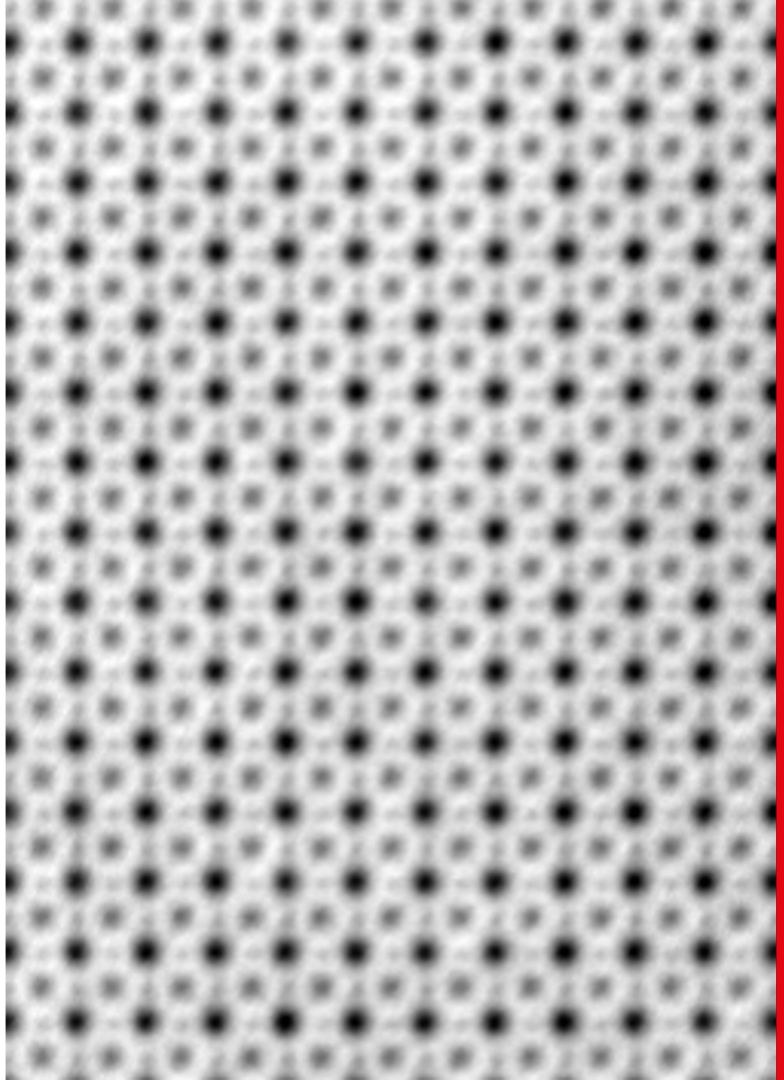


Transmission Electron Microscopy (TEM)

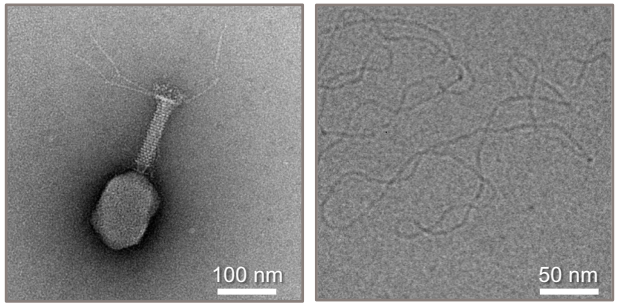
CCMX Summer School 2023

Emad Oveisi

Applications of TEM

Biology and life science

a

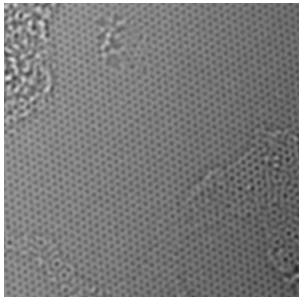


T4 virus [a]
A bacteriophage that infects Escherichia coli bacteria

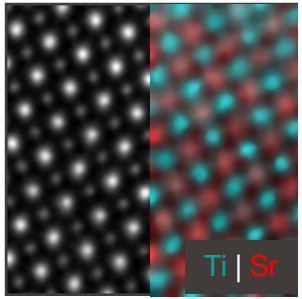
DNA molecule [a]

Chemistry & materials research

c



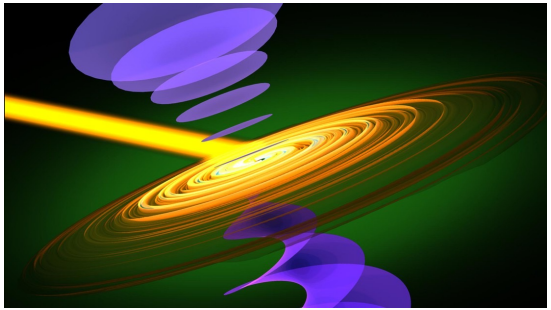
d



Imaging and spectroscopy at atomic resolution [c,d]

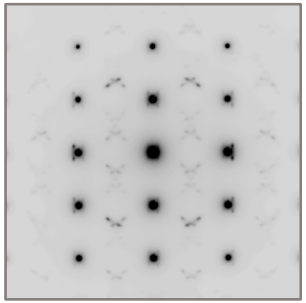
Fundamental physics

b



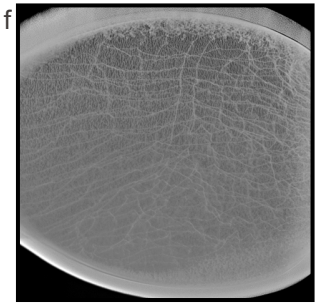
Ultrafast electron vortex beams [b]

e



Crystallographic studies [e]

f



Analyzing crystal defects [f]

[a] Images courtesy of Dr. D. Demurtas, CIME-EPFL.
[b] Vanacore et al., Nature Materials (2019).

[c] Huang et al., Nat. Comm. 2018.
[f] Oveisi et al., Ultramicroscopy (2017).

[e] Oveisi et al., Scripta Mat. (2013).
[d] Bencan et al., Nat. Comm. (2021).

Contents

Introduction to TEM

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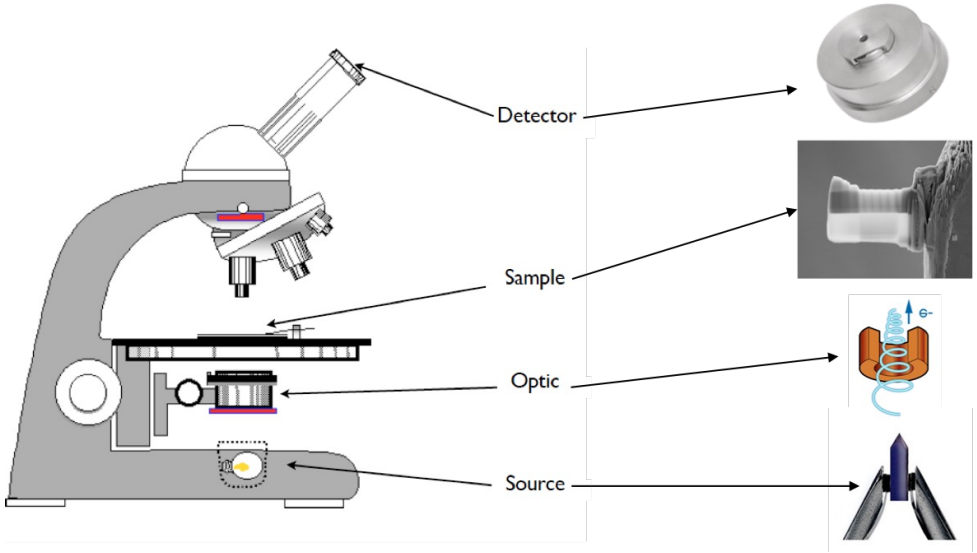


Electromagnetic lenses with variable focus

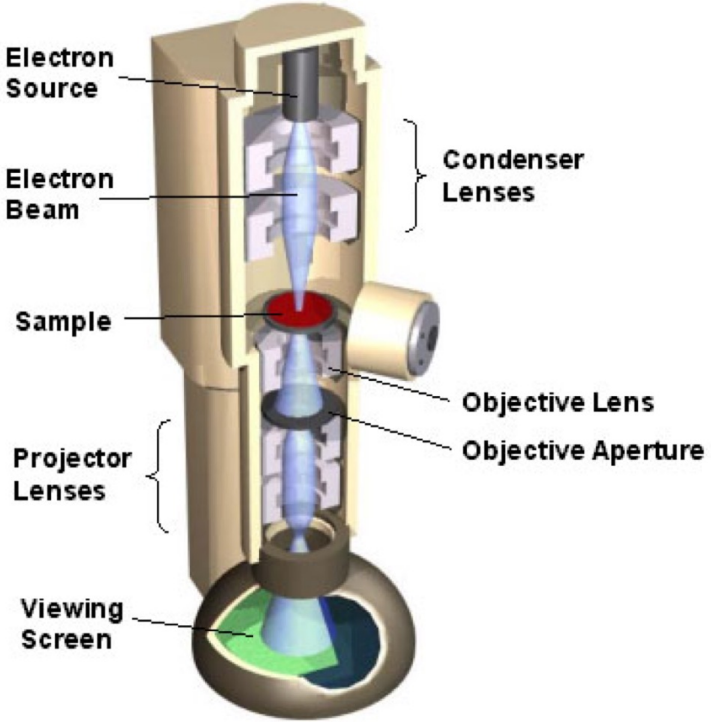
Visible light

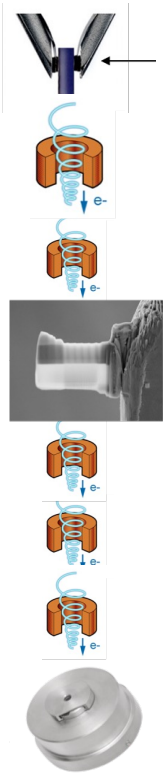
Fast electrons
100–300 keV
(0.5–0.8 *c* !)

Wave-particle duality

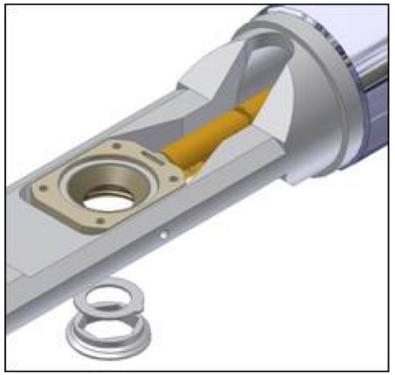
Can consider analogous to projection light microscopy, but with better resolution

Introduction to TEM





Double Cs-corrected FEI Titan-Themis @ CIME-EPFL



Specimen should be electron transparent: several nanometers thick

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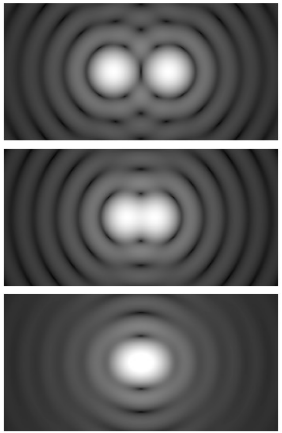
Why fast electrons?

The resolution of an optical microscope is defined as the minimum distance between two point sources (e.g. objects) such that their presence can be distinguished in the image.

Abbe's definition of maximum resolution of an optical system states that the smallest feature resolved is limited by diffraction.



Airy Diffraction Disks

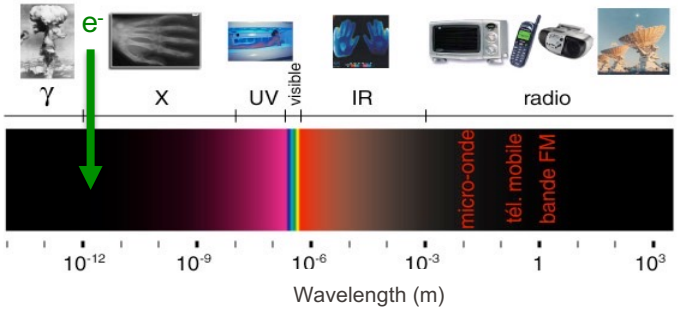
Visible light: λ ≈ 300-700 nm → resolution around half of the λ

Electrons: λ = h . c / E : Wave-particle duality

@ 200 keV: λ = 0.025 Å << interatomic spacing*

$$r = \frac{1.22\lambda}{2\sin\theta} \approx \frac{0.61\lambda}{\theta}$$

For the 200 keV TEM around < 1 Å resolution possible



*Aberrations & instabilities ➔ Actual resolution around 25-50 λ, but atomic/sub-Å resolution possible in TEM

Why fast electrons?

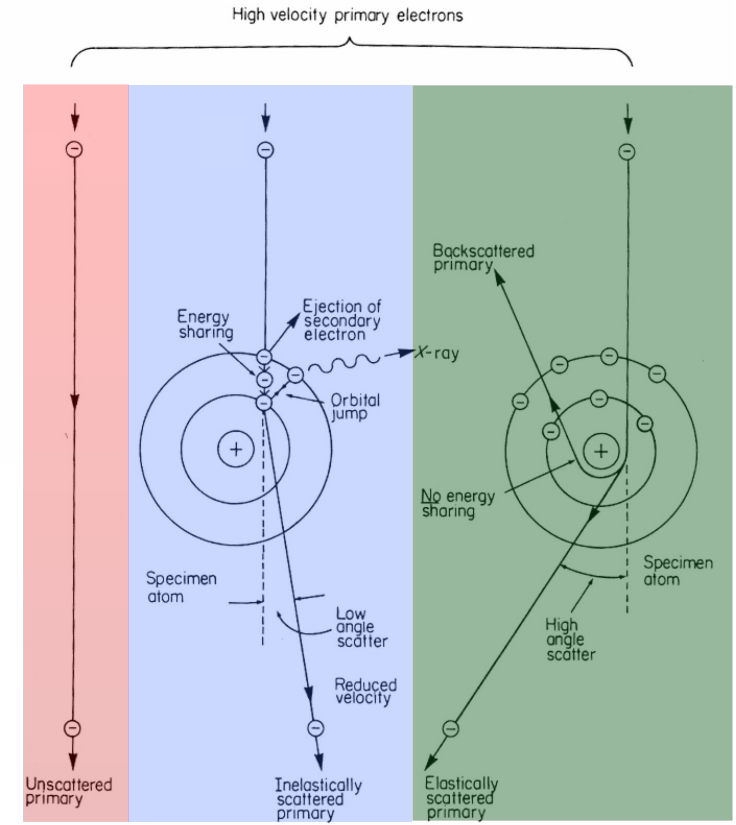
	Advantageous	Disadvantageous
Visible light	Not very damaging Easily focused Eye detector	Long wavelengths (400 nm)
X-rays	Small wavelength (Angstrom) Good penetration	Hard to focus Damage sample
Neutrons	Low sample damage Small wavelength (pm)	How to produce? How to focus?
Electrons	**Small wavelength (pm)** **Can be focused to a sub-Å size probe** **Wave-particle duality**	**Damage sample** **Poor penetration** (< x00 nm)

High energy electrons have a short wavelength
Easy to produce high brightness electron beams
Easy to manipulate: focused
Interact strongly with matter

Electron microscopes are used not only for obtaining good resolution images but also:

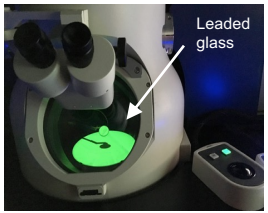
- **can be used as a diffractometer (TEM and EBSD)**
- **for chemical analyses (SEM and TEM)**
- **for imaging/measuring strain field in the sample (SEM and TEM)**
- **etc.**

e- matter interactions



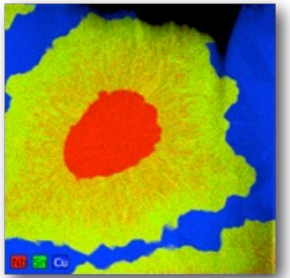
Inelastic events: The result is a **transfer of beam energy** to the specimen atom (**energy loss**) and a potential expulsion of an electron from that atom as a **secondary electron (SE)**.

If the vacancy due to the creation of a secondary electron is filled from a higher level orbital, an X-Ray or Auger characteristic of that energy transition is produced.



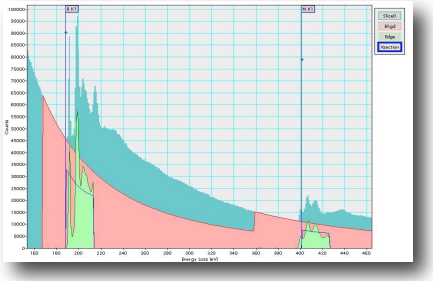
That's why TEMs are shielded!

Characteristic X-rays



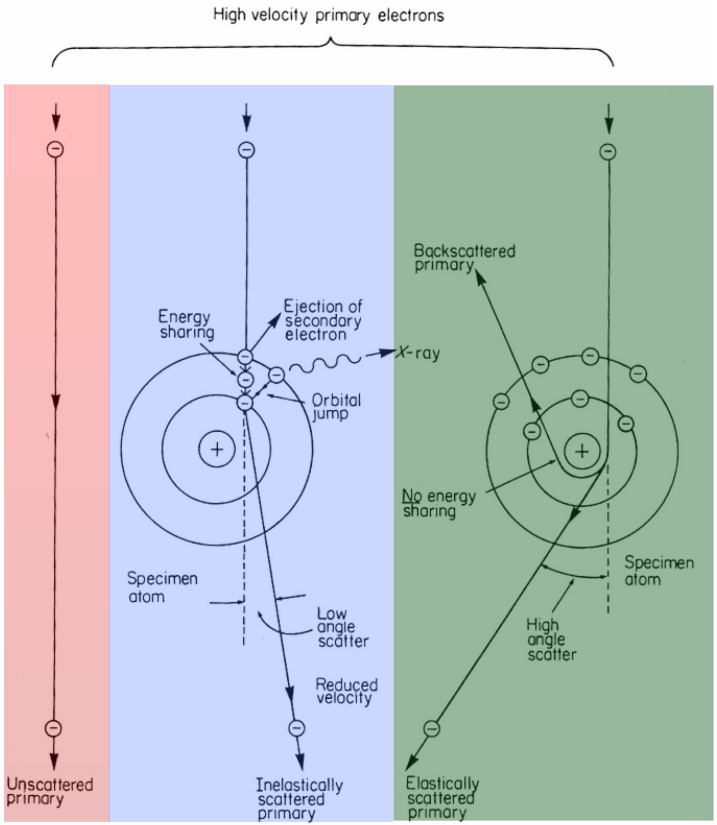
Chemical composition

Electron energy-loss spectrum



Chemical composition, band-gap & optical properties

e- matter interactions

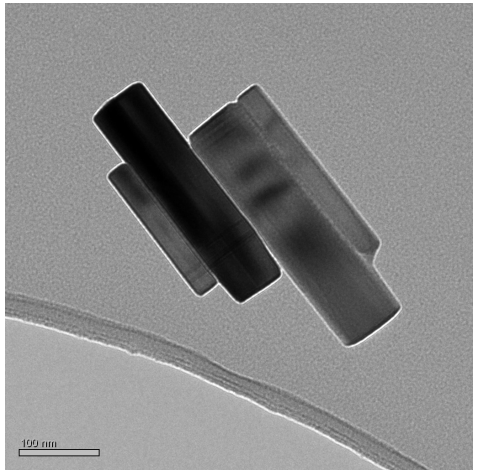


Elastic events occur when a beam electron interacts with the electric field of the nucleus or electron cloud of a specimen atom (Coulomb forces), resulting in a change in the direction of the beam electron **without a significant change in the energy** of the beam electron (< 1 eV).

Coulombic interaction within the electron cloud, Low-angle scattering
Coulombic attraction by the nucleus, Higher-angle scattering

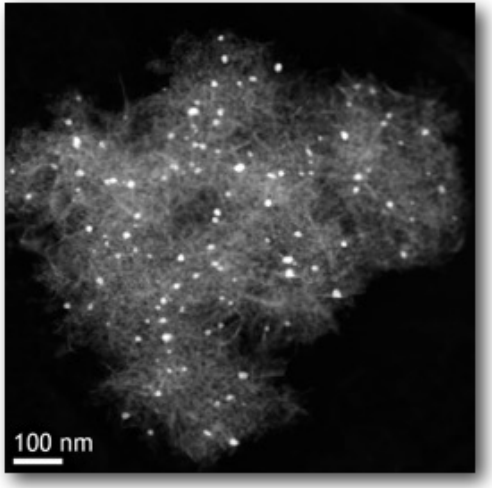
Thicker specimen or larger nucleus → More scattering

GaN nano wires



Mass/thickness contrast
Bright-field TEM image

Pt catalyst particles on Al_2O_3



Mass/thickness contrast
HAADF-STEM image

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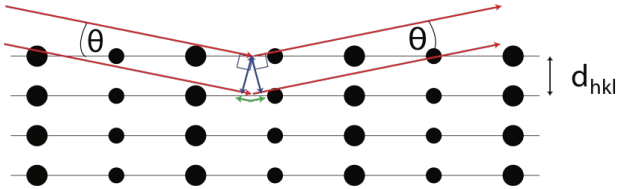
e^- as wave & Bragg scattering

For X-ray diffractometer



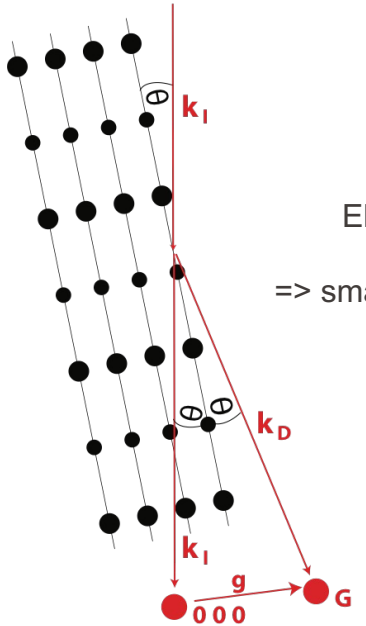
X-ray scattering:
Path difference between reflection from planes distance d_{hkl} apart
$= 2d_{hkl}\sin\theta$

=> Bragg law:
$n\lambda = 2d_{hkl}\sin\theta$

or

$\lambda = 2d_{nhnknl}\sin\theta$

For TEM electrons come from top, but otherwise geometrically the same



Electron diffraction: $\lambda \sim 0.001$ nm
therefore: $\lambda \ll d_{hkl}$
=> small angle approximation: $\lambda \approx 2d_{nhnknl}\theta$

=> Bragg diffraction at angle 2θ

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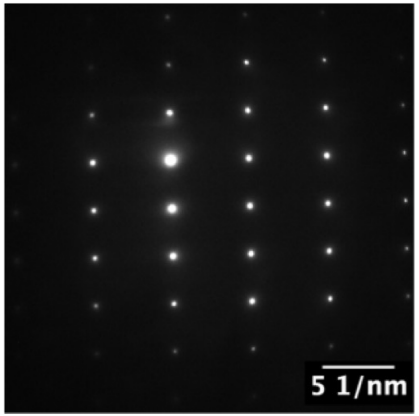
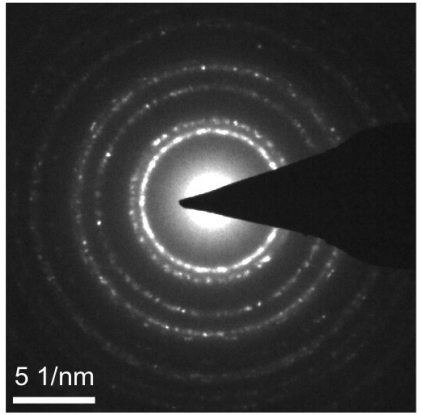
e⁻ as wave & Bragg scattering

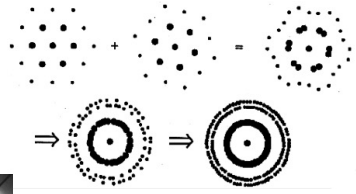
Electron diffraction pattern

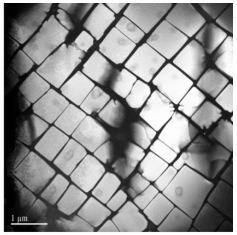


Spot pattern from a single crystal
Spots represent different (hkl) planes

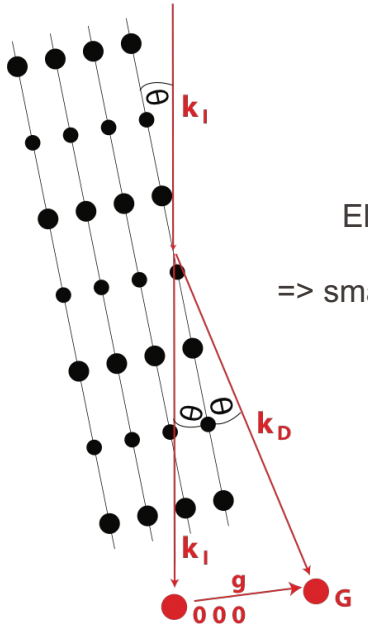


Ring pattern as many crystallites oriented differently in diffraction conditions





Therefore TEM gives image & diffraction!



Electron diffraction: λ ~ 0.001 nm
therefore: $\lambda \ll d_{hkl}$
=> small angle approximation: $\lambda \approx 2d_{nhnknl}\theta$

=> Bragg diffraction at angle 2θ

TEM operation modes

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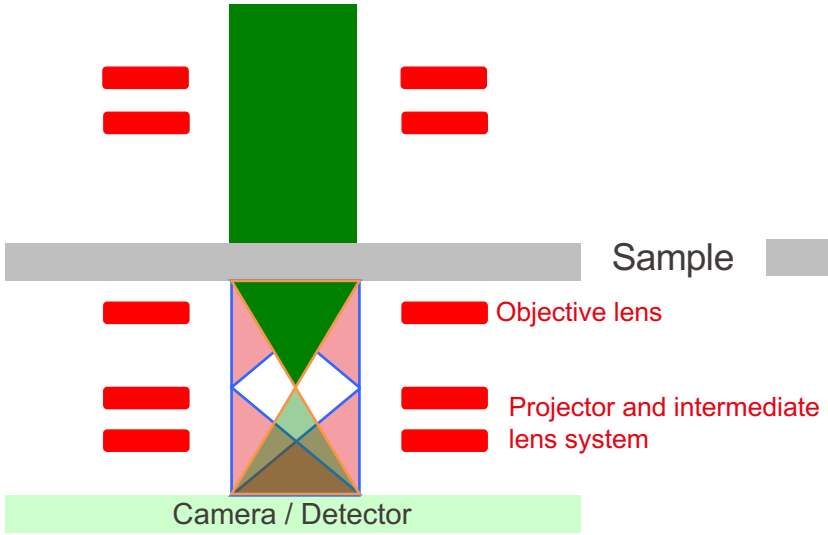
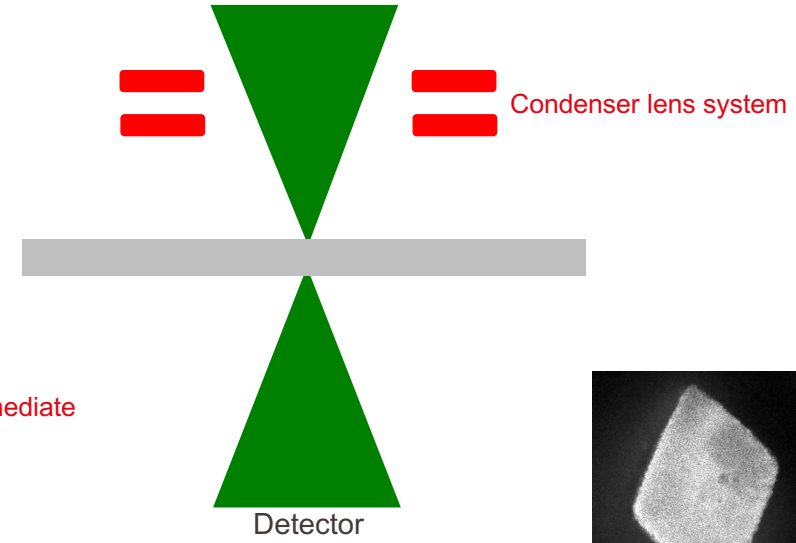
Conventional TEM

Electron beam (60-300 keV)



Scanning mode (STEM)

Electron beam (30-300 keV)



Fe-based metal-organic framework

2D projection of a 3D object

Image formation in TEM

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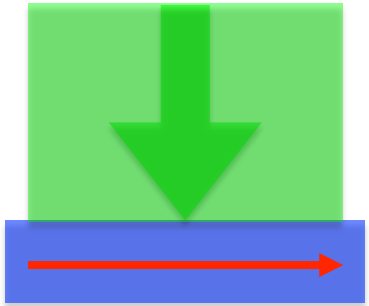
Parallel incident e⁻ beam; $\lambda \approx 0.02–0.03$ Å



Sample (thin, e⁻ transparent)

Objective lens

Focal plane

Image plane

Image formation in TEM

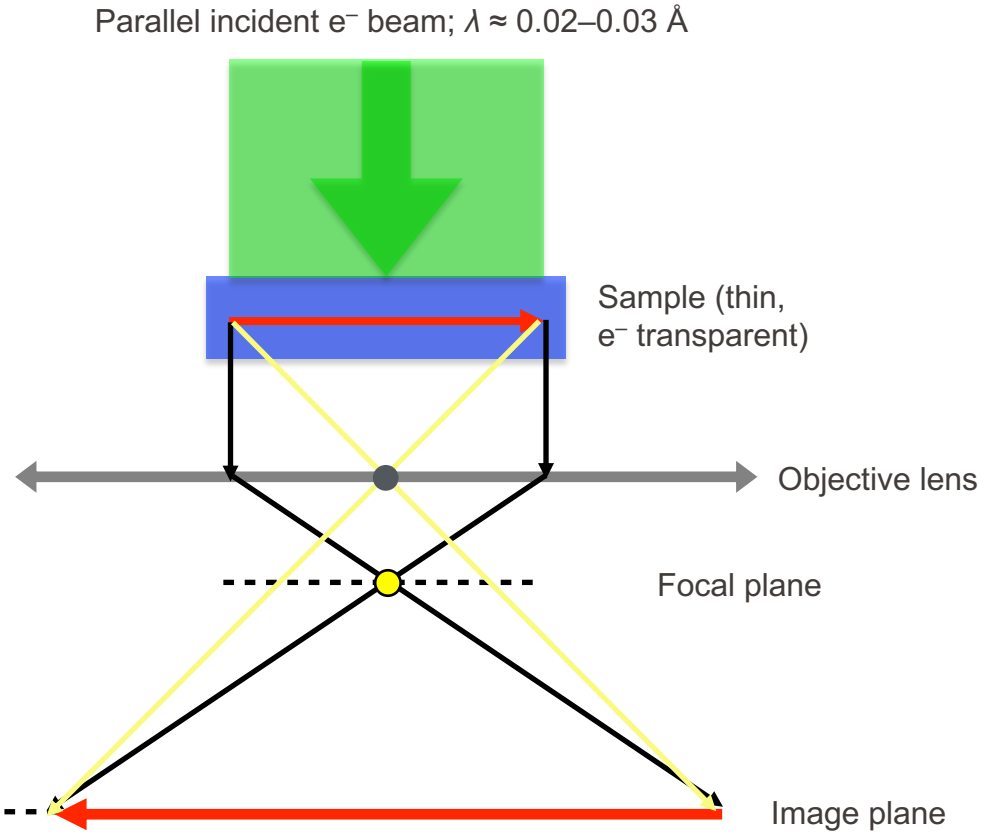


Image formation in TEM

Emad Oveisi

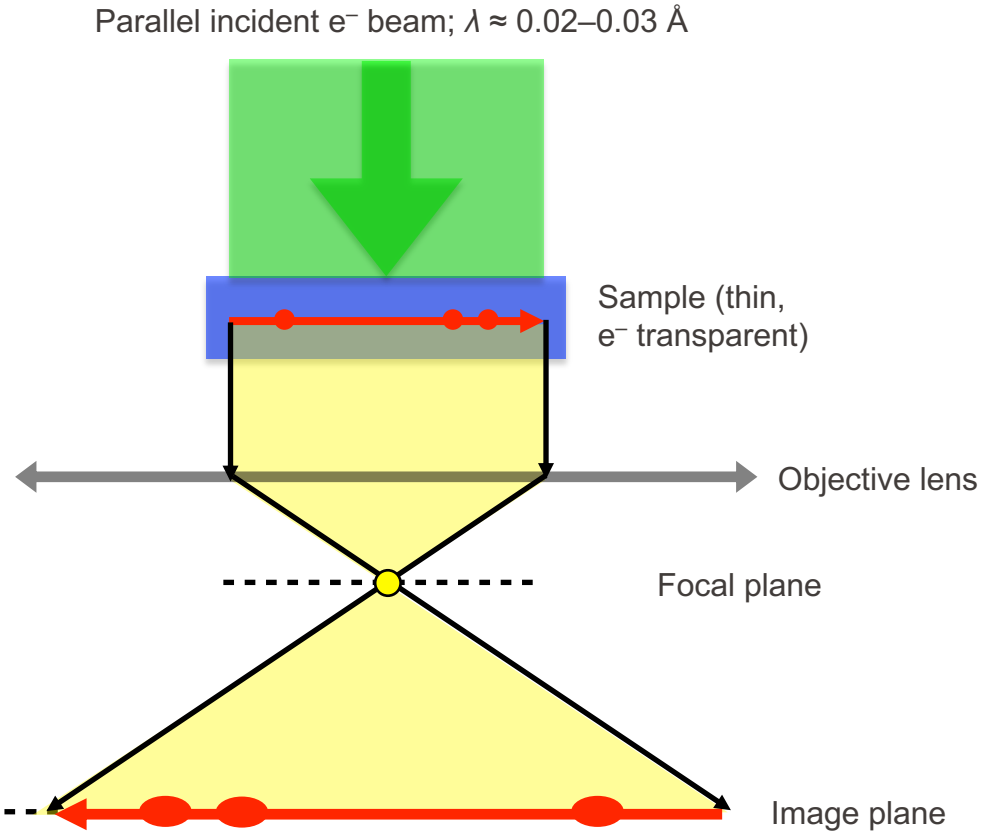


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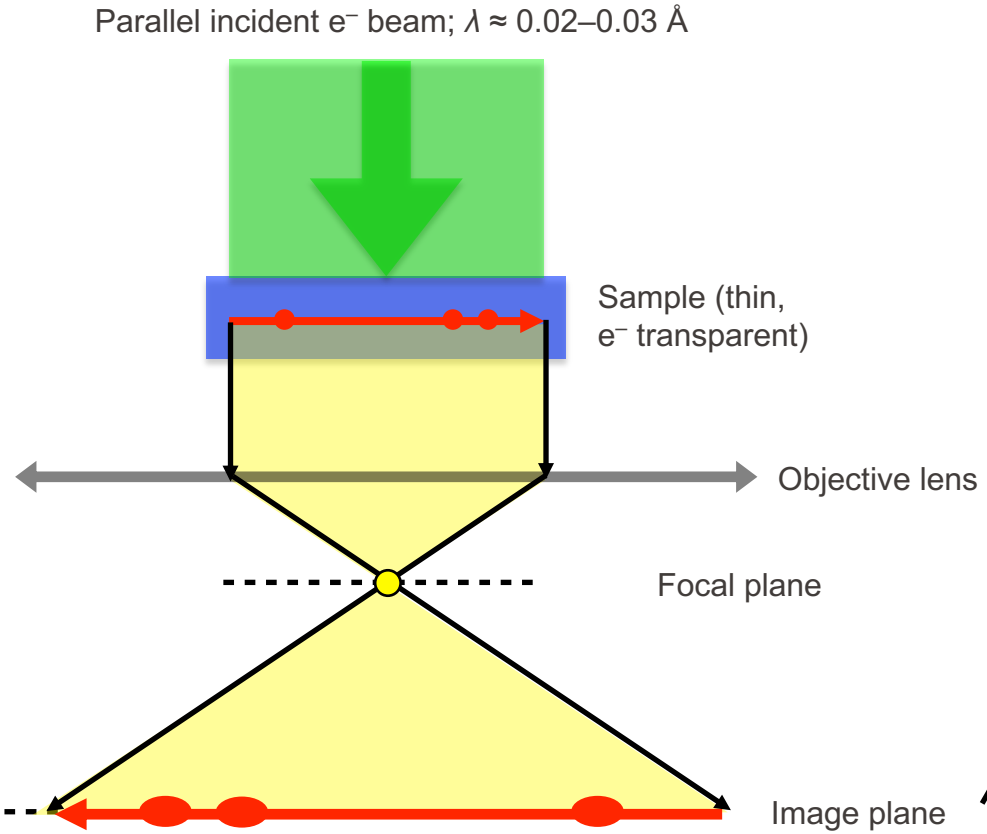
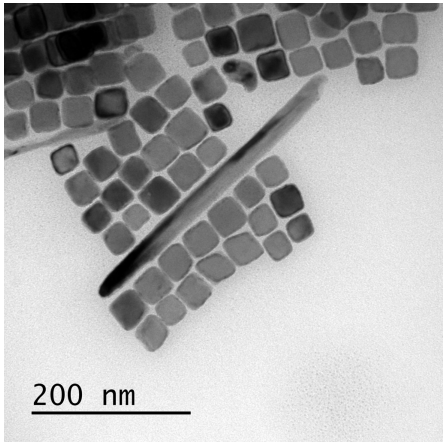
Image formation in TEM



Why some cubic particles appear darker than others?

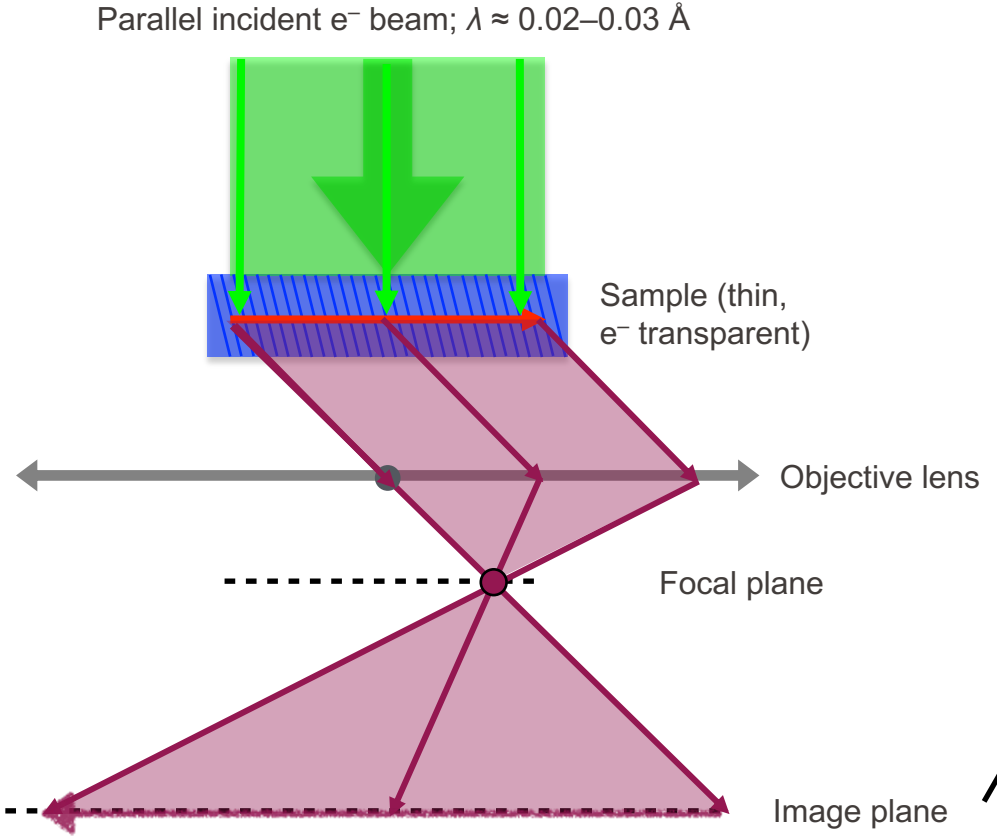
Cubic particles (≈ 40 nm) of Cu



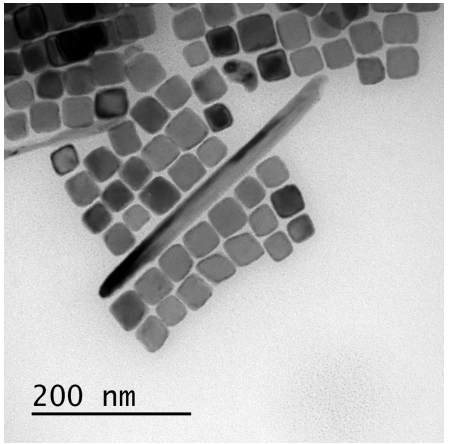
Bright-field image:
made by directly transmitted electrons

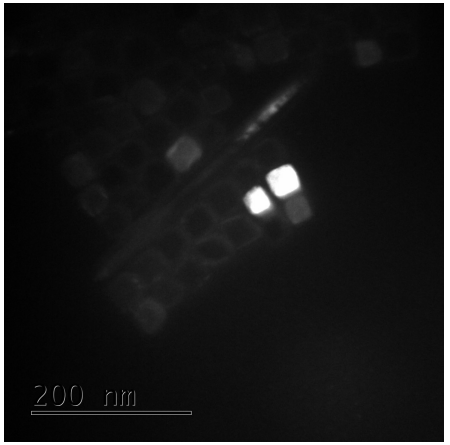
Image formation in TEM

Emad Oveisi



Bright-field image



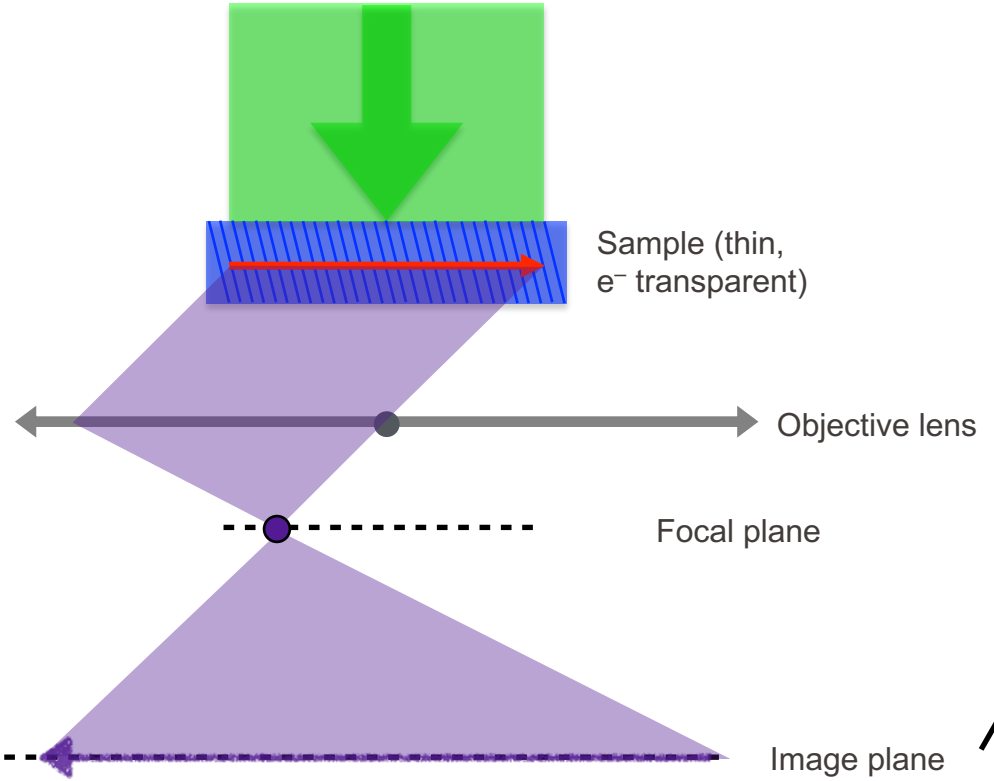


Dark-field image:
made by selected diffracted electrons

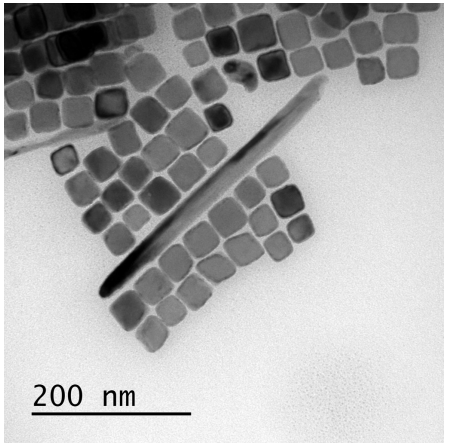
Image formation in TEM

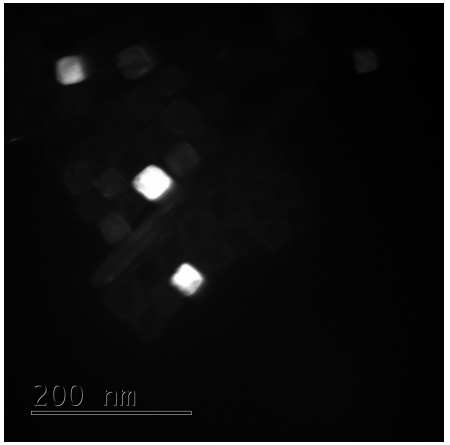
Emad Oveisi

Parallel incident e⁻ beam; $\lambda \approx 0.02–0.03$ Å



Bright-field image



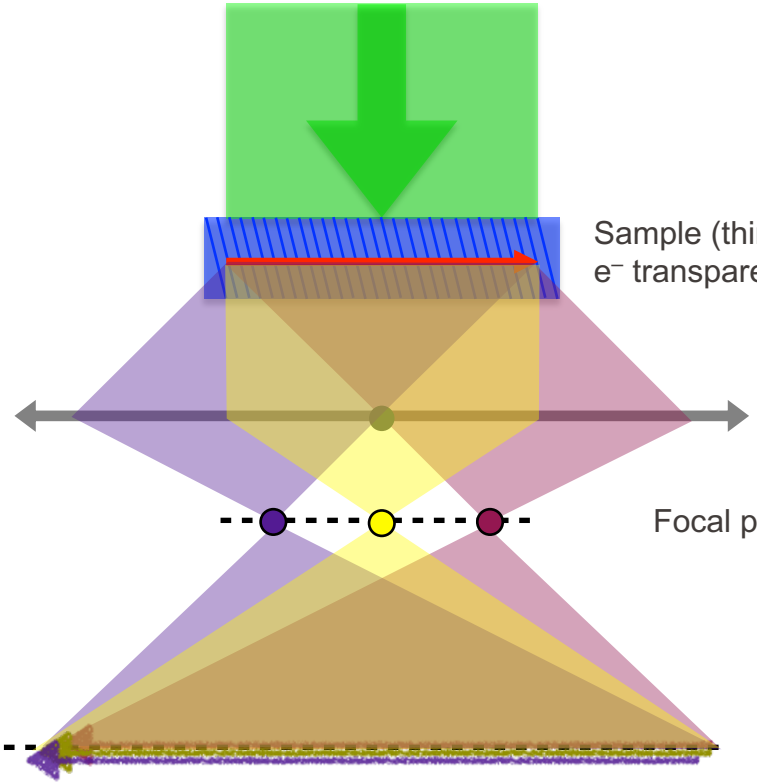


Dark-field image:
made by selected diffracted electrons

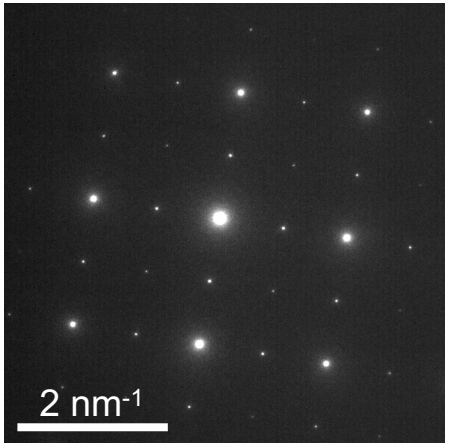
Image formation in TEM

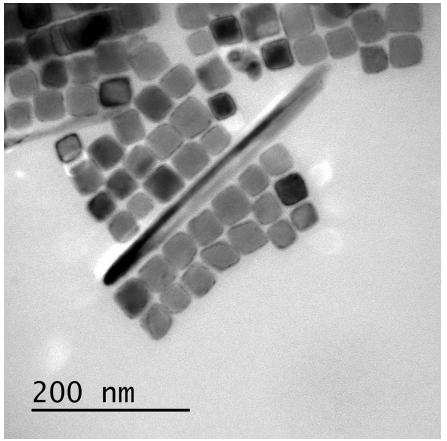
Emad Oveisi

Parallel incident e⁻ beam; $\lambda \approx 0.02–0.03$ Å



Electron diffraction pattern

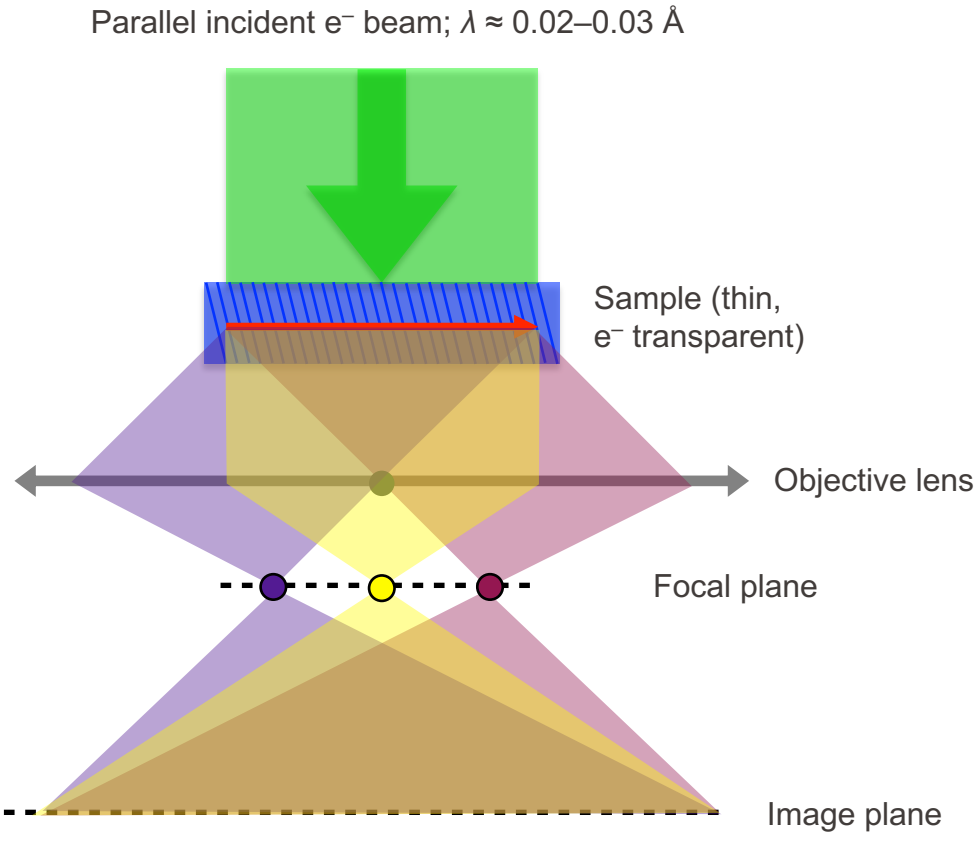




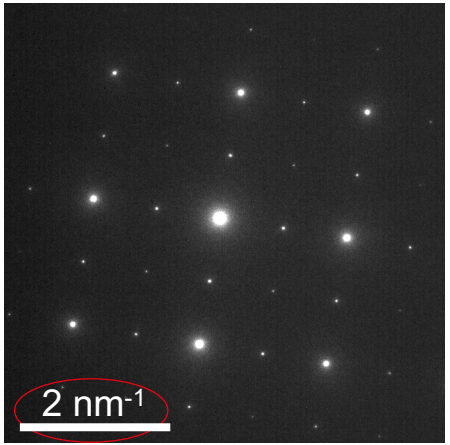
Ghost image:
Superposition of images made by direct and diffracted electrons

Image formation in TEM

Emad Oveisi



Electron diffraction pattern



- *In back focal plane of objective lens parallel rays focused to point*
- *Diffraction – coherent scattering – creates sets of parallel rays from different crystal planes*
- *Focusing of these parallel rays in back focal plane creates spots of strong intensity:* ***the diffraction pattern***

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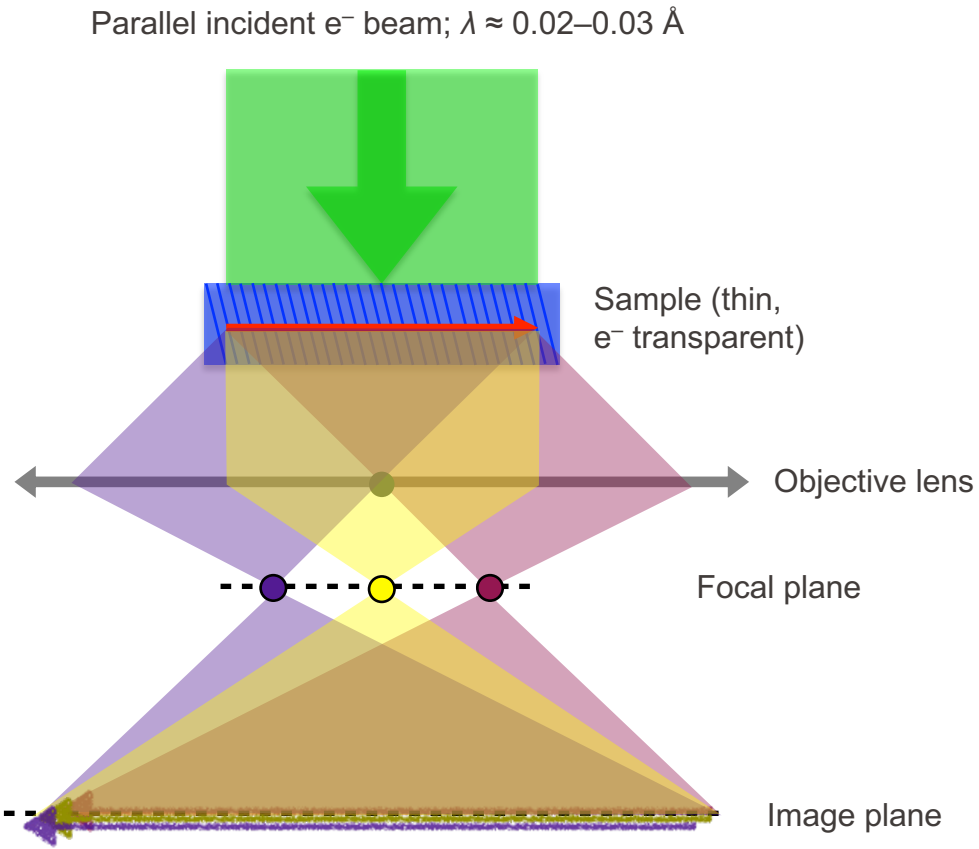
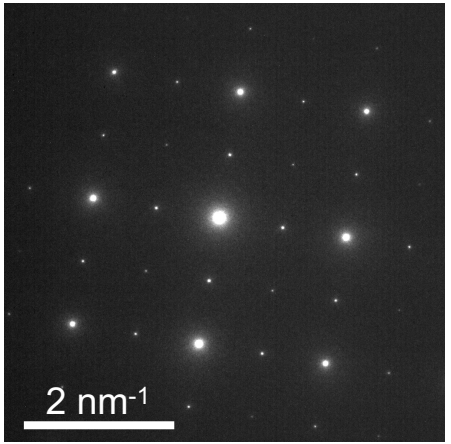
Image formation in TEM



Electron diffraction pattern



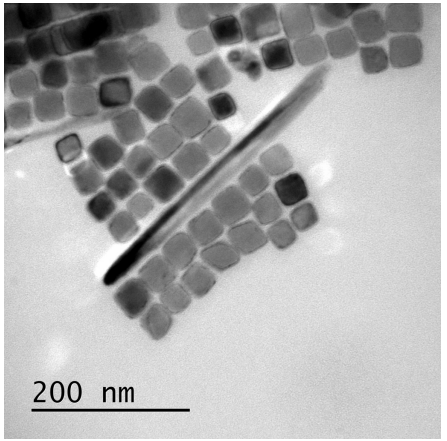


Image formation in TEM

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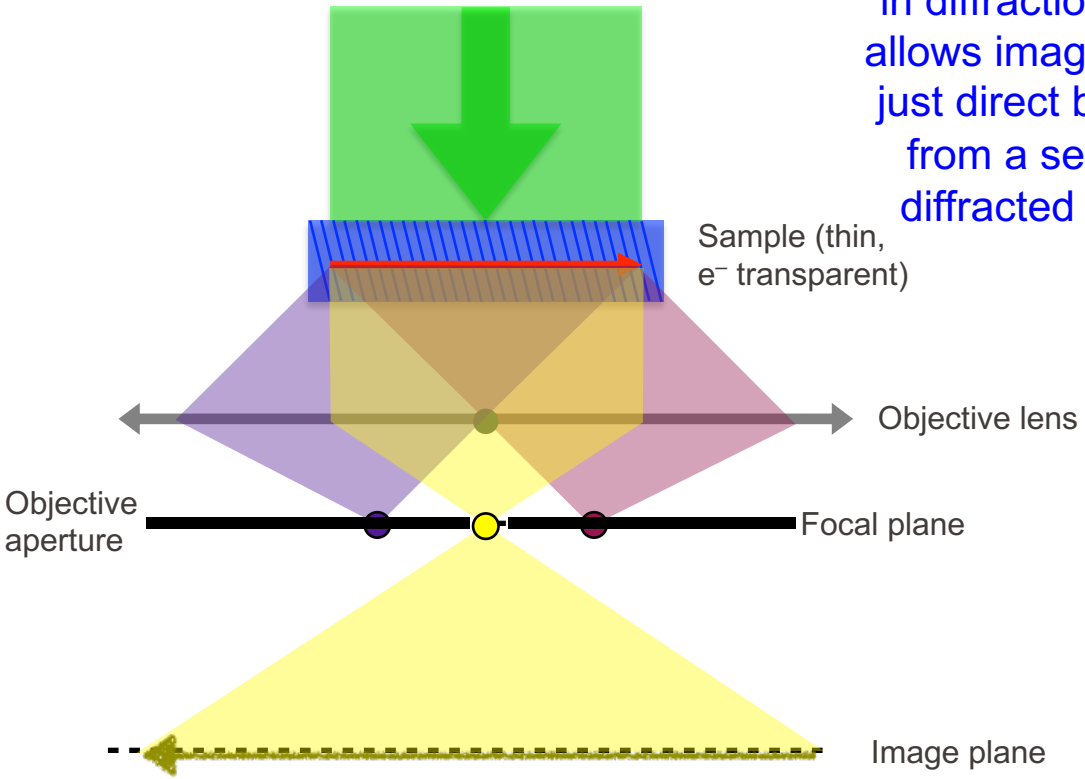
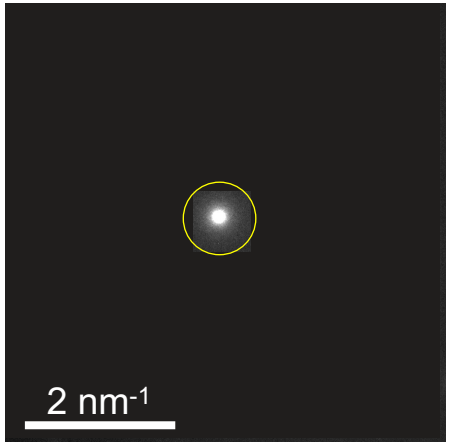
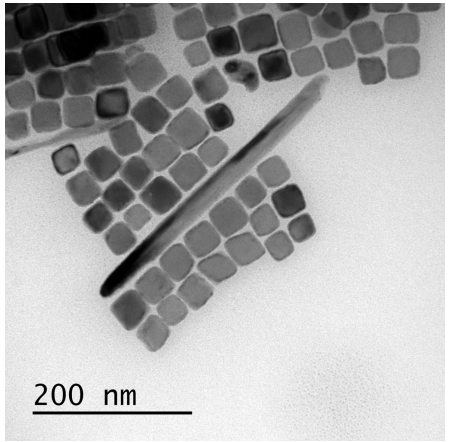


Insertion of the "objective aperture" in diffraction plane allows imaging from just direct beam or from a selected diffracted beam.

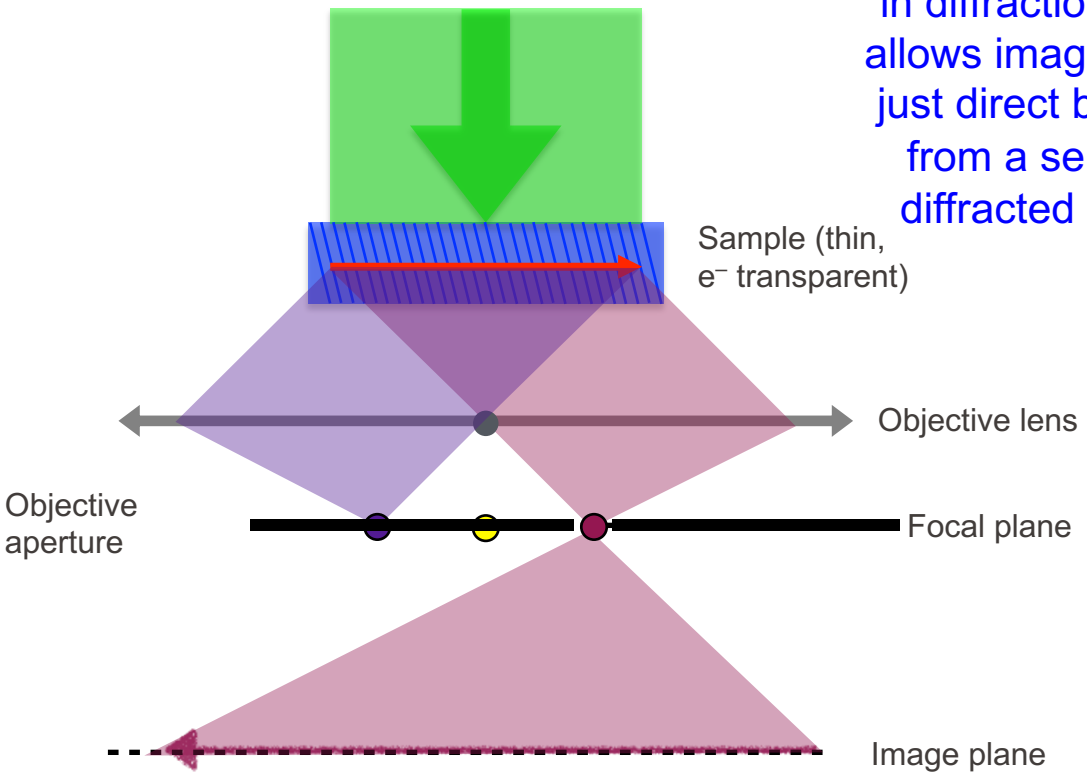
Electron diffraction pattern





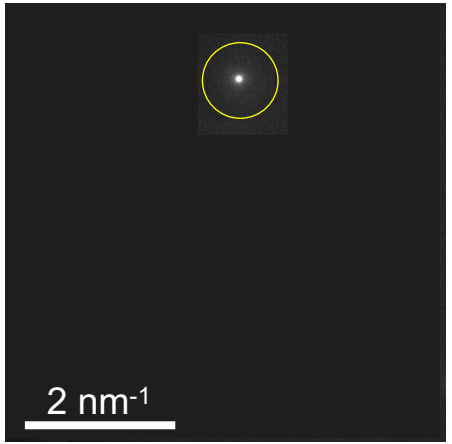
Bright-field image:
made by directly transmitted electrons

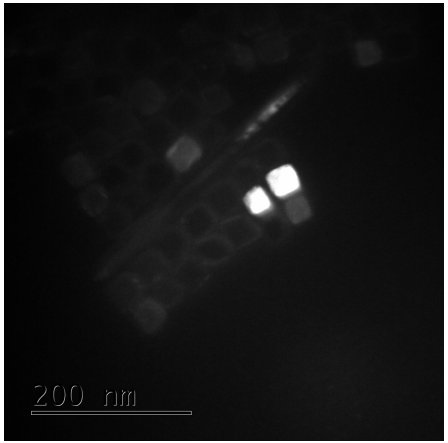
Image formation in TEM



Insertion of the "objective aperture" in diffraction plane allows imaging from just direct beam or from a selected diffracted beam.

Electron diffraction pattern





Dark-field image:
made by selected diffracted electrons

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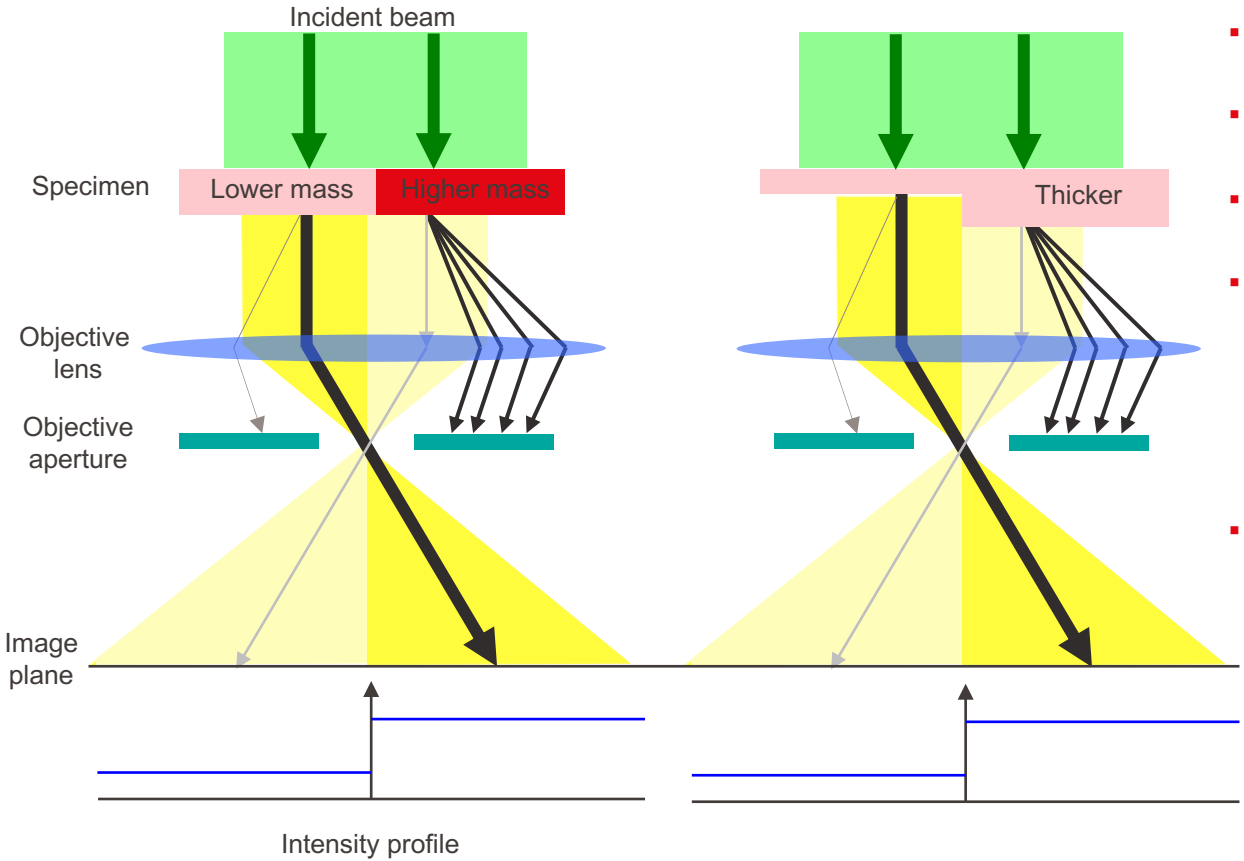
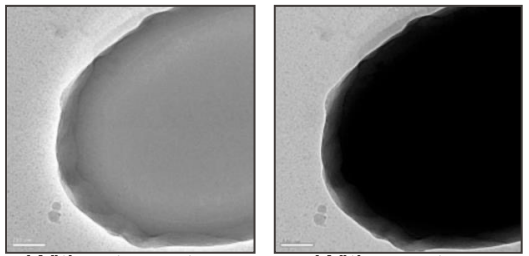
Image/Contrast mechanisms

- **Image formation in TEM**
  - Image and diffraction modes
  - Bright- and dark-field modes
  - High-resolution TEM
- **Image contrast in TEM**
  - Mass-thickness contrast
  - Diffraction contrast
  - Phase contrast

Mass-thickness contrast

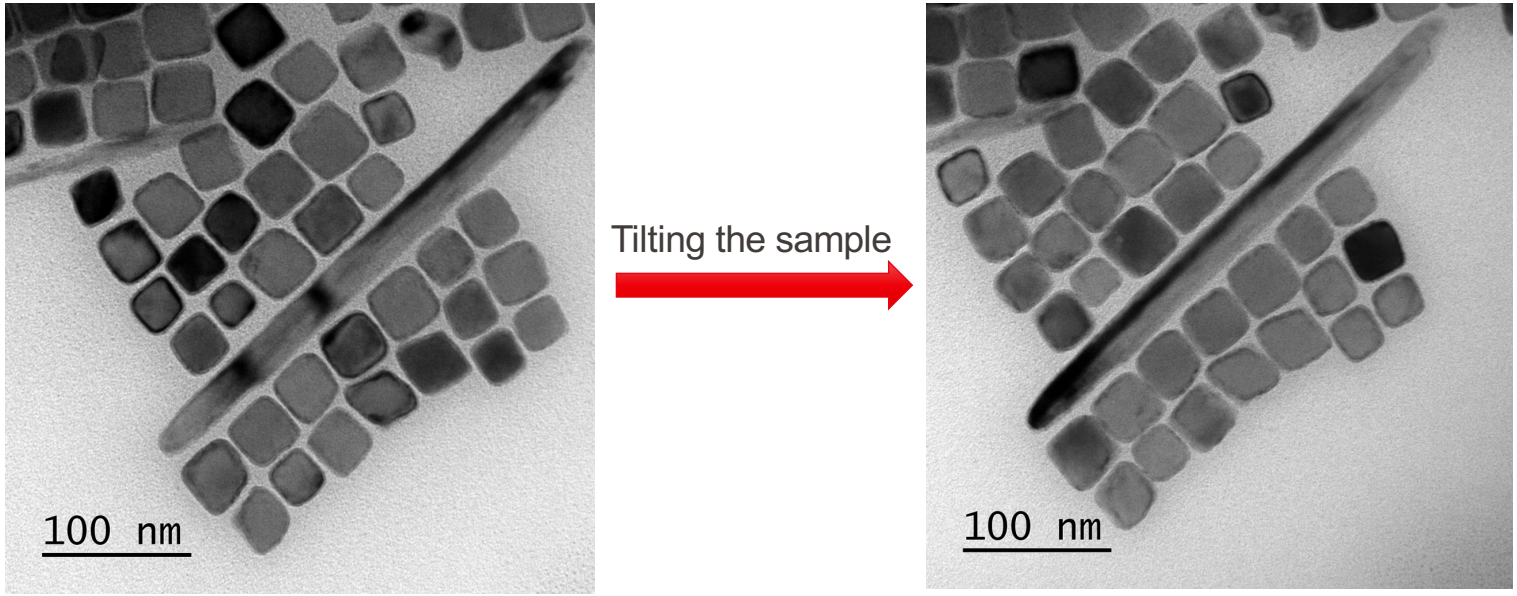


- Areas of higher mass/thickness scatter electrons more than others.
- Electrons are captured by the aperture and lost from the beam path.
- Areas of higher mass thickness will therefore appear dark in the image.
- This is known as:
  - **mass thickness contrast**,
  - **scattering contrast**,
  - **aperture contrast** or
  - **amplitude contrast**!
- Applies to both Crystalline and Amorphous materials.



Without aperture | With aperture

Example: Cupper nano particles



Why some of cubic particles appear darker than others?

Note that contrast changes when tilting the specimen

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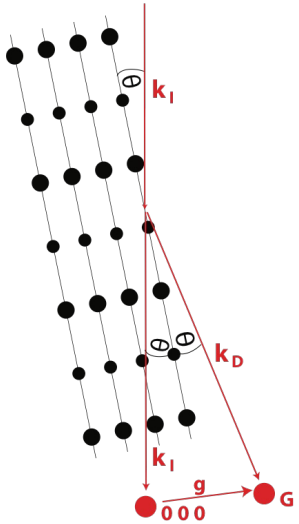
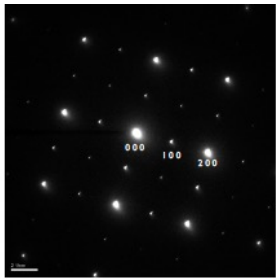
Image/Contrast mechanisms

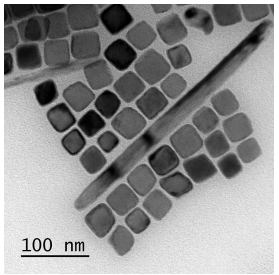


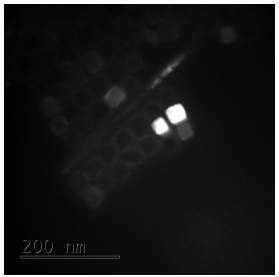
Path difference between reflection from planes distance d_{hkl} apart
$= 2d_{hkl}\sin\theta$

Bragg law: $\lambda = 2d_{nhnknl}\sin\theta$

→ Bragg diffraction at angle 2θ

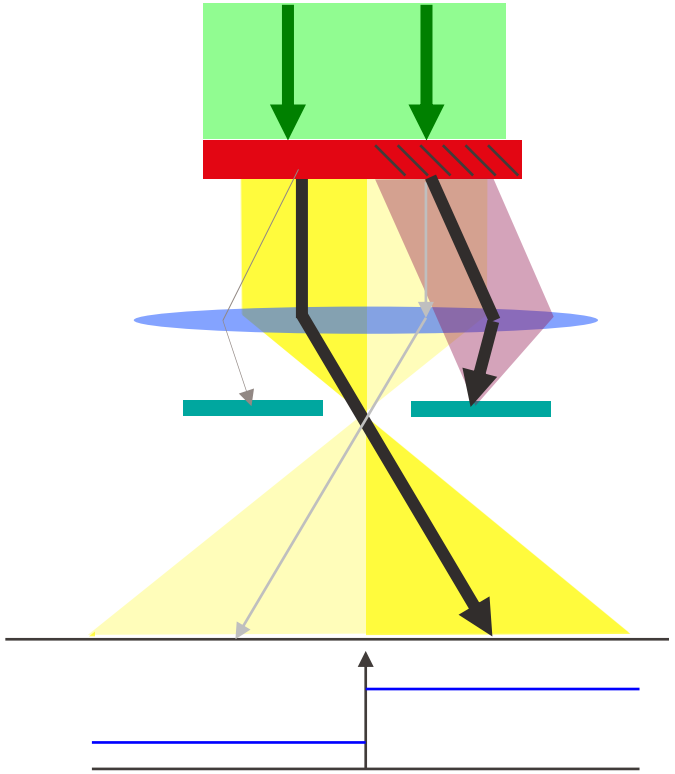






TEM gives image & diffraction information!

Diffraction contrast



Applications of electron diffraction | Determine crystalline state

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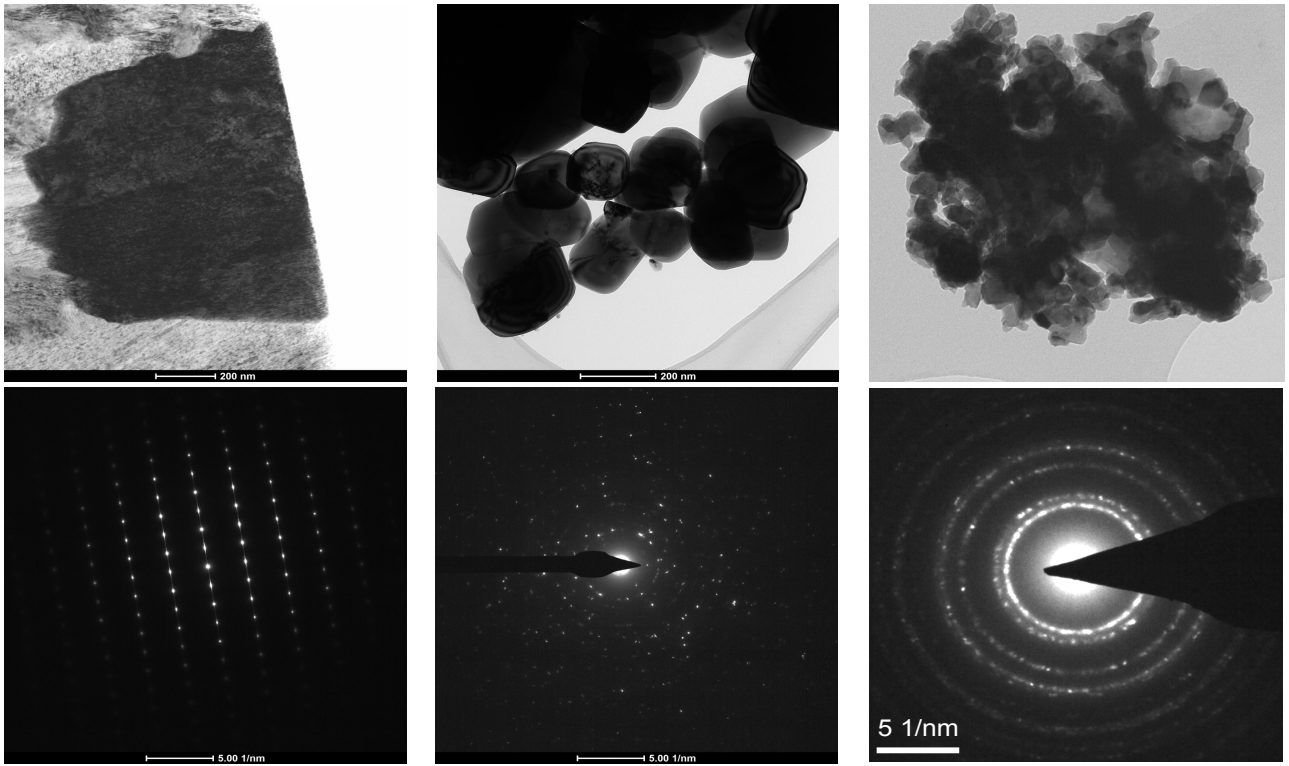

Diffraction contrast imaging

Principle of diffraction contrast imaging:

Typically we use an objective aperture to select either the direct beam or a specific diffracted beam in the back-focal plane.

If the diffraction condition changes across the sample the intensity in the selected beam changes; the intensity in the image changes correspondingly.

In other words we make a spatial map of the intensity distribution across the sample in the selected beam: *it is a mapping technique.*

In this way we can image changes in crystal phase and structural defects such as dislocations

As an example such TEM imaging was a key piece of evidence proving the existence of dislocations

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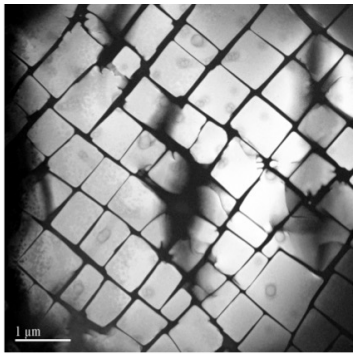
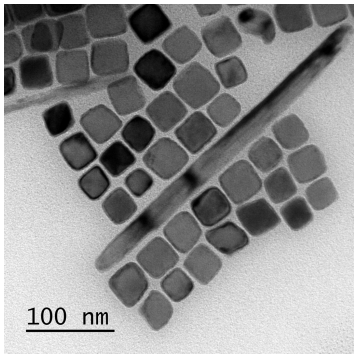
Diffraction contrast imaging

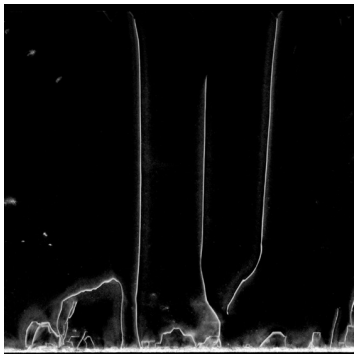
Examples of diffraction contrast image



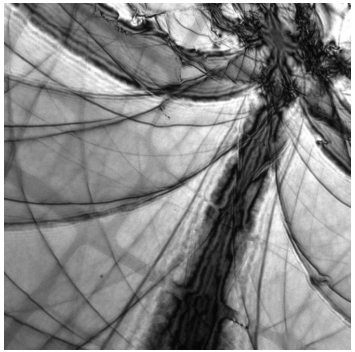
Dark-field (DF) image
Precipitates in NiAl super-alloy



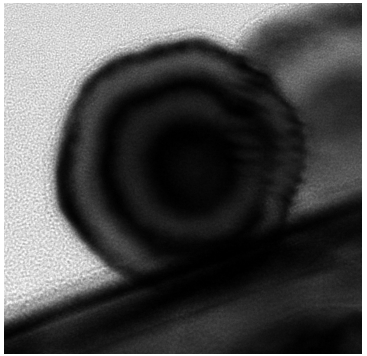
Bright-field (BF) image
Cu Nanocubes



Weak beam dark-field image
Dislocations in GaN



Bright-field image of bent regions in NiAl supper-alloy = Bend contours



Thickness fringes in BF-TEM image of a Cu nanocube

Applications of electron diffraction | Crystal phase discrimination

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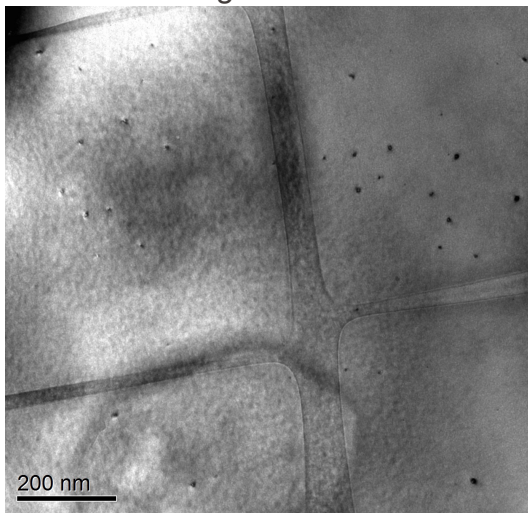
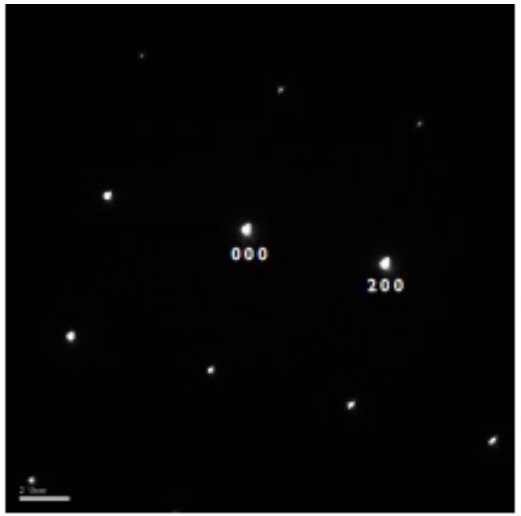
Example: Ni_3Al-based superalloy

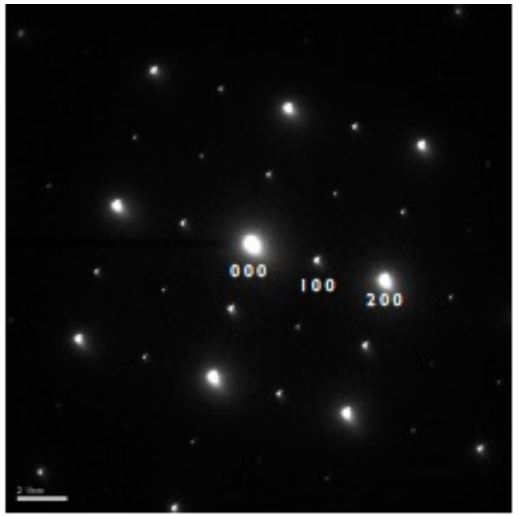
Bright-field

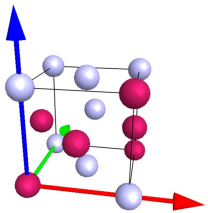


γ-phase matrix: FCC
(Ni, Al disordered on sites)



γ'-phase precipitate: primitive cubic
(Ni on face centres, Al on corners)





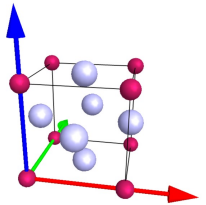
Which reflection to use to discriminate the the γ'-phase precipitate from the matrix?

a) {200} of γ'-phase precipitate

b) {200} of γ-phase matrix

c) (000) direct beam

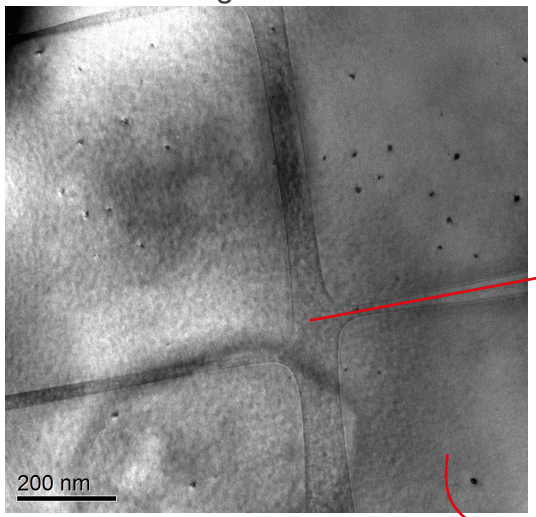
d) {100} of γ'-phase precipitate



Applications of electron diffraction | Crystal phase discrimination

Example: Ni_3Al-based superalloy

Bright-field



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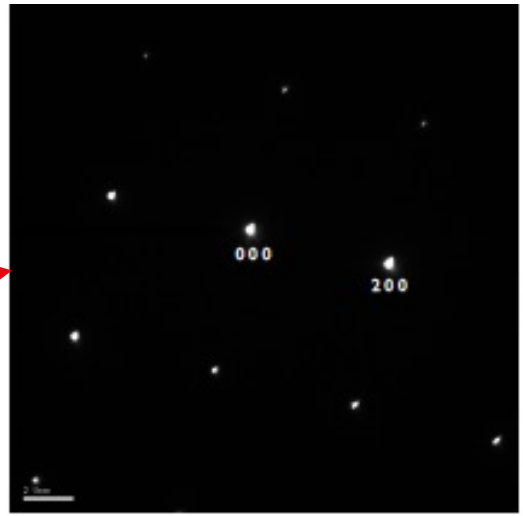

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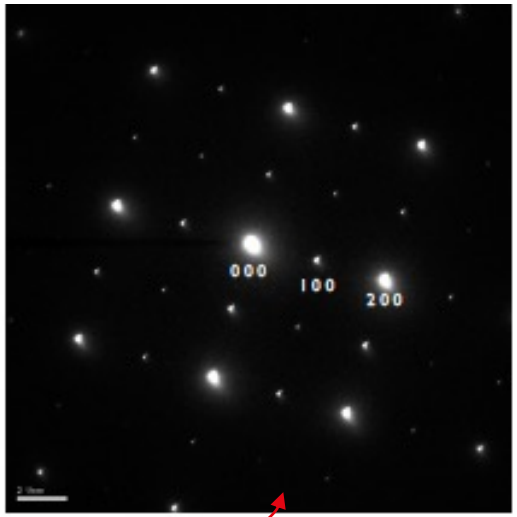

Note: EDX can't discriminate these two crystal phases!

Applications of electron diffraction | Crystal phase discrimination

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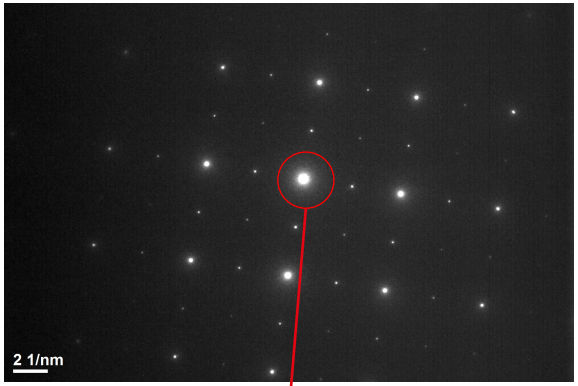
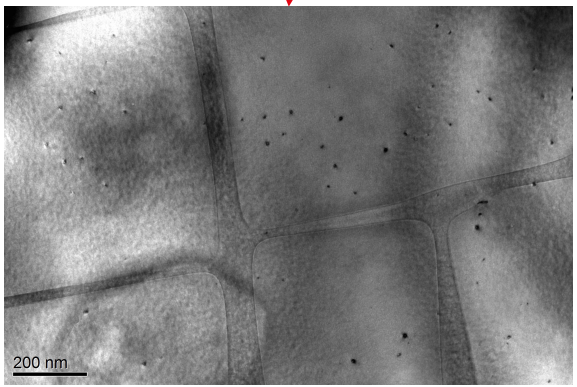
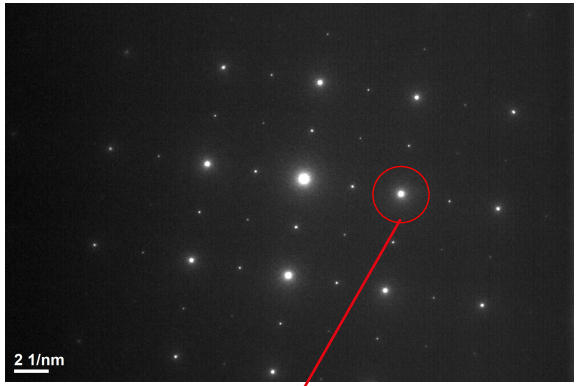
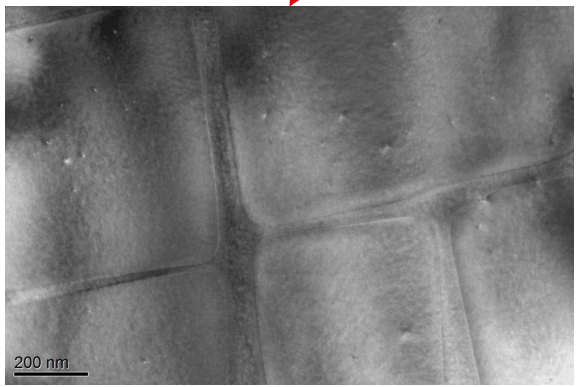
Example: Ni_3Al-based superalloy



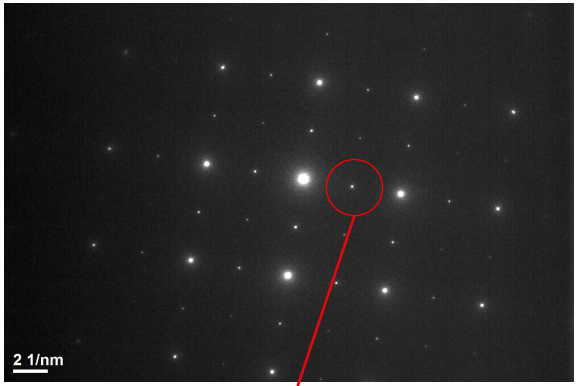


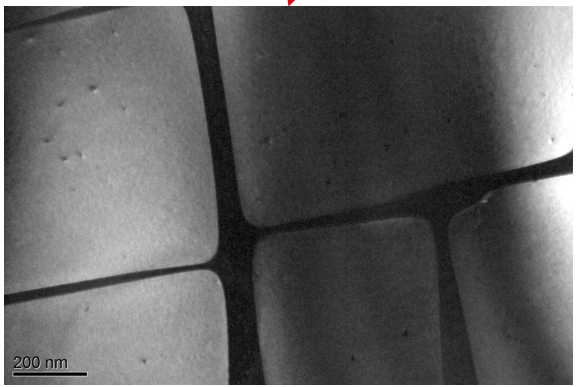
Bright-field image





Dark-field image **g** = (2 0 0)





Dark-field image **g** = (1 0 0)

Applications of electron diffraction | Crystal phase discrimination

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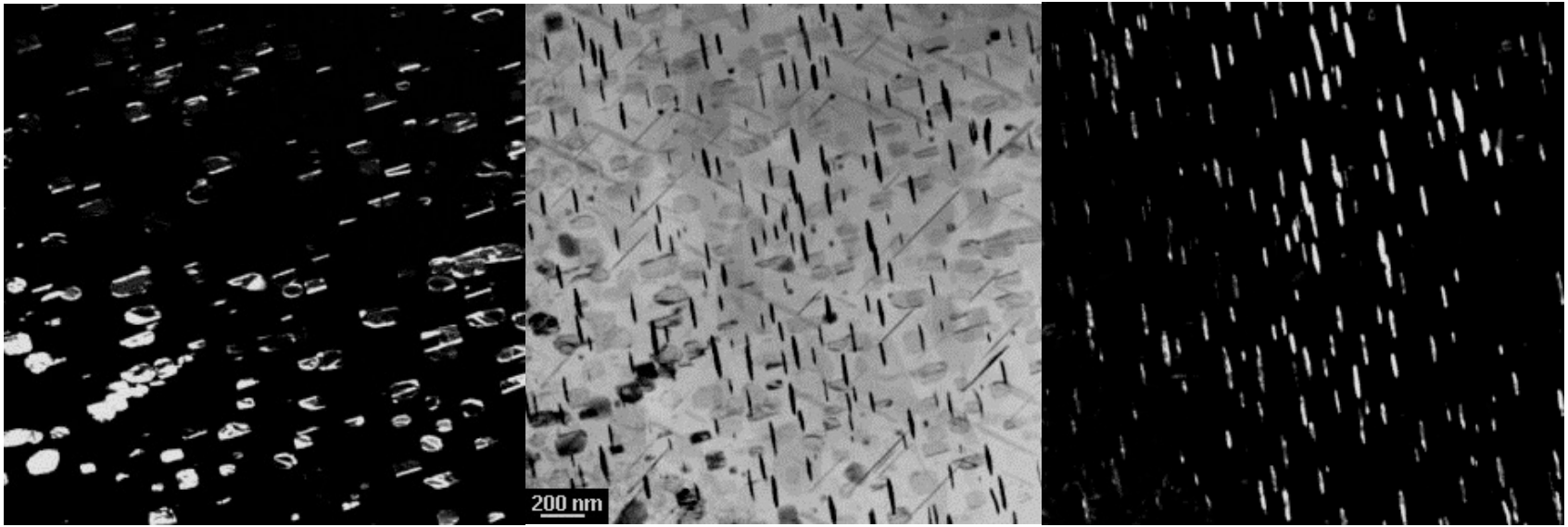
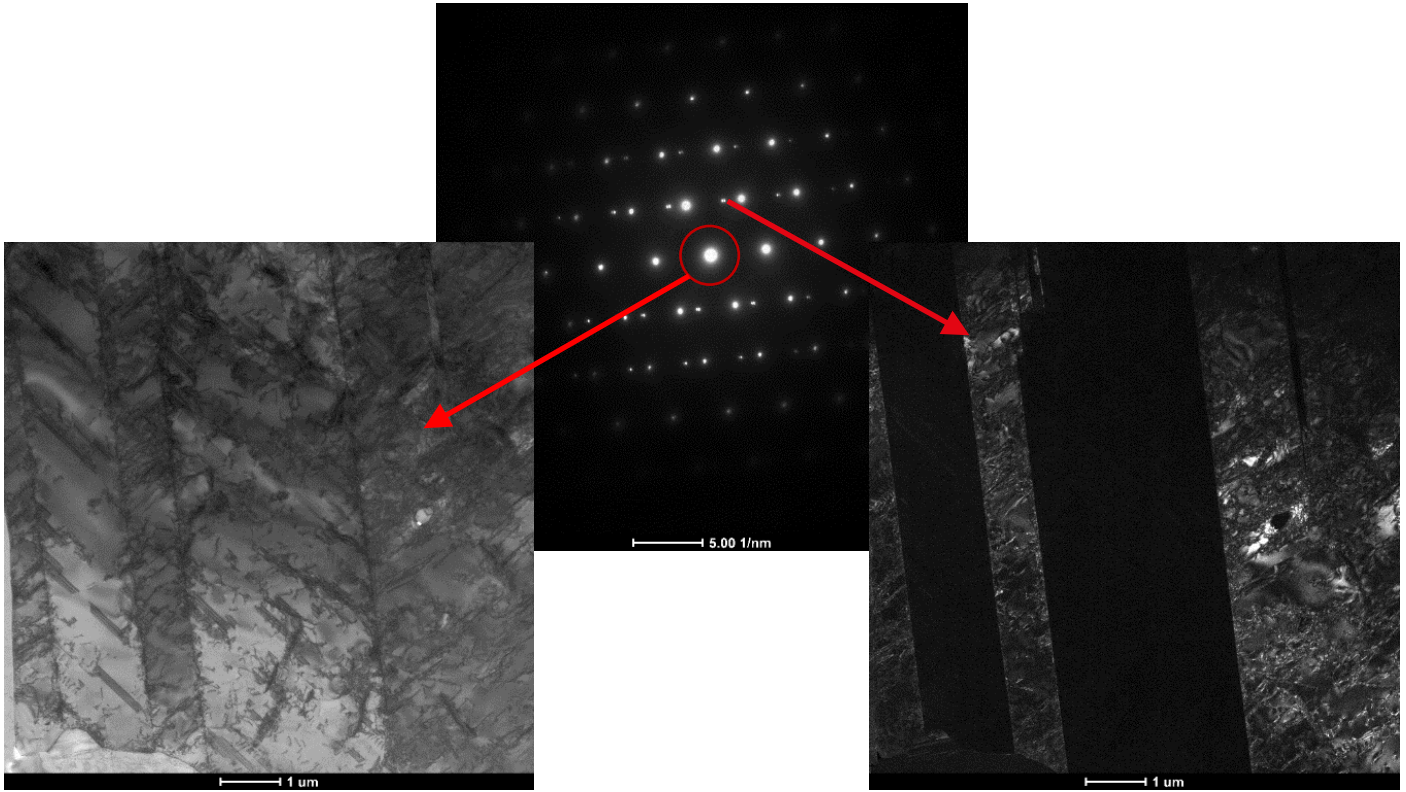
Example: Aluminum alloy containing precipitates with preferential growth direction



Orientation relationship between the matrix and precipitates can be determined

Twinning in electron diffraction



Applications of electron diffraction | Crystal's defect imaging/analysis

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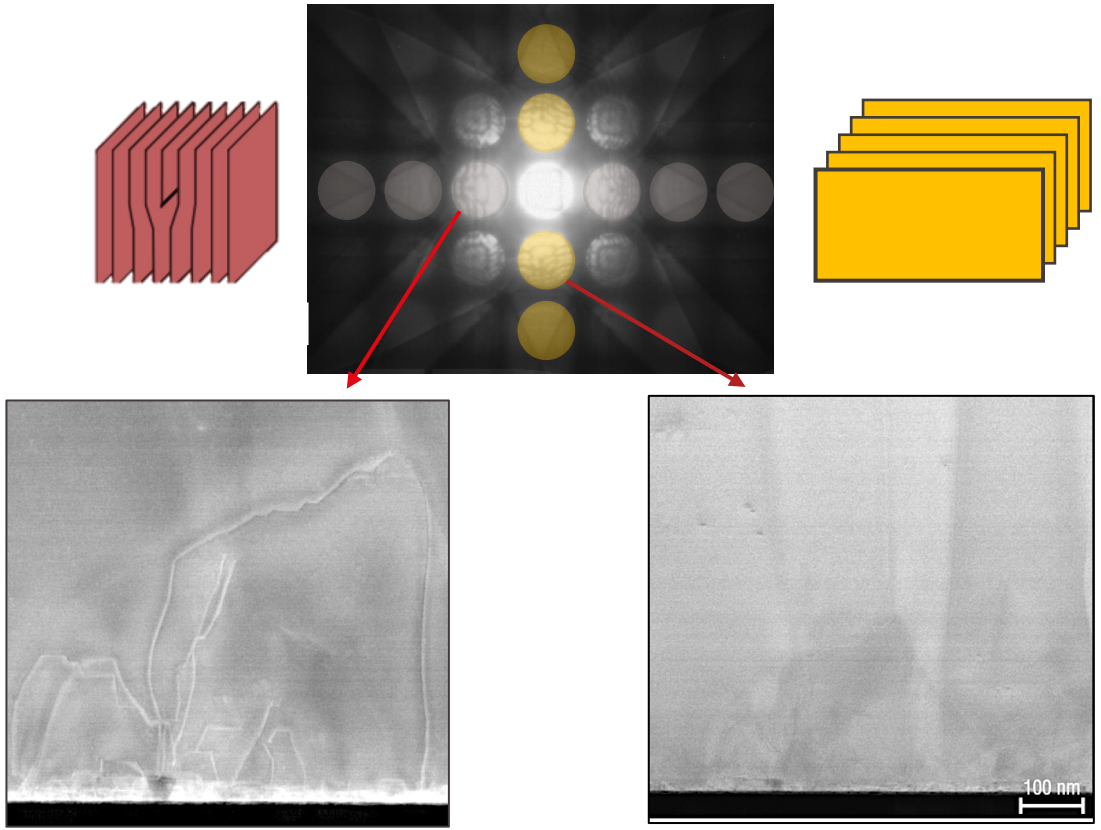
Exemple: Dark-field images of dislocations in GaN thin film on sapphire



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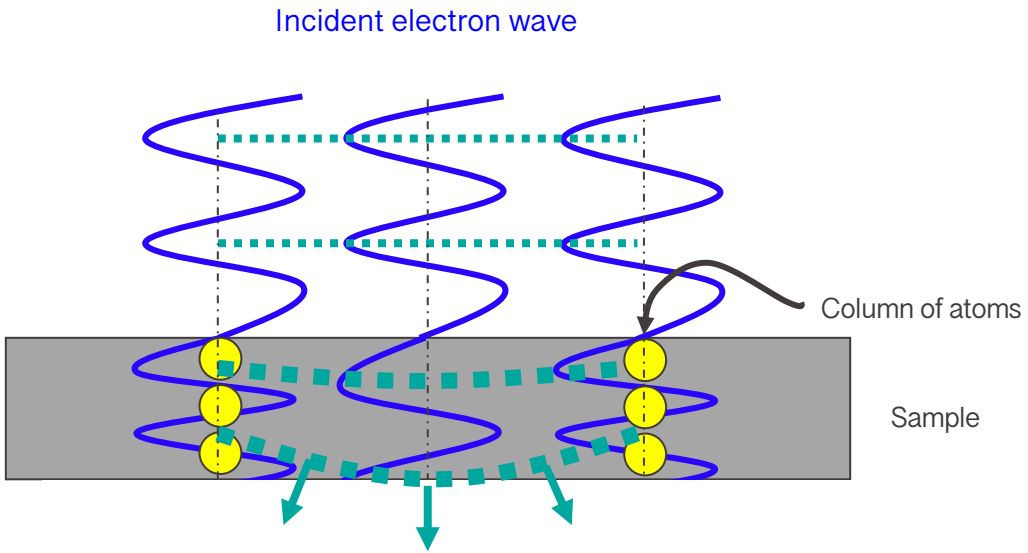
- **Image formation in TEM**
  - Image and diffraction modes
  - Bright- and dark-field modes
  - High-resolution TEM
- **Image contrast in TEM**
  - Mass-thickness contrast
  - Diffraction contrast
  - Phase contrast

Phase contrast | High-resolution TEM

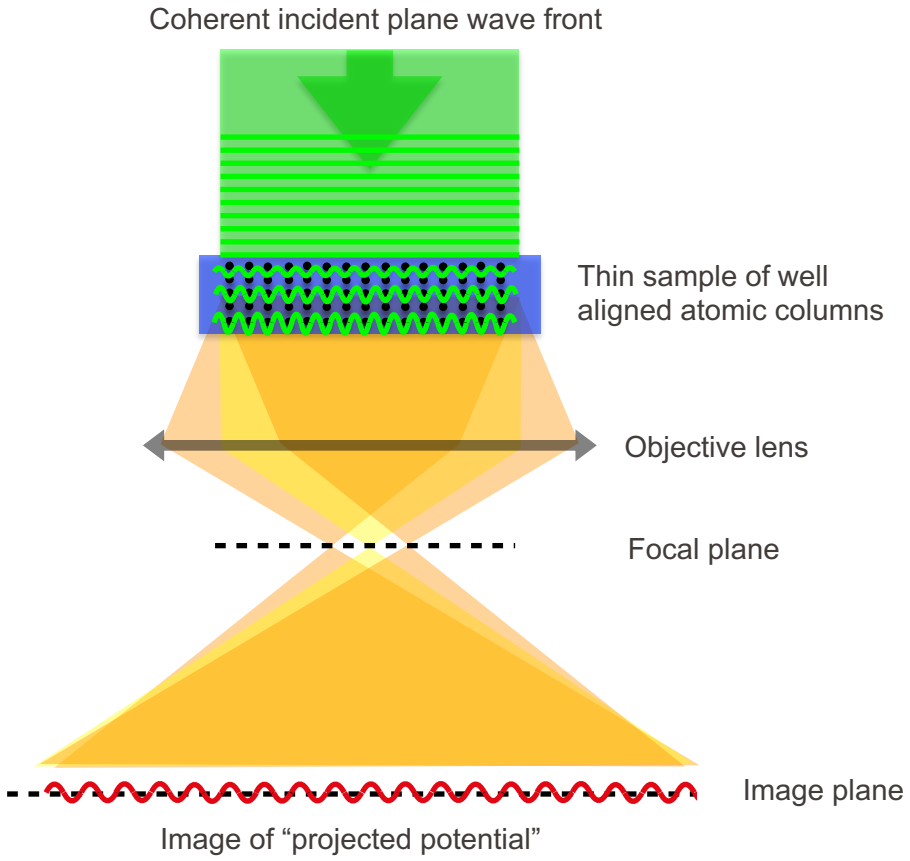
What happens when an electron wave passes through the sample?



Variations in the projected potential produce local relative phase shifts of the electron wave. The wave front therefore bends as wave travels through medium.

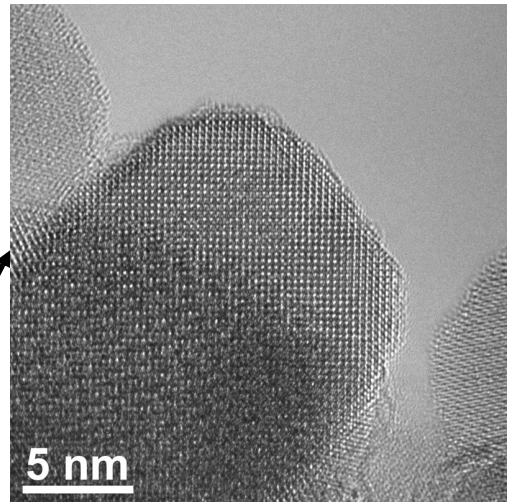
The direction of propagation of the electron may change! ⇒ Diffraction!

Phase contrast | High-resolution TEM



Due to changes in sample thickness and orientation, as well as lens imperfections, image interpretation is complex.

Resist the temptation of interpreting the spots as atoms!

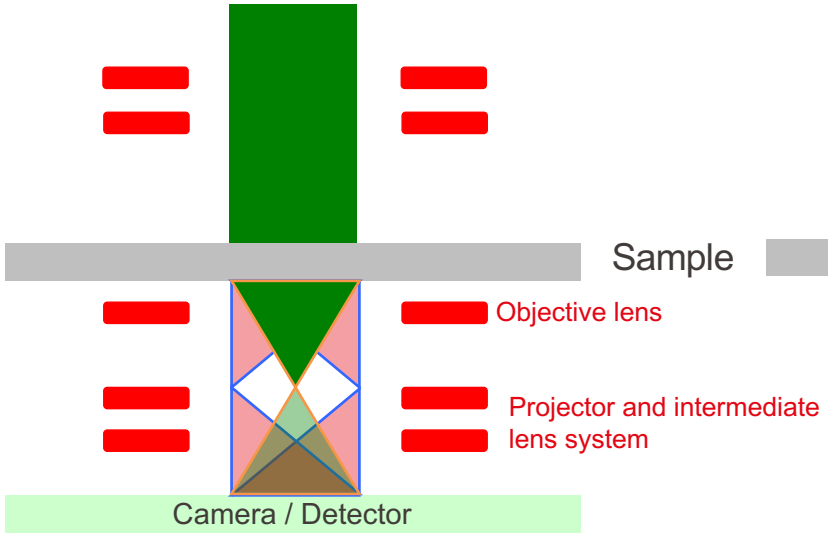


Lattice fringes in iron oxide nanoparticles

TEM operation modes

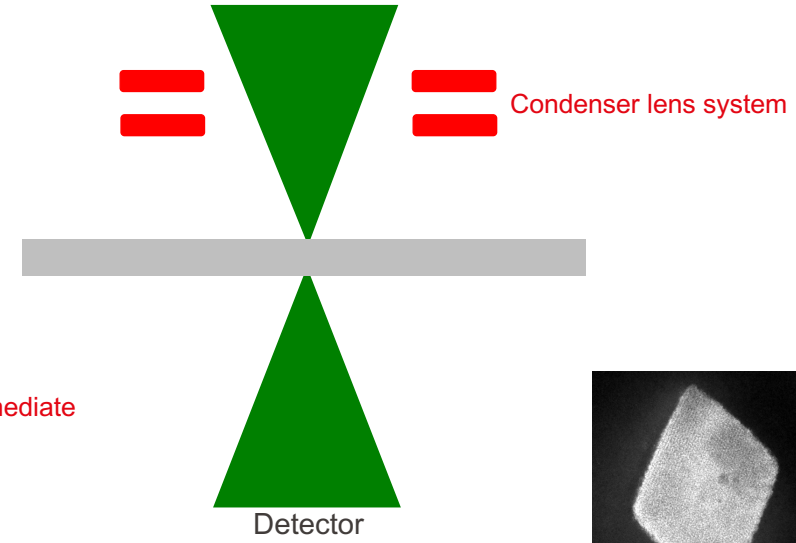
Conventional TEM

Electron beam (60-300 keV)



Scanning mode (STEM)

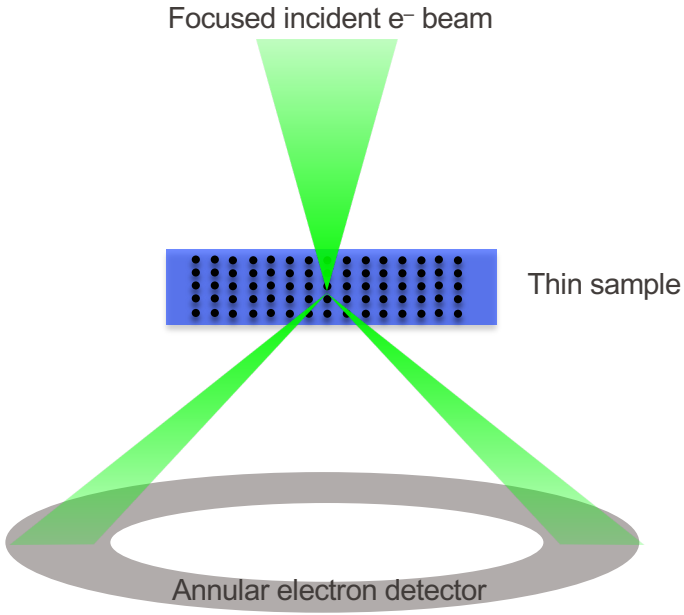
Electron beam (30-300 keV)

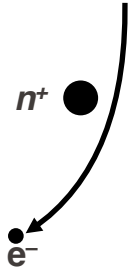


Fe-based metal-organic framework

2D projection of a 3D object

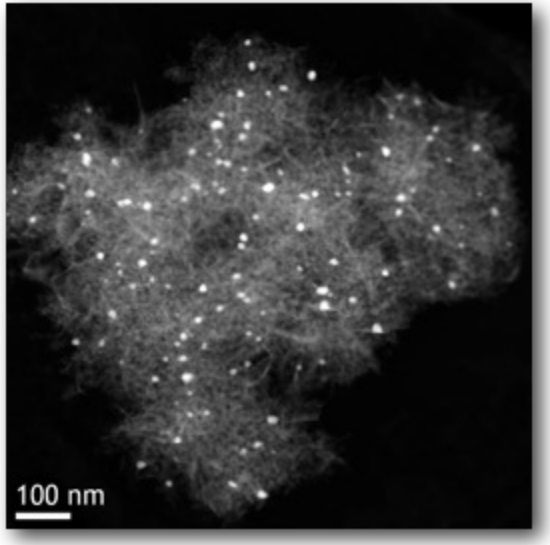
Scanning TEM (STEM) with focused probe





Rutherford-type scatteringcfrom nucleus (thermal diffuse scattering)

High-angle annular dark-field (HAADF) image

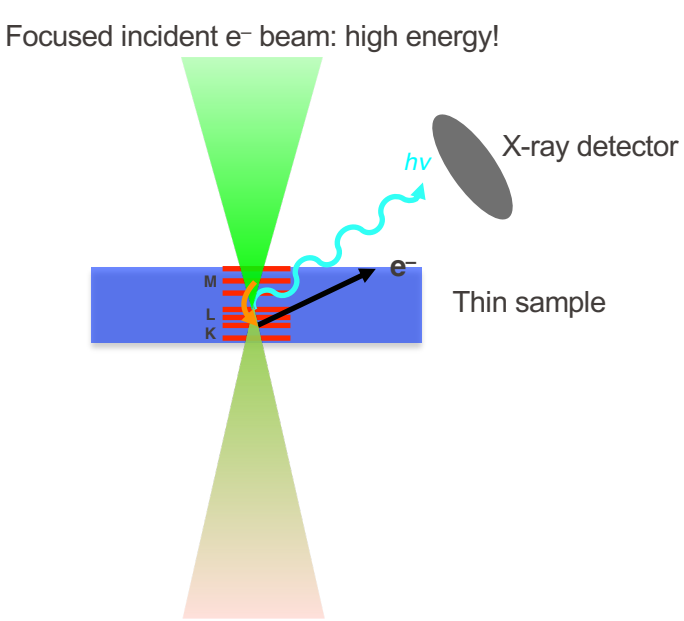


Pt catalyst particles on Al_2O_3

Mass/thickness contrast image

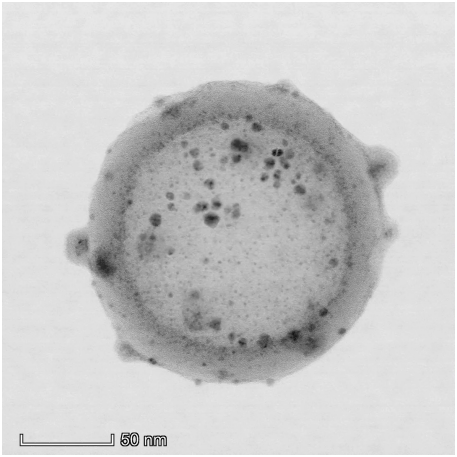
- Incoherent Rutherford-type scattering deflects transmitted e⁻ to high angle of scattering
- Larger nucleus / thicker the specimen → More scattering
- Map image intensity as function of probe position *(x, y)*
- Image intensity: $I(x, y) \propto t * Z^{1.6–2}$

Ionisation and energy-dispersive X-ray (EDX) mapping



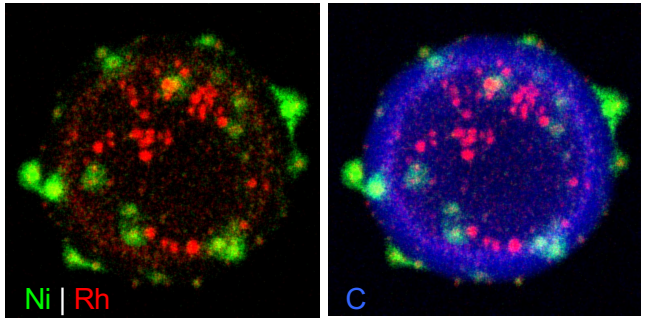
Transmitted beam suffers energy loss → Used in EELS

Bright-field STEM image



Ni-Rh catalytic nanoparticles in carbon shell

- High energy incident electrons (ballistic) → Ejection of inner shell e⁻
- Upper shell e⁻ descends
- Transition energy emitted as X-ray;
  Characteristic of element | $E_{X\text{-}ray} = E_{Upper\ shell} - E_{Inner\ shell}$



Map X-ray intensity as function of probe position (x,y)

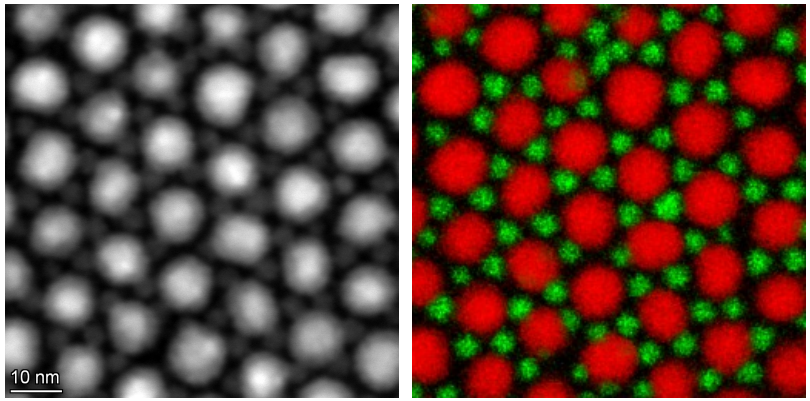
STEM-EDX

With fast mapping STEM-EDX has become a regular feedback tool for materials synthesis

Sample characterized in a couple of minutes/hours

With aberration-corrected STEM, atomic resolution EDX became possible

Example : Assembly of Fe_3O_4 and Cu particles



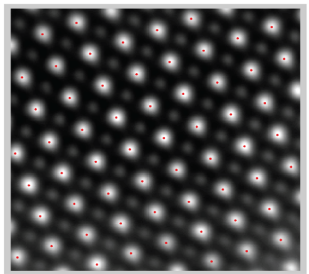
HAADF image

Map of Cu and Fe

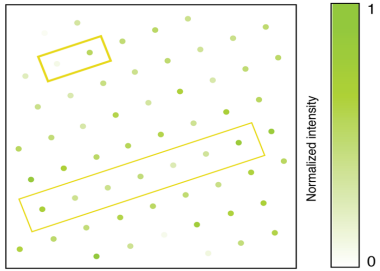
In collaboration with Chethana Gadiyar LNCE, EPFL

J. Am. Chem. Soc. 2020, 142, 37, 15931–15940

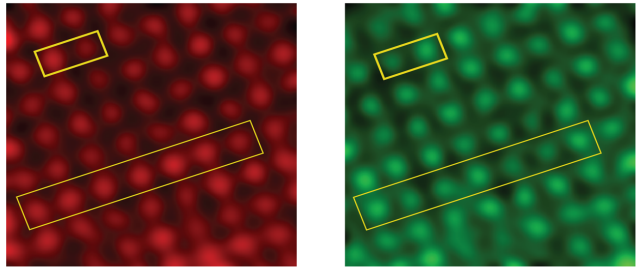
Example: $(Ba_xSr_{1-x})TiO_3$



HAADF image



Intesnsity map



Map of Sr and Ba

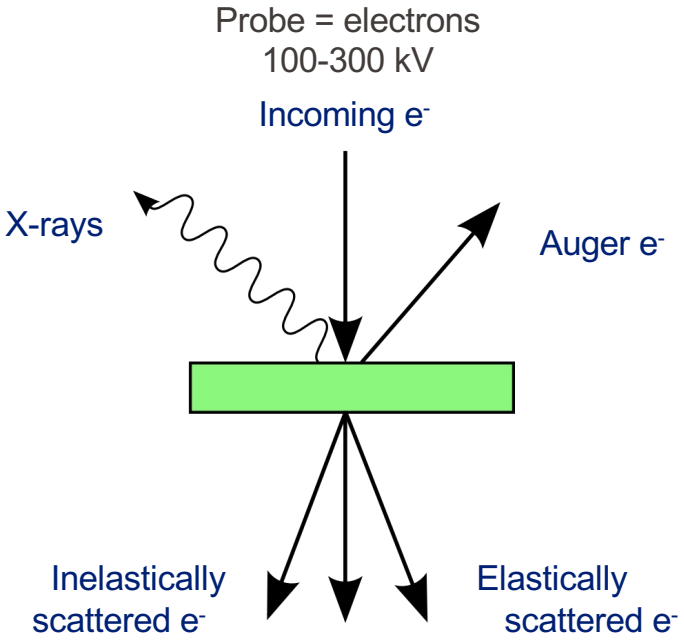
In collaboration with Prof. Dragan Damjanovic, EPFL

Nature Communications 2021.

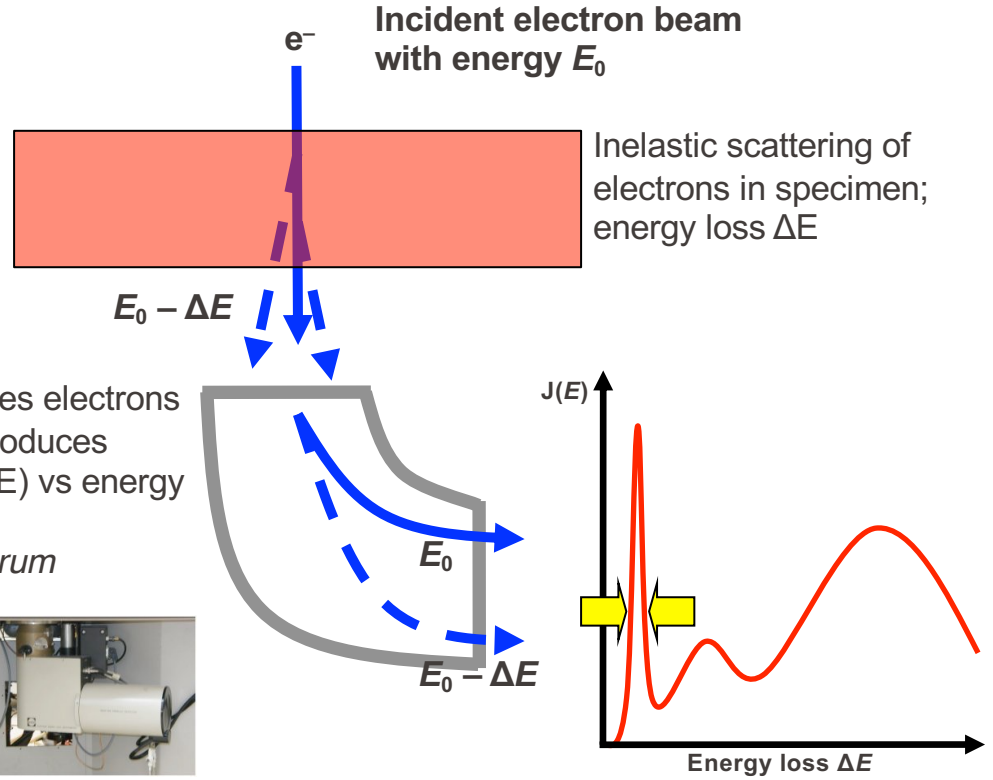
Electron energy-loss spectroscopy (EELS)

Emad Oveisi

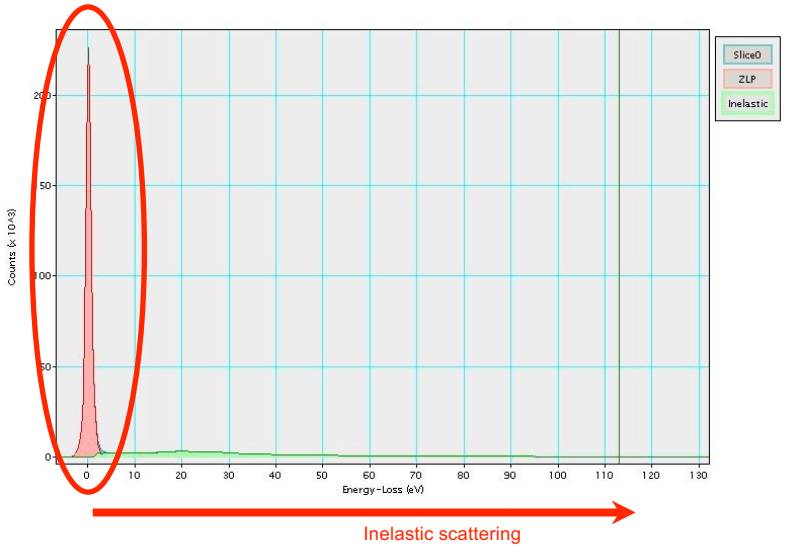
The TEM specific microanalysis technique!



Principle of energy-loss spectrometry



The EELS spectrum | Zero loss

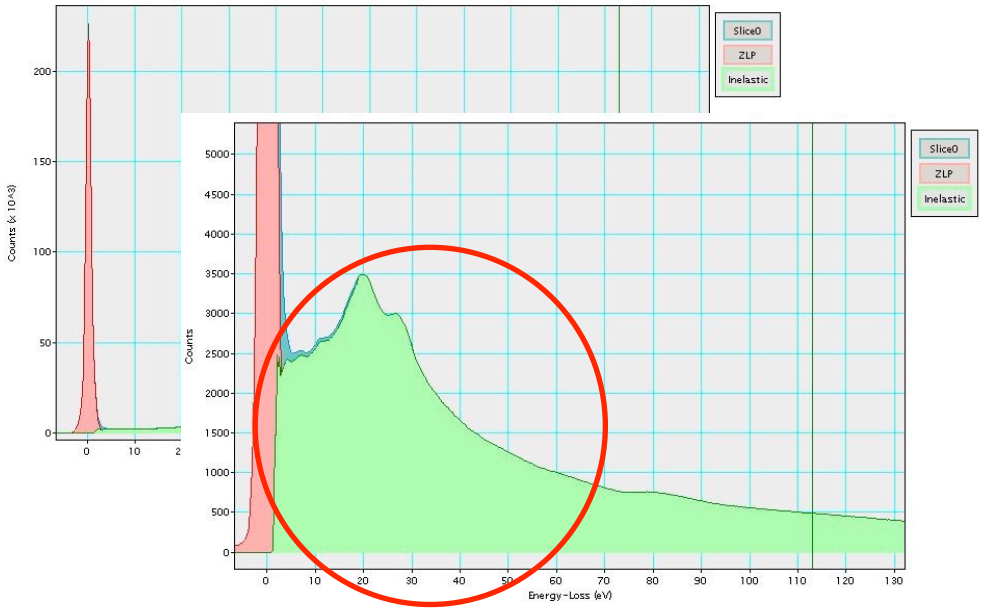


Zero-loss: Elastically-scattered e^-

Information about speciemen thickness

Emad Oveisi

The EELS spectrum | Low-loss

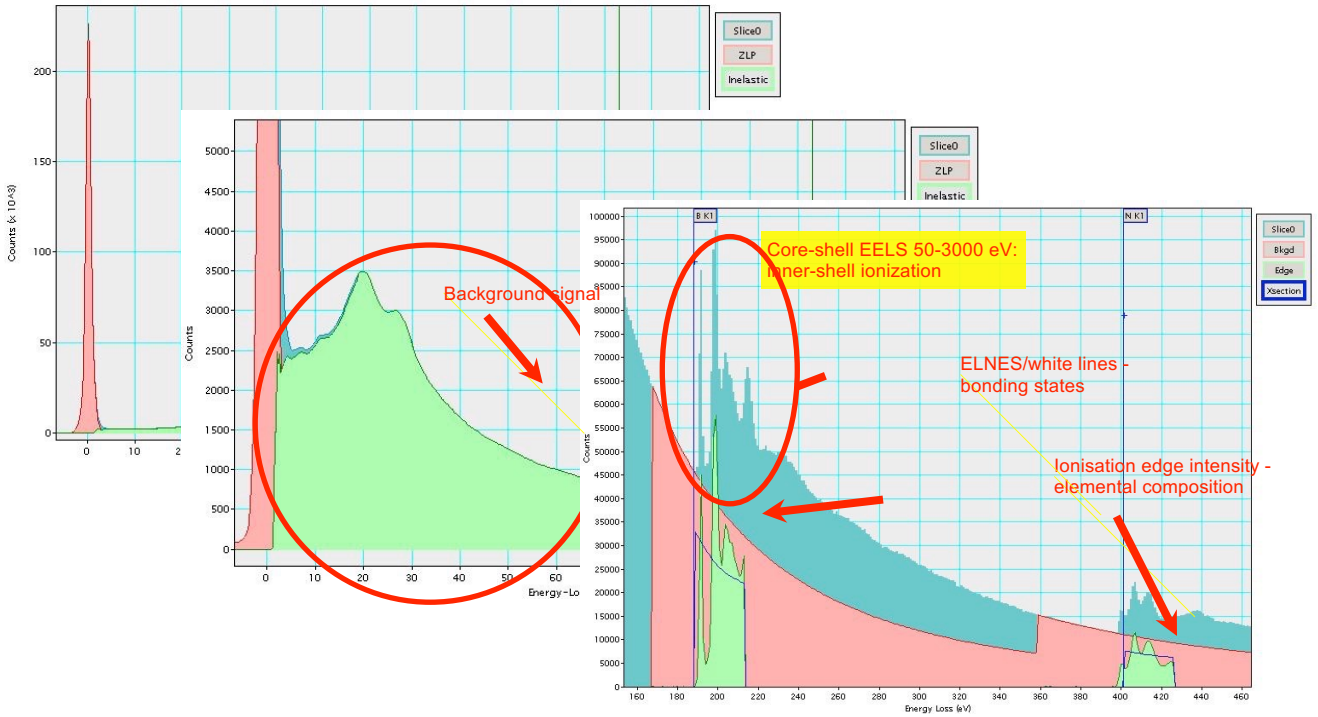


Interaction with outer electrons (conduction/valence)

Low-loss spectrum 0-50 eV: valence excitations - plasmons (bulk & surface),
band gaps, optical properties

Emad Oveisi

The EELS spectrum | Core-loss

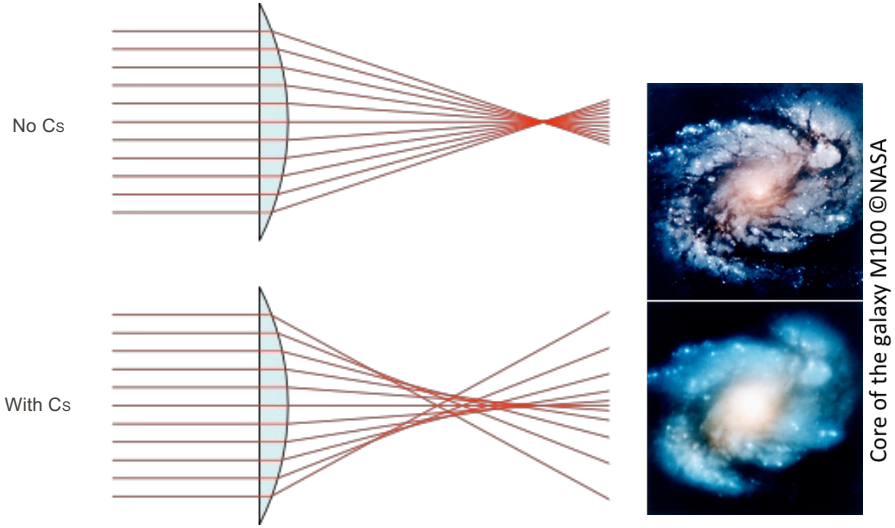


Interaction with core electrons

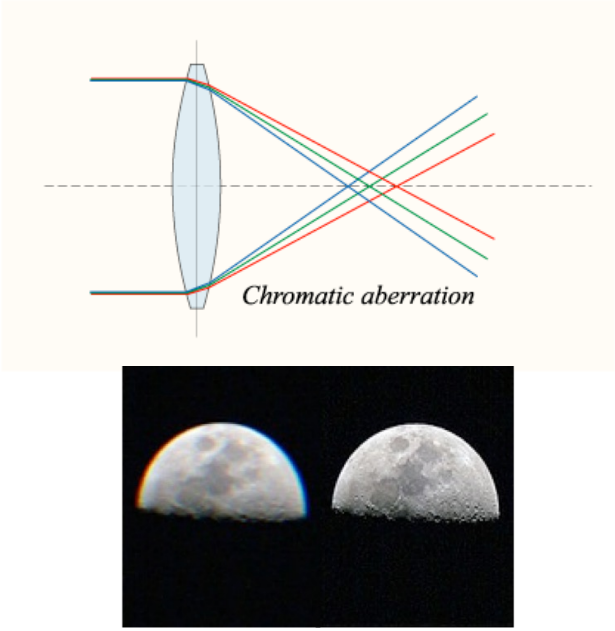
Core-loss spectrum 50-3000 eV: inner shell ionization
ELNES/white lines: Bonding state
Ionisation edge intensity: elemental composition

Limitation to spatial resolution: Lens aberrations

Electromagnetic lenses in TEM column are toroidal
Lenses inherently convergent
→ Spherical aberration (C_S) and Chromatic aberration (C_C)



Parallel rays that pass through the central region of the lens focus farther away than the rays that pass through the edges of the lens. Results in multiple focal points and thus a blurred image.



Lens cannot focus all energies (wavelengths) to the same convergence point.

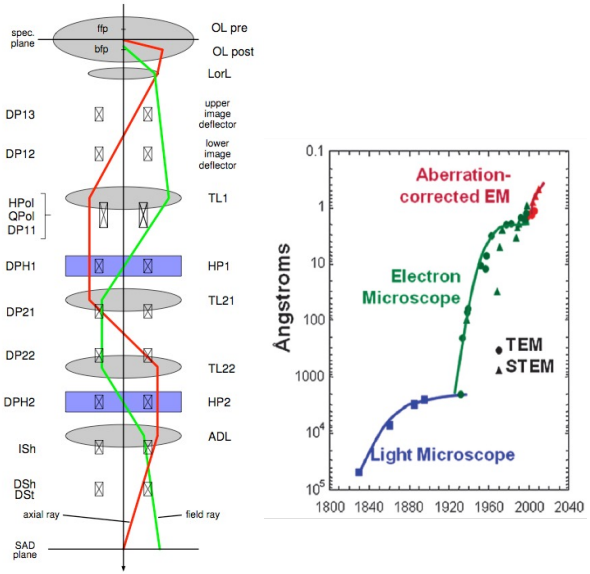
Resolution in TEM limited by aberrations, especially Cs

Emad Oveisi

Cs - aberration correction

Combination of standard radially-symmetric convergent lenses with multipole divergent lenses (e.g. tetrapoles, hexapoles) to tune Cs

Like “glasses” for TEM (or the Hubble)



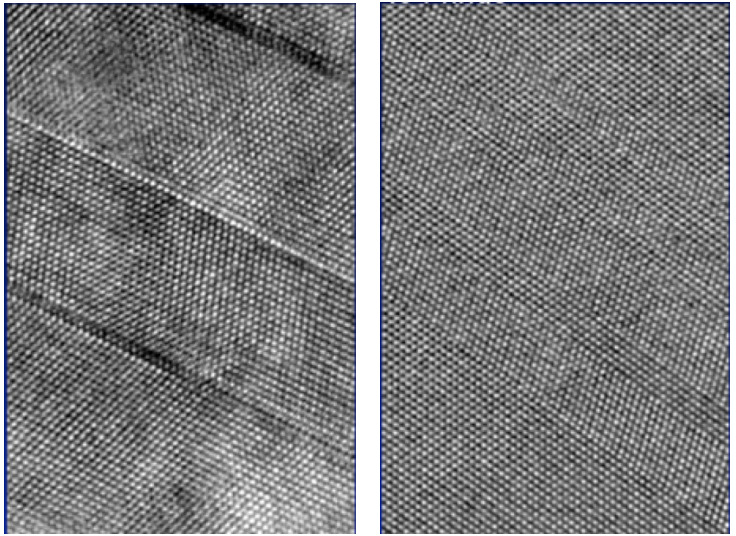
CEOS corrector

→ *Resolution jumps to sub-Å!*

Example: Σ3 grain boundaries in Al

Uncorrected

Cs-corrected



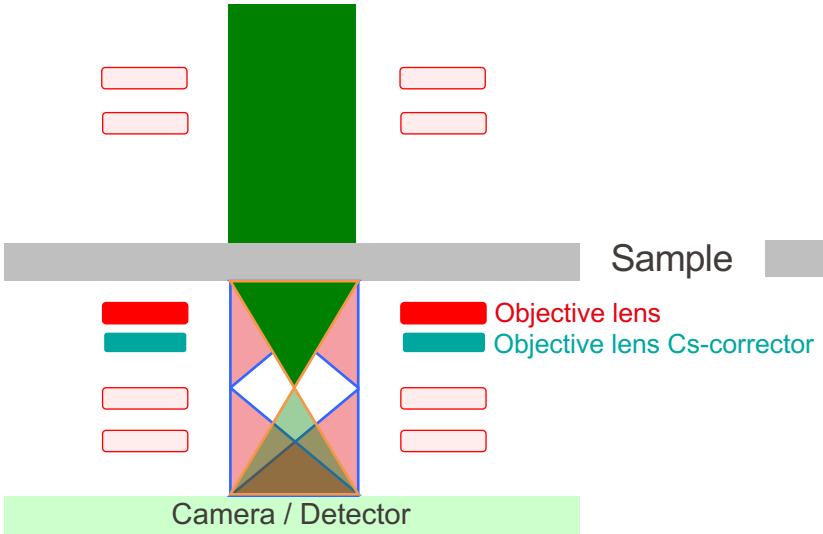
Images: Oikawa, JEOL

Cs - aberration correction

Emad Oveisi

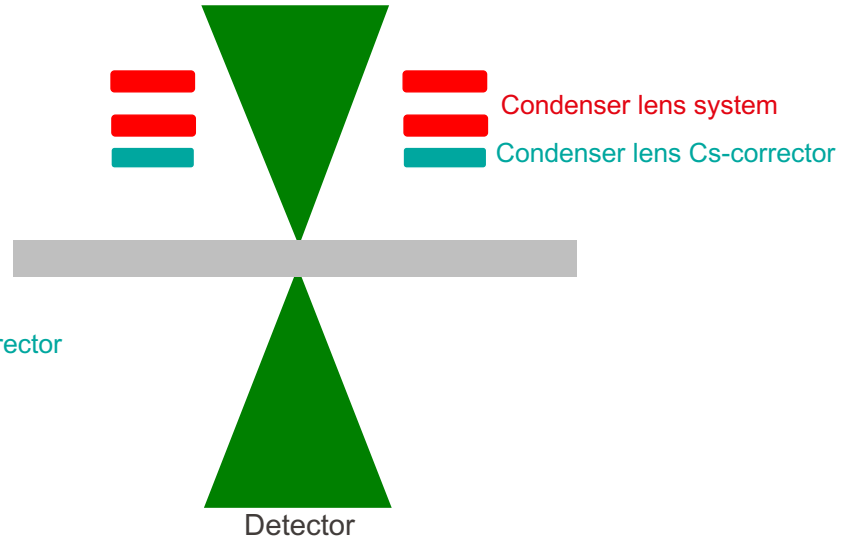
TEM (Image-corrected)

Electron beam (60-300 keV)



STEM (Probe-corrected)

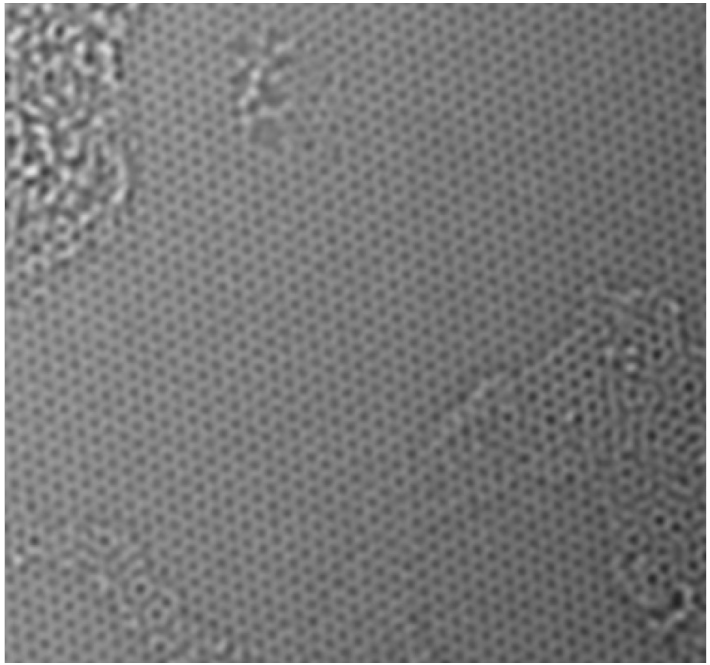
Electron beam (30-300 keV)



A double Cs-corrected S/TEM has both image and probe correctors

Cs - correction: Image-corrected low kV HR-TEM

- Cs-correction and monochromatic illumination to reduce Cc allow sub-Å resolution even at lower kV where electron wavelength λ is greater
- Beam sensitive, low contrast materials can be imaged
- Here a monolayer graphene is imaged with 80 keV, monochromated e^-



- Collaboration with group of Prof. Agrawal, EPFL

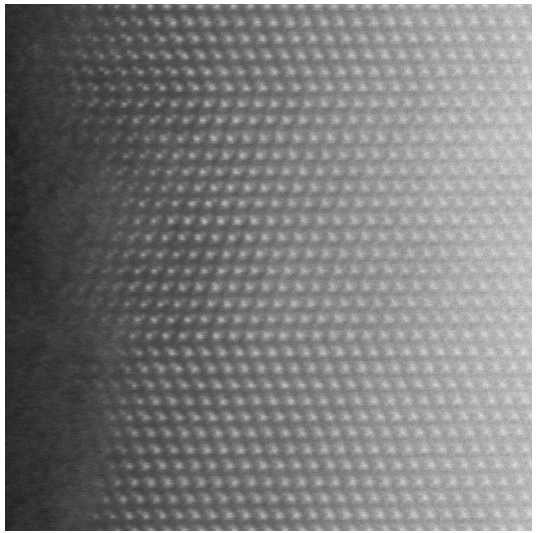
S. Huang et al., Nat. Comm. 2018.

Cs - correction: Probe-corrected HR-STEM

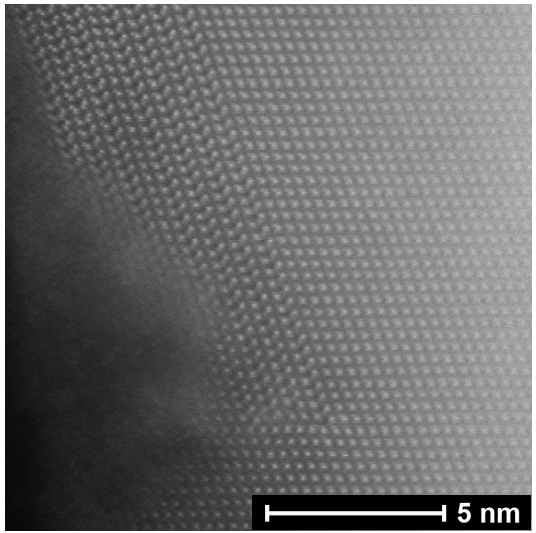
Example: (Al)GaAs nanowires imaged at 300 kV in STEM mode

HR-STEM with sub-Å Cs-corrected probe:

- Simple (quantitative) atomic column contrast
- Reduced sensitivity to small changes in sample orientation
- Image either in focus or out of focus (no contrast reversals with defocus, thickness)



FCC twinning

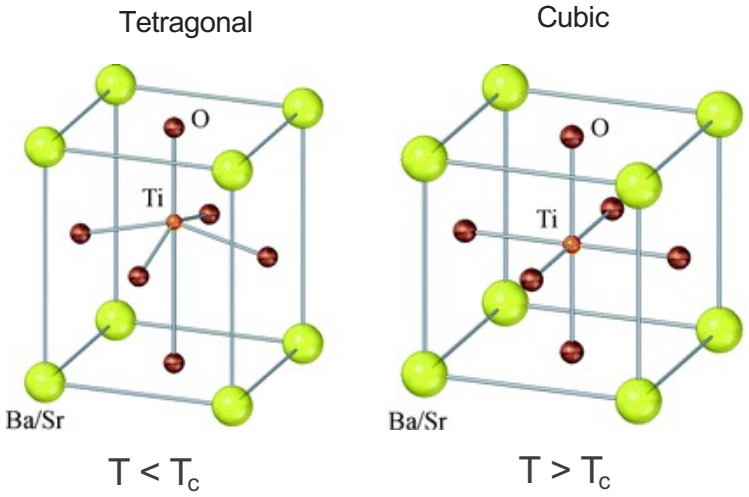


HCP inclusion and dislocation

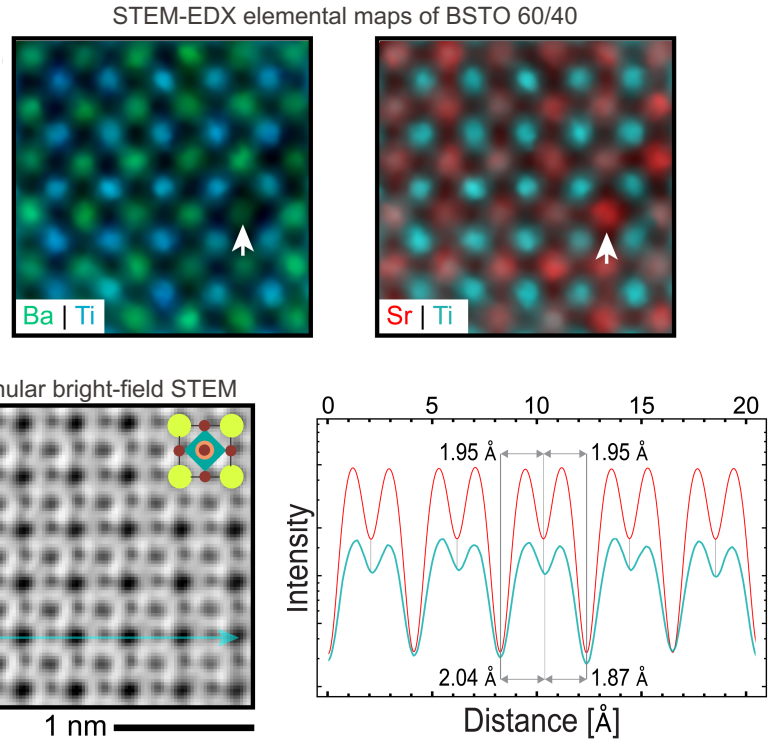
Images courtesy of D. Alexander, EPFL.

Cs - correction: Probe-corrected HR-STEM

Atomic scale symmetry and polar nanoclusters in the paraelectric phase of ferroelectric materials



Shift of ions positions below Curie temperature causes polarization



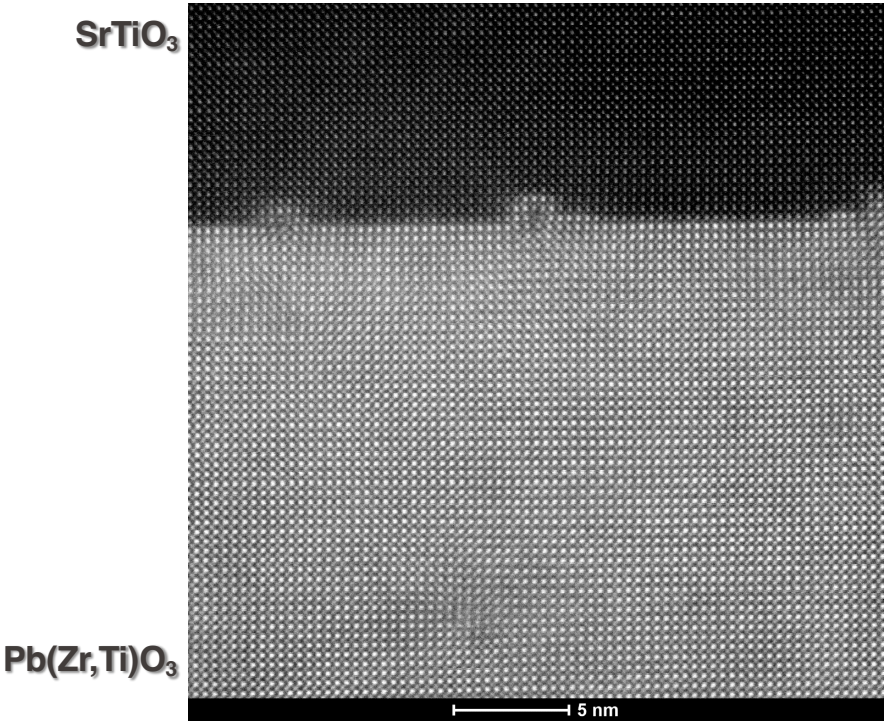
- Collaboration with group of Prof. Damjanovic at EPFL and Jozef Stefan Institute (Slovenia)

Nature Communications (2021).

Cs-correction: probe-corrected HR-STEM

Emad Oveisi

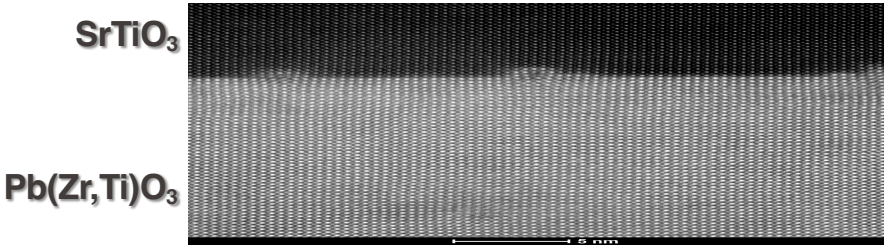
Cs-corrected atomic resolution HAADF-STEM of perovskite interface (300 kV)



Cs-correction: probe-corrected HR-STEM

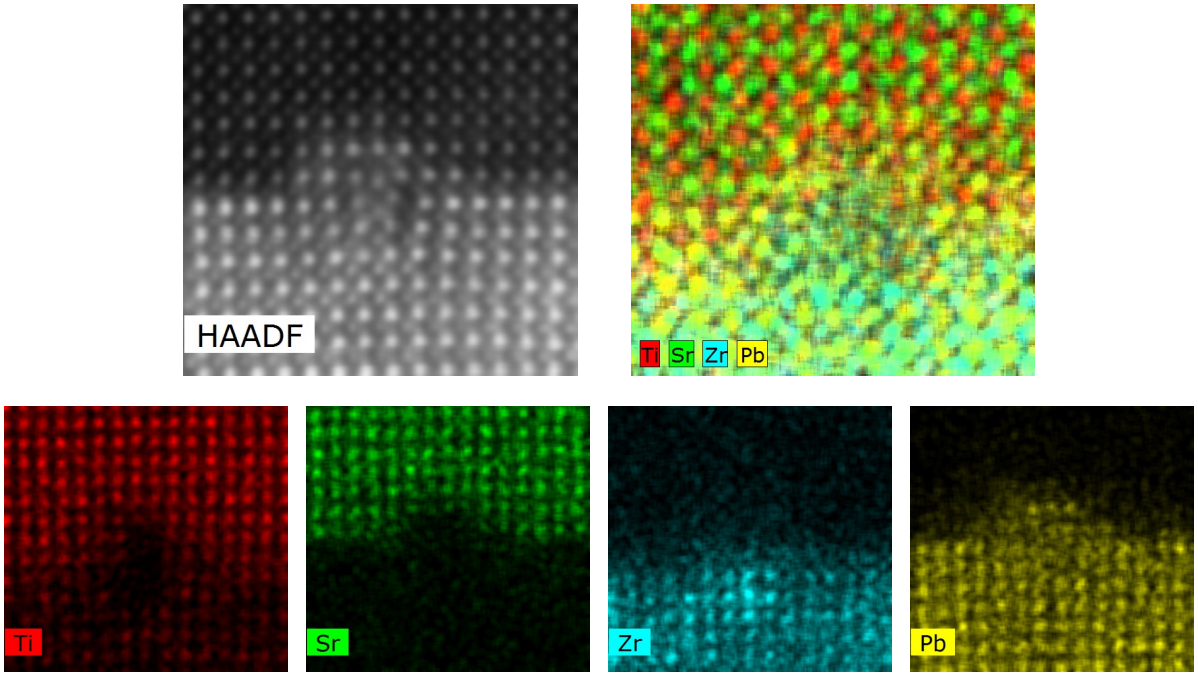
Emad Oveisi

Cs-corrected atomic resolution HAADF-STEM of perovskite interface (300 kV)



Cs-correction: probe-corrected HR-STEM

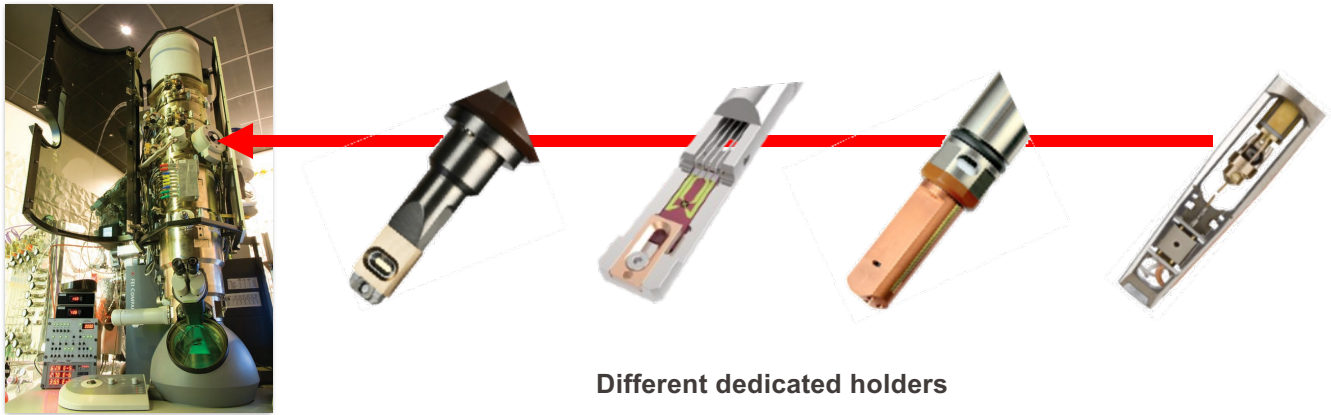
Atomic resolution STEM-EDX mapping reveals diffusion of Pb into strained region of misfit edge dislocation



TFS and CIME in collaboration with LC-EPFL

In situ TEM

Stimuli	Possible techniques	Information
• gas • liquid • temperature • magnetic field • current/voltage • mechanical load • light • …	• (S)TEM imaging • diffraction • electron energy-loss spectroscopy • energy-filtered TEM • energy-dispersive X-ray spectroscopy • Lorentz microscopy • holography • …	• microstructure • crystallography • chemistry • diffusion/migration • optical properties • electric fields • magnetic fields • …



Different dedicated holders

In situ TEM

Liquid phase processes
Hydration and other liquid phenomena
Crystal growth from the liquid phase
Corrosion

Phase transformations
Solid state phase transformations
- Crystallization and melting
- Structural transformations
- Solid state reactions

Solid state transformations in nanostructured materials
- Size-dependent transformations in embedded nanoparticles
- Shape and phase transformations in free-standing nanoparticles
- Sintering of nanoparticles

Elastic and plastic deformation
Microscopic phenomena during plastic deformation
- Deformation of polycrystalline materials
- The effect of gas environment on deformation
- Grain boundary motion
- Deformation phenomena in single crystals
- Deformation of multilayers

Relaxation of epitaxially strained materials
Mechanical properties of nanostructures, thin films and surfaces
- In situ indentation and straining of thin films
- Mechanical properties of nanostructures
- Surfaces: Tribology and Nanomanipulation

Surface reactions and crystal growth
Modification of surface structure
Oxidation and other chemical reactions at surfaces
Growth of nanostructures
- Carbon nanotubes

Thin film growth and defect formation
- Epitaxial growth
- Polycrystalline growth

Crystal growth on patterned substrates

Domain wall motion and flux dynamics
Magnetic domain switching
Flux motion in superconductors
Switching phenomena in ferroelectrics

Correlation of structural and electronic properties of materials
Electrical measurements on TEM samples: samples as devices
Electrical measurements on individual nanostructures

Beam induced processes
Electron beam induced phenomena
- Interaction with the vapour above the specimen
- Formation of point defects
- Beam induced transformations, surface reactions, growth
- Radiation enhanced dislocation motion

Hole drilling
Ion implantation
- High energy ion accelerators
- FIB in the TEM

Science of Microscopy, ed. Hawkes and Spence, 2006.

In situ TEM: Advantages and limitations

- A window into the behavior of materials under real processing conditions.
- A continuous view of a process, which may take the place of multiple post-mortem measurements.
  - It is easier to catch a transient phase or observe a nucleation event.
  - Specific and detailed kinetic information, such as the motion of individual dislocations under known stress, or growth rates of individual nanocrystals.
- This unique information comes at the cost of increased experimental complexity.
- Expensive: machine time, holders

Summary

TEM is now not one technique that can be easily summarised, but is split into many specialisms. In essence this is due to the many types of interaction of the electron beam with the atoms of a sample.

Modern TEM instruments are arguably the most versatile analytical tools. Many other possible uses (e.g. imaging of magnetic domains, optical plasmon mapping, in-situ studies) exist..

Aberration-correction and improved instrument stability give sub-Å resolution in TEM and STEM. With lower beam voltages lighter elements can be analysed without beam damage.

Faster, more sensitive spectrometers give unprecedented access to composition, chemistry and physics of materials.

Computer interfaces and software allow acquisition and processing of large datasets.

With the latest instrumentation, the sample and specimen preparation are often the limiting factor!

Some useful literature

Transmission Electron Microscopy by D.B. Williams and C.B. Carter (Springer)

Large Angle Convergent Beam Electron Diffraction by J.P. Morniroli

Aberration-corrected imaging in transmission electron microscopy: an introduction by R. Erni

Scanning Transmission Electron Microscopy by S.J. Pennycook and P.D. Nellist (eds) (Springer)

Diffraction Physics by J.M. Cowley (North Holland/Elsevier)

Science of Microscopy by C.W. Hawkes and J.C.H. Spence (eds) (Springer)

Transmission Electron Microscopy: Physics of Image Formation and Microanalysis by L. Reimer