**Cs-TEM vs Cs-STEM**

Duncan Alexander  
EPFL-CIME

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**FEI Titan Themis @ CIME EPFL**

- 60–300 kV
- Monochromator
- High brightness X-FEG
- Probe Cs-corrected: 0.7 Å @ 300 kV
- Image Cs-corrected: 0.7 Å @ 300 kV
- Super-X EDX detector
- GIF Quantum ERS energy filter
- Dual-channel, Ultrafast STEM-EELS  
  - Lorentz mode  
  - Biprism for holography  
  - Piezo stage  
- Tomographic acquisition
Limitation to spatial resolution: aberrations

Electromagnetic lenses in TEM column are toroidal
Lenses inherently convergent
=> spherical aberration ($C_S$) and chromatic aberration ($C_C$)

Resolution in HR-TEM limited by aberrations, especially $C_S$

Cs-correction (STEM and TEM)
Combination of standard radially-symmetric convergent lenses with multipole divergent lenses (e.g. tetrapoles, hexapoles) to tune $C_S$
Principle of aberration correction

- Compensate Cs and other distortions with equivalent but opposite components to add together with aim of giving ideal spherical wavefront

![Diagram showing trajectory and aberration function contributions in a quadrupole-octupole corrector.](image)

CEOS aberration corrector

- CEOS aberration corrector used for imaging correction in CTEM also used before sample as probe-corrector for STEM; sextapole-round lens-sextapole design. This is an “indirect” corrector type; ~30 power supplies but higher power and water cooling needed.

Indirect-action correctors, in which the aberration to be corrected is acted on by a combination of aberration arising as a by-product of a lower-order aberration intentionally created in the corrector. The undesirable lower-order aberration coefficients are typically of the same order of magnitude as the coefficients of the aberrations to be corrected. For the small angles used in electron microscopy, this means that the effect of the lower-order aberrations on the beam are very strong. These aberrations, therefore, need to be canceled very precisely, typically by using an equivalent optical element that cancels the undesirable low-order effect while at the same time increasing the desirable higher-order effect.
Nion aberration corrector

- First STEM aberration corrector installed on VG by Nion (Krivanek); quadrupole-octopole design. This is a “direct-action” corrector type as now used on Nion UltraSTEM: ~70 power supplies needed but low power and which can fit onto printed circuit boards.

Current correctors

- CEOS: C₅-corrected, “C₅ optimised”: CETCOR, CESCOR, D-COR
- CEOS: C₅-C₅ corrector (NCEM TEAM 1.0, Julich Titan Pico)
- CEOS: B-COR aplanatic optimised for far off-axis rays
- JEOL: unique C₅-C₅ corrector (CCC project)
- JEOL: Dodecapole C₅ corrector (“Grand ARM”)
- Nion: C₅-C₅ corrected
Understanding resolution in EM

• For Cs-TEM need to understand concepts of:
  • Contrast transfer function (CTF)
  • How to use CS to optimise CTF
  • Difference between point resolution and information limit
  • Properties of the camera (MTF), sample drift, “Stobbs factor”…

• For Cs-STEM need to understand concepts of:
  • Probe size, shape, brightness, depth of field (DOF)
  • Optical transfer function (OTF); STEM first to achieve 0.5 Å res
  • Scan (in)stabilities, detectors

Cs-corrected HR-TEM “interferometry”

Example: Σ3 grain boundaries in Al

Uncorrected

Cs-corrected

Images: Oikawa, JEOL

Reduced delocalisation in phase contrast image
CTF curves: uncorrected microscopes

- Negative phase of CTF $\Rightarrow$ black atom contrast (as in JEMS)

Cs-TEM: effect on CTF

- Higher information limit from shifting spatial and temporal envelopes
- Done by improved stability of instrument + monochromatic beam
- Here show negative Cs ("white atom" contrast)
- Adjust Cs, defocus to give one wide CTF pass band to information limit
Cs-TEM example: (Al$_x$Ga$_{1-x}$)As nanowire

Sample courtesy of Yannick Fontana, Anna Fontcuberta-i-Morral, LMSC

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Cs-TEM [1 1 0] GaAs simulation

Defocus: – 2 nm steps starting from +5 nm

 Cs-TEM: defocus effect on CTF

• Small change in defocus $\Rightarrow$ large change in contrast of high spatial frequencies!

300 kV Titan:
- Cs: –10 μm (adjustable!)
- Defocus: -4.4 nm
- Point resolution: ~0.7 Å
- Information limit: 0.7 Å

300 kV Titan:
- Cs: –10 μm (adjustable!)
- Defocus: 3.6 nm
- Information limit: 0.7 Å

300 kV Titan:
- Cs: –10 μm (adjustable!)
- Defocus: -0.4 nm
- Information limit: 0.7 Å
Cs-STEM example: (Al$_x$Ga$_{1-x}$)As nanowire

HAADF imaging: directly interpretable contrast on atomic structure; camera-like focus and no delocalisation!

Benefits of aberration correction
Analytics – STEM-EELS
Atomic resolution core-loss STEM-EELS mapping (Nion UltraSTEM)

More recently: atomic resolution EDX, EFTEM – but are they as interpretable?

Imaging organic molecules – Cs-(S)TEM
Cs-TEM (80 keV beam): imaging of molecule as weak phase object

Cs-STEM (200 keV beam): imaging of molecules by HAADF; need very clean (un-contaminating) sample
Measurement precision – Cs-TEM

Is science prepared for atomic-resolution electron microscopy?
Knut W. Urban

The efforts of microscopists have given aberration-corrected transmission electron microscopy the power to reveal atomic structures with unprecedented precision. It is now up to materials scientists to use this power for extracting physical properties from microscopic atomic arrangements.

Measuring precision – Cs-STEM

Mapping Octahedral Tilts and Polarization Across a Domain Wall in BiFeO₃ from Z-Contrast Scanning Transmission Electron Microscopy Image Atomic Column Shape Analysis

Albrecht Böttcher,1,2 Oleq S. Ovchinnikov,3 Hye Jung Chang,3 Mark P. O’Dea,2 Pu Yu,1 Jan Siedel,1 Eugene A. Efremov,1 Anna N. Morozova,1 Ramamurthy Ramesh,1 Stephen J. Pennycook,1 and Inge V. Kalia1

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The move to lower kV

- Before Cs-correction highest resolution by minimising $\lambda$ (MeV instruments with $\lambda < 1$ pm)
- Light materials (graphene, nanotubes, ...) suffer knock-on damage. Some thresholds:
  - Bulk graphene: 86 keV
  - Graphene edge atom: 36 keV
- Therefore need low kV – 80 kV max but 60 kV better – which have long wavelengths
- Aberration correction now mandatory for atomic resolution
- Notable projects: Suenaga CCC project (30 kV aim), Ute Kaiser’s Salve project (20 kV aim), both with combined Cs-Cc correctors; new UltraSTEM (20 – 100 kV range)

Doped graphene, BN monolayer – Cs-STEM

- Analysis of monolayer materials: low kV essential to prevent knock-on damage; here 60 kV used (knock-on threshold for bulk graphene ~86 kV) with Nion UltraSTEM
- Medium-angle ADF (MAADF) gives intensity $I = Z^{1.7}$ but with increased signal intensity compared to true HAADF image. (This intensity is needed for imaging single atom by single atom; $\beta = 58–200$ mrad.) Direct atom assignment by intensity.
Doped graphene, BN monolayer – Cs-TEM

Experimental analysis of charge redistribution due to chemical bonding by high-resolution transmission electron microscopy

We have shown that it is possible to obtain insights into the charge distributions in nanoscale samples and non-periodic defects from HRTEM measurements. For our examples of the nitrogen substitutions in graphene and hBN layers, we can assign experimentally observed contrast features to details in the simulated electron distribution. We can detect a single nitrogen substitution atom in graphene, which is possible only because of the electronic effect. In the case of hBN, the charge redistribution leads to a loss of the elementary contrast difference. Instead, the ionic character of the material is experimentally confirmed for the single layer. One key ingredient here is the extraordinary stability of the samples under the low-voltage electron beam, which allows us to obtain extremely high-signal-to-noise ratios from long exposures. The precisely defined, ultrathin sample geometry enables a straightforward analysis. The DFT-based TEM image calculation is irreplaceable for the interpretation of experimental results in these materials, and can provide insights beyond the structural configurations.

Cs-TEM of dislocations in graphene

Dislocation-Driven Deformations in Graphene

The movement of dislocations in a crystal is the key mechanism for plastic deformation in all materials. Studies of dislocations have focused on three-dimensional materials, and there is little experimental evidence regarding the dynamics of dislocations and their impact at the atomic level on the lattice structure of graphene. We studied the dynamics of dislocation pairs in graphene, recorded with single-atom sensitivity. We examined stepwise dislocation movement along the zigzag lattice direction mediated either by a single bond rotation or through the loss of two carbon atoms. The strake fields were determined, showing how dislocations deform graphene by dislocation and screw dislocation, and lattice rotations.

Fig. 2. Strain field mapping. (A) HRTEM image of a dislocation pair in graphene. GPA was applied to the HRTEM image in (A) to determine the phase map, (IC) [010] [001] [010] and (IF) rotation (in radians). The color scale for (IC) to (IF) is shown in IC, with a range of 0.3 to 0.4 V (V1 = c.1 V). For the phase map in (IF), the color map is black to white (G).
Studies of monolayer MoS$_2$

2010: Cs-TEM, 80 kV, TEAM 0.5 microscope

Imaging MoS$_2$, Nanocatalysts with Single-Atom Sensitivity**

Christian Kielovski, Quentin M. Ramasse, Lars P. Hansen, Michael Brener, Anna Carlson, Alistair M. Mclean, Henrik Tappe, and Sig Dalong

Figure 2. a) Reconstructed phase of the aberration-corrected exit wave.

Figure 3. A close-up of the phase image obtained from the region near the bottom edge of the MoS$_2$ nanocrystallite (partly contained in the white box of Figure 2). A red-green-blue color scale was applied to improve readability. The upper region is assigned to a double-layer MoS$_2$ slab, while the lower region is a single-layer slab. More generally, the chemical composition of individual columns can unambiguously be determined. A kink from a Mo$+2$ column to a Mo column is indicated along the surface step from the double-layer to the single-layer MoS$_2$.

Studies of monolayer MoS$_2$

2011: Cs-STEM, 60 kV, SuperSTEM

Atomic-Scale Edge Structures on Industrial-Style MoS$_2$, Nanocatalysts**

Lars P. Hansen, Quentin M. Ramasse, Christian Kielovski, Michael Brener, Erik Johnson, Henrik Tappe, and Sig Dalong

Figure 4. High-resolution STEM image of a MoS$_2$ nanocrystallite supported on a graphite support. The inset is a fast Fourier transform (FFT) of the image and shows hexagonally arranged spots at the 0.27 nm and 0.16 nm lattice distances, corresponding to the MoS$_2$ (100) and (110) lattice planes, respectively. Moreover, a hexagonal set of lattice distances at 0.21 nm, corresponding to graphite (100), is also revealed. At the low-indexed edges, line segments may be present as indicated by white circles.

Figure 5. a) Ball models of the MoS$_2$ structure with Mo and S atoms indicated. The Mo atoms are colored for improved visibility. The arrow points to a single sulfur atom that terminates the Mo edge as designated by an intensity analysis. b) Ball models (top and side views, respectively) of different...
Titan Themis Cs-STEM: CVD monolayer MoS$_2$

“Large-area MoS$_2$ grown using H$_2$S as the sulphur source”
Dumitru Dumcenco et al. 2D Materials 2(4) 2015

- 80 keV beam; even if below knock-on threshold can have beam-induced chemistry with residual gas molecules in column because not UHV (e.g. water etching).
- UHV or sample heating can be essential to good work!

Other limits
**Cs-TEM**

- Harder to align precisely on zone axis (need to flip from diffraction to image)
- Interpret via: focal series reconstruction; negative Cs imaging; simulation
- Easy to obtain fringe image but precise Scherzer focus potentially challenging
- Contrast inversions with thickness remain; but can image very thick samples
- Damage: beam intensity spread, but total dose may be higher
- Coherent imaging: CTF determines resolution limit
- Atomic column analytics with (Cc-corrected) EFTEM less proven
- Camera properties important (MTF, “Stobbs factor”)
- Picometer measurement precision
- Dynamics studies 25 fps easy, 1000 fps now possible (good for ETEM)
- Can still image samples which contaminate, e.g. organic molecules

**Cs-STEM**

- Easier to align precisely on zone axis (always in diffraction mode)
- Interpret via HAADF/MAADF/BF/ABF/iDPC image
- Very limited DOF but very precise focus; camera-like focus
- Arguably thickness insensitive: sample first nms of thickness
- Damage: strong local intensity, but total dose may be lower
- Incoherent imaging: OTF determines resolution limit for HAADF
- Atomic column analytics with STEM-EELS; STEM-EDX also works
- Scan instabilities and detector noise important; need very stable scan
- Equally good precision
- Slower, but possible to follow movement of single atoms
- Need contamination-free samples only UHV possibility

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