes that occur at the electrode/electrolyte interfaces in functioning nanobatteries created inside the microscope. In this presentation, I will focus on the use of wide range of *Operando* STEM methodes to analyse Li-ion batteries and understand the molecular level interactions occurring at the liquid/solid and solid/solid interfaces, focusing on cycling efficiency[1], side reactions, anode stability, controlled formation of a solid-electrolyte interphase (SEI) layer[1] and wide range of dynamic phase transformations occurring in Ni-rich cathodes. In addition, I will discuss new imaging methods[2-4] currently under development to improve our understanding even further, and demonstrate how they can be applied to beyond Li-ion energy storage technologies and many other materials challenges in the future.

[1] B. L. Mehdi et al.,*NanoLetters* (2015)

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INTELLIGENT MICROSCOPY: A PATH TOWARD TAILORED MATERIALS AT THE ATOMIC SCALE

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Abstract

Abstract In situ transmission electron microscopy (TEM) and electron energy-loss spectroscopy (EELS) are powerful tools for the observation of real-time materials processes. Specifically, in situ TEM is a reliable source for probing dynamic phenomena in order to gain a predictive understanding of myriad materials and use this knowledge to tailor future, improved systems. The development of radiation hard direct detection (DD) electron sensors has enabled improvements in the quality of in situ data for TEM imaging, and recently, we have demonstrated that DD provides far-reaching benefits for EELS [1]. With these improvements, critical issues in both physical and biological sciences can be addressed. Additionally, dynamic ionic or defect-dependent phenomena can be quantified with the combination of in situ methods and the DD EELS system [1-3].

This talk reviews recent work in using in situ investigation of chemistry-property relationships and coordination chemistry in a rapidly emerging family of 2D materials, MXenes. Their general formula is $M_{n+1}X_nT_x$, where M is a transition metal, X is C and/or N, n = 1–4, and T represents the surface termination (-OH, -F, and -O). This talk highlights thermal control of -F termination in $Cr_2TiC_2T_x$ and offer a first step toward tuning MXene surface termination to control metallic conductivity, work function, charge storage properties, and superconductivity [2,3]. Additionally, work in a wide variety of systems, including local structure evolution in high entropy alloys and metallic glasses, with profound implications in developing thermally stable, damage tolerant materials, with implications in critical applications in energy and aerospace industries.

Finally, an outlook on emerging time resolved studies and key challenges for "big data" will be presented [4]. In particular, a major challenge in understanding complex phase transformations is obtaining high resolution spectra at rapid time scales. Even with direct detection, spectra can be highly noisy. To combat this, use of machine learning for rapid acquisition EELS will be presented in the context of accuracy of identification of oxidation state and coordination chemistry. This work will be discussed as a foundation for future integration of ML at high speeds during in situ TEM experiments.

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CONVERGENT BEAM DIFFRACTION AT X-RAY FREE-ELECTRON LASERS

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Abstract

With multilayer Laue lenses, it is now possible to construct diffractive lenses for X-rays with an imaging resolution approaching 1 nm [1,2]. This is smaller than the unit cell of macromolecular crystals, and thus opens up intriguing possibilities for structure determination, as pointed out by Spence et al [3]. The intense X-ray beam formed by focusing to such dimensions would rapidly cause radiation damage but this can be circumvented with femtosecond-duration FEL pulses (which also possess the necessary coherence). Convergent-beam diffraction is well known in electron microscopy [4] and its application to thin crystals gave rise to ptychography [5,6]. Its application to (thick) three-dimensional crystals in a "diffraction before destruction" [7] experiment poses some nice challenges which will be discussed.

Other methodologies long available in TEMs also provide inspiration for FELs, such as point-projection imaging which gives a particularly simple and robust method to record highly magnified holograms of objects, for single-particle imaging.

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ELECTRON-BEAM MANIPULATION OF LATTICE IMPURITIES

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Abstract

Covalently bound impurity atoms in crystal lattices can be manipulated using the atomically focused electron probe of an aberration-corrected scanning transmission electron microscope. This has revealed inspiring new perspectives for top-down atomic engineering, with the potential to surpass existing techniques in both versatility and capabilities.

Manipulation was first realized for incidental silicon impurities in single-layer graphene. Elastic backscattering of a probe electron from a moving C nucleus [1] causes the Si to *directly exchange* places with one neighboring C atom via an out-of-plane displacement [2], and such dynamics can be controlled by directing the focused electron beam at the desired atomic site [3]. Our manipulation rate is nearly on par with any atomically precise technique [4], and such control is also possible in single-walled carbon nanotubes [5].

Phosphorus dopants in graphene can be manipulated with difficulty [6], and there seem to be physical limits on what is feasible as significantly heavier Ge impurities cannot [7]. The curious replacement of irradiated impurities by C atoms has also emerged as a practical hurdle for the further scaling of the technique. However, similar irradiation-induced atomic dynamics have been observed for many impurity elements [8], and based on our modeling, several transition metals also appear as promising targets.

Perhaps even more excitingly, the electron-beam manipulation of Bi dopants in bulk silicon was recently reported [9], although the precise mechanism was left unclear. We have now applied our established *ab initio* modeling methodology [10] to address this question, revealing a novel type of non-destructive mechanism we call *indirect exchange*. Further, we demonstrate that the promising nuclear spin qubit Sb can likewise be manipulated.

Support from the European Research Council (grant 756277-ATMEN) and computational resources provided by the Vienna Scientific Cluster (VSC) are gratefully acknowledged.

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EXTREME LIGHT-MATTER INTERACTIONS IN THE ULTRAFAST TRANSMISSION ELECTRON MICROSCOPE

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Abstract

Over the past few years, quantized interactions between coherent free electrons and femtosecond laser pulses have shown intriguing new prospects for light-matter interactions.

The talk will present theory and experiments of free electrons in laser-driven (ultrafast) transmission electron microscopy. Our experiment achieved what is, in many respects, the most powerful nearfield optical microscope in the world today. We resolve photonic bandstructures as a function of energy, momentum, and polarization, simultaneously with capturing the temporal dynamics and spatial distribution of the photonic modes at deep-subwavelength resolution. Recently, we used these new capabilities to observe coherent free-electron interactions with light trapped in photonic crystals and with polariton wavepackets in 2D materials.

I will explain how such capabilities enable creating a free-electron qubit, and discuss the new capabilities that such electrons provide for probing quantum materials.

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STRUCTURAL DISORDER CHARACTERIZED BY ABERRATION CORRECTED SCANNING TRANSMISSION ELECTRON MICROSCOPY: REAL-SPACE 2-D PARTIAL PAIR CORRELATION FUNCTIONS

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Abstract

The ability to design the composition and microstructure of electronic ceramics for emerging technological applications requires sophisticated characterization techniques that can provide quantitative information about local structure and chemistry. Such structure quantification is particularly important to the fundamental understanding of properties in many electronic ceramics, where local heterogeneities associated with dopants or intrinsic lattice defects give rise to local inhomogeneities in charge, strain and polarization. Herein we use aberration-corrected scanning transmission electron microscopy (STEM) imaging to quantify short- and medium-range lattice disorder on a sublattice basis, specifically focusing on the tetragonal tungsten-bronze (TTB) structured $Ba_5SmSn_xNb_{1-x}O_{30}$, which is a relaxor ferroelectric. We address the quantification challenges presented by the structural complexity of TTBs (including closely spaced atomic columns and 5 distinct cation column types in the <110> projection) by developing a new algorithm for simultaneously locating and indexing all atom columns in a lattice image. The method utilizes a reference lattice generated from a unit cell structural projection, from which deviations from the ideal lattice positions are quantified. From the fitted data, we then compute real–space partial pair correlation functions (p-PCFs) for each cation sublattice. Instead of radially averaging the data, which is typical for p-PCF analysis, we retain the vector in formation in 2-D p-PCFs. These correlation functions provide a robust method for quantifying short- and medium-range structural displacements, including incommensurate modulations, on particular sublattices. We demonstrate in that the incommensurate modulation in $Ba_5SmSn_xNb_{1-x}O_{30}$ is only associated with one of the cation sublattices. We feel that this approach will be generally useful for quantifying local- to long-range order in structurally complex materials.

NANOSCALE FUNCTIONAL CHEMISTRY AND OPTO-ELECTRONIC RESPONSE OF ORGANIC CRYSTALS

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Abstract

Vibrational spectroscopies offer of the ability to interrogate chemical bonds directly. Conventionally used IR and Raman spectroscopies probe molecular vibrations and phonons in large materials volumes, capturing details of functional groups and lattice dynamics, while tip-enhanced scanning near-field implementations have enabled vibrational spectroscopy approaching 10 nm spatial resolution but limited to surface studies. Recent advances in electron energy loss spectroscopy in the scanning transmission electron microscope (STEM-EELS) have enabled the observation of the spectral signatures of phonons and molecular vibrations at much shorter lengthscales. In molecular systems, where long range interactions have enabled 'aloof' spectroscopy with reduced electron beam induced damage, the broadband energy window accessible in high resolution EELS also makes it possible to detect the optical signatures from the mid-infrared to the UV energy range. However, understanding variations in bonding or optical response at the nanoscale in molecular and metal-organic solids requires measurements with higher spatial sensitivity than afforded by the simple aloof beam approach. Here, we develop strategies for high spatial and energy resolution STEM-EELS to fingerprint the optical properties and molecular vibrations of beam-sensitive crystals at the nanoscale.

Organic and metal-organic framework (MOF) glasses show significant promise in applications from LEDs and photovoltaics to photocatalysis but their macroscopic optoelectronic properties are often only linked to microscopic optical properties and atomic structure in statistical descriptions of volumes much larger than the molecule or unit cell. In contrast, high-energy-resolution STEM-EELS provides a nanoscale fingerprint of these optical properties and of characteristic molecular vibrations in MOFs. In particular, as a result of tetrahedral ligand-field splitting at Co metal centres, d-d transitions directly reflect the local coordination [1], while intra-ligand and metal-to-ligand transitions in the vibrational signal can be mapped with nm precision to distinguish nanoscale regions of distinct coordination chemistry. [2]

Perylene diamide (PDI) represents a major class of n-type organic semiconductors exhibiting particularly long exciton lifetimes that are extremely attractive for optoelectronic applications. Their observation, however, is a great challenge due to severe damage under electron beam irradiation even at moderate rates. Here, we use STEM EELS measurements in a spatially correlated near-aloof geometry, to record spatially resolve spectra in the vicinity of structural features such as point or line defects in PDI nanobelts. The beam-positioning strategy mitigates beam damage and enables the observation of the vibrational and optical characteristics of the nanobelts and changes therein at extended defects.

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CRYSTALLOGRAPHIC INFORMATION IMAGING (CII), HOW FAST DETECTORS AND MODERN COMPUTER ALOGRITHMS HAVE CREATED A NEW FORM OF ELECTRON MICROSCOPY

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Abstract

Four-dimensional diffraction datasets (4D-DDs) collected using the scanning electron nanodiffraction (SEND) [1] or 4D scanning transmission electron microscopy (4D-STEM) [2] techniques have gained increasing popularity in the electron microscopy community for their versatility in both electron diffraction and imaging. The advantage of having 4D-DDs over traditional 2D imaging techniques comes from the rich information captured in diffraction patterns, which can be related to the sample structure, defects, electric and magnetic fields. Experimental acquisition of large 4D-DDs has been significantly improved with the development of fast electron pixel array detectors. Now, 4D-DD analysis to disentangle various diffraction signals and extract information in a fast and efficient way has become the central focus of this new form of microscopy.

Here, we focus on the essential aspects of diffraction analysis, namely, diffraction pattern indexing, measurement of Bragg peak positions and examination of diffuse scattering. We demonstrate how these tasks are automated using novel techniques and modern computer algorithms and applied the analysis of tens to hundreds of thousands diffraction patterns for crystallographic information imaging (CII). In the process, we introduce how machine learning can aid such analyses. We highlight the potential of CII through application examples of nanostructure orientation and phase mapping [1], precision strain field analysis [3, 5], Cepstrum based electron imaging [4]. Further, we show how combination of these analyses enables the structure determination of highly complex materials, such as multi-principal component alloys.

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THREE-DIMENSIONAL IMAGING BY STEM DEPTH SECTIONING

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Abstract

Following the developments of geometric aberration correction, atomic-resolution imaging and spectroscopy become indispensable tools in the fields of physics, chemistry, and materials science. The spatial resolution in scanning transmission electron microscopy (STEM) is now in deep sub-angstrom realm of 40.5 pm [1]. However, such high spatial resolution is only valid in the projected lateral two dimensions, and the depth resolution along the axial direction is still far from atomic resolution. One of promising approach is STEM depth sectioning with larger illumination angles [2,3], where we acquire a focal-series of atomic-resolution STEM images in a single projection. We have recently installed a Delta-type 5th order geometric aberration corrector in JEM ARM300CF, and the flat region of Ronchigram is stunning enhanced up to 70 mrad in semi-angle [1,4]. To utilize such large illumination angles, it is prerequisite to accurately correct lower orders of geometric aberrations such as 2-fold, 3-fold astigmatisms and axial coma. We therefore developed a new method to efficiently minimize residual aberrations by using atomic-resolution STEM images [5], rather than the conventional Ronchigram algorithms. By implementing such automated geometric aberration correction, it becomes possible to perform STEM observations at sub-angstrom lateral resolution, even with the large illumination angle of 63 mrad. To evaluate the depth resolution with the larger illumination angles, we observed isolated single cerium (Ce) dopants embedded within a crystal of cubic boron nitride (*c*-BN) via ADF-STEM depth sectioning. We then achieve the depth resolution of 2.14 nm, with the illumination angle of 63 mrad [6]. On a basis of the identified three-dimensional locations of single Ce dopants, we also determine the Ce-Ce partial pair distribution function in a considerably longer distance up to 8.5 nm, which is much larger than the coherent conventional diffraction experiments (2 or 3 nm at most).

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DIMENSIONALITY REDUCTION AND UNSUPERVISED CLUSTERING FOR ELECTRON ENERGY-LOSS SPECTROSCPY-SPECTRUM IMAGE

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Abstract

A novel combination of machine learning algorithms is proposed for the differentiation of distinct spectra in a large electron energy loss spectroscopy spectrum image (EELS-SI) dataset. For clustering of the EEL spectra including similar fine structures in an efficient space, linear and nonlinear dimensionality reduction methods are used to project the EEL spectra onto a low-dimensional space. Then, a density-based clustering algorithm is applied to distinguish the meaningful data clusters. By applying this strategy to various experimental EELS-SI datasets, differentiation of several groups of EEL spectra representing specific fine structures was achieved. This allows to investigate particular fine structures by averaging all of the spectra in each cluster. Also, the spatial distributions of each cluster in the scanning regions can be observed, which enables investigation of the locations of different fine structures in materials. By applying these machine-learning algorithms, we differentiate various fine structures in an experimental EELS dataset and extract representative spectra without any physical background or prior knowledge. Furthermore, this strategy can be applied to any hyperspectral dataset as well as an EELS-SI one, e.g., energy-dispersive X-ray spectroscopy and cathodoluminescence datasets. Here, a low-loss EELS dataset of gold nanoparticles and EELS datasets of amorphous carbon layers as well as high-k materials are used to respectively evaluate the practicability of this strategy.

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MULTISLICE PTYCHOGRAPHY WITH STRUCTURED ILLUMINATION

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Abstract

Perhaps the most powerful application of ptychography to date has been its combination with tomography for 3D imaging [1]. Isotropic resolutions approaching 10nm are now possible at X-ray wavelengths using this technique, which has the added benefit of recovering a full complex refractive index map of the sample.

The compromise in reaching these impressive resolutions is a stringent limit on sample thickness, which reduces at a rate proportional to the resolution squared. This limit arises due to diffraction and multiple scattering within the sample, which violate the geometric optics assumption underpinning tomographic reconstruction.

One idea to circumvent this limit is multi-slice ptychography [2]. Here, diffraction and multiple scattering are accommodated by modelling the sample volume as a series of thin sections, each of which obey the rules of geometric optics, and modelling the propagation of light between the sections using wave optics.

In this talk I will explain multi-slice ptychography, the algorithms it uses to recover images, and how it can be combined with tomography to realise high-resolution 3D imaging of thick samples. I will focus on recent work using structured illumination to realise efficient, high-throughput 3D ptychography, with examples from visible light [3], X-ray [4] and electron [5] experiments.

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SELF-CALIBRATING METHOD FOR EFFECTIVE ABSORPTION CORRECTION OF ELEMENTS WITH MULTIPLE X-RAY LINES IN TRANSMISSION ELECTRON MICROSCOPY

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Abstract

Energy-dispersive X-ray spectroscopy (EDXS) uses the intensity of the characteristic lines of chemical elements relative to a standard to measure the chemical composition of samples in either scanning electron microscopy (SEM) of bulk or transmission electron microscopy (TEM) of thinned sections. Key for quantification are a knowledge of the fluorescence yield, the detection efficiency and the absorption within the sample. While for SEM of bulk samples these can all be determined and modelled to some reasonable accuracy, self-absorption is a considerable problem in TEM of thin foil samples where absorption within the sample depends on thickness, orientation, geometry and surface roughness of the thin foil sample and on the detector take-off angle. While some of these parameters can be measured and then absorption be modelled, the related error bars can be big. As a result, EDXS of stoichiometric samples does often not reproduce the nominal chemistry. For GaAs and for InAs cleaved wedges it has been shown [1] that the measured group III/V ratio deviated considerably from unity and that an absorption correction for the thickness known could even increase errors.

A method has been developed to provide more accurate X-ray microanalysis in a (scanning) transmission electron microscope by an internal self-consistent absorption correction based on the presence of at least one heavier element with multiple X-ray lines (K and L lines, or L and M lines, i.e. $Z > 20$).

Developed originally for compound semiconductors such as InGaN and InGaAs [2-4], the method has also been applied to SiGe where it provided 1-2 at% accuracy for two different alloy specimens that were up to 1µm (!) thick [5]. It has also been successfully used to quantify X-ray maps by applying the fitting to every point in maps.

Crucially, the traditional k-factor of an X-ray line for thin film analysis and its corresponding absorption correction factor or not modelled independently as function of absolute specimen thickness but their product (called 'effective k*-factor') relative to some other element within the sample is plotted as function of a K/L line ratio for a heavy element that can actually be directly measured from an experimental spectrum. Whether an increased K/L line ratio is due to a thicker sample, a smaller take-off angle or a thicker detector window becomes completely irrelevant; its value calibrates the absorption correction to be applied to get consistent results for quantification of K- and L-lines.

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SEQUENTIAL PLAN-VIEW TEM IMAGING OF POROUS GAN

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Abstract

Transmission electron microscopy (TEM) is a central technique for the characterisation of materials at the atomic scale. However, it requires the sample to be thin enough to be electron transparent, imposing strict limitations when studying thick structures in plan-view.

Here we present a method for sequential plan-view TEM that allows one to image complex structures at various depths. The approach consists of performing an iterative series of front-side ion milling followed by TEM imaging. We show it is possible to image how the sample properties vary with depth up to several microns below the surface, with no degradation of the sample and imaging conditions throughout the experiment [1].

We apply this approach to 3D cavities in mesoporous GaN distributed Bragg reflectors, in which it was previously demonstrated that threading dislocations in the sample acted as vertical channels for the porosification process [2], but the evolution of the pore morphology with depth was still unexplored. Here we demonstrate the ability to characterise the morphology of the pores, local crystal features and chemical composition through the multilayer structure.

The same workflow can be applied to a variety of complex micron-scale systems which are by nature too thick for standard TEM analysis, and can also be adapted for profiling samples in cross-section.

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DETECTING PLASMONS' INTERACTIONS AND STRAIN IN NANOSTRUCTURES

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Abstract

With the developments of electron monochromators and improvements in energy resolution using numerical techniques, electron energy loss spectroscopy has enabled the detection of plasmonic (and more recently phonon) resonances with exquisite sensitivity.

In this presentation we discuss recent examples of visualization and modeling of resonances in very simple, but very elegant structures, and demonstrate the origins of the highly active optical response of metallic nanostructures [1-4] and conductive transparent oxides [5].

We also show how we can take advantage of a new methodology, the STEM Moiré Geometric Phase Analysis (SMG), built on the well-recognised principles of Moiré interferometry, implemented in STEM to quantitatively deduce strain fields in complex nanostructures [6,7]. With this powerful quantitative technique, it is possible to expand the field of view in strain measurements to several microns in size and obtain reliable information while maintaining reasonable precision. With this technique, it is possible to detect strain in multilayers stacks consistent with other well-established techniques, such as dark-field electron holography, so that the depth sensitivity of the strain measurements can be investigated.

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CLUSTERING AND PRECIPITATE NUCLEATION IN AL ALLOYS STUDIED BY SPED AND HAADF-STEM

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Abstract

Age-hardenable aluminium (Al) – Al-Cu, Al-Mg-Si and Al-Mg-Zn – alloys are important structural materials for construction and automotive applications due to properties like high strength/weight ratio and good formability. Chemical composition and thermo-mechanical history of the alloy will to a large extent decide its microstructure and therefore physical properties. Our research group at NTNU and SINTEF in Trondheim has over a long period worked together with the Norwegian light metal industry on studies of precipitates and microstructure in these Al alloys [1]. Precipitation starts out with nanoscale clustering of solute elements and vacancies in the supersaturated, metastable Al solid solution. Solving the structure and quantifying the distribution of these clusters are important to design better alloys. We have solved the structure of the clusters in Al-Mg-Zn alloys [2]. These are essentially partial substitutions of Mg and Zn on the cubic aluminium sites organised in a truncated cube octahedral (TCO) shell. The basic TCO units arrange with respect to each other to form larger clusters. The structure determination was done mainly by high angle annular dark field scanning transmission electron microscopy (HAADF-STEM) imaging. The scanning (precession) electron diffraction (S(P)ED) technique makes it possible to do more quantitative studies and also get the structure of clusters in Al-Mg-Si alloys where Z contrast is much less. Doing SPED and quantitative convergent beam electron diffraction (QCBED) of the clusters, using our new Merlin direct electron detector, give promising results. This presentation will show our last work in combining advanced analysis of HAADF-STEM and S(P)ED to get more information about the early stage clusters in Al alloys [3].

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DEFECT STRUCTURE AND PROPERTIES IN CUBIC NITRIDES

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Abstract

Gallium nitride (GaN) is an immensely successful material for the fabrication of light emitting diodes (LEDs) for use in solid state lighting, but LEDs emitting in the green and amber spectral region still have poor efficiencies. Whilst the solid state lighting industry has gone from strength to strength despite these limitations, other potential applications of LEDs demand significant improvements to the efficiency of longer wavelength devices. The accelerating development of microdisplays for virtual and augmented reality (VR and AR) requires high brightness, high efficiency, fast-switching microLEDs. This need is not met even in the red spectral region where traditional III-V materials used for larger devices show poor performance at the required very small device sizes.

The low efficiency of green and amber LEDs based on GaN is partly due to the large internal electric fields arising in strained quantum wells, which reduce the radiative recombination rate. To eliminate these fields, non-polar nitrides have been considered, but the high indium contents required to achieve green emission in the wurtzite system have proved difficult to achieve. Metstable zincblende GaN in the ⟨001⟩ orientation provides an alternative route to non-polar, field-free quantum wells, and has a lower bandgap than wurtzite GaN, reducing the required indium content. However, challenges arise in achieving high phase purity and in controlling the formation of crystal defects. Stacking faults on the zincblende {111} planes are effectively monolayer insertions of the stable wurtzite crystal structure, with a low energy, making them very difficult to eliminate.

High resolution transmission electron microscopy on zincblende GaN nucleation layers allows us to deduce that stacking faults and their associated partial dislocations originate from the dissociation of perfect dislocations, and by direct nucleation of partial dislocations loops from the surface. The inclination of the stacking faults to the growth direction allows defects on different {111} planes to meet and annihilate, reducing defect densities through the thickness of the film, but in zincblende LED structures a significant number of stacking faults still intersect the InGaN active region. High resolution scanning transmission electron microscopy and energy dispersive X-ray spectroscopy reveal indium segregation adjacent to the stacking faults, and atom probe tomography shows that these indium rich regions form quantum wire structures. The emission from these quantum wires is highly polarised, which may have potential applications in the development of polarised LEDs for three-dimensional VR and AR displays.

ATOM-SCALE DEVICES MADE BY ELECTRONS

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Abstract

Electron beams can modify materials with atomic precision to make new devices with atom-scale components and control. I will describe TEM and AC-STEM-sculpted devices, characterization, and experiments where we push device functionality and the lower limits of device size [1-6]. Experiments are performed *in situ [4,5] and ex situ [1, 2, 7]*. *In situ* experiments include the study of electron flow (in metals and 2D materials including graphene, molybdenum disulfide, phosphorene), as a function of their structure as they are reshaped to zero width, and gated *in situ* [4,5]. *Ex situ TEM* experiments include all-electronic analysis of molecules by pushing them through tiny holes made by electron beams in the TEM and AC-TEM [2,9,10]. Single and multiple sub-nm holes in 2D membranes allow transport of ions and gas molecules through them and studies of fundamental physics laws at scale ~ 0.1 nm. Image analysis alongside AC-STEM allows correlated fabrication and analysis done simultaneously *in situ* to engineer atomic devices [7-10].

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UNCOVERING SOLID-STATE NUCLEATION MECHANISMS IN TITANIUM ALLOYS USING MATERIALS CHARACTERIZATION AT THE NANO SCALE

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Abstract

A number of nano-scale structural instabilities, which form in various metastable β titanium alloys, have been characterized. These include the athermal ω phase with hexagonal structure, O' phase with orthorhombic structure, O" phase with an ordered orthorhombic structure, and incommensurate modulated nanodomains. Importantly, it has been discovered that these instabilities may play a role in nucleation events in solid-state transformations in these alloys. Hence, these various nano-scale structural instabilities need to be taken into consideration when designing novel metastable β titanium alloys to optimize the mechanical performance by microstructure engineering. In this talk, these various metastable phases will be introduced, and the evidence for their role in solid-state nucleation in these alloys will be presented. A unique feature of metastable β titanium alloys is the ability to produce extremely refined distributions of the alpha phase in a beta matrix, which has very significant implications regarding mechanical properties. The underlying factors influencing the production of these very refined phase distributions have been determined and these results will be presented.

CORRELATIVE STEM-EELS AND RAMAN SPECTROSCOPY STUDIES TO OPTIMIZE GOLD NANOPARTICLES FOR EARLY CANCER DETECTION

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Abstract

There are various ways in which nanotechnology can assist in early cancer detection, oftentimes utilizing sensitive physical properties of nanomaterials to detect them attached to tumors or circulating cancer cells. The group with whom we collaborate has successfully employed gold nanoparticles contained within a silica shell as a triple modality detection agent [1]. One of the properties utilized is surface-enhanced Raman spectroscopy (SERS) whereby the gold nano-spheres significantly enhance the Raman signal from an organic dye when exposed to an illuminating laser beam, which has been incorporated into a working endoscopic system [2]. While this works well, to our knowledge there has never been any systematic study as to the influence of nanomaterial structure parameters such as size, shape, seperation, coating etc. on the strength of the Raman signal, and hence its utility in detecting small tumors. In this paper, we describe an approach to study the effect of these parameters in order to establish the optimum conditions to generate the highest possible Raman signal.

An array of gold nanoparticles of various size, shape and separation is fabricated from a vapor-deposited gold thin film utilizing standard electron lithographic processes. When a Raman dye is spread over the array, Raman imaging shows the variations of signal and hence the parameters giving rise to maximum signal. Nanoparticle size is seem to be a critical feature. The plasmon resonances and energies are then determined across the array using electron energy loss spectroscopy (EELS) in a scanning TEM (STEM) [3] and the individual spectra are then correlated with the Raman signal from the exact same nanoparticle structures. By this procedure we can establish the critical parameters which yield the highest Raman signal, which leads to the systematic design of the most effective SERS nanoparticles [4].

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ATOMIC-RESOLUTION STRUCTURE IMAGING OF ELEMENTAL AND COMPOUND SEMICONDUCTORS: DEFECTS, INTERFACES AND INTERMIXING

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Abstract

Elemental and compound semiconductors have band-gap energies that span wavelengths from far-infrared (IR) to near-ultraviolet (UV), keeping these materials in heavy demand for an ever-widening range of optoelectronic and photonic devices. With lattice parameters that range from 0.648 nm (CdTe) all the way to 0.357 nm (C), observations with aberration-corrected electron microscopes comfortably enable atomic-resolution structure imaging when viewed in low-index zone-axes. Thus, detailed atomic arrangements associated with many different types of structural defects can be directly obtained. Moreover, composition profiles on similar length scales can, in principle, also be achieved when using probe-corrected instruments, with the proviso that the analysis should not be limited by atomic displacements that might take place during the period of observation. This talk will provide an overview of atomic-resolution structure imaging (and some spectroscopy) of defects and interfaces observed in heterostructures consisting of elemental and/or compound semiconductors. The widespread nature of interfacial intermixing in these compound systems will also be emphasized.

Acknowledgments: The author gratefully acknowledges ongoing collaborations with the groups of Esperanza Luna and Achim Trampert (Paul-Drude Institut, Berlin), Christiana Honsberg and Yong-Hang Zhang (Arizona State University), and Jacek Furdyna and Xinyu Liu (Notre Dame University). Use of facilities in the John M. Cowley Center for High Resolution Electron Microscopy is also acknowledged.

DYNAMICAL SCATTERING IN PICOMETRE STEPS AND OTHER ADVENTURES IN 4D-STEM

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Abstract

The pioneering works of the distinguished scientists we are celebrating at PICO this year have led us to a wonderful new era in microscopy. As just one example, dynamical scattering, convergent beam electron diffraction, aberration correction and fast detectors underpin the power of the modern scanning transmission electron microscope (STEM) experiment.

In modern STEM, we now have exquisite control over each part of the experiment, from the form of the incident electron wavefield, to the angular resolution with which we can detect it after scattering within the specimen. This talk will explore how we can tune the the incident wavefield and specimen orientation to tailor the scattering within the specimen, so that specific information is imparted to the scattered electron wavefield. We can then collect that information with matching detector geometry using a fast, pixelated detector. In this way, we can generate images with contrast optimised to features of interest.

The talk will illustrate this approach with applications to understanding structure-property relationships in functional materials using data from a double-aberration-corrected FEI Titan^3 80-300 FEG-TEM fitted with an EMPAD pixelated detector.

DEVELOPMENT AND APPLICATION OF LASER ILLUMINATION SYSTEM INTEGRATED INTO TEM

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Abstract

Illuminating a specimen with a laser introduced into a TEM enables observation of photon induced phenomena at the nanoscale. Laser introduction by dedicated TEM holders has been used mainly for specimen heating [1]. However, the geometric constraints of such systems create limitations on specimen shape and orientation. In this study, we have developed a laser illumination system (Luminary Micro), which can focus the laser onto the specimen though the upper hole of the objective lens pole piece in a TEM column [2-3].

A laser optics unit, attached to an accessory port on the condenser lens section, magnifies, focuses, and directs the laser output onto the specimen along a path that is almost coaxial to the electron beam. The port is fitted with a vacuum-sealed glass window through which the laser beam is transmitted from the optical unit to the internal mirror that is installed between the objective lens pole piece and the illumination lens (condenser mini lens), and then directed onto the specimen. The operation and alignment of the laser is fully remote-controlled, and the laser focus position at the specimen can be precisely controlled and measured using a special alignment tool. The type of laser can also be changed according to the desired wavelength and power.

Using this newly developed system, experimental application data are demonstrated. Using a specimen in which multiple metal phases in WC/Co alloy with Ag/Cu wax which is tool steel, the melting phenomenon of each metal phase due to laser heating was observed. The power of the laser for heating a specimen can be adjustable by changing the laser output. As a result, each metal layer in the specimen was melted depending on the power of the laser. Even the WC area of the specimen changed its shape, indicating that the local heating temperature reached around 2800 degrees.

The Luminary Micro system enables us to use almost any kind of sample holder for heating specimens and/or analysing interactions between materials and lasers in TEM and STEM. The system allows these experiments be carried out using standard imaging conditions and specimen geometries. The system's integration with the TEM itself, enables advantageous compatibility with many holder types including: cooling holder, indentation holder, biasing holder, tension holder, high inclination holder, and more. This versatility means various material science experiments, that were previously not feasible, can now be realised. By combining the system with a cooling holder, it may be possible to realize a wide range of temperature conditions, from about -170 to several thousand-degrees.

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EXPLORING 2D MATERIALS WITH HIGH-RESOLUTION, HIGH SPEED, HIGH EFFICIENCY, AND LOW DOSE

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Abstract

The availability of aberration-correction, monochromation and high-acceptance angle spectrometers in state-of-the-art electron microscopes makes it possible to obtain information about the local atomic and electronic structure of the investigated sample with high spatial resolution and high energy resolution. The ability to detect both spectra and electron diffraction patterns with a fast direct electron detector allows every electron to be counted without any readout noise and with high efficiency, since comparatively few electrons are thrown away due to the use of apertures or lossy detection. In this presentation we will show how a Dectris ELA direct electron detector [1] mounted to a Nion HERMES microscope [2] is being used to acquire EELS spectra with up to 6 meV energy resolution and zero-loss filtered diffraction patterns at more than 15,000 frames per second, corresponding to an acquisition time of about 17 s for a map of 512 x 512 diffraction patterns or spectra. The 4D-STEM data sets are then used to recover the local electrostatic potential and charge density with deep sub-Ångstrom spatial resolution [3,4], while the EELS data - being highly quantitative after having removed relativistic contributions to the spectrum and recovering the local dielectric function [5] - is used to correlate local electronic structure with it, particularly at defects.

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SPECTRA-ULTRA, THE LATEST DEVELOPMENTS

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Abstract

Spectra, launched less than 2 years ago, has been very well received across the global scientific community, most probably because this highly versatile HR-(S)TEM system managed to achieve the highest commercially available STEM resolution using a wide analytical pole-piece (the S-TWIN with a 5.4 mm gap). This enables a variety of complex experiments like *in-situ*, 3D tomography and many other techniques that simply can't be made compatible with a dedicated narrow UHR pole-piece. Noteworthy is also that this statement of best resolution is true for all accelerating voltages between 30 and 300 kV.

Now major exciting new capabilities are added to the Spectra family with the launch of its latest sibling: The Spectra-Ultra. The "best resolution on a wide analytical pole-piece at all high tensions" is still the Ultra's mantra but now changing the accelerating voltage can actually be done in a matter of minutes instead of hours. Being able to operate the microscope at different high tensions during a single microscopy session opens new opportunities for optimization of the different experiments. Traditionally the acceleration voltage is optimized for the imaging mode and other signals such as EELS and EDX are then acquired at that same voltage, irrespective of the fact that EDX benefits from lower HT's, because of higher ionization cross sections and that EELS prefers higher HT's to avoid multiple scattering. As more materials classes are being investigated with electron microscopy, beam damage is increasingly becoming the (resolution) limiting factor and optimizing the accelerating voltage to the experiment will become the norm. With Spectra Ultra probe high tension becomes an adjustable parameter in the same manner that probe current is.

The same vision of making the best use of every electron that interacts with the sample also led to a revolutionary new EDX geometry yielding a massive 4.45srad solid angle. Compared to our trusted Super-X, Ultra-X is 6 times faster which is equivalent to say that similar EDX data can be obtained with just a sixth of the total dose and when combined with a lower accelerating voltage for EDX acquisition the minimum required dose can further be reduced. Results obtained with Ultra-X will be presented here and elsewhere [1].

Spectra-Ultra inherited the same Spectra family gun options: the X-FEG/Mono, X-FEG/Ultimono and X-CFEG. The "X" stands for extreme brightness. Even though brightness is a simple concept, measuring it and comparing specifications turns out to be tricky. Over time other approaches like "Coherent Current" have been proposed [2]. Here an approach based on combined probe resolution-current specifications will be presented.

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UNRAVEL LOW-DIMENSIONAL MATERIAL'S CONTRAST AND PROPERTIES

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Abstract

Two-dimensional materials exhibit properties, which can differ strongly from those of the bulk counterparts. They offer unique opportunities for new and miniaturized electronic and optical devices, which properties may even alter with changes of single atom's positions. *In situ* high-resolution TEM setups can simultaneously image and functionalize the material under study. Here we present recent results using the unique chromatic and spherical aberration-corrected *SALVE* instrument [1] and discuss contrast peculiarities. In EFTEM mode, allowing the use of most elastically and inelastic scattered electrons without the loss of beam intensity resulting from monochromation, we show using core-energy-loss electrons that elastic contrast is preserved even in images of a one light-atom thin sample – graphene [2].

Further we discuss the formation and identification of defects in a variety of two-dimensional van der Waals inorganic heterostructures and organic crystals [3 - 7]. We understand the role of electronic excitations on the example of MoS_2 imaged at voltages from 20-80kV and show that the formation of vacancies is possible at electron voltages nearly half of the knock-on threshold and quantify the damage. Moreover, our results from low-loss EELS and 3D diffraction provide insight into layer-number-dependent property changes which allow to exactly determine the numbers of layers in selected 2D van der Waals crystals. On the more fundamental base we show that differentiating between the bond nature of two metal atoms is now possible [8] and moreover show that the chiral structure of the SWNT can be utilized for assembling helical molecular motor applied as atom carrier for nucleation of metals confirming the two-step nucleation mechanism [9].

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RECENT ADVANCES FOR 4D STEM AND EELS DATA ACQUISITION: STELA AND K3

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Abstract

In today's real-world materials science problems, usually the solution comes from not only one, but the combination of multiple types of experiments and various sources of information; in the high-speed competitive design and production of new electronic devices at the nanometer scale, the ability to accurately measure the mechanical properties, and spatially align and correlate data from multiple sources is imperative to form a clear picture of the interfacial phases present. Scanning transmission electron microscope (STEM) diffraction imaging technique uses a parallel or convergent electron beam to generate a diffraction patterns at each scanning point; Using a high-speed CMOS camera, these two-dimensional diffraction patterns can be recorded for a two-dimensional (2D) STEM rastered area, which results in a 4-dimensional (4D) STEM diffraction data cube.

The introduction of hybrid-pixel electron detection technology (such as recently released Gatan Stela camera) allows acquisition of 4D STEM datasets, at low keV, with high speed. With the Stela camera, diffraction imaging and 4D STEM workflows in Gatan Microscopy Suite (GMS) are now fully integrated with hybrid-pixel electron detection. This single interface is not only optimized for data acquisition and processing but streamlines the workflow to shorten your time to results. Hardware synchronized 4D STEM eliminates data loss during acquisition, and pre-optimized 4D STEM tools along with DigitalMicrograph® and Python scripting allow for the highest quality diffraction data acquisition and processing, irrespective of the user level of expertise and complexity of their experiment. Stela utilizes a hybrid-pixel electron detector that employs electron counting to minimize noise and uses on-the-fly digitization for the highest dynamic range. Low-noise, high dynamic range, along with its fast frame rate (>16,000 pixels/s), and optimized performance at low kV (<80 keV), make Stela the best electron diffraction camera for a variety of advanced 4D STEM studies.

In the case of EELS data acquisition, we have discussed and proven previously that electron counting mode, using the K2 camera first and then the K3 camera, nearly eliminates the instrumental noise, while reducing point spread function (PSF). The ability of counting nearly eliminates all the instrumental sources of noise. This effectively leaves the shot noise as the limiting source of noise. In addition, the sharp PSF allows spectra to be acquired over larger energy ranges while maintaining energy resolution and greatly reduced spectral tails. The K3 camera is capable of counting electrons at a speed of 3000 spectra per second with increased dynamic range compared to its predecessor the K2. Other important capabilities such as DualEELS that were missing with the K2 camera are now available with the K3.

STRUCTURAL NANOTOPOLOGY AND 'MOLECULAR GYMNASTICS' OF HERPESVIRUS GLYCOPROTEINS

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Abstract

Herpesviruses are large (~200 nm diameter) enveloped DNA viruses. Herpes simplex virus type 1 (HSV-1), the prototype species of the alphaherpesvirus family, has been the first pleomorphic virus studied by electron cryo tomography (cryoET) [1]. Already this early cryoET study revealed the complexity of the viral envelope membrane being densely studded with a variety of glycoproteins, of which HSV-1 encodes at least 12 different kinds. Furthermore, they occupy different conformations, such precluding a comprehensive structural analysis of the dense 'forest' of viral glycoproteins and their relative nanotopology. HSV-1 glycoproteins play a crucial role in viral entry – the first step in the viral 'life' cycle, with four glycoproteins assumed to comprise the fusion machinery – as well as in virus assembly. X-ray crystallography of isolated glycoprotein ectodomains has provided significant insights into their structures. Complementary, we have used cryoET and a range of experimental investigation systems – from virus-cell interactions to the analysis of full-length glycoproteins embedded in native membranes – to provide structures in the functional context. Combining this with mutations informed by molecular dynamics simulations we have been able to stabilize the metastable and hence hitherto elusive prefusion conformation of glycoprotein B, the herpesvirus fusion protein, opening the path to new interventions [2]. Recently, we have introduced signpost DNA origami nanostructures (SPOTS) coupled to aptamers as a new tool for probing and surveying proteins on membrane surfaces [3]. We demonstrate that they can be used to confidently identify proteins of interest independent of their conformation and thus facilitate structure determination by cryoET sub-volume averaging. SPOT multiplexing provides a promising next path to chart the viral surface and to provide critically needed mechanistic level knowledge on the relative positions and functional clustering of viral glycoproteins.

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TOOLS AND TECHNOLOGIES FOR VISUAL PROTEOMICS WITH CRYO-ET – A SHORT RETROSPECTIVE OF 20 YEARS WITH WOLFGANG BAUMEISTER

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Abstract

Today, we see breathtaking results of isolated molecular structures with atomic or near-atomic resolution obtained by single-particle cryo-electron microscopy (cryoEM) on an almost daily basis. In fact, also " the virus" and its components that keep us from being present in person at this year's PICO meeting appeared weekly in more and more detail (and in color, of course) and even made it to a front page of the New York Times.

However, structures that are isolated from their environment can only tell part of the story. It is much more informative to study them in their native context within the cell and get a picture not only of the structure itself, but also of its associations and interactions with the cellular proteome (and thus of how "the virus" really does its malicious work). Doing this is called visual proteomics (who do you think coined that term?), and by using cryo-electron tomography (cryoET) we are able to get even down to molecular resolution. But there are still both bottlenecks and obstacles that hinder their application and, moreover, the throughput needed to better quantitatively describe what we visualize in tomograms. So, what are the key issues that need to be addressed? First and foremost is sample preparation. TEM, as we all know, requires electron transparency, and since resolution scales with thickness, we need to ensure that the best compromise is made between achievable resolution and volume to be studied. It can be said that focused ion beam (FIB) milling of frozen hydrated samples is nowadays the method of choice to obtain such thin samples from cells that can be frozen directly. However, larger samples, like tissues that need to be frozen under high pressure, are the most interesting. Here, preparation is still in its infancy and alternatives need to be explored, some of which we will cover in this talk.

Wouldn't it be extremely cool to actually take a biopsy and examine it directly via cryoET? But a microscope that allows us to first distinguish between healthy and diseased cells and then image specific areas of interest with molecular resolution, that is, to bridge all scales without any preparation, is science fiction. Nevertheless, we have combined several techniques and methods into a workflow that could make this at least partially possible: seamless navigation with cryoCLEM, lift-out lamella preparation with a cryoFIB, and imaging with cryoET.

This talk will hopefully give you a glimpse into the beautiful (and colorful) world of cryoET and visual proteomics, and a true picture of how hard and cumbersome it can be, what hardware and software solutions are needed that make it happen, and what our prospects are for the future. It will also feature important milestones ("from formerly uncharted territories") in my 20-year long journey of working with Wolfgang Baumeister.

LAMIN FILAMENTS, MUTATIONS AND THE NUCLEAR LAMINA

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Abstract

The nuclear lamina, mostly composed of intermediate filaments proteins termed lamins, is primarily responsible for the mechanical integrity of the nuclei in multicellular organisms Lamins, are also involved in many nuclear activities. Structural analysis of nuclei revealed that lamins form 3.5 nm thick filaments often interact with nuclear pore complexes. Point mutations in the *LMNA* gene, encoding A-type lamins, have been associated with at least 15 distinct diseases collectively termed laminopathies, including muscle, metabolic and neurological disorders, and premature aging syndrome. Although we have previously provided a view on lamin organization, we still lacking a high-resolution structure of lamin filaments. This is a fundamental information that is needed in order to understand how point mutations in lamins cause distinct diseases, laminopathies. Taking the advantage of recent developments in cryo-ET, we looked into the organization of in situ assembled nuclear lamins, in health and disease. The organization and assembly of lamin filaments will be discussed.

ELECTRON CRYO-MICROSCOPY OF BIOMOLECULES AT ATOMIC RESOLUTION

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Abstract

Single particle electron cryo-microscopy (cryo-EM) is a mature methodology for routine structure determination with detailed features equivalent to those obtained by X-ray crystallography at comparable resolutions. There are more than 4,000 cryo-EM structures better than 4 Å resolution deposited to the protein databank as in early 2021. Cryo-EM has the resolving power to visualize atomic details of biomolecules. We utilize standard 300 kV transmission electron microscopes (Titan Krios) to image macromolecular complexes in our Stanford-SLAC Cryo-EM Center. We have found to be readily feasible to record sufficient images of vitrified apoferritin in less than ¾ a day for reconstructing its structure at ~1.27-1.34 Å resolution with either a K3 camera with an energy filter or a falcon 4 detector without an energy filter [1]. A quantitative validation analysis of the maps substantiates the resolvability of all atoms except hydrogen in all the amino acids, water molecules and metal ions. Such capability is not always achieved for all macromolecules because of their compositional and/or conformational heterogeneity. Nevertheless, advanced data processing method can be used to sort out the structure variants of many macromolecular complexes such as membrane channel [2] from which novel chemical properties of the macromolecules can be derived at near atomic resolution.

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COHERENT AND INCOHERENT IMAGING OF BIOLOGICAL SPECIMENS WITH ELECTRONS AND X-RAYS

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Abstract

Since radiation damage is proportional to fluence, radiation damage limits the spatial resolution of biological structures determined by either X-ray or electron scattering. If only elastic scattering is used for structural information then electrons are superior as the ratio of elastic to energy deposited by inelastic scattering is higher than for X-rays. For soft X-rays in the water window below the O K edge photoabsorption contrast might be better than elastic scattering for distinguishing different biological materials. Phase contrast elastic scattering is most effective in the hard X-ray region up to about 10 keV. Radiation damage limits spatial resolution for most X-ray imaging to 10-20 nm. Local molar concentrations of Na, K and Ca ions can be determined at somewhat lower spatial resolutions using relevant absorption edges. At higher energies Compton scattering is superior and is only limited by the fluence available from the light source, since energy deposition is small.

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ATOMIC RESOLUTION STRUCTURE DETERMINATION BY CRYO-EM – WHERE ARE THE LIMITS?

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Single particle cryo electron microscopy (cryo-EM) has developed into a powerful technique to determine 3D structures of large macromolecular complexes. Due to improvements in instrumentation and computational image analysis, the number of high-resolution structures is steadily increasing. The method cannot only be used to determine high-resolution structures but also to study the dynamic behavior of macromolecular complexes and thus represents a very complementary method to X-ray crystallography. Furthermore, the maximum attainable resolution by cryo-EM has constantly improved in recent years. Most of the high-resolution structures are still in the 3 Angstrom resolution regime but some have even crossed the 2 Angstrom barrier. We have recently installed a new prototype electron microscope which is equipped with a monochromator and a next-generation spherical aberration corrector. This microscope is optically superior to the currently commercially available instruments and can therefore be used to test the resolution limits in cryo-EM. We have used the test specimen apoferritin to determine its structure at 1.25 Angstrom resolution [1] which is sufficient to visualize for the first time individual atoms clearly separated in the density map.

Recently, we managed to use this microscope not only to improve the resolution of the very stable and rigid protein apoferritin. We also obtained significant improvement in resolution for other more dynamic macromolecular complexes for which one could have expected that the microscope itself may not be a major resolution limiting factor.

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EFFICIENT SAMPLE SCREENING AND AUTOMATION-SETUP IN CRYO-EM

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Abstract

One of several important parameters for success in cryo-EM experiments is imaging particles embedded in the right ice-thickness. Considering that the current plunge-freezing methods still fall short of delivering consistent ice thickness over multiple frozen grids, each automated acquisition session in cryo-EM still requires a considerable amount of time finding the optimal regions on the cryo-EM grid to collect data from. Several strategies are explored to speed this process up, without the need of offline image processing.

SCANNING TRANSMISSION ELECTRON MICROSCOPY FOR CRYO-PRESERVED SAMPLES IN LIFE SCIENCES AND MATERIALS SCIENCES

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Abstract

The high vacuum environment and the charged irradiation in a TEM require a careful stabilisation of many specimens of interest. Cryogenic fixation provides an ideal matrix for radiation sensitive samples in which the organic macromolecules, biological samples or inorganic compounds are suspended without the need for dehydration, ideally in a suddenly arrested state frozen in time and temperature. While conventional cryo-TEM has possibly reached the limits of physical optics in the quest for the structure refinement of proteins at high resolution, the flexibility of STEM detection modes can enhance contrast for specific features or optimise the sensitivity to the specimen. A flurry of STEM techniques ranging from imaging to nanobeam diffraction open most exciting possibilities for cryo-observation at low doses.

We describe our efforts so far to introduce STEM for cryo-microscopy and tomography of biological specimens, organic crystals, hybrid organic-inorganic materials, biominerals and for colloidal chemistry. Application examples range from mass density measurements over thick sample tomography to high-resolution detection of single metal atoms that are embedded in macromolecules and surrounded by vitreous ice. For the detection of metal ions or small metal clusters ADF-STEM offers high sensitivity. Contrary, the bright-field has plain benefits for thick specimen tomography, demonstrated for cell biology, for the three-dimensional imaging of crystallisation dynamics of protein molecules in solution or organic/inorganic nanoparticle superlattices in colloidal chemistry.

New approaches based on pixelated and multi-segmented STEM detectors for low-dose imaging are under development. On one hand, phase contrast methods, such as ptychography and variations of differential phase contrast with segmented detectors, will be essential to enhance the high resolution information in STEM. On the other hand, the additional momentum resolution of segmented detectors will improve depth of field over a large field of view while maintaining spatial resolution, vice versa, and will therefore be particularly suited for tomography of thick samples. Facilitated autofocusing schemes enhance the throughput and dose efficiency of automated tilt series recording. Our recent advances towards higher throughput in cryo-STEM tomography, based on a multi-segment solid state detector, a custom implementation of a flexible scan generator, and a scalable eight-channel array of non-multiplexed digital to analog converters, will be reported. Together with an integration into the SerialEM platform we leverage the capabilities of the multi-channel setup for automated workflows.

ZOOMING IN ON CELLS AND MOLECULES WITH CORRELATIVE LIGHT AND ELECTRON TOMOGRAPHY

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Abstract

In correlative light and electron microscopy (CLEM), imaging modalities are combined to study cellular processes. Fluorescence light microscopy enables the imaging of dynamic events in relatively large fields of view while electron microscopy can reveal structural molecular arrangements in relatively narrow fields of view. During the last decade a variety of methods to image the 3D morphology of cells and tissue were realized. These approaches resolve interpretable morphological details in the 2-10 nm range, providing a wealth of information on the cellular architecture and processes. Examples of these methods include array tomography, serial section block-face scanning electron microscopy as well as dual-beam scanning electron microscopy. More recently, dedicated approaches for 3D CLEM that are compatible with these methods were developed, providing practical ways to target structures in a cell or tissue.

For imaging structures with molecular resolution, specimens need to be vitrified to ensure structural preservation, effectively limiting the size of the biological system that can be studied to 200 μm for specimens prepared with high pressure freezing, and to 5-10 μm when using plunging freezing. In addition, for high resolution imaging with tomography, a thin lamella needs first to be extracted from the cellular volume using focused ion beam milling. During the last few years, the required instrumentation matured, effectively transforming this challenging workflow into a practical approach to perform structural biology studies in the cellular context. Currently, novel methods and technologies are proposed, aiming at targeting and imaging molecular structures in their cellular context using a combination of CLEM and cryo electron tomography.

Here we will discuss the multi-scale light and electron microscopy methods that we have developed, explored and used, in the framework of studying the biology and mechanisms related to intracellular virus-induced replication organelles. The methods include volume imaging with TEM and SEM [1] and super resolution cryo CLEM [2]. We will also discuss our recent discovery of a molecular pore in the coronavirus replication organelle, for which the use of dual-beam focused ion beam milling in combination with cryo electron tomography and subtomogram averaging was vital [3].

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STRUCTURAL INSIGHTS INTO PROTEIN BIOGENESIS AT THE ENDOPLASMIC RETICULUM

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Abstract

A large fraction of protein synthesis and further protein biogenesis occur at the Endoplasmic Reticulum (ER). The ER is the starting point of the secretory pathway, which is utilized by approximately 30% of the human proteome. The majority of these protein are co-translationally inserted or translocated across the ER membrane by the ER translocon complex. Protein biogenesis in the ER involves additional processes, many of them occurring co-translationally while the nascent protein resides at the translocon complex, including recruitment of ER-targeted ribosome-nascent-chain complexes, glycosylation, signal peptide cleavage, membrane protein topogenesis and folding. We have dissected this machinery in the native membrane by cryo-electron tomography, providing insights into signal-peptide (SP) accommodation and glycosylation. Using cryo-EM single particle analysis we reveal the molecular determinants for subsequent cleavage of the SP by the signal peptidase complex (SPC). Our studies highlight the integration of *in vivo*, *ex vivo* and reconstitution approaches to obtain mechanistic insights into cellular processes by cryo-EM approaches.

EXPLORING THE STRUCTURE AND FUNCTION OF MICROBIAL MOTILITY: CHEMOTAXIS AND INTERMICROBIAL TRANSPORT

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Abstract

Most motile bacteria contain a highly sensitive and adaptable sensory system composed of clusters of chemoreceptors. This chemosensory system is used to detect changes in nutrient concentrations, allows the cells to navigate towards preferential environments and is also involved in host infection by some pathogenic bacteria. While it is one of the best understood signaling systems to date, unraveling structure and function of the bacterial chemotaxis system remains challenging. High-resolution analysis using methods are inherently limited to structural fragments and rely on specimens taken out of their natural environment. Thus, they lack the larger context of the native system. Here, we use cryo-electron tomography (cryoET) to study the three-dimensional architecture of the bacterial chemoreceptor arrays. We identified species-specific characteristics of this system of selected pathogens, such as in Treponema denticola (periodontal disease) and Vibrio cholerae (cholera). However, not all bacteria are motile, but can still benefit from chemotactic behavior: we have gained insight how non-motile bacteria can benefit from chemotactic microbes that share the same environment.

ATOMIC STRUCTURE AND FE-SEGERAGTION IN Σ13 [0001] TITANIUM TILT GRAIN BOUNDARIES

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Abstract

Grain boundaries (GBs) due to their five degrees of freedom have a large parameter space and are therefore difficult to be completely (structurally and chemically) characterized. However, the advent of aberration corrected scanning transmission electron microscopy (STEM) has revolutionised this field. Recently, direct observations of complex GB structures have been reported [1]. Analytical techniques like energy dispersive X-ray spectroscopy (EDS) and electron-energy loss spectroscopy (EELS) allow for chemical characterization down to atomic scale. Moreover, correlative microscopy with combination of STEM and atom probe tomography (APT) have been used to quantify GB segregation and thereby calculate GB energy [2]. However, GB structures in titanium (Ti) and other commercially important hcp metals are scarcely explored. The hcp-bcc allotropic transition in Ti makes fabrication of its bicrystals to form specific GBs particularly challenging.

Therefore, we present novel template based thin film deposition pathways for obtaining columnar grains containing tilt GBs of Ti using high power pulsed magnetron sputtering on MgO and $SrTiO_3$ substrates. Electron backscatter diffraction in a scanning electron microscope is used to investigate the texture evolution and global GB character. Films deposited at 600□C predominantly have (0001) basal plane texture. Interestingly, most GBs have □13 (27.8□ / [0001]) coincidence site lattice (CSL) orientation with maze-like GB planes. STEM imaging of these GBs lifted-out site specifically using focused ion beam reveals the seemingly curved GBs to be faceted into segments that are 20-50 nm long. Most of these facets are symmetric □13 {25-70} with ~5% asymmetric {10-10}/ {11-20} segments. Additionally, the structural units that constitute the GB are observed using atomic resolution STEM and are found to be partly consistent with theoretical predictions [3]. The structure also matches well with the 5-7-5 structural units that are observed in hexagonal 2D materials [4]. STEM-EDS revealed preferential segregation of Fe to specific {25-70} facets, due to a difference in coincident site lattice density. Atomically resolved EDS shows that Fe atomic columns are trapped in a cage-like Ti surrounding. The influence of Fe segregation on the atomic structure and migration of the GBs will be discussed.

Authors acknowledge the funding from KSB Stiftung.

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INTERFEROMETRIC STEM FOR PHASE IMAGING AND INTERACTION-FREE MEASUREMENTS

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Abstract

Imaging individual beam-sensitive materials, such as biomolecules, at atomic resolution with negligible damage has been a long-standing goal in electron microscopy, but it necessitates a significant dose decrease from current methods. The Elitzur and Vaidman quantum bomb detector[1] is an interferometric quantum protocol proposed to reduce interactions in electron microscopy[2,3]. Specialized apertures featuring holographic beamsplitting gratings are used to implement interferometry inside a scanning transmission electron microscope (STEM)[4,5]. STEM-holography uses a single phase grating to diffract the electron beam into two paths, which traverse the specimen plane, recombine and interfere. With a probe corrector, the separated electron paths can be focused to Angstrom lengthscales[4]. The aberrations can also be removed holographically[6]. Here we present a 2-grating interferometer that recombines the beams post-specimen, forming a scanning 2-grating electron Mach-Zehnder interferometer (2GeMZI). To highlight the capabilities of the 2GeMZI, we use it to efficiently make interaction-free measurements of an opaque sample with electrons[7] and achieve quantitative phase imaging of a latex nanoparticle. Interaction-free measurements involve a single input quantum traversing two paths of an interferometer and self-interfering destructively at an output. Obstructing one path disrupts the interference and results in an *increase* in events at the output. Our 2GeMZI features two nanofabricated diffraction grating holograms[8], functionally used as beamsplitters, aligned such that the output destructively interferes. To demonstrate an interaction-free measurement with electrons[7], we insert an aperture edge to block one interferometer path. This wavefunction disruption causes an increase in output events: the signature of the interaction-free measurement. We also employ the scanning capabilities to image a latex nanoparticle on graphene, recovering the phase with a resolution of σ_{ph}=200 mrad. The 2GeMZI is particularly effective due to the abilities to tune path separation, arbitrarily apply phase shifts between paths, access individual paths, scan the probes, and provide real time phase information, all while using a conventional bright field detector. Realizing interaction-free STEM will advance low-dose imaging of radiation-sensitive samples.

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DEFECT DYNAMICS AND SELF-HEALING IN LEAD-FREE DOUBLE PEROVSKITE NANOCRYSTALS

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Abstract

Self healing materials are capable of restoring their original structure and functionality after exposure to a damaging process. Semiconductors rarely exhibit such properties, and accumulation of structural defects typically lead to degradation of materials and material performance. High efficiency of metal-halide perovskite devices is assigned to defect tolerance and facile annealing. Here, we report defect tolerance and crystal structure self-healing in double perovskite nanocrystals. In situ TEM experiment is utilized to create void defects in perovskite nanocrystals, and capture their dynamics and structural self-healing in real-time. We discover an inaccessible surface volume for the voids, limiting their dynamics to the inner core crystal volume. Removal of the surface ligands resulted in relaxation of this constrain, outer diffusion of the voids, and a self-healing of the double perovskite crystal lattice. We measure the influence of ligand passivation on void diffusion, going down several atomic layers below the surface. This study provides a first real time observation to self-healing and reconstruction processes of perovskite crystals.

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IN-SITU ATOMIC-RESOLUTION TEM STUDY OF PHASE-TRANSITION AND ENERGY-STORAGE PATHWAY IN ANTIFERROELECTRIC $PBZRO_3$

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Abstract

Benefit from the ultrafast charging/discharging rates, superior reliability, high voltage endurance and broad operating temperature,[1,2] dielectric-based electrostatic capacitors are serving as critical components for many electrical power systems such as wearable/portable electronics and collection of fluctuating energy sources. *In situ* investigation of structure-property relationship offers a promising approach for comprehending the structural features that are responsible for the energy storage behavior. However, the ultrafast charging and discharging processes that finished in milliseconds or less in normal parallel-plate capacitors, bring great challenges to resolve the nature of the phase-transition process.[3,4] Here, we report an *in-situ* atomic-resolution transmission electron microscopy (TEM) study of the energy storage process in antiferroelectric (AFE) $PbZrO_3$ by using electron beam irradiation as an external stimulus. By imaging light oxygen and heavy atoms using negative spherical aberration imaging (NCSI) technique, our study reveals that the unit-cell volume shrinks and then expands during the orthorhombic (AFE_O)-to-monoclinic (FE_M) and FE_M-to-rhombohedral (FE_R) phase transitions. In particular, quantitative tracking of oxygen octahedral rotation reveals a novel FE-ferrodistortive transient phase,[5] which is characteristic of Pb antiparallel displacements and a cycloidal polarization order, and bridges the AFEO and FEM phases. In oxygen-and-Pb deficient $PbZrO_3$,[6] our time- and atomic-resolution study further reveals point-defect-induced unit-cell-wise energy storage pathway. These results point out a new territory for exploring exotic ferroelectric phases in nonpolar dielectric materials and offer a straightforward approach for visualizing energy storage process in defect-engineered dielectric ceramics.

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PTCU OCTAHEDRAL ELECTROCATALYST FOR PROTON EXCHANGE MEMBRANE FUEL CELL: STRUCTURE-PERFORMANCE-DURABILITY STUDY

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Abstract

The use of PtCu nanoparticles in electrocatalysis has been reported in the last few decades for several oxidative reactions. However, the application of oxygen reduction reaction (ORR) for proton exchange membrane fuel cell (PEMFC) operation was limited by the insufficient catalytic activity achieved, and complications of the stability of the material. The main challenge is to reduce the loading of platinum meanwhile increasing the efficiency and durability of the Pt-based catalysts. We have designed, synthesized, and characterized a Pt-based nanomaterial that shows impressive efficiency towards ORR, 0.767 $mAcm^{-2}_{Pt}$ and 0.58 Amg^{-1}_{Pt} the specific and mass activity respectively (4.5 and 3.3 higher than the Pt/C-JM benchmark) that not only preserved, but increase the electrocatalytic activity after 10,000 cycles under accelerated degradation test (ADT) acidic conditions. After ADT, hollow octahedral Pt-rich nanoparticles achieved 1.02 $mAcm^{-2}_{Pt}$ and 0.64 Amg^{-1}_{Pt} toward ORR.

PtCu nanoparticles were prepared via solvothermal synthesis[1], which subsequently revealed octahedral morphology with Pt-rich skin enhances the electroreduction on its surface. We use advanced electron microscopy techniques to understand the stability process. We found a relationship between the particle structure and the electrochemical performance. The sample was dealloyed via an electrochemical etching process, revealing Pt-rich hollow octahedral morphology in all the cases. The durability of the catalyst in acidic medium has been investigated for diverse conditions including, under oxygen and argon saturated atmosphere and different potential ranges. We track the structure evolution of the nanoparticles by several analytical and electron microscopy techniques such as identical location transmission electron microscopy. The electrochemical performance was analyzed by using the rotating disk electrode technique.

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SINGLE-SHOT AUTOMATIC RECOVERY OF DISLOCATION 3D STRUCTURES IN TEM AND QUANTIFICATION OF THE RECONSTRUCTION

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Abstract

Stereo imaging in scanning transmission electron microscopy (STEM) combined with reconstruction methods applied to dislocation structures is nowadays a relevant topic in the field of electron microscopy and material science [1]. It allows to reveal the 3D distribution of the curvilinear crystal defects by acquiring just two images under different viewing angles [2]. The recent development of the "tilt-less" imaging technique advanced further the possibilities of the 3D stereo imaging [3]. It allows single-shot simultaneous acquisition of stereo-pairs, eliminates lateral shifts and significantly decreases the imaging time. On the other side, currently existing post-processing algorithms for 3D reconstruction remain manual and time-consuming.

Modern deep learning technologies enabled us to develop fast and automatic method for 3D reconstruction of STEM stereo pair images of dislocations [4]. With our 3D contour matching convolutional neural network (CNN) we can trace and match dislocations from both views and obtain 3D reconstruction without any prior information about the dislocation structure. We successfully applied this approach to the STEM images acquired simultaneously with the "tilt-less" technique on a segmented annular STEM detector.

To optimize the imaging conditions for dislocation detection and the 3D reconstruction by CNN we present tools from Topological Data Analysis (TDA) to analyze the topological correctness of the outputs and compare then to the ground truth. We use the topological descriptors [5] to build topological distances between the outputs with different imaging conditions (detector acceptance semi-angle and deviation parameter, $\mathbf{s_g}$) and the ground truth and choose the settings with the highest topological correctness.

Our novel and fully automatic approach for 3D stereo reconstruction demonstrated high precision and allowed to significantly reduce the time of the experiment and post-processing compared to the conventional tomography.

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THREE-DIMENSIONAL CHARACTERIZATION OF NANOPARTICLES WITH SIMULTANEOUSLY ACQUIRED BRIGHT-FIELD, DARK-FIELD AND SECONDARY ELECTRON SIGNALS IN STEM

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Abstract

As a result of the importance of nanoparticles for applications that include catalysis and plasmonics, their three-dimensional (3D) characterization is crucial. Nanoparticles are commonly studied using electron tomography in the transmission electron microscope (TEM), which is based on the acquisition and analysis of two-dimensional projections of a three-dimensional object viewed from different directions in transmission geometry. However, some nanoparticles lack mirror symmetry, making it desirable to use a technique that is capable of studying their surfaces precisely. Although secondary electrons can be used to provide topological information, conventional secondary electron microscopes typically have a spatial resolution that is too poor to characterize the surfaces of the smallest nanoparticles.

In this work, bright-field (BF), dark-field (DF) and secondary electron (SE) signals were recorded simultaneously during tilt series scanning TEM (STEM) of a chiral gold nanoparticle [1] using a Cs-corrected HF5000 STEM (Hitachi, Japan) operated at 200 kV. In total, 121 images were acquired over a sample tilt range of -60° to +60° with a tilt increment of 1° without dynamic focus. Tomographic reconstruction of the BF and DF images was performed using a SIRT algorithm with the ASTRA toolbox, while STEM SE images were reconstructed using freely available photogrammetric reconstruction tools [2,3].

The benefits of comparing the three imaging modes for tomographic reconstruction will be discussed as an approach for interpreting both volumetric and surface features [4].

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INTRODUCING PHYSICAL MODELLING IN NMF DECOMPOSTION OF EDXS SPECTRUM IMAGES

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Abstract

Energy Dispersive X-ray Spectroscopy (EDXS) spectrum imaging is a useful tool for local chemical analysis. In many cases, the studied materials are composed of several spatially overlapping phases. To perform a local chemical quantification, one has to retrieve the pure spectra of each phase. Machine learning algorithms such as vertex component analysis [1] or non-negative matrix factorization (NMF) [2] are especially suited for this unmixing task as the EDXS spectrum images are noisy. However, these algorithms do not include any prior knowledge on the physics of EDXS. Therefore, the obtained decomposed "spectra" are rarely estimated correctly, thus, preventing a chemical quantification.

To overcome this issue, we propose an algorithm similar to NMF but based on a physically informed factorization model. The data are approximated by the product *DA* with $D = GP + B$. The columns of *GP* are the phase specific linear combination of the spectra of each chemical element that could appear in the phases. *G* is the matrix whose columns are the tabulated characteristic X-rays lines for each element [3,4], and *P* the matrix whose columns are the amount of each element in the phases. B is the matrix of the phase specific continuum X-rays [5,6]. The phase abundances for a given pixel are contained in the columns of *A*. Given that the abundances are proportions, we constraint *A* to be non-negative and each column to sum to 1. Similarly, P is constrained to be non-negative. EDXS data are well approximated by a Poisson distribution, we use a Poisson likelihood to tie the model and the observations.

To test the algorithm, we use an EDXS physical models, based on a real-case sample, to construct artificial data that closely resembles experimental data. Specifically, a mixture of spatially overlapping bridgmanite ((Mg,Fe)SiO3, Brg), ferropericlase ((Mg,Fe)O, Fp) and/or Ca-rich perovskite (CaSiO3,Ca-pv) [7] was designed. This unmixing problem is challenging as the phases share some chemical elements. The performance of our algorithm is compared to that of NMF by calculating the angles between the reconstructed phase spectra and their ground truths. In this quantitative comparison, NMF yields 7.8°, 15° and 9.1° for Brg, Fp and Ca-pv, respectively, while with our algorithm we obtain 7.7° for Brg and 7.2° for the other phases.

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ATOMIC STRUCTURE AND ELECTRON MAGNETIC CIRCULAR DICHROISM OF INDIVIDUAL ROCK SALT STRUCTURE ANTIPHASE BOUNDARIES IN SPINEL FERRITES

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Abstract

The antiphase boundaries (APBs) that are commonly observed in spinel ferrite films hinder their applications as high frequency inductors and microwave and spintronic devices, as a result of their influence on the physical properties of the materials. However, it is challenging to correlate magnetic properties with atomic structure at individual APBs due to the limited spatial resolution of most magnetic imaging techniques. Here, we use aberration-corrected scanning transmission electron microscopy and electron magnetic circular dichroism (EMCD) technique to measure the atomic structure and magnetism of a single APB in $NiFe_2O_4$. Intensity analysis of APB in high angle annular dark-field scanning transmission electron microscopy images suggests a new type APB with rock salt structure interlayer in $NiFe_2O_4$ with a relative translation of (1/4)*a*[011]. High-spatial-resolution EMCD have been used to experimentally demonstrate reductions of 46.8%±8.2% and 38.8%±14.5% in Fe and Ni EMCD strengths in APB, respectively, compared to perfectly ordered $NiFe_2O_4$. DFT calculations and dynamical diffraction calculations suggest that the reduced EMCD strengths result from the fact that Fe ions at the APB interlayer are antiferromagnetically coupled with each other, whereas Ni ions show a significant decrease in magnetic moment. Our combined approach of using element specific EMCD under high-spatial-resolution and first-principles calculations is applicable to studies of a broad spectrum of other defects in magnetic materials.

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SCHLIEREN IMAGING OF MAGNETIC FIELDS WITH HOLLOW-CONE ELECTRON BEAMS

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Abstract

We have been using the recently developed hollow-cone Foucault imaging method [1, 2] for studying magnetization structures, such as magnetic domains and domain walls. The hollow-cone Foucault imaging has two modes depending on the inclination angle of the illumination beam, bright-field Foucault mode and dark-field Foucault mode; Angle inclination is performed by using deflectors above the specimen and by inclining electron beam circulating with respect to the optical axis. The advantage of this imaging method is that it can simultaneously visualize both magnetic domains and magnetic domain walls under the infocus condition.

We have also found a specially controlled mode, a schlieren imaging mode [3], between these Foucault modes. Although the schlieren imaging is a method developed in the middle of the 19th century to qualitatively visualize distributions of refractive indices of transparent media, such as air, it is still used today as a method for visualizing the difference in refractive indices [3, 4] and also for visualizing the sound shock waves generated by high-speed flying objects [5].

In this study, we have extended the schlieren imaging method to directly observe magnetic fields outside specimens by using the hollow-cone illumination in 200-kV transmission electron microscope (JEM-2100F) [1]. The specimens were $Fe_{0.88}Ga_{0.12}$ alloys (at%) having large magnetostriction at room temperature [6]. Their thin films (250 nm in thickness) were prepared by a focused ion beam instrument (NB-5000).

To date, we have obtained only qualitative data on magnetic field leaking out to the vacuum area from the specimen. We are working on further developments of the hollow-cone schlieren imaging method to obtain more quantitative and clearer magnetic field characteristics in the vacuum area.

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MEASURING THE MEAN INNER POTENTIAL OF BERNAL GRAPHITE USING OFF-AXIS ELECTRON HOLOGRAPHY

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Abstract

The mean-inner-potential (MIP) of a crystal is the average electrostatic Coulomb potential within a crystal with respect to vacuum. The MIP is a fundamental material property which reflects chemical-bonding and crystallographic surfaces [1]. We conducted off-axis electron holography experiments on highly-oriented-pyrolytic-graphite (HOPG) in a transmission-electron-microscope to measure the MIP from nanometer-scale volumes of Bernal graphite oriented with respect to the electron beam, along the principal axis or directions in the basal plane. These MIP were related to mean orbital electron radii and diamagnetic susceptibilities in perpendicular planes. Such intrinsic property measurements are challenging because of defect induced interfaces at basal planes [2, 3]. Indeed, our structural examination of HOPG show stacking faults and planar rotations around the principal-axis, such that measuring intrinsic properties requires probing a volume of $\sim 102\times102\times102 nm^3$. Experiments on individual Bernal graphite crystals with (0001) basal, or (1-100), (2-1-10) prismatic planes, resulted in MIP of 10.16±0.40V, 11.37±0.35V, 12.66±0.41V, respectively [4]. First-principles calculations from crystalline slabs [4] confirm these anisotropic measurements with 11.72V, 13.65V, 14.56V, respectively. Additionally, these experiments enabled to measure the mean free path for inelastic scattering in graphite of 197keV electrons at a collection angle of 18mrad resulting in 150.6±2.0 nm. These measured MIP enable to determine projected mean radii of electron orbitals and volume susceptibilities (SI), assuming spherically symmetric charge distribution, at 0.704±0.015Å, $(-1.99\pm0.08)\times10^{-5}$; 0.744±0.015Å, $(-2.23\pm0.07)\times10^{-5}$; 0.785±0.015Å, $(-2.48\pm0.08)\times10^{-5}$. The measured orbital radii and diamagnetism in the basal plane are comparable to expected values for carbon σ-bond hybridization [5-6]. Increased MIP on prismatic planes is related to s-orbital components, which decrease due to delocalized electrons between basal planes.

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ABOUT THE ATOMIC STRUCTURE OF DIFFERENT PHASES WITHIN ONE GRAIN BOUNDARY IN PURE CU

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Abstract

Grain boundary engineering is a wide research field in which grain boundaries (GBs), the interfaces between two crystals, and their impact on materials properties are studied. Most research focus on adjusting the materials microstructure e.g. by changing the amount of specific GBs as twin GBs to improve the materials properties. However, only few experimental studies investigate the atomic structure of different high angle GBs, which can describe a GB and is, together with its thermodynamic properties, finally responsible for the impact of a GB on materials characteristics. For each specific GB, not only one but multiple stable and metastable states can occur [1]. Thus, by heating or applying strain, GBs can undergo phase transformations and co-existences of different GB phases, also termed "complexion", may be possible [2].

In our study, we investigate a ∑37c $[1\,1\,\bar{1}]\{1\,10\,11\}$ symmetric tilt GB in copper using aberration-corrected HAADF STEM. Two different phases, termed "domino" and "pearl", are observed, connected by a phase junction. Whereas the "domino" phase consists of a structural unit which is every second time rotated parallel to the GB plane, the "pearl" phase can be described by three subunits. Both atomic structures will be discussed in detail. Having a look at areas with a length of 300nm, we could make out multiple phase junctions, which separated 10 to 40 nm long "pearl" phase segments from 20 to over 100 nm long "domino" segments. According to our molecular dynamics (MD) simulation, the "domino" phase has a lower GB energy at room temperature compared to the "pearl" phase. However, at elevated temperature, the quasi-harmonic approximation approach reveals a phase transformation under ambient conditions, so that the "pearl" phase is energetically favorable. As the experimentally investigated sample was exposed to a heat treatment (400°C for 3h), the "pearl" phase may have been formed at elevated temperatures and got kinetically trapped during cooling. Both phases are still observed when the GB plane inclination varies up to 11° off the symmetric case, showing that a phase transformation is not limited to a specific GB configuration.

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HAADF STEM SIMULATION: A KEY TO SOLVE A METAL COMPOUND STRUCTURE

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Abstract

Transmission electron microscopy image simulation is a powerful tool to support image interpretation, and eventually solve materials science puzzles. In the present work we tackled the crystallographic structure of a ternary metallic compound made of calcium, magnesium and zinc, whose identity is disputed since some time. Postulated to be a trigonal phase $Ca_2Mg_6Zn_3$ in 2002 on the base of SADP and diffraction simulations [1], many other structures have been proposed since then. In 2016 we proposed a new phase, a hexagonal $Ca_2Mg_5Zn_5$, on the base of X-rays and TEM diffraction [2], whose composition incidentally coincides with the one proposed in 1933 by Paris [3]. However, doubts remain, in particular in distinguishing it from the known isostructural phase $Ca_3Mg_{11}Zn_4$ [4]. Here we apply image simulations to interpret HAADF STEM images obtained in such a compound [5,6], using the simulation softwares xSTEM [7], Dr. Probe [8] and jEMS [9], in order to come to a conclusion on this ternary phase.

The approach allowed confirming the $Ca_2Mg_5Zn_5$ crystallographic phase. The comparison between xSTEM, Dr. Probe and jEMS results indicate that the inclusion of the frozen lattice model in the simulation scheme seems essential to obtain a good match to the experimental image for thick areas of the sample. This may relate to the way the electrons are absorbed by channelling on the atomic columns of the crystal. Simulations results will be presented and discussed here.

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MULTIPLE POLARIZATION ORDERS IN INDIVIDUAL TWINNED HAFNIA COLLOIDAL NANOCRYSTALS

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Abstract

Hafnia (HfO_2) undergoes a martensitic transformation from a high temperature tetragonal phase to a low temperature monoclinic phase, which can result in crystallographic twinning. The atomic structures of such twin boundaries have been poorly understood since they were first reported in the 1960s [1]. Recently observed ferroelectric behaviour in HfO_2 thin films [2] promises a new generation of non-volatile memories. However, the fact that high-angle annular dark-field scanning transmission electron microscopy reveals Hf atoms and not O atoms [3] means that the ferroelectric-structure relationship is not well understood.

In our recent work [4], we applied the negative spherical Cs imaging (NCSI) technique to image both Hf and O atoms in colloidal nanocrystals of HfO_2. Our results show that twin symmetry involves a two-fold screw operation and not a reflection or 180° rotation. Crystallographic twinning creates interesting polarization structures at the twin boundaries, with nm-sized ferroelectric and antiferroelectric phases that potentially allow much higher information storage density. The observed correlation between twinning and ferroelectric and antiferroelectric phases is based on their symmetry, providing general guidance for discovering new ferroelectric and antiferroelectric phases in materials that are not limited to oxides.

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ATOMICALLY RESOLVED TOMOGRAPHIC RECONSTRUCTION OF NANOPARTICLES FROM SINGLE PROJECTION: INFLUENCE OF AMORPHOUS CARBON SUPPORT

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Abstract

Nanostructures have a wide range of applications due to their unique geometry and arrangement of atoms. For a precise structure-property correlation, information regarding atomically resolved 3D structures of the nanostructure is of utmost benefit. Though modern aberration-corrected transmission electron microscopes can resolve atoms with sub-angstrom resolution, an atomic-scale 3D reconstruction of the nanostructure is a challenge using tilt series tomography due to high radiation damage. Instead, inline 3D holography-based tomographic reconstructions from single projection registered at low electron doses are more suitable for defining atoms positions at nanostructures. Nanostructures such as nanoparticles are generally supported on amorphous carbon film for TEM experiments. However, neglecting the influence of carbon film on the tomographic reconstruction of the nanoparticle may lead to ambiguity. In order to address this issue, the effect of amorphous carbon support was quantitatively studied using simulations and experiments, and it was revealed that increasing thickness and/or density of carbon support increases distortion in tomograms.

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RESOLUTION AND CONTRAST IN A LIQUID CELL SCANNING ELECTRON MICROSCOPE PLATFORM

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Abstract

Multi-modal in situ investigations based on electron microscopy has immense potential to inform us about the dynamical structure and chemistry of materials under working conditions [1]. Although transmission electron microscopes (TEM), especially with aberration correction, is able to provide very high spatial resolution information about the morphology of functional materials, the scanning electron microscope (SEM) has its own advantages in terms of being more amenable towards the addition of complementary capabilities. Here, we implemented a two-window liquid cell setup similar that is used in the TEM holders for the SEM to explore its possibility as an electrochemical imaging platform [2]. We will show that with such this geometry, we are able to resolve 30 nm Cu_2O nanocubes in several hundred nanometres thick liquid with a resolution of a few nanometres using both backscattered and transmitted electrons imaging.

To better understand the limits of image resolution and contrast as a function of liquid layer thickness, we also simulated the electron beam trajectories using Monte Carlo simulations. The simulation results show good agreement with the experimental results [2]. First, the simulations highlight the dependence of image contrast on the liquid thickness. In particular, the transmitted electron images collected at high scattering angles show contrast inversion at liquid thickness above a few hundred nanometres. The contrast inversion is related to the difference in angular distributions between the electrons scattered by the liquid and the sample, which leads to a systematic relation with the atomic number of the sample and the liquid thickness. Second, for the backscattered electrons, the image resolution and contrast is primarily limited by the background noise that is caused by electron scattering in the liquid layer. Results from dynamical studies of cubic Cu_2O electro-deposition will also be presented [3].

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DIRECT ELECTRON IMAGING OF DISLOCATION DYNAMICS AT NANOSCALE

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Abstract

A major strength of transmission electron microscopy (TEM) is being able to examine defects and their dynamics. However, correlating structural dynamics with properties remains as a challenging problem, especially in nanocrystalline materials, which are widely used for their special properties, but there are no standard characterization techniques because of their small sizes. TEM with its high spatial and temporal resolution is an ideal tool to meet these challenges. Here, we utilize in-situ electron imaging to monitor dislocation activities and structural evolution and correlate with mechanical responses in nanocrystalline (NC) molybdenum (Mo) nanopillars. We focus on the inhibited dislocation activities by the reduced grain size and the resulted Hall-Petch strengthening. Mo selected as BCC metals shows distinctive size dependent mechanical behaviors upon the grain refinement from the more extensively studied FCC metals.

The fine microstructure and structural activities in NC Mo nanopillars upon deformation are determined based on a novel approach we developed using a combination of in-situ electron imaging and scanning electron nanodiffraction [1]. Nanopillars were fabricated from a nanocrystalline Mo film by the focus ion beam method (FIB). A highly sensitive picoindenter was used to conduct compression testing on the nanopillars and simultaneously record the mechanical response from the material. By recording the image video during deformation, we can monitor dislocation activities using the activity intensity method as described in Ref [2]. Before compression, SEND was performed on the nanopillars to obtain local structure information from the diffraction dataset, which helps the identification of nano-grains and dislocation characters.

Using the described approach above, we have tested nanopillars of ~200 to 300 nm in diameters. Our findings demonstrate that grain boundaries (GBs) play an essential role that contribute to the strength and plasticity. Dislocation with edge and screw characters are both observed, and their motions are restricted by GBs. Together, the results provide critical insights into strengthening via grain refinement in a refractory metal.

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REVEALING BILLION-YEAR-OLD SECRETS OF A METALLIC METEORITE

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Abstract

Iron meteorites provide a window into the billion-year thermomagnetic history of the solar system, while furnishing insight into the formation of Fe-Ni phases that are kinetically unattainable on terrestrial timescales. The slow cooling conditions that they experience can result in the formation of tetrataenite ($L1_0$ order, AuCu-I type), whose alternating layers of Fe and Ni atoms are stacked parallel to the tetragonal *c* axis, which is also the easy direction of magnetocrystalline anisotropy. In this work, we use correlative experimental and theoretical methods that span different length scales to reveal a highly intricate distribution of crystallographic phases in the slowly-cooled meteorite NWA6259. We discover a previously hidden tetragonal Fe-Ni nanophase, alongside Ni-poor and Ni-rich precipitates of cubic symmetry formed within the chemically-ordered tetrataenite matrix. Aberration corrected scanning transmission electron microscopy imaging is used to study the interphase structure and the corresponding strain fields of the precipitates and matrix. The distinct magnetic nature of the precipitates and their effect on the magnetic structure are studied by using a combination of quantitative magnetic imaging [1,2], micromagnetic simulations and bulk magnetometry measurements. Fresnel defocus imaging and off-axis electron holography are used to reveal distinct magnetic domain structures in specimens prepared with the magnetic easy axis parallel and perpendicular to the electron beam direction. The observations provide new insight about the archetypal Fe-Ni phase diagram, as well as implications for the development of new types of advanced permanent magnets.

The authors acknowledge Laura H Lewis and Michalis Charilaou for their collaboration during this study. We are grateful for financial support from the DFG through CRC/TRR 270 and the European Research Council through ERC□2019□SyG project 856538.

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QUANTITATIVE APPLICATION OF MOIRE SAMPLING IN SCANNING TRANSMISSION ELECTRON MICROSCOPY

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Abstract

The concept of Moiré sampling refers to the intentional formation of aliasing artifacts (also called Moiré fringes) when undersampling of a periodic signal [1]. In Scanning Transmission Electron Microscopy (STEM), the scanning grid of the electron beam raster can be used to undersample the crystalline lattices and form a STEM Moiré hologram [2]. The STEM Moiré hologram embeds, in the Moiré fringes' spatial arrangement, structural information of the sample such as a deformation field. However, the STEM Moiré hologram is an aliased version of the crystalline periodicities and requires a reconstruction step before retrieving the structural information.

In this study, a reconstruction method based on the Moiré sampling theorem [3] is presented to recover an oversampled micrograph from a STEM Moiré hologram recorded on a InP/$InAs_{0.35}P_{0.65}$/InP epitaxial stack. The reconstruction approach is then applied to map the deformation field using the STEM Moiré GPA strain characterization technique [2]. The experimental results are furthermore compared to Dark-Field Electron Holography (DFEH), another strain characterization method also build on Moiré interferometry [4]. A small mismatch is observed between the two techniques and is related to their respective contrast mechanisms. The STEM Moiré GPA method is based on the STEM contrast mechanism with a very narrow dept of field (few nm) and an electron probe sensing the surface of the thin lamella, while DFEH is based on the Conventional Transmission Electron Microscopy contrast mechanism considering the entire thickness of the same lamella. Both methods are, therefore, sensing different depth sections of the sample. The electron transparent thin foil is subjected to strain relaxation causing a non-uniform strain distribution along the lamella thickness. When comparing experimental data to the Finite Element Method simulations for modeling the strain relaxation, both STEM Moiré GPA and DFEH techniques are confirmed to accurately measure the deformation in the sample with respect to their characteristics (depth-of-field, location sensitivity) [5,6].

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PRESENTATION OFF-AXIS ELECTRON HOLOGRAPHY OF NEEL-TYPE MAGNETIC SKYRMIONS IN MULTILAYER SAMPLES

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Abstract

Magnetic skyrmions are swirling spin structures, which are foreseen as information carriers in novel technologies that combine fast logic operation and non-volatile memory [1]. Multilayers of heavy metals and ferromagnets have been shown to host Néel-type magnetic skyrmions at room temperature, as a result of the presence of broken inversion symmetry and a strong Dzyaloshinskii–Moriya interaction at their interfaces [2]. Néel-type magnetic skyrmions can be observed using Lorentz transmission electron microscopy by tilting the sample with respect to the incident electron beam direction [3]. The technique that is used most often for such observations is Fresnel defocus imaging.

Here, we use off-axis electron holography [4] to study Néel-type magnetic domains and skyrmions in an Ir/Fe/Co/Pt multilayer sample that was sputtered directly onto an electron transparent membrane. The preparation of the sample, the reconstruction of the recorded off-axis electron holograms and the influence of sample tilt angle on the resulting electron optical phase images are discussed. The evolution of the magnetic domain structure in the presence of applied magnetic fields and the relative contributions of the out-of-plane and in-plane components of the magnetic fields of skyrmions to recorded phase images are discussed. Good agreement is found between experimental phase images of individual Néel-type magnetic skyrmions and simulated images based on theoretical models [5].

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A RETROFITABLE LASER-FREE ULTRAFAST PULSER FOR TIME RESOLVED ELECTRON MICROSCOPY

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Abstract

Time-resolved electron microscopy with an extreme space-time resolution (STR) in the range between 10^{-23} and 10^{-20} m·s continues to gain great interest. During the last decade, photocathode-based UEMs (ultrafast electron microscopes) have enabled scientists for the first time to examine dynamic phenomena at a time scale of 100fs, but at the cost of poorer spatial resolution and beam coherence. Recently, Euclid Techlabs, in collaboration with scientists at Brookhaven National Laboratory and the National Institute of Standards and Technology, has succeeded in bringing a novel laser-free UEM to the market. Based on a patented award-winning beam chopping technology [1, 2], the UltraFast Pulser™ (UFP) can be installed on any TEM to provide picosecond electron pulses for time-resolved electron microscopy. The UFP uses the state-of-the-art RF technology making the repetition rate of electron pulses variable from 1 Hz to 12 GHz, with pulse widths from single electron events to native instrument (DC beam) operation [3 ,4]. The UFP system eliminates any need to modify the electron emitter, thus maintaining the native TEM specifications of spatial resolution and beam coherence. Several experiments have been conducted since 2020 [5, 6]. More opportunities and new research territories enabled by this technology will be explored.

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QUANTITATIVE ANALYSIS OF MAGNETIC STATES IN AN ARTIFICIAL SPIN ICE BY OFF-AXIS ELECTRON HOLOGRAPHY

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Abstract

Frustration refers to the inability of a system to simultaneously satisfy competing interactions and can give rise to complex physical properties [1]. In this work, we studied an artificial spin ice that consisted of elongated single-domain patterned nanomagnets, which provided frustration by design. The dynamics of artificial spin ices is driven by the magnetostatic coupling between the magnets, giving rise to emergent excitations such as chiral dynamics [2].

We used off-axis electron holography in the transmission electron microscope to quantitatively measure the magnetic phase shift induced by the sample [3]. The microscope was operated in magnetic-field-free conditions using a Lorentz lens, while the conventional microscope objective lens was used to apply magnetic fields of up to 1.5 T to the sample. The phase shift of the electron wave was measured using an electron biprism and interpreted using a model-based iterative reconstruction algorithm to retrieve the projected in-plane magnetisation. The three-dimensional magnetic stray field surrounding the sample was calculated and the experimental results were compared with micromagnetic simulations.

The permalloy nanomagnets were patterned into a chiral ice geometry on a SiN membrane using lift-off lithography. Magnetic interactions between them were studied by applying in-plane magnetic fields to the sample. The resulting magnetic fields were recorded at remanence. The measurements revealed a zigzag magnetization distribution, with stray fields that became more complex at the edges of the array, indicating an asymmetric energy landscape. The results showed that the pattern of stray magnetic stray field in the plane of the particles is different from that integrated in a direction perpendicular to the plane of the sample, emphasizing the need to take its three-dimensional nature into account.

This study is supported by the DFG through CRC/TRR 270 and the European Research Council through ERC-2019-SyG project 856538.

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SHAPING ELECTRON WAVE FUNCTIONS WITH QUANTUM LIGHT

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Abstract

Generally, the study of phenomena that occurs at ultrashort time scales relies on the use of probe pulses of even shorter durations, such as those provided by femtosecond and attosecond light sources. Since electrons can potentially reach much shorter durations (a typical beam of 10^5 eV is characterized by an oscillation period of 0.02 attoseconds), they present themselves as a promising candidate for the study of ultrafast physics. Additionally, electron pulse compression is also of primal importance in free-electron lasers where the N^2 superradiance emission produced by N electrons produces characteristic ally high intensities. Stimulated by these possibilities, the manipulation of the free electron density matrix using light has attracted a strong interest in different contexts including imaging, spectroscopy and light generation.

In parallel, the advent of photon-induced near-field electron microscopy (PINEM), a technique which synchronizes fs laser illumination with electron pulses at the sample, has shown to be capable producing strong electron-light coupling by exploiting the evanescent components of the near-field, in contrast to the negligibly small interaction probabilities in free space. This strong coupling results in multiple gain and loss features in the electron spectra. PINEM experiments have so far relied on coherent light sources (i.e., lasers), for which the measured spectra are well reproduced by assuming sample bosonic excitations that are coherently populated with a large number of quanta. Interestingly, this technique, performed with laser pulses, has been combined with free electron propagation over a few millimeters to yield a temporal compression of the electron density which transforms into a series of pulses with duration down to the attosecond regime. While the aforementioned studies make use of laser pulses, thus inducing the electron to interact with a coherent state of the light field, an extension to quantum optical fields has been recently predicted to introduce quantum effects in the electron spectra [4]. As a consequence, quantum light represents a new opportunity to manipulate the electron wave function by adding a new degree of freedom.

In this work [1], we show that a wide range of electron statistics can be explored by employing quantum light in the interaction with free electrons. In particular, we study how the focusing properties of the optically-modulated electrons differ from the coherent case when different quantum states of light, such as squeezed states and minimum phase uncertainty states, are employed. Furthermore, we find that the interaction with phase-squeezed and minimum-phase-uncertainty light states produces faster compression of the electron density, while amplitude-squeezed light gives rise to ultrashort double-pulse electron profiles whose shape and relative distance can be tuned with the light intensity and the squeezing parameter. As an application, we also demonstrate that the interaction of such modulated electrons with a target produces a Poissonian distribution of sample excitations independent of the electron shape, but where the off-diagonal elements are strongly dependent on the statistics of the light used to modulate the electron. Together with the fundamental interest in studying the new plethora of phenomena, we envision the application of quantum light in the control of electron compression and in the generation of light with nontrivial statistics.

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HOW MUCH CAN INELASTICALLY SCATTERED ELECTRONS CONTRIBUTE TO ELECTRON CRYOTOMOGRAPHY OF BIOLOGICAL SPECIMENS?

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Abstract

Electron cryotomography (cryoET) is an application of bright-field transmission electron cryomicroscopy (cryoEM) used to determine three-dimensional structures of cells and large macromolecular complexes [1]. The samples used for cryoET are often thicker than 1000 Å, meaning many 300 keV electrons suffer at least one inelastic scattering event in transit through the specimen. The current practice is to remove these electrons with an electron energy filter. Given the advent of practical chromatic aberration correction [2], there is a potential improvement in signal by incorporating these inelastically scattered electrons in the image in the context of an achromatic objective lens. Unfortunately, inelastically scattered electrons also have reduced spatial coherence. This study evaluates the conditions necessary to best use inelastically scattered electrons for biological imaging and determines the potential improvement in signal afforded by achromatic imaging of the thick biological specimens of interest in cryoET. To understand the loss of spatial coherence caused by the specimen, we measured the loss of signal from inelastically scattered electrons through a thick carbon specimen by quantifying the fading of the 111 reflections from gold nanoparticles as a function of defocus on different thicknesses of carbon. The results from these studies were also confirmed using a C_c corrected microscope (PICO) [3]. The results show that inelastically scattered electrons can be used to enhance the signal in phase contrast images of thick specimens. Our measurements using model carbon specimens indicate C_c correction could increase the number of biological processes accessible to cryoET.

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MACROMOLECULAR ORGANIZATION OF MEMBRANE-ASSOCIATED ATG18 OLIGOMERS

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Abstract

Atg18 is a core protein in macroautophagy that is a highly conserved eukaryotic pathway critical for the cell's stress response and homeostasis. The precise structural role of Atg18 in the autophagy process remains to be established. Atg18 belongs to the PROPPIN family consisting of seven WD40 repeats that form a beta propeller motif. Atg18 has been shown to bind Atg2 and Atg9 and thereby participates in the elongation of phagophores. Atg18 has a conserved PIP/PIP2-binding motif (FRRGT) that constitutes a specific membrane anchor. We demonstrate how Atg18 can form filaments in low-salt solution by solving the helical structure to near-atomic resolution using electron cryo-microscopy. The oligomerization sites are conserved even under high-salt conditions, as we demonstrate by single particle analysis of this small 50 kDa protein. We reconstituted Atg18 with liposomes and elucidated the macromolecular organization of Atg18 oligomers on cellular membranes using subtomogram averaging. The structure reveals that oligomerization motifs present in helical filaments and in solution are also observed between juxtaposed membranes, demonstrating how Atg18 may function to remodel and extend phagophore membranes in concert with the larger autophagy machinery.

PSPA ADOPTS AN ESCRT-III-LIKE FOLD AND IS CAPABLE OF REMODELING BACTERIAL MEMBRANES BY FUSION AND FISSION

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Abstract

The phage shock protein A (PspA) is a member of the PspA/IM30(Vipp1) protein family. The PspA/IM30(Vipp1) family is wide spread among the bacterial phyla and has also been identified in chloroplasts as well as in some archaea, where it is involved in maintaining the integrity of cellular membranes. A hallmark of this protein family is the formation of MDa-sized homooligomeric ring and rod structures. Here, we present the 3.6 Å resolution cryo-EM structure of PspA assembled in helical rods, which reveals that PspA adopts a canonical ESCRT-III fold. Using cryo-EM and cryo-ET, we show that PspA interacts with small unilamellar bacterial membrane vesicles by fusion and fission events, resulting in µm-sized vesicles with distorted shapes and internalized membranes. These activities are mediated by PspA rods engulfing lipid bilayers and amorphous PspA assemblies acting as lipid transfer zones. As similar membrane remodeling activities are well-established for ESCRT-III proteins, we suggest that bacterial PspA belongs to the evolutionary ancestry of ESCRT-III proteins.

CAPTURE OF CO-TRANSLATIONAL MULTI-DOMAIN PROTEIN FOLDING WITH CRYO-EM

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Abstract

Cryo Electron microscopy (cryo-EM) is a powerful technique to solve three dimensional structures of macromolecules. Since the implementation of direct electron detectors, it is possible to solve structures of large proteins (> 50 kDa) with near atomic resolution. Despite the major advances of cryo-EM, this technique is still limited by the size of particles, which is critical to reach high resolution reconstructions.

In order to overcome this problem, small or medium sized proteins can be coupled to large references like a ribosome (~ 2.5 MDa). Taking this strategy into account, we designed a construct with a modified arrest peptide (SecMstr) at the C-terminal end of the medium sized multi-domain protein, namely the phosphoglycerate kinase (PGK ~ 45 kDa). The arrest peptide is responsible for keeping the nascent polypeptide chain attached to the ribosome. All together, we were able to demonstrate that it is possible to obtain an intermediate resolution cryo-EM density which corresponds well to the expected fold of native three-dimensional structure of this medium sized protein.

At this point, the resolution is still far from atomic resolution (~ 6 – 10 Å). However, these promising results open up new opportunities for further improvements and could represent a more general strategy of solving small protein structures using cryo-EM.

PRESENTATION TITLE TO BE CONFIRMED

Raimond Ravelli

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Abstract

Abstract to be handed in later.

A MILLIKELVIN SCANNING TUNNELING MICROSCOPE IN ULTRA-HIGH VACUUM WITH ADIABATIC DEMAGNETIZATION REFRIGERATION

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Abstract

We present the design and performance of an ultra-high vacuum (UHV) scanning tunneling microscope (STM) that uses adiabatic demagnetization of electron magnetic moments for controlling its operating temperature in the range between 30 mK and 1 K with the accuracy of up to 7 μK [1]. The time available for STM experiments at 50 mK is longer than 20 h, at 100 mK about 40 h. The single-shot adiabatic demagnetization refrigerator (ADR) can be regenerated automatically within 7 hours while keeping the STM temperature below 5 K. The whole setup is located in a vibrationally isolated, electromagnetically shielded laboratory with no mechanical pumping lines penetrating through its isolation walls. The 1K pot of the ADR cryostat can be operated silently for more than 20 days in a single-shot mode using a custom-built high-capacity cryopump. A high degree of vibrational decoupling together with the use of a specially-designed minimalistic STM head provides an outstanding mechanical stability, demonstrated by the tunneling current noise, STM imaging, and scanning tunneling spectroscopy measurements all performed on atomically clean Al(100) surface.

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EARLY RESULTS FROM THE X-RAY PERIMETER ARRAY DETECTOR (XPAD) ON THE ARGONNE PICOPROBE

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Abstract

Evolving the Argonne p steradian detector [1], the X-ray Perimeter Array Detector (XPAD) combined with a custom electron optical pole piece (ZTwin) has improved upon the performance of conventional solid state x-ray detectors in Argonne's Analytical PicoProbe Electron Microscope which is located in the ANL Materials Design Laboratory and achieved First Light on December 17th of 2020. Using a 60 mm^2 SDD system in a Tecnai F20 as a reference level (1x), we have measured that the *XPAD* improvement is greater than 20 times the 60 mm^2 SDD system and more than 6x the performance of a Quad 30 mm^2 system in a Talos FX20, both exceeds the performance metrics of the original p steradian detector, reaching Argonne's design target. Due to the increased collection efficiency of the *XPAD*, there is also a corresponding improvement in the minimum detectable mass fraction where a ten fold improvement has been measured [2]. Additional work is in progress to further delineate the performance metrics and will be discussed [3].

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NANOSCALE THERMOMETRY OF SOLIDS AND LIQUIDS USING IN-SITU STEM

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Abstract

Thermal management and heat dissipation are important limitation towards faster and more compact electronic devices. Specifically, interfaces, defects and surfaces can act as heat traps and sources of local strain due to a mismatch in the thermal expansion coefficient, which will lead to device failure or the formation of temperature induced defects. While TEM provides an excellent probe to measuring the local atomic and electronic structures, there is no direct approach to determine the temperature on a similar length scale. To date, nanoscale thermometry is carried out either through scanning probe microscopy-based (SPM-based) techniques like scanning thermal microscopy or non-contact optical methods, such as Raman, fluorescence, and luminescence thermometry.

In this contribution, we utilize atomic-resolution imaging and electron spectroscopy in an aberration-corrected scanning transmission electron microscope (STEM) to characterize the thermal properties of 2D materials, including pristine and allowed transition-metal dichalcogenides, as well as graphene liquid cells [1] during in-situ heating and cooling experiments. We use the aberration-corrected JEOL ARM200CF at the University of Illinois at Chicago, equipped with a cold-field emission electron source and a Gatan Continuum GIF. In-situ experiments will be carried out using the Protochips Aduro and Gatan cooling stages, which provide the stability to perform atomic-resolution analysis in the temperature range of 90 K - 1000 K.[2] The experimental data is then compared to our first-principles modeling results.[3,4,5]

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EFFICIENT PARTICLES SIZE DISTRIBUTION CHARACTERIZATION WITH TEM: RECENT ADVANCES IN EXPERIMENTS AND ANALYSIS AUTOMATION

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Abstract

Transmission Electron Microscopy (TEM) has been widely used on particles size distribution characterization, particularly with nanostructured samples and in cases that require complementary high-resolution techniques – such as chemical mapping or localized diffraction analysis. Among the numerous applications examples are the quality control on catalysts synthesis and the metallic alloys development by precipitates optimization.

Traditionally, particles size distribution characterization with TEM involves lengthy imaging experiments to support a fair statistical representation of the sample, and extensive image analysis for features identification and measurement. For this reason, TEM is often regarded as an ineffective and expensive approach for the purpose and only recommended in case alternative techniques such as Scanning Electron Microscopy (SEM), X-ray diffraction (XRD) techniques and Dynamic light scattering (DLS) are not applicable.

This presentation features recent instrumentation advances that allow for automated TEM imaging and chemical mapping experiments, and simultaneous data analysis by customizable algorithms for flexible features detection. Results indicates successful unattended TEM imaging experiments and data analysis with high yield (>6000 particles / 10 minutes), and advanced nanoparticles characterization by combining scanning TEM (STEM) imaging, X-rays Energy Dispersive Spectroscopy (EDX) mapping, and Artificial Intelligence (AI) data analysis methods already integrated to an easy-to-use workflow.

DETERMINATION OF 3D STRAIN FIELDS BY DARK-FIELD ELECTRON HOLOGRAPHY UTILIZING DYNAMIC DIFFRACTION

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Abstract

Semiconducting quantum dots (QD) have three dimensional strain fields. These strain fields influence the optical and electrical properties. The phase measured by means of dark-field electron holography (DFEH) contains information about the displacement fields [1]. Inhomogeneous displacement fields in electron beam direction cause a non linear phase behaviour under dynamical diffraction conditions like already shown in previous papers [2, 3]. This phase depends on the specimen thickness, the excitation error, the extinction length and the depth of the inhomogeneous displacement field in the specimen. This dependency also creates a possibility to determine the structural properties of heterostructures in electron beam direction.

To determine the depth of $In_{0.80}Ga_{0.20}As$ QD surrounded by GaAs in the specimen dark-field holograms are recorded using the (400)-reflection under variation of the excitation error. The measured phases are compared with multi-beam calculations using spherical precipitates as a model. We assume the displacement fields of the spherical precipitates are comparable to the QD's displacement field in the wider area of the dot. These calculations are done by numerical propagation of the Darwin-Howie-Whelan equations for the same excitation conditions as found in the experiment. In order to determine the depth of the precipitate the phases obtained for different excitation errors have to be unique for every depth in the specimen. By comparing the measured and calculated phases the best fit is found and the depth of the QD is determined.

We additionally investigated the influence of surface relaxation effects in quantum well (QW) structures. The dark-field phase of the (220)-reflection is measured for a 45° inclined $In_{0.25}Ga_{0.75}As$ quantum well embedded in GaAs. The QW has a larger lattice constant than the GaAs, thus relaxation effects at the specimen surfaces are expected. Comparing the measured dark-field phases a better match is found for calculations additionally including the surface relaxation effects. This shows the importance of the consideration of relaxation effects in the holographic analysis of specimens with strained surfaces.

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ATTOLIGHT MÖNCH ADD-ON FOR (S)TEM

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Abstract

The Mönch is a (S)TEM add-on that has been designed to serve as a Cathodoluminescence (CL) detector with unprecedented signal-to-noise ratio and spectral resolution. It is used to measure simultaneously ultra high-resolution images and emission hyperspectral maps of individual nano-particles, quantum dots or atomic defects. The design behind the Mönch has a solid publication track record, which includes reports on nano-plasmonics, quantum nano-optics, simultaneous measurements of Cathodoluminescence and Electron Energy Loss Spectroscopy (EELS), non-linear spectroscopy of individual quantum wells and more [1]. Among the most recent examples, a combination of EELS and CL revealed nanometre resolved optical properties of h-BN/WS2/h-BN heterostructures, which were directly correlated to chemical and structural information [2].

Current developments, which include a collaboration with Jülich Forschungszentrum, aim at going even further by extending the capabilities of the Mönch to provide a versatile platform that can be used either to collect or inject light. Electron Energy Gain Spectroscopy (EEGS) [3] is an example of measurements that were performed by injecting light with the Mönch.

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CONTINUOUS-WAVE PHOTONIC CHIP-BASED TEMPORAL PHASE PLATES FOR TEM

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Abstract

The optical shaping of free-electron beams enables a broad range of applications, from efficient free-space acceleration [1] and attosecond bunching of electrons [2] to the implementation of laser-driven phase plates [3,4] and beam splitters [5]. Despite recent progress towards phase-matched and high-efficiency coupling [8,9], inelastic electron light scattering (IELS) typically requires femtosecond high-intensity laser pulses, prohibiting active beam control in state-of-the-art continuous-beam electron microscopes.

Here, we demonstrate IELS on a CW-pumped Si_3N_4 microresonator with a *Q*-factor of $>10^5$, achieving an unprecedented high coupling to a continuous electron beam.

In a custom modified Schottky-field-emission TEM [10], a continuous electron beam interacts with the optical whispering gallery mode confined in a fiber-coupled Si_3N_4 microresonator chip (fabricated in the photonic Damascene process [11], linewidth of ~70 MHz and free spectral range of ~1 THz for the quasi-TM fundamental mode). When the CW laser is tuned to a resonance of the cavity, the initially narrow energy distribution is significantly broadened up to about 150 eV, indicating a strong temporal sinusoidal phase modulation. The interaction strength between the electrons and the evanescent cavity field is spatially mapped and we discuss the potential application as a versatile temporal phase plate for electrons.

We envision a new class of active electron-optical elements for TEM based on IELS, building on state-of-the-art integrated photonics.

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COMBINING QUANTITATIVE ADF STEM WITH SIN_X MEMBRANE-BASED MEMS DEVICES: A SIMULATION STUDY WITH PT NANOPARTICLES

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Abstract

Computer simulations are used to assess the influence of a 20nm thick SiN_x membrane on the quantification of atomic-resolution annular dark-field (ADF) scanning transmission electron microscopy images of Pt nanoparticles. The discussions include the effect of different nanoparticle/membrane arrangements, accelerating voltage, nanoparticle thickness and the presence of adjacent atomic columns on the accuracy with which the number of Pt atoms in each atom column can be counted. The results, which are based on the use of ADF scattering cross-sections, show that an accuracy of better than a single atom is attainable at 200 and 300 kV. At 80kV, the scattering in a typical SiN_x membrane is sufficiently strong that the best possible atom counting accuracy is reduced to +/- 2 atoms. The implications of the work for quantitative studies of Pt nanoparticles examined using SiNx membranes will also discussed.

QUANTIFICATION OF LONG-RANGE ELECTRIC FIELDS BY COMBINING 4DSTEM AND MULTISLICE SIMULATIONS

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Abstract

Many electronic devices, like transistors and batteries, have built-in nano-scale electric fields, whose quantitative characterization is desired. Four-dimensional scanning transmission electron microscopy (4DSTEM) facilitates measuring those fields by determining the diffraction pattern's shift in the center of mass (COM) [1, 2] which is approximately proportional to the electric field [3]. The challenge here is that the existence of a COM shift does not prove the existence of a field, because of the COM's sensitivity to other factors.

The objective of this study is to quantify COM-influencing factors in order to measure built-in electric fields via 4DSTEM and to predict optimal experimental parameters. To accomplish this, STEM simulations including nano-scale electric fields are carried out using the STEMsalabim code [4] which is based on the multislice algorithm [5]. This approach facilitates the analysis of individual parameters and reveals their effect on the COM. In a first case study, a GaAs p-n junction is investigated, at whose characteristic depletion region an electric field forms [6]. To determine the influence of the built-in field, the corresponding voltage is included in STEM multislice simulations. Simulation series are done for the three cases of including only atomic scattering, only scattering at the built-in voltage, and a combination of both. From the simulated COM then the electric field and the electric potential are calculated.

Our results show the capability of the presented method to correctly include the deflection of electrons due to long-range electric fields in STEM simulations. Furthermore, the dependence of the measured field on the specimen thickness, the probe convergence angle, and other factors is investigated which allows to propose optimal experimental conditions. This study shows the necessity for complex image simulation studies and is a promising approach to improving the quantitative measurement of nano-scale electric fields via 4DSTEM. The simulated results will be compared to experimental COM data sets acquired using a pnCCD camera in a double C_S-corrected Jeol JEM 2200FS.

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IDENTIFYING SUPPORT EFFECTS IN CO OXIDATION WITH AU

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Abstract

When comparing commonly used oxide catalytic supports such as alumina and titania, numerous material properties are changing including symmetry, surface chemistry and electronic structure. As a result, it is very difficult to assign one or two of these as the undisputed driver of performance changes observed in catalysis. In this work, we overcome this hurdle using structurally similar $LnScO_3$ (*Ln* = La, Sm, Nd) nanoparticle supports that are synthesized to have cuboidal shapes [1,2]. These supports were observed to have a similar, Sc rich, surface reconstruction [3] using a combination of aberration corrected transmission electron microscopy (acTEM) and X-ray photoelectron spectroscopy. Depositing Au nanoparticles onto these supports and using them to catalyze CO oxidation, we observe no change in the activation energy of the reaction but do find a difference in the rate. Additional acTEM characterization indicates thermodynamic similarities in these nanoparticles as well, as the degree of particle wetting is similar across all samples.

We attribute the static activation energy to the structural, thermodynamic, and chemical similarities observed at the active sites. The rate differences are connected to the strength of CO_2 binding to the support surface as demonstrated by temperature programmed desorption measurements on the blank supports. The ability of these supports to dilute the number of independent variables as demonstrated through advanced characterization lays a foundation for future studies with various reactions and metals to give insight towards which material properties dictate catalytic performance.

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MEASUREMENT OF STRAIN- AND DOPING CONCENTRATION-DEPENDENT ELECTROSTATIC POTENTIAL CHANGE IN GAN WIRE USING OFF-AXIS ELECTRON HOLOGRAPHY

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Abstract

The group-III nitride semiconductors have been widely used in lighting devices. With the higher size demands for integrated devices, the group-III nitride semiconductors would suffer strain while working. Strain-induced band gap change in group-III nitride semiconductors have been investigated at both tensile and compressive strain [1, 2]. The change of surface state and donor ionization energy under strain could induce the change of electrostatic potential. The change of all parameters induced by strain is a lack of systematic investigation.

A Si-doped GaN buffer epitaxially grown on a c-plane GaN substrate, between which there is a thin layer of higher Si-doped GaN. For off-axis electron holographic TEM measurement, a wire with a rectangular cross-section was prepared in a focused ion beam (FIB) system. The longitude direction of the wire is the c-axis of wurtzite GaN. Using a NanoFactory holder, mounted with an electrochemically etched tungsten needle, the GaN wire can be bent in-plane. In the bent wire, one side is under tensile strain, while the other side is under compressive strain. The phase shift of both sides can be obtained by reconstructing the holograms. Then the electrostatic potential can be calculated.

The higher Si-doped GaN layer has a higher phase shift, forming a peak compared to GaN buffer and GaN substrate. The peak height (the difference between the peak and GaN buffer) varies at tensile and compressive strain, indicating that the change of electrostatic potential at different doping concentration has different values. The peak height under strain is higher than unstrained status, and the tensile side is higher than the compressive side, indicating that (1) under strain, higher doping concentration induces larger change on electrostatic potential, (2) the electrostatic potential changes to the same direction at both tensile and compressive strain, and (3) the electrostatic potential changes larger under tension than compression. With theoretical calculation, we reveal how much donor ionization energy changes. The donor ionization energy goes closer to, even into conduction band under strain. Tensile strain, as well as larger strain, induces more significant ionization energy change.

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HOW TO MAKE ELECTRON HOLOGRAPHY QUANTITATIVE: DEMONSTRATED EXEMPLARY FOR NITRIDE SEMICONDUCTOR INTERFACES

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Abstract

We present a methodology to quantify polarization and electron affinity changes at interfaces by combining scanning tunneling spectroscopy, off-axis electron holography in transmission electron microscopy (TEM), and self-consistent calculations of the electrostatic potential and electron phase change. We use a precisely knowngrown-in doping structure to calibrate the surface potential of the TEM lamella and thereby achieve a quantitative analysis of electron phase changes measured by off-axis electron holography. Using this calibration, we deduce quantitatively polarization and electron affinity changes for $Al_{0.06}Ga_{0.94}N/GaN$ and $In_{0.05}Ga_{0.95}N/Al_{0.06}Ga_{0.94}N$ interfaces. The latter interface reveals, as expected, biaxial relaxation as well as polarization and electron affinity changes. However, at the $Al_{0.06}Ga_{0.94}N/GaN$ interface anomalous lattice relaxations and vanishing polarization and electron affinity changes occur, whose underlying physical origin is anticipated to be total energy minimization by the minimization of Coulomb interactions between the polarization-induced interface charges.

SINGLE EVENT DETECTION IN TRANSMISSION ELECTRON MICROSCOPY FOR SPECTROSCOPIC COINCIDENCE EXPERIMENTS

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Abstract

Electron energy loss spectroscopy (EELS) and energy dispersive spectroscopy (EDX) are two broadly used techniques in transmission electron microscopy which are sensitive to the chemical composition of the investigated materials. Both measurement techniques share the fact that excitations of atomic states are involved. Indeed, for every X-ray emitted there was at least one electron that gave part of its energy to excite the atom in the first place, and therefore one could imagine that they convey very similar information. However, current EDX detectors have a relatively low energy resolution (100 eV) where for EELS energy resolutions of 1eV are easily accessible. On the other hand, EELS suffers from a large background signal making it hard to detect low concentration elements. Since the two signals originate from the same process, the information on the time-of-arrival of both electrons and x-rays can be exploited in order to select only electrons and X-rays coming from the same process hence significantly reducing the background signal. The idea of observing such temporal correlations has previously been applied by Kruit *et al.*[1] making use of real time event filtering and serial EELS acquisition. In the present work, we demonstrate the capability of detecting the single electron events with a time resolution of 1.6 ns by using a Timepix3 detector[2], which was placed at the end of a spectrometer. The single x-ray detection was performed using a Super-X-EDX detector setup where a digital pulse processor was used to detect the time-of-arrival and energy. This setup shows the potential to improve the signal-to-background ratio of the EEL spectrum and revealing the inelastic scattered electrons coming from low abundance elements in a matrix of majority elements [3]. This is achieved by selecting only the electron events which have an X-ray detected inside the time coincidence window, which is determined by the time resolution of the detection setup. This method is different from previous setups as every single event is detected and timestamped. After the acquisition, the offline data treatment can be performed giving not only access to the time correlated data but also to the conventional EEL and EDX spectra. Finally, the setup is improved in order to synchronize the incoming electron beam with the two detectors giving the ability to add spatial information to the coincidence data [4]. We will demonstrate the latest results on this setup while extrapolating its possibilities for future materials science.

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ATOMICALLY RESOLVED 3D STRUCTURAL RECONSTRUCTION OF SMALL QUANTUM DOTS

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Abstract

The semiconducting Quantum Dot (QD) has potential applications in light-emitting diodes, single-photon sources and quantum computing due to shape-dependent (opto) electronic properties. Atomic resolution 3D-structure determination is important in understanding growth kinetics and improving device performance. 3D-reconstruction of large QDs was reported using characterization techniques like atomic force microscopy, atom probe tomography and tilt series electron tomography but, still, atomic resolution tomography of QD specially sized below 10 nm is a challenge. Inline-3D-holography is an emerging and promising technique to perform atomic resolution tomography at low electron dose. In the present study, atomically resolved 3D structures of QDs were reconstructed using Inline-3D-holography, implemented on InN QDs (<10 nm) grown on a Si substrate. The residual amorphous glue distorts the exit surface geometry; hence an error correction method was proposed. This is the first experimental evidence of pre-pyramid shaped 3D structure of QD sized below 10 nm that supports theoretical predictions.

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EXPLORATION OF ATOMIC STRUCTURES IN Σ3 [111] ALUMINIUM TILT GRAIN BOUNDARIES

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Abstract

Polycrystalline material are comprised of an intricate network of numerous types of grain boundaries (GBs). GBs are the interfaces separating individual crystals and their atomic structure differs from the adjoining bulk crystals. The distinct atomic nature of GBs controls their properties like mobility, cohesive strength and sliding resistance, which in turn have an enormous impact on bulk properties. Therefore, understanding the structure of these interfaces and the way they influence properties can be a key to engineer novel alloys.

During grain growth of Al thin films on sapphire, it was indicated that Σ3 [111] tilt boundaries in domains with orientation relationship II (ORII) have a higher mobility than that in ORI [1]. One possibility for such behaviour is the existence of multiple states of the incoherent Σ3 twin boundaries that may lead to different interfacial properties, which ultimately affect grain boundary migration at high temperature. However, a direct correlation between the atomic structure of different structural states of Σ3 [111] tilt GBs and their effect on properties is still lacking.

In the present study, the microstructural features (grain size, grain orientation, CSL type and distribution) of Al thin films on sapphire were characterized by electron backscatter diffraction. In-plane site specific TEM specimens from pre-examined thin films were extracted by plasma FIB to study the local atomic structure of Σ3 [111] (211) GBs from both ORs by using advanced transmission electron microscopy (TEM) techniques. Scanning TEM (STEM) reveals that despite having the same macroscopic degrees of freedom, two different atomic motifs of the symmetric incoherent Σ3 tilt boundary are observed for two ORs. The structure of $\Sigma3_{ORI}$ and $\Sigma3_{ORII}$ can be divided into sub-units □E□ and □G□, respectively. The E unit exhibits a square shape and the G unit adopts a hexagon shape. The asymmetric variant of $\Sigma3_{ORII}$ revealed a large number of facets having length greater than 50 nm. These large facets consists of two different types of disconnections (Type A and Type B) along the boundary while $\Sigma3_{ORI}$ is comprised of a single disconnection (Type A) along the boundary and no large facets are observable. The different structural states of incoherent Σ3 [111] twin boundaries will be explored in terms of their microscopic degrees of freedom and implications on interface properties will be discussed.

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XRD AND TEM STUDY OF THE QUASICRYSTALLINE PHASE IN PELLETS AND METAL-MATRIX COMPOSITES CONSOLIDATED USING SPARK PLASMA SINTERING

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Abstract

The production of quasicrystal powders by gas atomization has been commercialized. Among various quasicrystals synthesized, Al-Cr-Fe and Al-Cu-Cr-Fe quasicrystals are very promising materials for coatings and reinforcements in composites due to their high hardness, good wear and corrosion resistance and the relatively low cost of their constituent metals [1]. As these quasicrystals are metal based, the behavior of bonding between the quasicrystalline reinforcement phase and the metal matrix is superior to that for ceramic phase reinforced metal-matrix composites. However, long duration sintering can result in phase changes and decrease the beneficial properties of the quasicrystals in the composites.

In our study, we employed spark plasma sintering to shorten the heating duration for the Al-Cr-Fe and Al-Cu-Cr-Fe quasicrystal powders and thus minimize the chance of phase changes when fabricating quasicrystalline reinforced composites. The phases of the sintered compacts were analyzed using x-ray diffraction. The microstructure of the quasicrystalline phases was further studied using high resolution transmission electron microscopy.

Experimental results showed that even when the heating duration was as short as 30mins during spark plasma sintering at 650ºC, phase changes could still occur. X-ray diffraction patterns showed peaks from the decagonal Al-Cr-Fe phase while HRTEM images revealed that in decagonal Al-Cr-Fe quasiperiodic planes were periodically stacked along the 10-fold axis with a periodicity of about 1.2nm. Spark plasma sintering of icosahedral Al-Cu-Cr-Fe and pure Al blended powders was conducted at 450ºC for 10mins and no phase change was detected using X-ray diffraction or high resolution electron microscopy. This implies that spark plasma sintering is effective in fabricating quasicrystal reinforced Al-based composites.

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PHASE RELATION AND MORPHOLOGY OF TITANIA POLYMORPHS IN EXTREME HYDROTHERMAL CONDITIONS

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Abstract

Nanocrystalline titania has recently attracted much attention due to its modified material properties and chemical reactivity [1]. Four polymorphs of TiO_2 existing at ambient conditions are rutile, anatase, brookite and TiO_2-II, the only quenchable high-pressure phase with the α-PbO_2 structure. The most common and studied of the four crystal structures are anatase and rutile, with brookite being commonly overlooked mostly due to the difficulty in its preparation and purification [2], and TiO_2-II due to the need of high pressures for its synthesis [3].

In this work we present the results of our study of four titania polymorphs phase relations in extreme hydrothermal conditions (2 MPa to 6 Pa and 100 – 300 °C) and their morphologies after recovery. So obtained titania was characterised by transmission electron microscopy, powder X-ray diffraction, Raman and FTIR spectroscopies. Aberration-corrected STEM EELS in combination with principle component data treatment were used in order to understand the nanostructure of particles.

Pure nanocrystalline brookite was afforded at 2 GPa and 100 °C with the mean particle size of 6.3 nm. Aberration-corrected STEM EELS revealed that all samples synthesized up to 1 GPa at 100 °C had core-shell situation with brookite being the inner and anatase the outer layer of the spherical particles. Rutile–TiO_2-II phase boundary was extended towards lower p-T ranges and pure TiO_2-II was afforded at as low as 5 GPa and 285 °C with mean particle diameter being 50 nm.

The conditions used in this work are viable for industrial production settings and can be utilized at large-scale synthesis. This opens up an opportunity for two more titania polymorphs for common use in industrial applications.

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MODULATION OF CATHODOLUMINESCENCE EMISSION BY INTERFERENCE WITH EXTERNAL LIGHT

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Abstract

Narrow bandwidth light sources, such as laser sources, represent an excellent tool to probe optical excitations in atoms, molecule and nanostructures with high spectral selectivity. However, the free-space light dispersion imposes severe limitations to this kind of sources when far-field optics is employed unless more elaborated schemes are used. In this scenario, electron beams hold the promise of overcoming such limit thank to the evanescent components, carried by the electric field produced by a charged moving particle, which are able to excite optical transitions with a spatial resolution only limited by the electron lateral size, now reaching the sub-Ångström regime in state-of-the-art electron microscopes. Unfortunately, since such electric field spans a broad range of frequencies, spectral selectivity is lost and quasi-monochromatic electrons need to be employed to retrieve energy loss spectra (EELS) by performing a post-selection of the scattered probes. Interestingly, the recent field of ultrafast transmission electron microscopy (UTEM) is attempting to reconcile techniques exploiting light and e-beams separately with the aim of converging toward an experimental method able to reach an unprecedent combined spatial, spectral and time resolution. In this regard, femtosecond laser pulses are sent to excite sample optical excitations in synchronization with the time of arrival of the electron, giving rise to an imaging technique with meV-fs-nm resolution, the so-called photon-induced electron microscopy (PINEM). Here, stimulated quanta exchange between the electron and the sample occurs, leading to a strong spectral structuring of the electron wave function. Such spectral modulation has also been combined by a subsequent electron free-space propagation to produce electrons whose density is formed by a train of attosecond pulses. In a broader context, the ability of shaping electron beams has stimulated recent theoretical studies predicting the possibility of achieving control over the density matrix of excitations supported by the sample via the interaction with modulated free electrons as well as the prediction that cathodoluminescence (CL), produced by PINEM-modulated electrons, bears coherence with the same modulating laser which may be revealed in an interferometric setup.

In our study [1], we analyze the modulation of the light intensity in the far-field produced by the interference between the scattered electric field produced by a dimmed laser and the one produced through the interaction of a previously modulated electron. We show that the total signal is composed by coherent and incoherent contributions. In addition, we find that by tuning amplitude and polarization of the external laser pulse and by modulating the electron by a PINEM interaction, the former can be completely cancelled leading to a partial suppression of the total emission probability measured in the far-field. We speculate that electron modulations approaching the point-particle limit lead to a complete suppression of the far-field emission. Our results may open new routes toward coherent control of optical excitations at the atomic scale as well as toward a spectroscopic method with a time resolution only limited by the detector spectral window.

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STRUCTURAL CHARACTERIZATION OF HFO_2 THIN FILMS FOR RESISTIVE SWITCHING MEMORY APPLICATIONS USING TRANSMISSION ELECTRON MICROSCOPY TECHNIQUES

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Abstract

HfO_2 is of interest for use as an insulating layer in ReRAM devices due to its low fabrication cost and high dielectric permittivity. Primary attention has focused on amorphous and polycrystalline HfO_2 films [1-3]. In contrast, single crystalline HfO_2 can exhibit multiple polymorphs and undergo transformations between different phases during growth [4].

Here, we use advanced transmission electron microscopy (TEM) to study local crystal structures and orientations in single crystalline HfO_2 layers in ReRAM devices. High-resolution TEM and high-angle annular dark-field imaging are used to determine the crystallinity and atomic arrangement in the layers, while annular bright-field imaging and integrated differential phase contrast are used to image oxygen atomic columns. Furthermore, position averaged convergent beam electron diffraction based on scanning nanobeam diffraction with a pixelated detector is used to distinguish between crystal structures that have similar atomic arrangements and the experimental results are compared with simulations.

Single crystalline HfO_2 thin films in two different devices with the layer sequences Pt/Ti/HfO_2/ITO/YSZ and Pt/HfO_2/ITO/YSZ are found to be single crystalline and epitaxial across the entire region of each device. The Pt/Ti/HfO_2/ITO/YSZ device contains three monoclinic structures in [100], [010] and [001] orientations, whereas the Pt/HfO_2/ITO/YSZ device contains orthorhombic and cubic-like phases, in addition to monoclinic [010] and [001] orientations.

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UNVEILING NANOSCALE OPTICAL PROPERTIES OF TMD MONOLAYERS USING COMBINED ELECTRON SPECTROSCOPY TECHNIQUES

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Abstract

In this contribution, we will present results on optical and structural properties of a monolayer of semiconducting transition metal dichalcogenide (TMD) encapsulated with hexagonal boron-nitride (h-BN), at the tens of nanometers scale. The strength of this work is the correlation of optical spectra at the nanoscale with structural and chemical maps. It connects information usually available separately from optical diffraction-limited techniques, such as photoluminescence (PL) [1], and from high-resolution techniques, such as electron microscopy [2] or scanning tunneling microscopy [3].

To achieve this, we used combined high spatial and spectral resolution techniques in a scanning transmission electron microscope (STEM). Electron energy loss spectroscopy (EELS) was used to obtain optical absorption (low-loss range) and chemical analysis (core-loss range). Cathodoluminescence (CL), a nanoscale counterpart of PL [4], was used to measure light emission at the tens of nanometers scale. The two types of spectral data were acquired in the same regions. This gave access to, for example, the local Stokes shift, which is the difference between absorption and emission energies. It can give information about the physical mechanism involved in the absorption and emission processes [1]. In addition to these spectral measurements, spatial information was gathered using whether atomically-resolved imaging or local strain measurements.

Using these combined techniques, we could measure localized emissions from excitonic complexes in WS_2 monolayers maintained at 150 K, with bright spots as small as few tens of nanometers. The localization of such bright spots was linked to the chemical environment, displaying small patches of contaminants (such as C, O, N, or Si), probably coming from sample preparation [5].

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DIRECT EVIDENCE OF AN Al ALLOYED SiN_x INTERLAYER WITHIN AN AMMONIA PREDOSED AlN/Si INTERFACE VIA STEM-EELS

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Abstract

Gallium nitride (GaN) and its alloys with Aluminium (Al) and Indium have shown great application within the electronic and optoelectronic fields due to their wide and tuneable band gap, high defect tolerance within devices, high breakdown voltage and high saturation velocity[1]. A cost effective route towards these devices has been to grow GaN on Silicon (Si) substrates via metalorganic vapour phase epitaxy (MOVPE)[2]. These structures show good performance compared to GaN devices on other substrates such as sapphire or silicon carbide, but do require an aluminium nitride nucleation layer and a significant amount of subsequent strain management within the buffer layers to enable a low wafer bow and to prevent wafer cracking[2].

Previous studies have shown that predosing the Si wafer surface with either trimethylaluminium (TMA) or ammonia before AlN growth can have an appreciable impact on wafer bow[3]. Many questions still remain as to the importance and role of the predose in the growth of the AlN and subsequent strain management. Previous electron microscopy studies have inferred the formation of both amorphous and crystalline silicon nitride (SiNx) interlayers, but with little compositional analysis due to the relatively large spatial resolution of electron dispersive x-ray spectroscopy (EDS)[3]. Other studies have used electron energy loss spectroscopy (EELS) which offers sub nm spatial resolution of the interface, and has been used to great effect for compositional analysis within TMA predose studies[4,5].

In this study we investigate a series of samples with different durations of ammonia predose via EELS, to not only probe the composition of the interface, but also to obtain chemical bonding information within this layer. Here we are able to show direct evidence of a SiN_x interlayer up to 5.3 nm thick with an ammonia predose time dependency, and for the first time highlight an Al alloying of the SiN_x[6].

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DIRECT IMAGING OF STRUCTURAL DEFECTS IN NMC CATHODE MATERIAL

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Abstract

To achieve the climate neutrality goal by 2050 the targeted development of electrical energy systems is of great importance [1]. Modern Li-ion batteries (LIBs) should provide high energy and power density together with optimal weight to volume ratio, be safe and cheap to maintain and propose long lifetime and stability [2,3]. To design the desired materials one should have an in-depth insight into the structural arrangements at atomic level.

Recently the development of layered Ni-rich Li[NixCoyMn(1-x-y)]O2 (NMC) cathodes has helped to reduce the cost and to increase the charge capacity of LIBs. However, these materials are known to have a very rich defect landscape. The so-called cation intermixing with formation of anti-sites often takes place in these layered cathode materials: cations are found to sit in the initial positions of Li. In extreme cases the amount of cations in Li position is close to the amount of cations in their initial columns- this effect is often observed next to the particles surface [4].

Here we demonstrate that HADF-STEM imaging is of great use to characterise the NMC at atomic level and to directly visualise the present defects. Beam sensitive materials can be imaged in detail using the lowered dose and short dwell time. HR HAADF-STEM allows to detect anti-sites where Ni, Co or Mn-cations take the Li sites. This structural modification can result in blocking of Li pathways during the charge and discharge followed by lowered performance of the batteries.

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COMBINING ETEM/ESTEM AND ELECTROCHEMICAL IMPEDANCE SPECTROSCOPY

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Abstract

In this project a transmission electron microscope (TEM) is transformed into a generic high-temperature electrochemical workstation. Here in situ TEM/STEM is combined with electrochemical impedance spectroscopy (EIS). EIS is today one of the most powerful characterization methods for distinguishing electrochemical resistances for different processes (electronic conductivity, ionic conductivity, gas diffusion, catalytic reactions etc.) [1]. With environmental TEM/STEM (ETEM/ESTEM) a specific nanoscale feature or interface can be investigated and correlated with EIS measurements while applying an electrical potential at elevated temperatures in a reactive gas environment. This is in contrast to conventional EIS experiments that only give access to averaging information of many grains or interfaces.

We are particularly focussing on solid oxide electrolysis cells (SOEC). This is a promising technology for green energy storage that converts electrical energy from wind or solar power to chemical energy, for example as H_2 and CO [2]. SOECs operate in reactive gasses (O_2, H_2O, CO_2) at high temperatures, typically ≥ 800°C). SOECs are composed of hard and brittle ceramics. This can make it challenging to manipulate 100 nm thin cells without breaking them. However, in situ characterization is needed because post mortem electron microscopy does not give direct insight into time, temperature and electrical potential dependencies of cell degradation.

We present different approaches for designing high-temperature electrochemical TEM experiments. Our model SOECs are composed of materials commonly used in state-of-the-art SOEC. The cells are prepared by pulsed laser deposition (PLD). In addition to full cells, we work with electrospun nanofibers representing electrode or electrolyte materials in an SOEC.

For the experiments, an ETEM is used in combination with custom-made and commercial heating/biasing TEM holders and a potentiostat for measuring EIS data as well as I-V response. We can observe segregation and grain formation, accelerated as a function of temperature, and dependencies on gas environment. Structural degradations are observed both as a response to heating and applied polarization.

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TAILORING OF SHAPE AND SIZE OF PLATINUM NANO-PARTICLES TO ENHANCE THEIR OXYGEN REDUCTION REACTION PERFORMANCE

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Abstract

High oxygen reduction reaction (ORR) activity has been considered for many years as the key to many energy applications. Herein, we demonstrate two approaches how to exceptionally improve the ORR of platinum nanoparticles (NPs). The first approach is based on combining theory and experiment to prepare Pt NPs with optimal size for the efficient ORR [1]. Optimal nanoparticle sizes are predicted near 1, 2, and 3 nm by computational screening. To corroborate our computational results, we have addressed the challenge of approximately 1 nm sized Pt nanoparticle synthesis with a metal–organic framework (MOF) template approach. The electrocatalyst structure will be demonstrated by a C_s-image corrected high-resolution transmission electron microscopy (HR-TEM) and its ORR activity by rotating disk electrode setup measurement. The measured mass activities (0.87 A/mg_{Pt}) are close to the computational prediction (0.99 A/mg_{Pt}). We report the highest up to date mass activity among pure Pt catalysts for the ORR within similar size range [1, 2]. The specific and mass activities are twice as high as the Tanaka commercial Pt/C catalysis (0.42 A/mg_{Pt}) [3]. The second approach is based on theoretical considerations that Pt NPs with a degree of surface defects, e.g., containing surface concavities, can significantly enhance the catalytic activity towards the ORR. Hence, we developed a one-step and up-scalable top-down approach to produce Pt/C catalyst (with ~3 nm Pt NP diameter) starting from a bulk Pt wire [4]. The ORR activity of the developed catalyst exceeds that of the commercial Pt/C catalyst, ~2.7 times in terms of specific (1.62 mA/cm^2_{Pt} at 0.9 V vs the reversible hydrogen electrode) and ~1.7 times in terms of mass activity (0.71 A/mg_{Pt}) [5]. Besides, the technique used here reduces the complexity of the synthesis (and therefore production costs) compared to state-of-the-art bottom-up techniques. The evidence of a high density of surface defects (including the surface concavities) of Pt NPs will be demonstrated with use of the C_s-image corrected HR-TEM, high-resolution scanning transmission electron microscopy (HR-STEM) and also STEM tomography, with which the surface defects of the very small nanoparticles could be observed.

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AUTOMATED MAPPING OF THE CRYSTALLOGRAPHIC SAMPLE ORIENTATION FROM DIFFRACTION PATTERNS IN MOMENTUM-RESOLVED STEM

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Abstract

In scanning transmission electron microscopy (STEM), sample tilt is a parameter that we want to control for optimum imaging conditions. However, residual tilt cannot always be eliminated because of the limited precision of the tilt stage and because it is partly due to the sample geometry at interfaces or bent specimens. In this study, we present an automated method for measuring local crystal orientation from convergent beam electron diffraction patterns. Using ultrafast cameras that record a diffraction pattern at each scan position (4D STEM), mapping of crystal orientation over a field of view becomes possible. The availability of tilt mapping has the potential to improve strain measurements [1], spectroscopy at interfaces [2], and quantitative studies of bent samples that require local sample tilt as input to simulations.

Central to our method is the detection of Kikuchi band crossings in diffraction patterns. The distance of Kikuchi band crossings to the central beam of the incident probe is a measure of out-of-plane inclination while a mutual change of azimuthal orientation of all bands around a crossing indicates in-plane rotation. Band positions are determined from peaks in azimuthal scans of an annular region at large scattering angles. This procedure has been implemented in a fast computer program and can be automatically applied to a series of patterns as recorded in a 4D STEM experiment.

Achievable accuracy and precision were investigated by simulations. For realistic parameters related to the detector and electron dose used, the method was found to be accurate within 0.1 mrad for the out-of-plane orientation and 5 mrad for the in-plane orientation for diffraction patterns averaged over a unit cell. The spatial resolution of this approach is limited by the lateral extent of probe propagation in the crystal. In case of e.g. [110] Fe, the probe extends laterally over 5 unit cells at 40nm sample thickness.

The method is applied to 4D STEM data from a twin boundary in steel using an Electron Microscope Pixel Array Detector (EMPAD) in a probe-corrected FEI Titan microscope operated at an accelerating voltage of 200 kV. The analysis provides a map of position-dependent local crystal orientation with unit cell sampling and reproduces the expected in-plane rotation of 70.53° between the twins. Such an analysis is possible within minutes and provides immediate insight into the presence of sample bending, strain fields, and stacking faults that were not possible prior to the advent of momentum-resolved STEM.

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MOIRÉ ANGLE DEPENDENT EXCITONIC ABSORPTION IN TWISTED BILAYER WSE_2 BY EELS

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Abstract

Atomically-thin layers of semiconducting transition metal dichalcogenides (TMDCs), such as tungsten diselenide (WSe_2), have attracted considerable interest because of new properties that can be obtained when artificially fabricated into van der Waals homo- or heterostructures. Due to interlayer coupling, bulk and multi-layered TMDCs are indirect gap semiconductors, while their monolayers exhibit a crossover to a direct band gap. It has been demonstrated that the interlayer coupling strength in homostructures is also sensitive to the twist angle in the cases of bilayer MoS_2 [1] and WS_2 [2]. In this work, the combined high spatial and spectral resolution of aberration-corrected scanning transmission electron microscopy (STEM) and monochromated electron energy-loss spectroscopy (EELS) in the low-loss regime are used to investigate the excitonic response of atomically-thin WSe_2, specifically in twisted bilayer WSe_2 as a function of moiré angle.

Few-layered WSe_2 flakes were mechanically exfoliated from a synthetic bulk crystal and transferred onto a Si_3N_4 TEM grid with periodic micron-sized holes. Atomically-resolved imaging has been performed on a Nion UltraSTEM200 operated at 60 keV and monochromated EELS performed on a modified Nion HERMES-S200 (also known as ChromaTEM) operated at 60 kV with the sample cooled using liquid nitrogen ($T \approx 150$ K). In addition to freestanding WSe_2 monolayers, fragments of bilayers and trilayers with variable twist angle between 0–30° are also routinely observed. Well-defined hexagonal moiré patterns with nanometer periodicity are evident in the high-angle annular dark-field (HAADF) images for the low twist angles. The excitonic absorption signatures of these nanometric twisted bilayers from low-loss EELS are compared to the WSe_2 monolayer and trilayer counterparts of zero twist angle. The spectra demonstrate a good general correspondence to the thickness dependence of the A, B, and C exciton resonances, namely a pronounced decrease in C exciton energy with number of layers [3]. Comparing different twist angles in the bilayers also show sizable blueshifts in the C exciton energy up to 200 meV, which subsequently alter drastically the spectral shape between the B–C excitonic transitions, with extremes between the zero-twist and towards the anti-aligned (28°) case suggesting underlying differences in interlayer coupling with respect to the moiré angle [4].

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UNDERSTANDING TRANSITION METAL DICHALCOGENIDE ABSORPTION LINE WIDTHS IN ELECTRON ENERGY LOSS SPECTROSCOPY

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Abstract

Two-dimensional transition metal dichalcogenides (TMDs) have attracted excitement for potential applications; however, localized states in these materials are not fully understood. Specifically, monolayer corrugation has an impact on the optical properties and its role of absorption linewidths increase is not fully characterized. It has been observed that h-BN encapsulation reduces graphene roughness. [1,2] A direct spatial correlation of corrugations to their effect on the optical properties has not yet been demonstrated. An ideal technique to map this change is electron energy loss spectroscopy (EELS), and typical exciton linewidths in suspended monolayers are above 50 meV. [3,4] It is larger than purely optical measurements for monolayer on substrates.[5] Encapsulated TMDs monolayers show sharper EELS absorption lines, around 20 meV, with spectra comparable to extinction spectra.[6] Is the corrugation alone responsible for this line width increase, as these experiments indicate, or are other effects playing a role?

We experimentally probed the relation between exactions absorption lines and corrugation in freestanding and h-BN supported monolayers using EELS and electron diffraction in a scanning transmission electron microscope. Experiments were performed on the ChromaTEM microscope, a modified Nion HERMES 200 equipped with an electron monochromator. To reveal the effect of corrugation, electron diffraction patterns were acquired with the sample tilted 28° with respect to the electron beam. For the freestanding monolayer, most of the diffraction spots get larger and fuzzier as a function of their magnitude, except those along the sample tilt axis, as observed for rough graphene [1,2]. For the supported monolayer, the diffraction spots are unchanged between the tilted and un-tilted configurations. In this contribution, we will discuss the structural differences between freestanding and h-BN supported monolayers and the possible impact on the observed optical exciton line width.

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OPERANDO TEM STUDY OF ALL-SOLID-STATE BATTERY INTERFACE

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Abstract

The liquid electrolytes that are typically used in traditional Li-ion batteries are flammable, especially at higher operating voltages and temperatures. By contrast, an all-solid-state battery (ASSB) makes use of a solid electrolyte, which reduces the risk of flammability [1]. However, the presence of the solid–solid electrolyte–electrode interface in ASSBs introduces different challenges from those of the traditional liquid–solid electrode–electrolyte interface [2]. First, in batteries containing liquid electrolytes, the entire surface of the electrode particles is wetted by electrolytes, whereas the electrode particles and solid electrolytes in ASSBs are connected primarily at point contacts, which are limited in terms of their numbers (as not all electrode particles are in direct contact with electrolyte particles); therefore, ionic transport is basically restricted, diminishing the specific capacity of these batteries. Decomposition reactions at electrode–electrolyte interfaces during battery cycling causing the formation of passivating layers as well as electrode volume changes during battery cycling result in the loss of contacts between the electrode and electrolyte particles, further decreasing direct ion exchange pathways. Second, inhomogeneous (de)lithiation through point contacts can induce strain, which affects electrode mechanical integrity leading to capacity fade.

With operando transmission electron microscopy by visualizing the solid–solid electrode–electrolyte interface of silicon active particles and lithium oxide solid electrolyte as a model system, we show that (de)lithiation (battery cycling) does not require all particles to be in direct contact with electrolytes across length scales of a few hundred nanometers. A facile lithium redistribution that occurs between interconnected active particles indicates that lithium does not necessarily become isolated in individual particles due to loss of a direct contact. Our results have implications for the design of all-solid-state battery electrodes with improved capacity retention and cyclability.

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INTERFACIAL DEFECTS IN THE LAYERED STRUCTURE OF A CHALCOPYRITE COMPOUND

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Abstract

$CuInS_2$ is a diamond-like semiconductor with the crystal structure of chalcopyrite that was first synthesized to explore the applicability to ternary compounds of the Grimm−Sommerfeld valence rules for tetrahedral coordination. [1, 2] The chalcopyrites have the diamond-like zincblende structure but with two cations ordered on sublattices. The Bravais lattice of the chalcopyrite is body-centered tetragonal, belonging to space group $I\bar{4}2d$. The direct energy band gap of $CuInS_2$ makes it a strong light absorber, with a value of 1.53 eV that lies at the optimum for efficient single-junction solar cells. Therefore, $CuInS_2$ has been explored for polycrystalline thin-film solar cells, beginning with a 3.6% efficient homojunction cell, then n-CdS/p-$CuInS_2$ heterojunction solar cells first reaching 7.3%,12 and later 11.4% power conversion efficiency, but far less than the theoretically achievable value of ∼30%. The cause of this low performance is not understood. Here, a single crystal grown from 1 mol % Cu-deficient melt was studied by using atomic resolution high-angle annular dark-field (HAADF) scanning transmission electron microscopy (STEM) and electron dispersive spectroscopy (EDS) (no beam damage issue). [2, 3] We observed that the $CuInS_2$ crystal has a lamellar structure which are alternating pseudotwins. The thickness of the lamellae ranges from several nanometers to hundreds of nanometers. The pseudotwin plane was identified as {112}. Periodic interfacial structures ("interphases") are observed to be inserted into the pseudotwin boundaries. The interfacial structure includes two bright atomic planes but with interplanar spacings different from those of the chalcopyrite $CuInS_2$ bulk. We used STEM-EDS to determine how much the composition of the interphases deviates from bulk $CuInS_2$ crossing through the interface. With respect to the bulk, the interfacial layers are depleted of Cu (down −7.9 at. % in the single layer and down −11.5 at. % in the double layer) and contain up to +8.6 at. % excess In and +3.0 at. % excess S. Thus, while the bulk contains 25.2 at. % Cu, the single and double interfacial phase layers contain only 17.1 and 13.7 at. % Cu, respectively, but 32.9 at. % In. The copper deficiency of the interfacial phase suggests that the interfacial phase accommodates the copper deficiency of the overall crystal. [4]

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SCULPTING PLASMONIC RESONANCES IN NANOPARTICLES BY INTELLIGENT ELECTRON BEAM CONTROL

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Abstract

Designing nanostructures with intentional optical properties is key task in nanophotonics and plasmonics, particularly as quantum technologies emerge. Generally, as-synthesized nanostructures retain specific dielectric properties based on fabrication methods and material properties. Accessing the entire plasmonic design space is challenging, and particularly, the inverse design problem is a major goal. With the aim of producing specific optical responses on the nanoscale, we utilize encoder-decoder neural networks[1] to decipher the plasmonic design space by learning the correlative relationship between geometry and plasmonic response. Here we demonstrate through automated and intelligent control of a converged electron beam in a scanning transmission electron microscope (STEM), monochromated electron energy loss spectroscopy (EELS) allows to retrieve nano-optical properties of as-synthesized nanoparticle geometries[2]. With this knowledge, a platform is provided for the electron beam to be controlled to dynamically modify, on the nanoscale, single or multiple nanoparticles geometrically and chemically. We demonstrate that both geometric and chemical modification lead to changes in plasmon resonance in both space and energy, and that by combining automation of beam control with the decoded design space learned from the encoder-decoder neural network, selective nano-optical properties can be realized, ultimately allowing a realistic path to quantum technologies using confinement of light at the nanoscale.

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DOSIMETRY IN TEM AND STEM

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Abstract

In the case of beam-sensitive specimens, it is desirable to know the electron dose D and the dose rate (current density J) that was used to acquire an image or spectral data. For defocused TEM illumination of diameter d , this dose is easily calculated as a fluence: J T, where T is the recording time and $J = I_b/A$, where I_b is the beam current and $A = (4/\pi)d^2$ is the irradiated area.

For small probes, the current density depends on the radial coordinate r within the probe. For a fully focused CFEG probe and assuming no lens aberrations, the current density is a diffraction-limited Airy disk: $J(r) = J(0)\ [2J_i(3.2r/d_h) / (3.2r/d_h)]^2$ where J_1 is a first-order Bessel function and d_h is the full width at half maximum. The resulting range of dose rate gives rise to several complications:

(1) If radiation damage causes an exponential dose dependence of some signal, with a characteristic dose D_c such that $S(D) = S(0)\ \exp(-D/D_c)$, the observed time dependence S(t) can be regarded as exponential only over a limited interval of exposure time T.

(2) Estimating D_c from the initial decay of the signal S(T) requires knowledge of some average current density J*, defined by $D_c = J^* T_c$ where T_c is a characteristic time. This quantity can be evaluated as $J^* = I_b/[4/\pi)(d^*)^2]$ if the probe current I_b and the effective beam diameter d* are known. For a Gaussian or Airy-disk J(r), taking $d^* = d_h$ gives rise to a substantial error but taking d* = full width at one-tenth maximum gives J* correct to within a few percent.

For sub-nm probes, some damage occurs outside the probe because of the delocalization of inelastic scattering and energy deposited by fast secondary electrons. Point spread functions for these effects have been calculated [1,2].

In the case of a raster-scanned image that is not undersampled, the average dose per frame can be estimated as $D = (I_b/A_f)T_f$, where A_f is the frame area and T_f the frame time. This approximation will not apply to undersampled or sparsely sampled images.

To allow comparison with x-ray data, the fluence D can be converted to MegaGray units: $D(MGy) = (0.1)\ S'(MeV\ cm^2/g)\ \ D(C/m^2) = E_{av}(eV)/[\lambda_i(nm)\rho(g/cm^3)]\ D(C/m^2)$, where S' is the stopping power per unit mass [3], $E_{av}(eV) \sim 7Z$ (for Z<30) is the average energy loss (in eV) per inelastic collision, $\lambda_i(nm)$ is the inelastic MFP in nm [4] and $\rho(g/cm^3)$ is the specimen density. D(MGy) should then be independent of electron accelerating voltage. However, discrepancies exist between calculated values of S' and those derived from EELS measurements of E_{av} and λ_i, suggesting a need for further investigation.

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VISUALIZING PLASMON-DRIVEN NANOPARTICLE TRANSFORMATIONS WITH OPTICALLY-COUPLED ENVIRONMENTAL TEM

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Abstract

Understanding and manipulating chemically-driven nanoparticle transformations is crucial for the efficacy of next-generation energy storage and catalytic materials. These transformations are generally dictated by macroscopic, extrinsic parameters like temperature and chemical environment. Local surface plasmon resonances (LSPRs) offer an alternative means to locally control nanoparticle dynamics due to their ability to confine light at the nanoscale. However, most techniques to track nanoparticle phase transitions have difficulty identifying differences in transient behaviour at the single and sub-nanoparticle level.

Here, we use in situ environmental transmission electron microscopy coupled with optical illumination to demonstrate how LSPRs enable spatially-modified phase transformation dynamics in nanoparticles [1]. Using the dehydrogenation of palladium hydride as our model reaction, we show how LSPRs transform a normally non-reactive facet into a reactive site. Using displaced aperture dark field imaging, we track the phase transition dynamics in individual nanoparticles under various external parameters like optical illumination and hydrogen pressure.

First, we find that without illumination, colloidally synthesized Pd nanorods undergo their phase transition starting from both tips. Next, to identify the role of the LSPR, we design a Au-Pd crossed-bar structure which creates a localized area of strong electromagnetic enhancement at the middle of the Pd nanorod, spatially separated from the favourable tip nucleation sites. Under illumination at the LSPR wavelength, Pd nanorods that are coupled to a plasmonic Au antenna demonstrate either nucleation of the dehydrogenated phase from only one tip, or nucleation from the middle of the nanorod near the electromagnetic hot spot. By varying illumination wavelength, we verify that these new dynamics are a result of LSPR excitation. Furthermore, we track the nucleation behaviour of over 20 Au-Pd pairs under resonant illumination as we vary the surrounding hydrogen pressure. Our results suggest that the middle nucleation phase transition mechanism is less energetically favourable than tip nucleation, which is further supported by molecular dynamics simulations.

Our proof-of-concept results demonstrate how in situ TEM can provide insight into new mechanisms, and provide a foundation for site-selective reactions via optical control.

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AG SEGREGATION INDUCED NANOFACETING TRANSITION OF A CU TILT GRAIN BOUNDARY AND ITS IMPACT ON PLASTIC DEFORMATION MECHANISMS

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Abstract

Structural materials contain many internal interfaces separating chemical homogenous areas of distinct crystallinity called grain boundaries (GBs). Their associated structure and energy determine material's properties and thus require profound understanding of these parameters. A few decades ago, the thermodynamical concept was introduced that GBs can undergo structural transitions either with conservation of all macroscopic degrees of freedom (congruent transitions) or by dissociating into new GB segments (faceting). However, direct experimental observations are rarely reported for metallic boundaries. Instead, indirect proof from diffusion experiments, abnormal grain growth and simulations has been mainly used to infer the presence of structural transitions, mainly in symmetric GBs. Especially transitions induced by changes of chemical composition at the grain boundary – for instance by segregation – are scarcely mentioned in literature, especially at the nano- and atomic scale. A correlation of the transition's impact on mechanical properties is not available.

We describe the observation of a nanofaceting transition of an asymmetric tilt GB in Cu upon Ag segregation by atomically resolved scanning transmission electron microscopy. The initially flat {110}/{410} boundary was found to dissociate into Ag-lean {230}/{100} asymmetric facet segments and Ag-rich symmetric {210} segments. Thus, a preferential segregation pattern of stable nanosized facets was found. Further, the influence of Ag solute excess concentration on the facet formation was studied by a diffusion couple. It was found that the asymmetric segment remains constant in size, while the symmetric facet segment increases with increasing solute excess. At the diffusion couple's interface a purely symmetric {210} boundary is found. Following the GB, short asymmetric facet segments are introduced and increase their density until a purely asymmetric GB is found close to the non-segregated reference state. Finally, we linked the observed faceting transition to a change in plastic deformation behavior by tensile straining GB containing bicrystals in situ inside the electron microscope. While pure Cu reference boundaries deform by local emission of partial dislocations resulting in the formation of a single deformation twin, the Ag segregated bicrystal shows numerous partial dislocation nucleation sites resulting in an increase of deformation twins.

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LIVE 4D-STEM PROCESSING AT 15'000 DETECTOR FRAMES PER SECOND FOR SERIES REGISTRATION

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Abstract

4D-STEM is arguably one of the current hot topics in electron microscopy. As a STEM technique it suffers from artefacts of the serial acquisition (especially at atomic resolution). However, registration of series of 4D-STEM maps has recently been demonstrated to be an efficient way to reduce those artefacts [1]. These series can account for massive amounts of data and it is difficult to estimate their quality before processing them.

We demonstrate results from live processing 4D-STEM data at over 15'000 detector frames per second using the newly developed functionality of the Nion Swift microscope software [2] and a Dectris ELA direct detector [3] mounted to a Nion HERMES microscope. Freely customizable virtual detectors are applied to the detector frames as they are streaming in and allow to display a plethora of live updating signals, like annular dark & bright fields, but also center-of-mass analysis of the data. The high frame rate allows to acquire e.g. a map of 512x512 scan points within 17s; close to the speed of traditional STEM imaging. Data sets are buffered on the hard drive and suitable maps as judged by the live output can be chosen for permanent storage. But it is also possible to only store live processing output which allows to record series that would be larger than disk space if raw data were to be saved. As the detector is mounted on the spectrometer, the data can be easily zero-loss filtered.

This functionality allows to efficiently acquire high quality series of 4D-STEM maps that can then be registered in post-processing. Application to different material systems will be shown and effects of energy filtering and different mask sizes for center-of-mass analysis will be demonstrated.

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MACHINE LEARNING ELECTRON DIFFRACTION

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Abstract

Four-dimensional diffraction datasets (4D-DDs) collected using the scanning electron nanodiffraction (SEND) [1] or 4D scanning transmission electron microscopy (4D-STEM) [2] techniques are getting increasingly popular in the electron microscopy community for their versatility in both electron diffraction and imaging and facilitated by the recent development of fast electron detectors. Finding faster and better ways to extract information from large 4D-DDs is now one of the major focuses to further advance this field.

Machine learning (ML) has revolutionized scientific research in recent years. In computer vision, deep learning is used to solve difficult problems, such as detection, classification, and segmentation [3]. In the field of electron microscopy, deep learning has been applied to, or proposed for, crystallographic analysis of electron image and diffraction data, single atom detection, automatic learning of microstructural features, etc.

Here, we describe three examples of using machine learning to obtain quantitative information about crystal lattice from 4D-DDs. The first is train measurement by using convolutional neural network (CNN) to measure diffraction disk positions in electron nanodiffraction patterns [4]. The precision and computing efficiency are demonstrated in the measurement of the strain fields in a Si-based FinFET device. The results are comparable with those calculated by the circular Hough transform method [5] and have better precision (~0.04%) in some cases.

The second example is precision crystal orientation mapping using a trained artificial neural network (ANN), where we determine small changes in crystal orientation within a crystalline grain [4]. We design a simple ANN model to be trained on the simulated dynamical diffraction patterns. A high angular resolution of 0.01° or better is achieved for precision orientation mapping, which we demonstrate on both single crystalline GaSb thin sample and polycrystalline UO_2 after irradiation.

The third type of ML based analysis uses CNN to classify different types of diffuse scattering in coherent electron nanodiffraction patterns. The training data for the CNN are labelled by Cepstral STEM imaging [6], where diffuse scattering due to severe lattice distortion is highlighted. Once trained, the CNN can be applied to large 4D-DDs for defect imaging and sensitive to hidden defects invisible to high-resolution imaging.

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OFF-AXIS HOLOGRAPHY OF POLARIZATION FIELDS IN INGAN/GAN WITH MONOLAYER RESOLUTION

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Abstract

Typical quantum wells (QW) in InGaN/GaN light emitting devices are around 2 nm wide and extend in the basal plane. In a 200 nm thick specimen a tilt of 10 mrad away from on-edge orientations already leads to a broadening of interfaces of 2 nm due to projection effects, thus is already too large to allow the investigation of such thin quantum wells. This forbids the observation of these quantum wells under kinematic conditions, since these require the specimen to be tilted by larger angles; for comparison the diffraction angle of (0002) beams at 300 keV is at 7.7 mrad.

We present off-axis electron holography on multiple 2.2 nm wide $In_{0.15}Ga_{0.85}N$-QW embedded in a GaN matrix under (0002) systematic row conditions, where the QW are edge-on. The holograms are recorded with a Cs-corrected setup making use of the additional lenses in the corrector as well as the projective system [1]. Additionally, the variations in dynamical diffraction are probed by systematically tilting the incident beam along the systematic row in a very limited angular range. An anisotropic reconstruction mask allows to preserve the high-resolution information up the (0002) reflections needed for monolayer-resolved measurements of the electrostatic potential, while simultaneously sufficient wide holograms of >200 nm are achieved to overcome artefacts resulting from the specimen edges.

Since the holograms are recorded under strongly diffracting conditions, we additionally performed multislice calculations for a range of incident beam tilts and specimen thicknesses [2]. Strain relaxation effects at the specimen surfaces were included in the simulation by obtaining atomic displacements from finite element strain calculations performed for each simulated thickness. The effect of the polarization fields were also included into the simulation.

Based on the exit wave calculations and experimental results we discuss the robustness of various measures, which can be used for estimation of the polarization field strength. It turns out that the phase step across the QW is a far superior estimate for the field strength than the slope of the exit wave's phase in the region of the QW. Especially, projection effects and limited instrumental resolution (e.g. as found for holography of such thin QW using the Lorentz lens) make the measures of the slope highly inaccurate.

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TOWARDS MEASUREMENT OF POLARISATION-INDUCED ELECTRIC FIELDS IN $PbZr_{0.2}Ti_{0.8}O_3$ BY MOMENTUM-RESOLVED STEM – IMPACT OF SYSTEMATIC ERRORS

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Abstract

Ferroelectric tunnel junctions are a focus of next-generation memories where polarisation represents a logical bit. (S)TEM methods that allow for quantitative mapping of polarisation-induced electric fields are desirable to improve such devices.

The advent of ultrafast cameras in STEM allows for high momentum and spatial resolution in aberration-corrected STEM. Especially, momentum-resolved STEM (MR-STEM) has enabled first moment-based imaging of subatomic electric fields in thin specimens [1,2] and electron ptychography [3]. Recently, polarisation-induced fields in semiconductors were mapped by unit-cell averaged MR-STEM [4]. The method assumes that long-range components of the electric field are dominant over atomic ones when averaging first moments over unit cells. The validity of this assumption depends on crystal symmetry and experimental parameters such as specimen thickness or tilt. This leads to a systematic error δ which we address here for polarisation mapping in $PbZr_{0.2}Ti_{0.8}O_3$ (PZT) in both experiment and simulation.

Experimental MR-STEM data from PZT has been evaluated with different methods and compared to comprehensive simulations. Using an isolated atom model with periodic boundary conditions in STEMsim [5], all non-zero first moments in unit-cell averages arise from dynamical scattering and contribute to δ, hence being caused by symmetry breaking due to atom displacements.

Experimental results from PZT were recorded with an aberration-corrected Titan at 300kV and a Medipix camera with a sample thickness around 10nm determined by comparing diffraction patterns with simulations. The formally calculated projected atomic electric fields cannot be related to the electric field directly due to dynamical scattering, but the atom positions can be resolved. In the unit cell average, the domains as expected from [6] can be seen. This matches the result of structure evaluations. A single domain seems to have an electric field around 90MV/cm. The systematic error is below 40MV/cm taking mistilt of up to 2mrad into account. This and further experimental results are critically examined by comprehensive simulations addressing mistilts and thickness dependencies.

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EFFECT OF PROBE DEFOCUS IN MOMENTUM-RESOLVED STEM EXPERIMENTS

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Abstract

With the recent implementation of fast electron cameras, the recording of scanning transmission electron microscopy (STEM) data with momentum resolution was enabled in dependence of the scan position, thus increasing the number of measurement dimensions to four. This trend can be pursued further by implementing the automated acquisition of focal series momentum-resolved (MR) STEM [1]. In that manner, the individual defocus-dependence [2,3] of both conventional and 4D-specific STEM signals, such as the first and second moments in diffraction space, may be investigated.

Using an aberration-corrected FEI Titan 80-300 microscope equipped with a Medipix camera [4], focal series MR STEM experiments are performed using an α-In_2Se_3 specimen. Alongside the high-angle annular dark field (HAADF) signal recorded using a dedicated Fischione detector and displaying Z-contrast, various STEM signals are extracted from the five-dimensional data. This includes signals mostly made of elastic and coherent components, such as the virtual bright field (BF) and annular bright field (ABF) imaging modes, but also the first moment, relevant for its direct relationship to the projected electromagnetic fields experienced by the electrons while going through a thin specimen [5], as well as the second moment. For each signal, image contrast can be estimated by calculating the standard deviation within the field of view, and plotted as a function of probe defocus, thus allowing to detect local maxima on the defocus axis. Comprehensive multislice simulations are performed to interprete the results.

As is observed both from experiment and simulation, STEM signals obtained by exploiting different regions of diffraction space possess a unique dependence to defocus. This is therefore a cause of mismatch in the defocus of maximum contrast observed for distinct STEM images exploited in a single analysis, which can be a source of error in such a multiple-signal approach. It is also shown that certain signals displaying two local contrast maxima, e.g. the second moment, can yield geometrical information such as the vertical positions of the interfaces between the specimen and vacuum. Findings are discussed with respect to several experimental imperfections such as mistilt, carbon contamination, aberrations and partial temporal coherence.

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FABRICATION OF THREE ELEMENT BOERSCH PHASE SHIFTERS AND THEIR CHARACTERIZATION BY VOLTAGE-CONTROLLED INTERFERENCE OF COHERENT HIGH ENERGY ELECTRON BEAM

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Abstract

Electron wavefront manipulation has been actively studied recently using static masks or using voltage-controlled devices [1]–[5]. Among those, multi-element Boersch phase shifter devices fabricated with scalable methods are promising candidaes to realize a programmable holographic synthesis of electron waves in two- and three-dimensions. The target applications include the phase modulation imaging and the aberration correction [1] to name a few. Recently, we reported the fabrication of a three-element Boersch phase shifter device and its successful operation in a simplified design without the top-shielding electrode. The devices were fabricated with electron beam lithography and dry etching processes that will easily be applied for devices with a large number of elements. However, the experiment demonstrated the beam deflection induced by exposed metal contact wires. Here, we report the fabrication of the three-element Boersch phase shifter including the top electrode. The experiment in a 200 keV transmission electron microscope demonstrated the voltage-controlled three-electron beam interference, wherein the parasitic beam deflection was significantly suppressed. Therefore, the device structure is compatible for a programmable multi-element device, a step forward to the arbitrary synthesis of electron wave.

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LOCALIZED SURFACE PLASMON RESONANCES IN SIERPIŃSKI FRACTAL NANOANTENNAS

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Abstract

Localized surface plasmon resonances (LSPR) are used to amplify the signals from small amounts of analytes in surface-enhanced Raman spectroscopy (SERS), improving detection limits down to the single molecule level [1]. Design of SERS substrates optimized for different tasks is still an active and ongoing field of research. For flexible platforms, fractal nanostructures present themselves as strong candidates based on their multiband response arising from their hierachical geometry [2,3].

We study Sierpiński fractal antennas of increasing fractal generation to understand the behaviour of the LSPR modes as the fractal generation, and thereby the geometrical complexity of the structure, is increased. We use electron beam lithography to fabricate our silver fractals, then characterize the LSPR response using monochromated electron energy loss spectroscopy in an FEI Titan microscope, complemented by finite-difference time-domain simulations.

A simple equilateral triangle (Generation 0 fractal) supports multipolar edge and cavity modes. As the fractal generation is advanced to Generation 1, by adding a single triangular aperture in the centre, the aperture causes energy shifts in some of the LSPR peaks and changes to the nanoscale electric field distribution of others. We are able to understand this behaviour in terms of the disturbance to the charge-current configurations of each unique mode, caused by the introduction of the aperture, and by coupling of the aperture to the modes of the equilateral triangle [2,4]. Interactions between the LSPR and additional apertures in successive fractal generations cause, for example, redshifting of the dipolar mode and modifications to the field configurations of the triangle's cavity modes.

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ANOMOLOUS CONTRAST IN HAADF STEM IMAGING OF DYSPROSIUM SCANDANATE

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Abstract

When recording scanning transmission electron microscopy (STEM) data from epitaxial thin films of $LaVO_3$ grown, under biaxial tension, on the (1 1 0) orthorhombic surface of $DyScO_3$ substrates [1], a surprising observation is made. If imaging on the [1 1 0] pseudo-cubic zone axis – corresponding to the $[1\ \bar{1}\ 1]$ and [1 0 0]/[0 1 0] orthorhombic axes of the $DyScO_3$ and $LaVO_3$, respectively – the high angle annular dark-field (HAADF) intensity of the $LaVO_3$ is equal to, or even greater than, that of the $DyScO_3$, despite the greater average atomic number of the latter (Z_{av} of 22.2 for $DyScO_3$ vs 20.8 for $LaVO_3$). This effect becomes more pronounced as the inner angle of collection on the HAADF detector is increased even though, according to general principles, this should move the recorded scattering intensity further towards a "*Z*-contrast" behaviour where image intensity $I \propto Z_{av}^{1.6-2}$. We reveal that the reason behind this anomalous contrast relates to the first order Laue zones (FOLZs) of the two materials. When imaging with a 300 keV electron beam on these zone axes, the $LaVO_3$ has a FOLZ of weak intensity at ~82 mrad scattering angle. In contrast, the $DyScO_3$ has a FOLZ at a lower angle of ~48 mrad, and having a remarkably strong intensity. Associated with this, for scattering angles higher than the FOLZ of $DyScO_3$, the $DyScO_3$ shows a lower scattered intensity than the $LaVO_3$, as observed by plotting radial distribution functions of position averaged convergent beam electron (PACBED) diffraction patterns from the two phases. When the HAADF detector inner angle is increased to select only this scattering regime, the anomalous contrast is therefore observed. Frozen phonon based PACBED and HAADF STEM simulations made using Dr. Probe confirm this result [2]. In order to study the partition of scattering, simulations are also made using the quantum excitation of phonons model of μSTEM [3]. It is found that, fundamentally, an intense *elastic* scattering of $DyScO_3$ into its FOLZ correlates into a net reduction of *inelastic* phonon-derived "thermal diffuse scattering" across the full angular scattering range.

The lower angle FOLZ of the $DyScO_3$ results from a doubling of the unit cell in the zone axis direction, as related to the tilt-rotation patterns of the oxygen octahedra of the *Pnma* perovskite structure [4]. STEM and PACBED data show that a 9–10 unit cell thick layer of the $LaVO_3$ at the film-substrate interface shares the same anomalous contrast and lower angle FOLZ as the substrate. Further studies suggest that this unexpected result is from a shared structural nature, driven by the transition of the in-phase octahedral rotation axis from being in-plane in the $DyScO_3$ substrate to out-of-plane in the $LaVO_3$ film bulk.

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REAL-TIME INTERACTIVE PTYCHOGRAPHY FROM ELECTRON EVENT REPRESENTATION DATA

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Abstract

The arrival of direct electron detectors (DED) with high frame-rates in the field of scanning transmission electron microscopy has enabled many experimental techniques that require collection of a full diffraction pattern at each scan position, a field which is subsumed under the name 4D-STEM. DED frame rates approaching 100kHz place stringent requirements on data transmission rates and computing infrastructure.

Current commercial DEDs allow the user to make compromises in pixel bit depth, detector binning or windowing to reduce the per-fame file size and allow higher frame rates. This change in detector specifications requires decisions to be made before data acquisition that may reduce or lose information that could have been advantageous during data analysis.

The 4D Camera, a DED with 87kHz frame-rate developed at Lawrence Berkeley National Laboratory, reduces the raw data to a linear-index encoded electron event representation (EER).

In this poster, we show with experimental data from the 4D camera that linear-index encoded EER for 4D-STEM phase-contrast imaging methods enables real-time, interactive phase-contrast from large-area 4D-STEM datasets.

We detail the computational complexity advantages of the EER and the necessary computational steps to achieve real-time interactive ptychography using commonly available hardware accelerators.

Real-time reconstruction with direct phase-contrast imaging methods allows fast data exploration during the experiment and generation of an accurate initial guess for probe aberrations in iterative reconstruction methods.

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[2] PMP acknowledges financial support from STROBE: A National Science Foundation Science & Technology Center under Grant No. DMR 1548924

REAL-TIME MEASUREMENT OF CHANGES IN ELECTROSTATIC POTENTIAL IN INDIVIDUAL TiO_2 NANOPARTICLES DURING RESISTIVE SWITCHING USING OFF-AXIS ELECTRON HOLOGRAPHY

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Abstract

Resistive switching phenomena in complex metal oxides have attracted great interest for applications in next-generation non-volatile memory devices. Recently, nanoparticles have been adopted as switching materials, both for use in smaller devices and as model systems for understanding resistive switching mechanisms in nanoscale materials [1,2].

Here, we investigate the origin of resistive switching behavior in individual nanoparticles that have an electrical bias applied to them *in situ* in the transmission electron microscope (TEM). We use current-voltage (I-V) measurements in the TEM to study resistive switching mechanisms in real time, while simultaneously applying off-axis electron holography to the same particles. The technique allows projected electrostatic potential distributions in materials to be recorded with nm spatial resolution [3], providing information about the role of heterointerfaces and conducting nanofilaments during switching processes.

We study TiO_2 nanoparticles that were synthesized using a sol-gel method and then vacuum annealed. We observe reproducible forming-free bipolar switching in individual nanoparticles that are contacted electrically in the TEM using a moveable electrical probe. I-V curves obtained from single nanoparticles are correlated with electrostatic potential measurements. Comparisons with simulations suggest that a conducting path is created close to the interfaces of the nanoparticle with both electrodes during soft forming, while the subsequent formation and annihilation of a conducting path at one interface is primarily responsible for SET and RESET processes.

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