Recent developments of Transmission Electron Microscopy for the characterization of nanostructures

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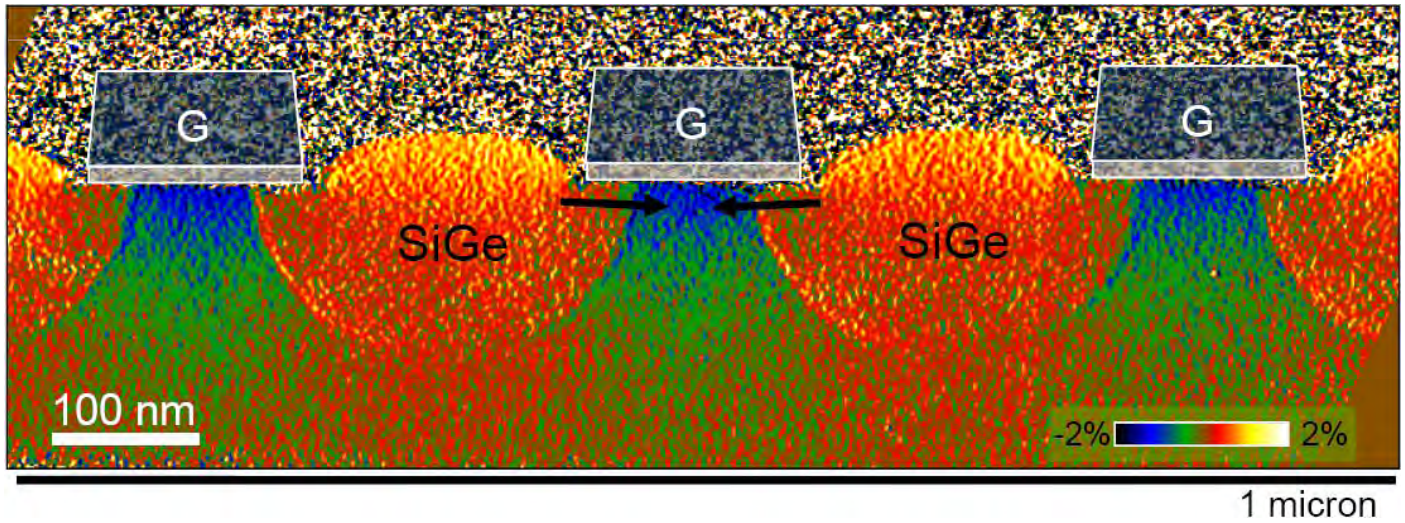
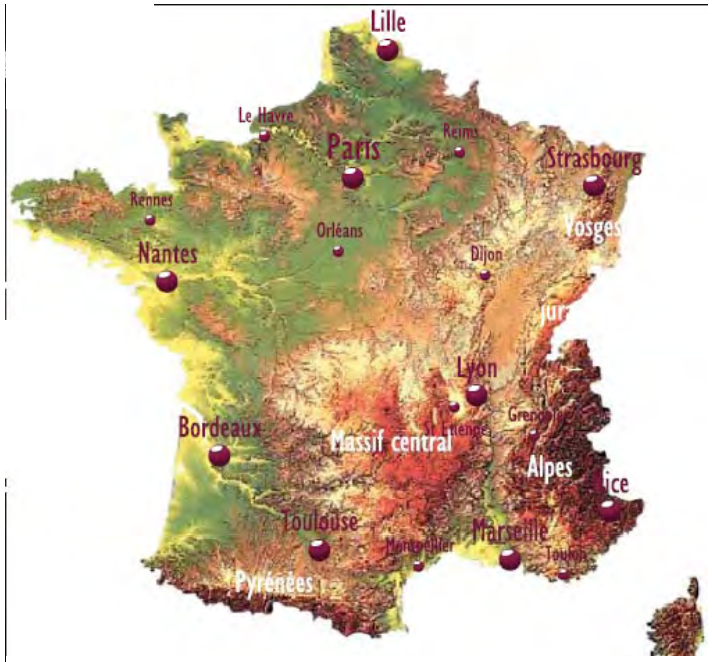
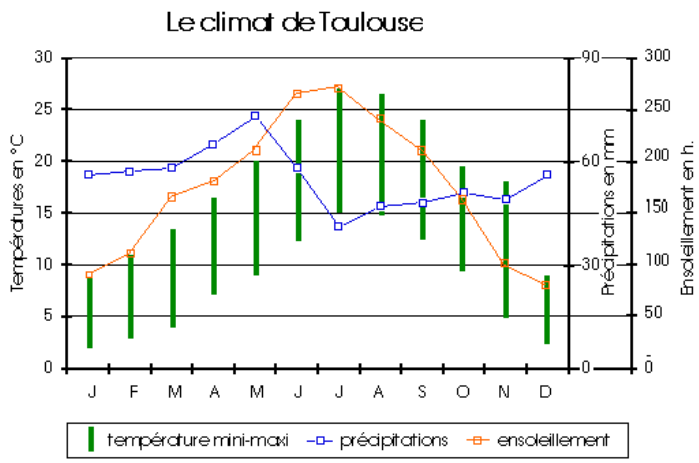


Where do I come from ?





Where do I come from ?

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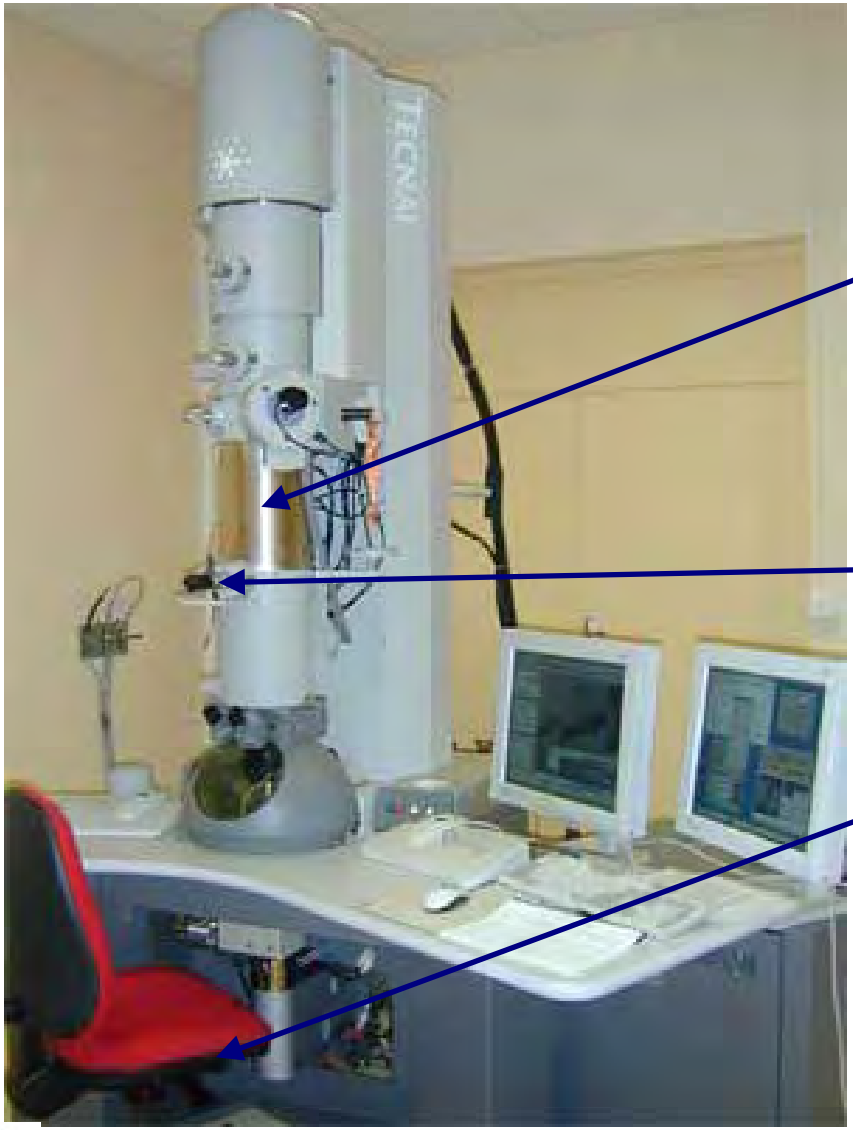
SACTEM Toulouse 200 kV FEG, TECNAI



Bright source of electrons
and small probe (down to 1 nm)

Cs corrector CEOS

0.12 nm point to point resolution
HREM without delocalisation

Holography prism
local electromagnetic fields

TRIDIEM Energy Filter GATAN

EELS spectrometer
0.7 eV energy resolution
1 nm spatial resolution
Image filtering
elemental mapping
with 1 nm spatial resolution

Outline

Basic theories and techniques

1) Imaging with electrons
2) Specificities of TEM
3) Imaging techniques
4) EELS and EFTEM
5) Holography
6) Sample preparation

TEM through examples

1) Dislocation loops in ion implanted Si
4) H-related platelets in semiconductors
6) Nanocrystals
7) Strain in layers and devices

Imaging with electrons

Three main reasons for "a microscopy using electrons"

-wave nature of electrons: when accelerated with a high potential V, λ=h/mv

$$\lambda = \frac{h}{\left[2m_0 eV\left(1+\frac{eV}{2m_0 c^2}\right)\right]^{\frac{1}{2}}}$$

V=300 kV,

λ=0.00197 nm

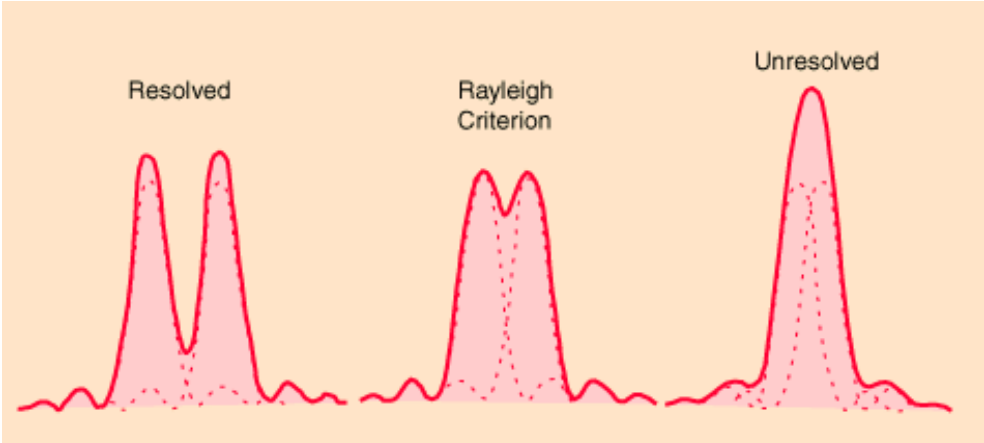
- lenses exist: a monochromatic parallel electron beam can be focused by a magnetic lens

- electron sources exist: electrons can be emitted by heating (thermoionic) or when applying an intense electron field (FEG)

Wavelength and resolution

- Very small wavelength => high resolution

- In practise, resolution limited by diffraction effects, mainly from the objective lens, which decrease with decreasing λ

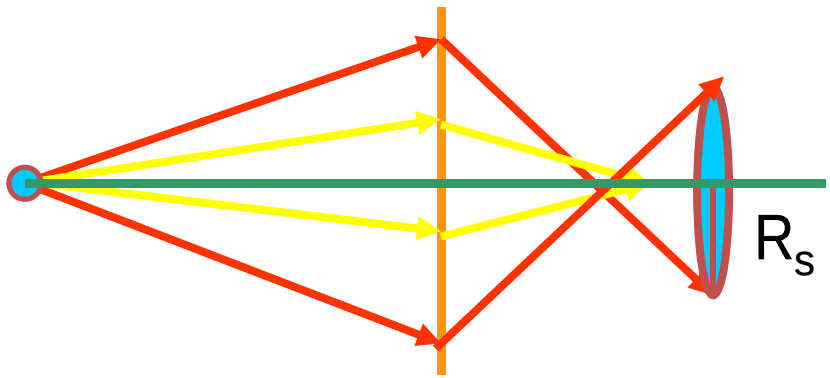


Rayleigh criterion

$$\boldsymbol{R}_{th} = 0.61\,\frac{\lambda}{\alpha}$$

α lens effective angular aperture, limited by spherical aberrations

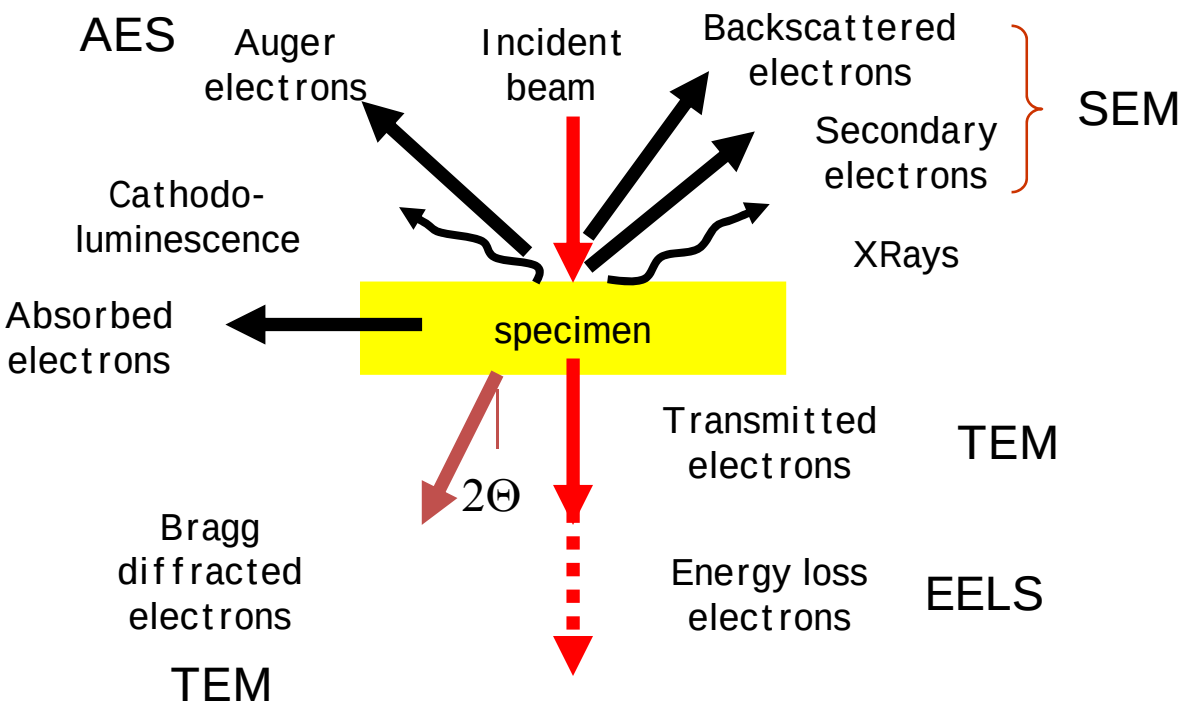
$R_s = C_s \alpha^3$



Cs~1 mm => R_s=0.17 nm
for V=400 kV

Information obtained by TEM

- Result from e/solid interactions
- electrons scattered from both electrons and nuclei of materials
- 2 contributions: elastic (small angles, no loss) / inelastic (with e, energy losses)



- very strong elastic interactions => very thin sample (<100 nm) => preparation step ?

Coherent diffusion: electron diffraction (1)

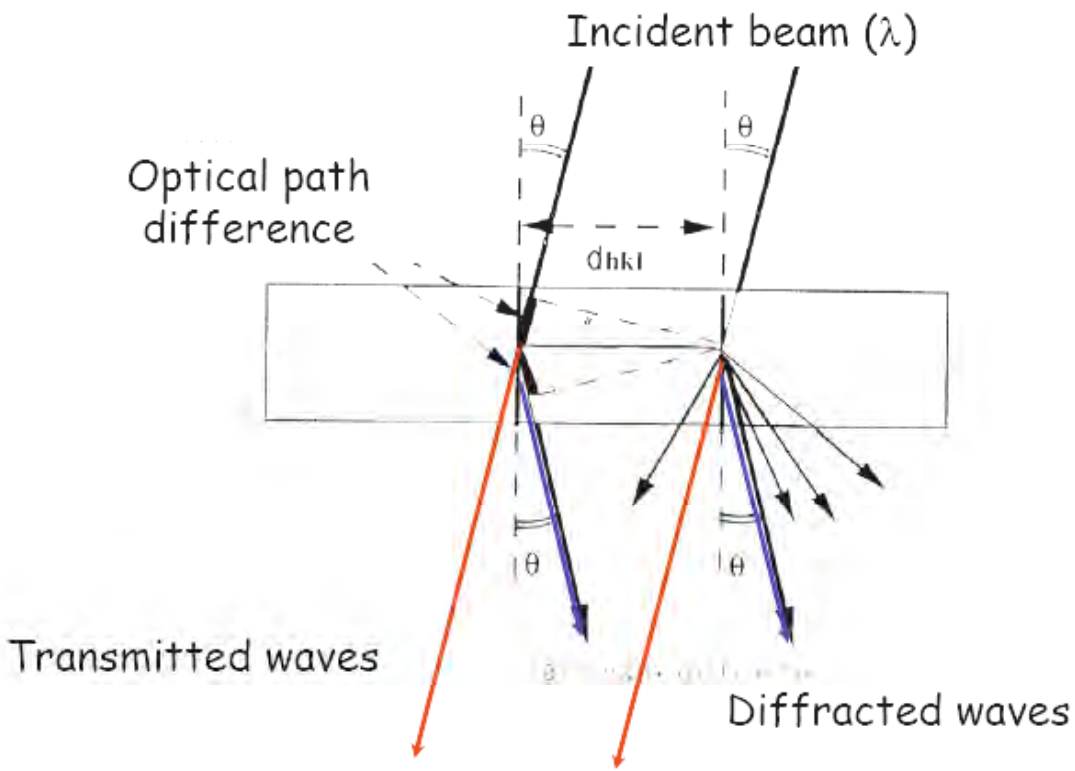
- 1 atom scatters everywhere, with I that decreases as angle increases
- periodical arrangement of atoms => I max in discrete directions

=>

Bragg's law

$$2d_{hkl}\sin\theta_B = n\lambda$$

θ_B incidence angle of one diffracted beam



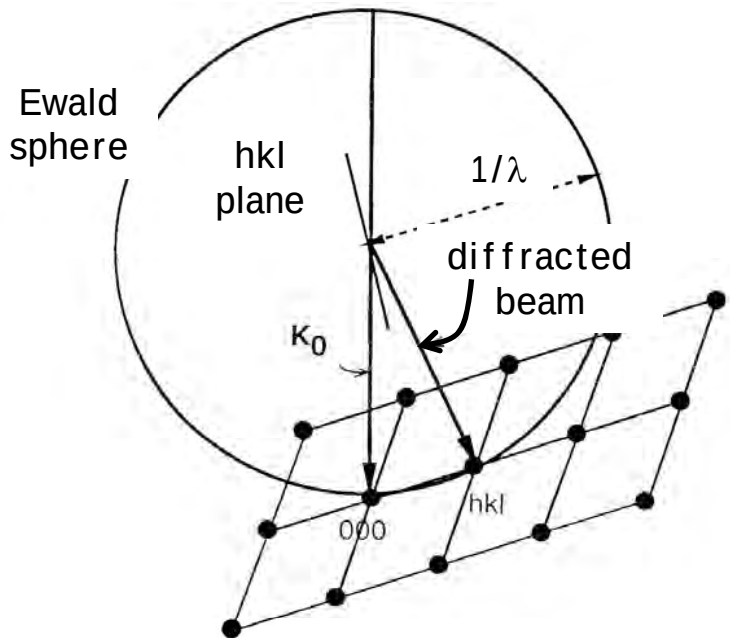
Coherent diffusion: electron diffraction (2)

- Reciprocal space

  $g_{hkl}=1/d_{hkl}$

- Diffraction pattern = intersection of the Ewald sphere ($1/\lambda$) with the reciprocal space of the crystal

- Spatial frequencies in the diffraction pattern: $u=1/d= 2\theta_B/\lambda$

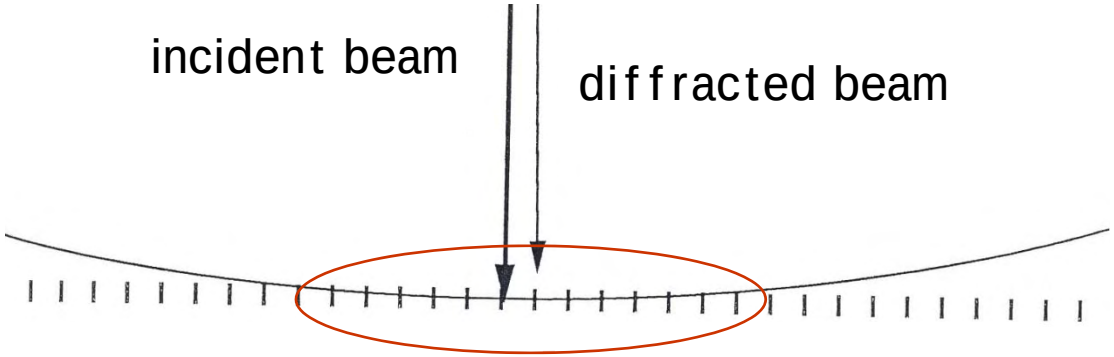


Coherent diffusion: electron diffraction (3)

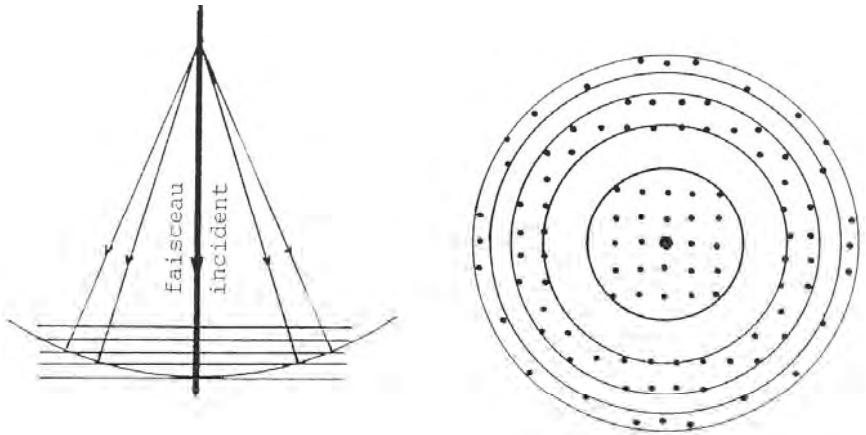
- For electrons at hundreds of KeV, $1/\lambda$ large

  => Ewald sphere almost flat

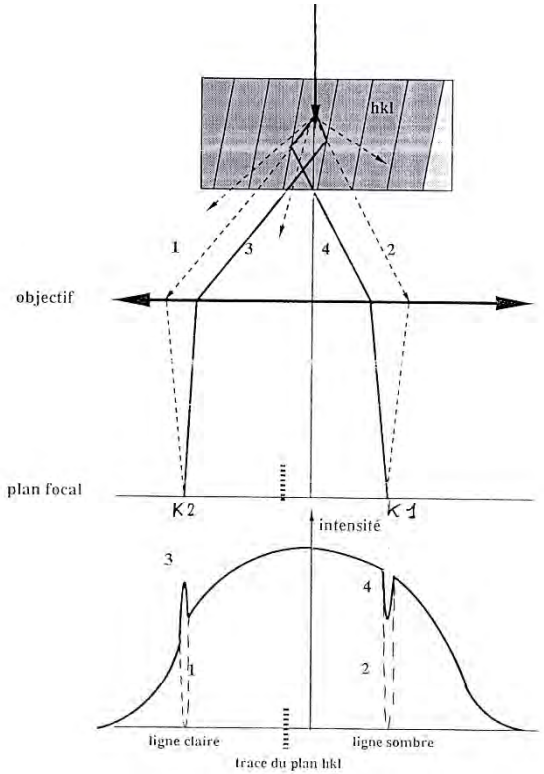
  => diffraction pattern = planar section of the reciprocal space

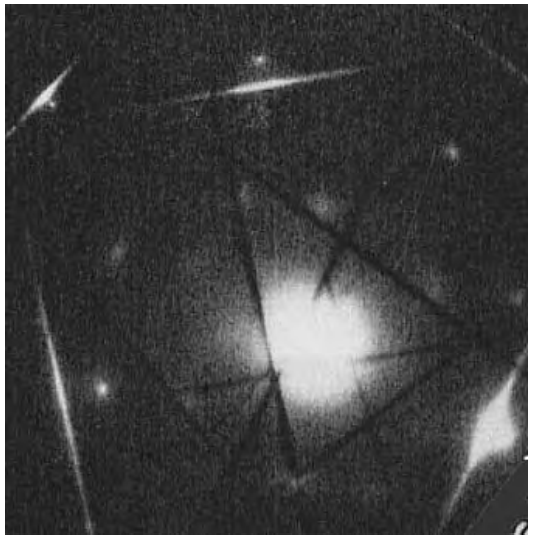


- Ewald sphere intersects different planes => circular regions named Laue zones



Coherent diffusion: electron diffraction (4)





- “thick” objects => elastic + inelastic diffusion => Kikuchi lines (b/w)
- each pair ⇔ (hkl), d(b,w)=1/dhkl
- position in the pattern very sensitive to the sample orientation
  => sample orientation / e-beam

Specificities of TEM

- Structural information from both the real space (image) and the reciprocal space (diffraction)

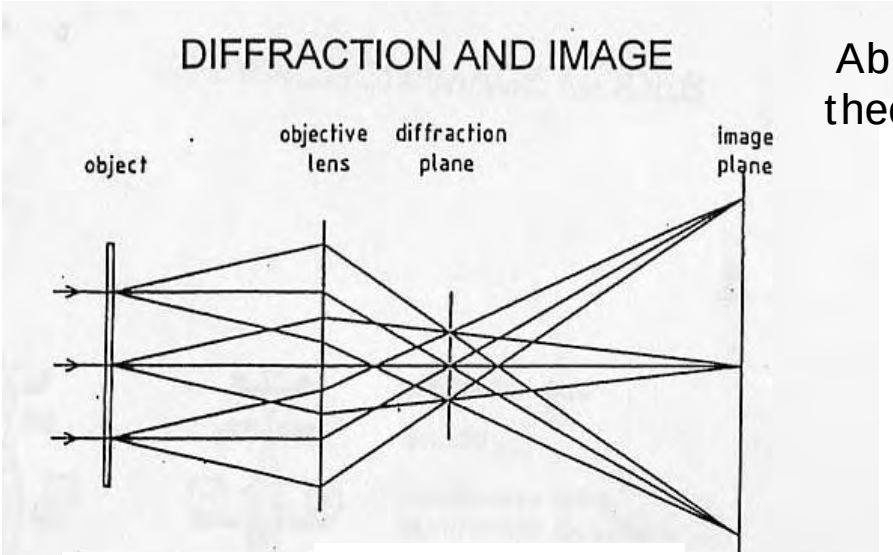
- Working with both modes is necessary for materials science because:

1) image contrast depends on diffraction conditions
2) diffraction pattern depends on sample morphology

How does it work?

Diffraction and image (1): a bit of theory

Incident beam ⇔ atomic potential
q(x,y) transmission function of the object (A, Φ)



Abbe theory

ψs wavefunction at the exit plane

$\psi_d(u,v)$= FT($\psi s(x,y)$)
intensity distribution

ψi=FT($\psi d(x,y)$)
First image

- Ray paths in an ideal lens !

Diffraction and image (2): a bit of theory

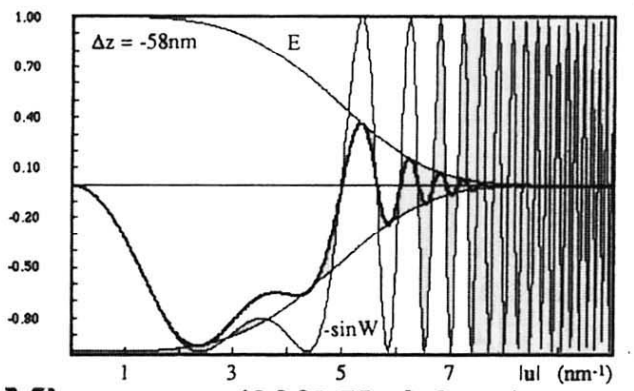
- Not as simple because objective lens not perfect (aberrations + aperture)

  - Aperture function $O(u,v) = 1$, for $(u^2+v^2)^{1/2} \leq uo$
    $= O$, elsewhere

  - function modifying the relative phases of the beam according to their positions in the back focal plane

$$\exp(i\chi(u, v))=\exp(i\pi\Delta \mathbf{f}\lambda(u^2+v^2)+i\pi/2\mathbf{C_s}\lambda^3(u^2+v^2)^2)$$

=> <u>Contrast transfer function</u> $T(u, v) = O(u, v) \cdot \exp\{i\chi(u, v)\}$



Microscope (300kV; 0,2nm)

Diffraction and image (3): a bit of theory

- Wave function in the back focal plane becomes

$\psi_d(u,v)=FT(\psi_s(x, y)).\mathbf{T(u,v)=} FT(q(x, y)).T(u,v)$

- Image wave function:

$\psi_i(x,y) = FT\{\psi_s(u,v)).\ T(u,v)\} = FT\{\psi_s(u,v)\}*FT\{T(u,v)\} = \psi_s(x, y)*t(x,y)$

$= q(x, y)*t(x,y)$

with $t(x,y)=\mathbf{FT(\ T(u,v))}$, the point response function of the objective lens

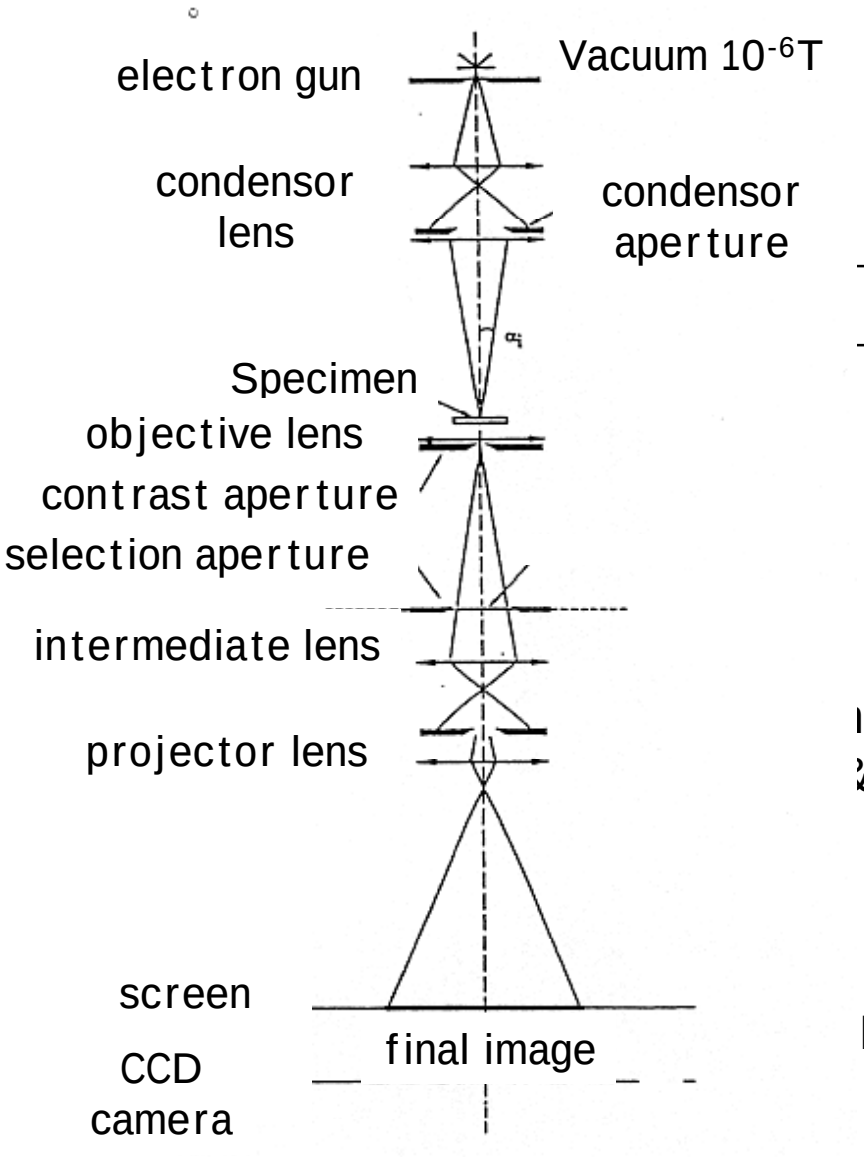
- Intensity distribution in the image:

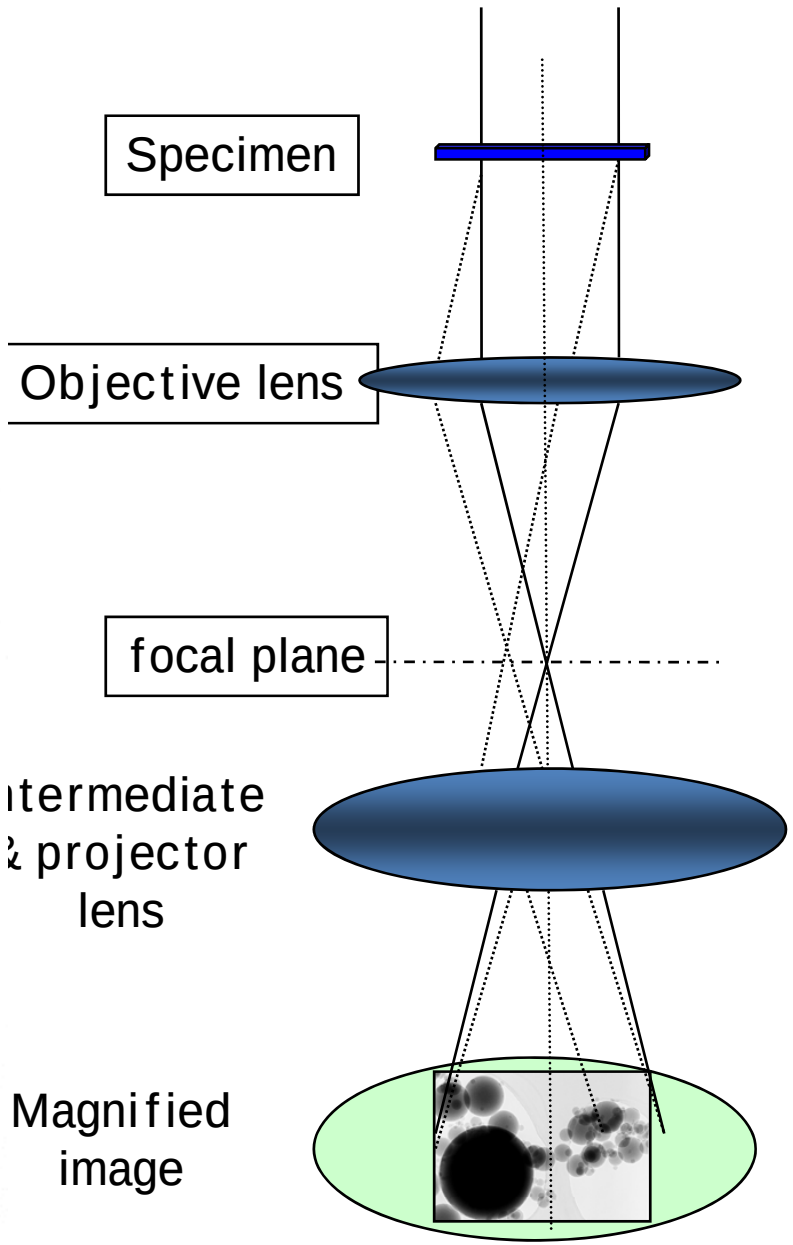
$$\boxed{I(x,y)= |\psi_i(x,y)|^2 = |q(x, y)*\mathbf{t(x,y)}|^2}$$

=> Relation between intensity distribution in the image / structure of the object is not direct

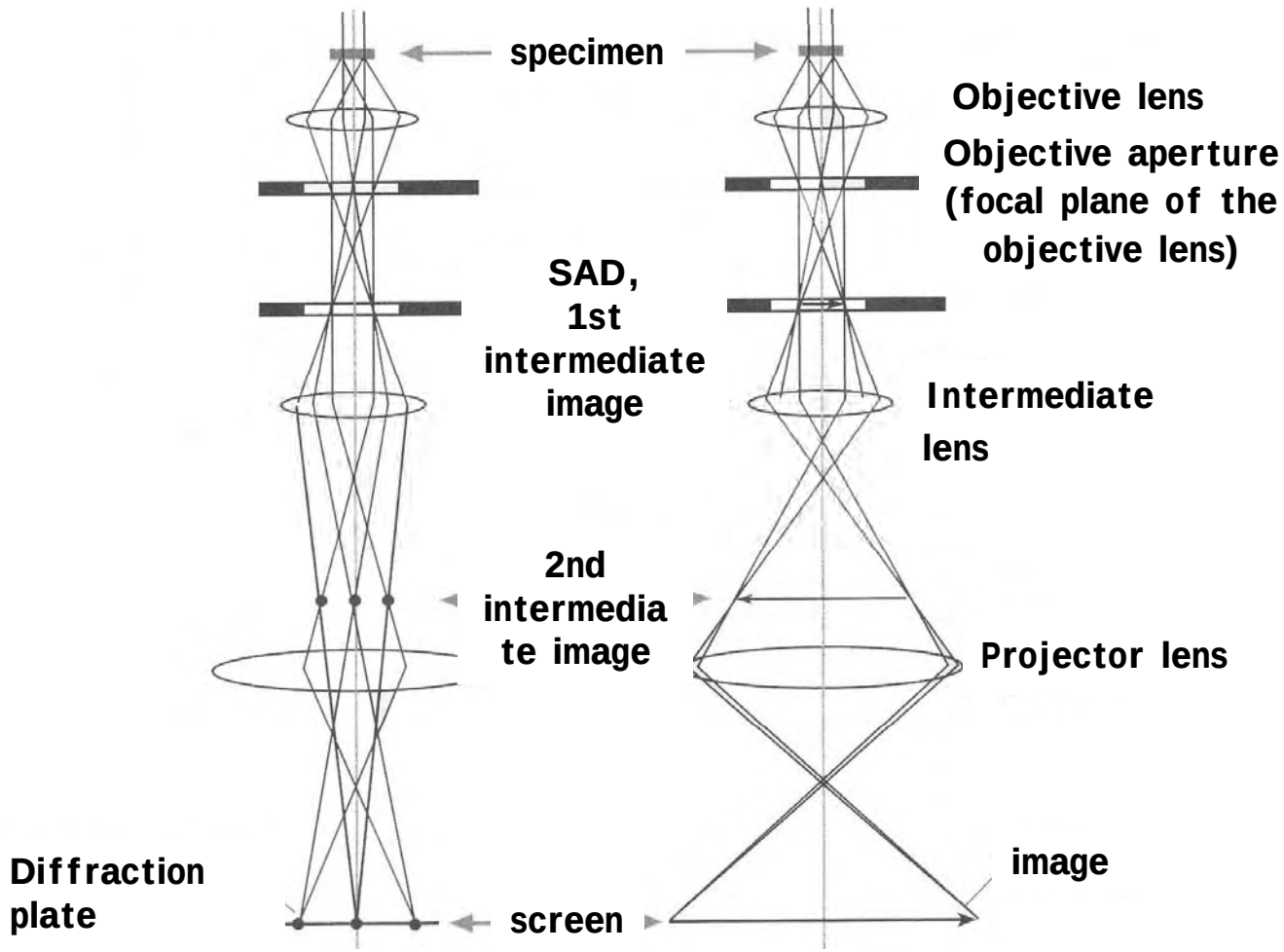
+ approximations (incident beam neither parallel nor monochromatic + chromatic aberrations)

Components of a TEM





Diffraction mode/image mode



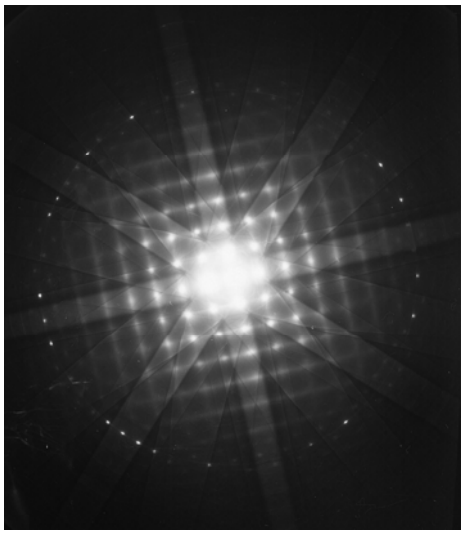
Diffraction/imaging mode: adjust the excitation of the lens to image on the screen either the focal/ image plane of the objective lens

Diffraction techniques (1)

Different purposes: identify unknown crystals / align a known crystal /e-beam => tilt the specimen/e-beam (double tilt holder)

Different techniques following the convergence of the beam or by limiting the diffracting zone:

- SAD: select in image mode a region by setting an aperture in the image plane of the objective lens + switch in diffraction mode (0.5 µm)



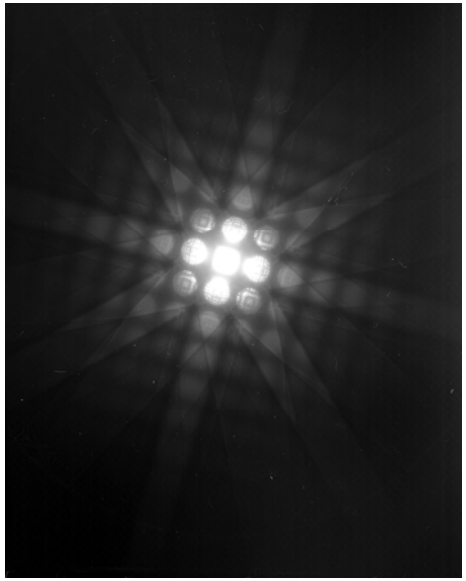
SAD
Si, B=[001]

Diffraction techniques (2)

- Micro and nano-diffraction: reduce the size of the illuminated area by adjusting the 2 condensor lenses

  => 10 nm (1 nm in nanoprobe mode)

  => precipitates (>10 nm in diameter) but irradiation



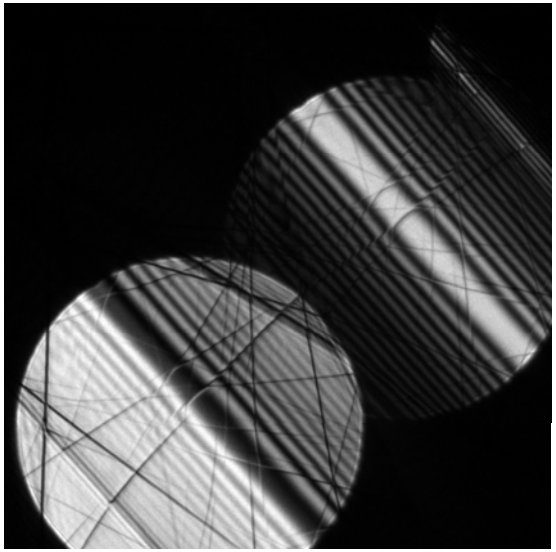
Micro-diffraction
Si, B=[001]

Diffraction techniques (3)

Convergent beam: the e-beam is focused on the sample with a very large angular aperture. Spots->disks with lines containing information relative to the propagation of electrons along the ≠ directions

=> bi-dimensional mappings of the I distribution = f(orientation of the crystal/e-beam)

=> sample thickness,
point group of the crystal (symmetries),
changes (10^{-3}) in inter-planar distances due to strain.



CBED
Si, close to [001]

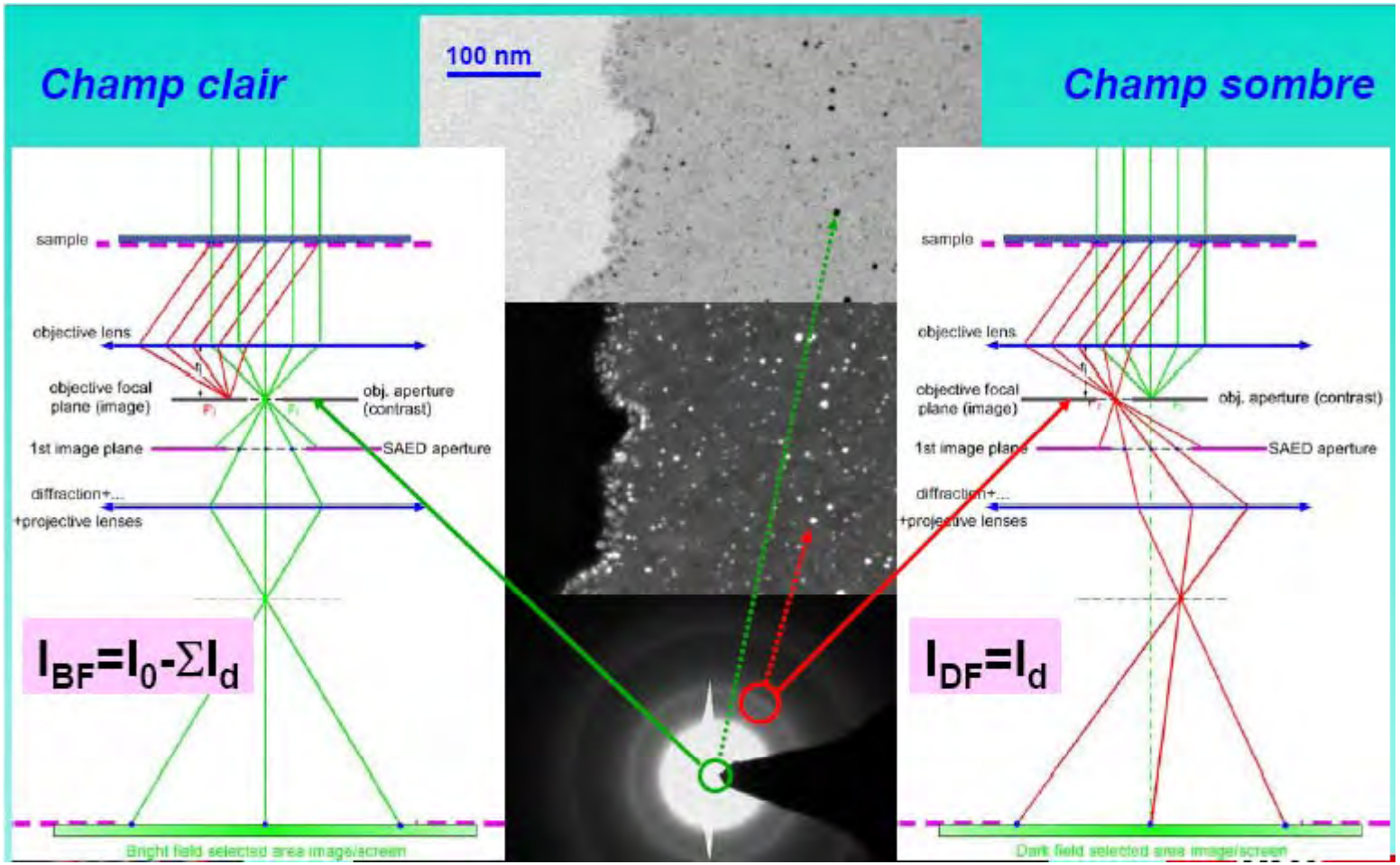
Imaging techniques

2 types, depending whether the e-beams emerging from the object interfere in the image plane or not,

- amplitude (diffraction) contrast => “only one” beam (no phase information)
  - “Conventional imaging”: Bright field, Dark field, Weak Beam Dark Field
    “Low” magnification (some x 10000) => “low” resolution, in the nm range

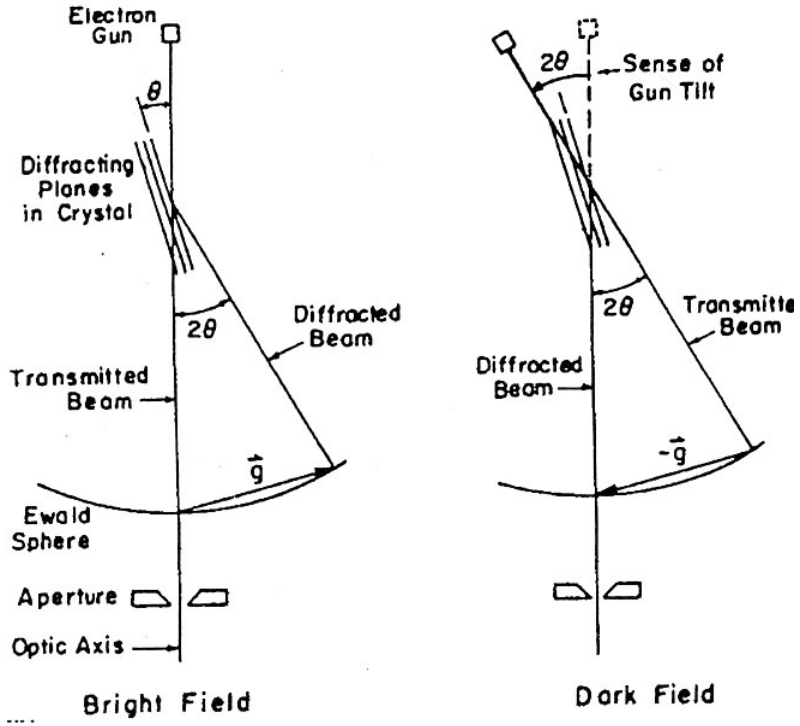
- phase contrast imaging
  - Fresnel Contrast (abrupt Z change)
  - High Resolution Electron Microscopy (HREM)
    High magnification (some x 100000) => atomic resolution

Diffraction contrast: Bright Field / Dark Dield



Diffraction contrast: 2 beams condition in a crystal

1) Excite 2 beams (T + D)
2) Select 1 beam with the objective aperture

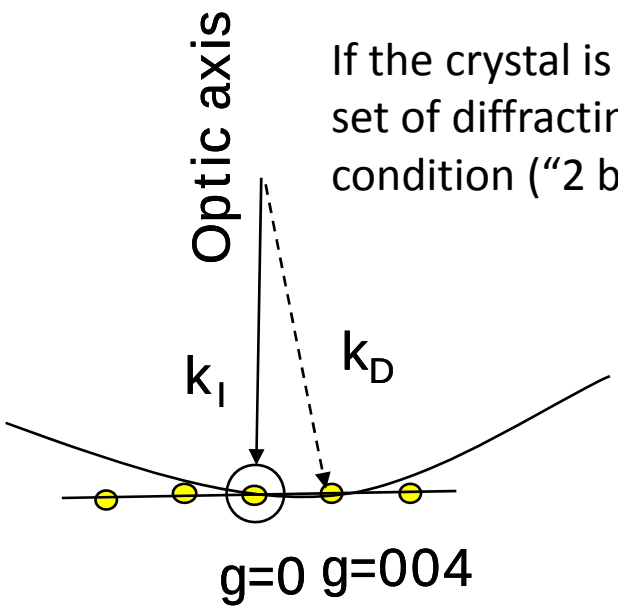


Bright field: select the transmitted beam => zones in the object which do not diffuse electrons are bright

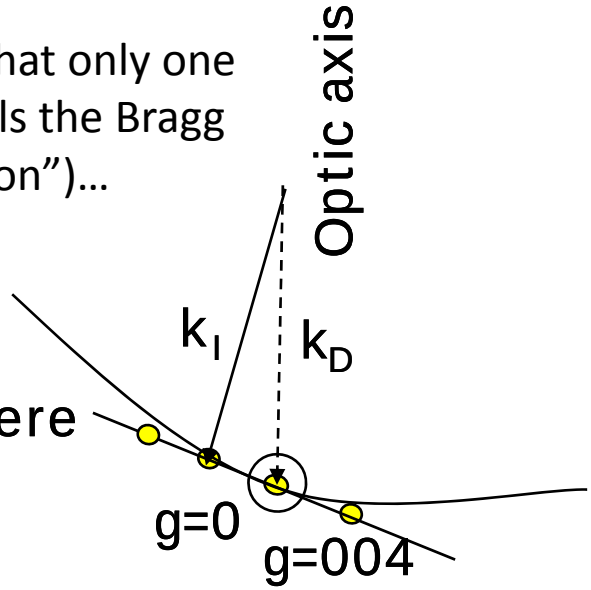
Dark field: select one diffracted beam => only the zones in the object from which these electrons originate are bright

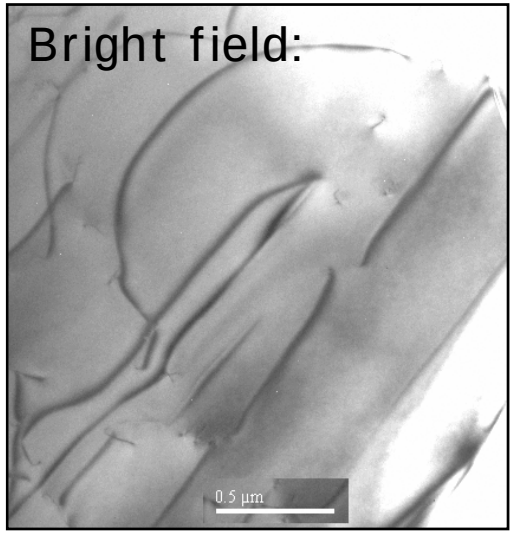
Diffraction contrast: 2 beams condition in a crystal

If the crystal is oriented so that only one set of diffracting planes fulfils the Bragg condition ("2 beams condition")…

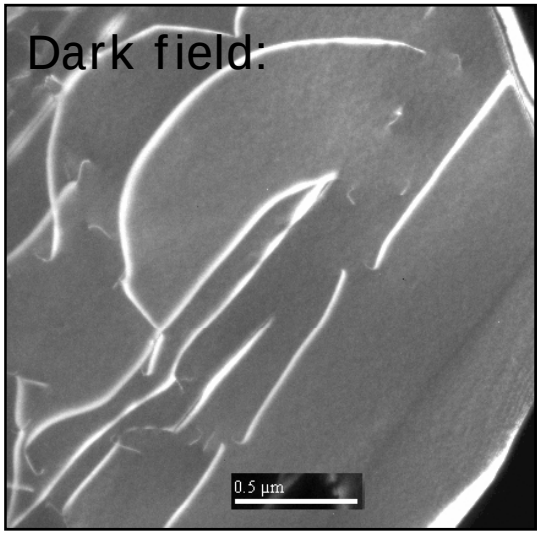


Ewald sphere





…BF/DF complementary



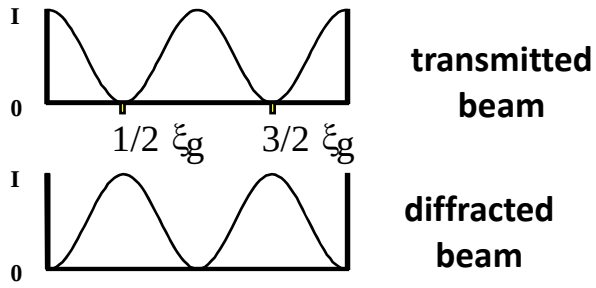
Contrast in conventional electron microscopy

Intensity of the transmitted/diffracted beams (=> contrast in the image) results from the variation of :

- orientation/e-beam
- atomic composition
- atomic structure
- strain
- specimen thickness…

Thickness fringes

•intensity of the transmitted (diffracted) beams at the exit surface of the specimen depend on its thickness (t)

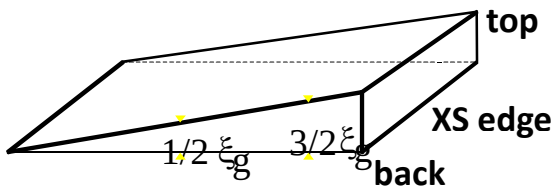


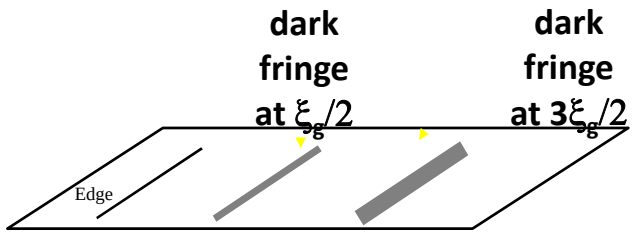
I oscillations from black to white when t increases: “thickness fringes”

From the first black fringe to the next t has increased by ξ_g (extinction distance)

ξ_g = f(crystal, ghkl, λ)

=> detect strain, estimate thickness





bright-field image

Thickness fringes

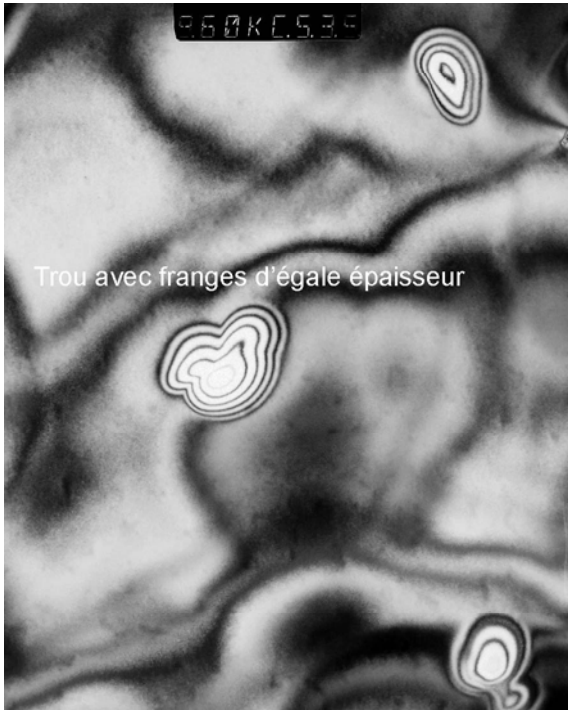


Image in Bright Field near a hole, with associated thickness fringes

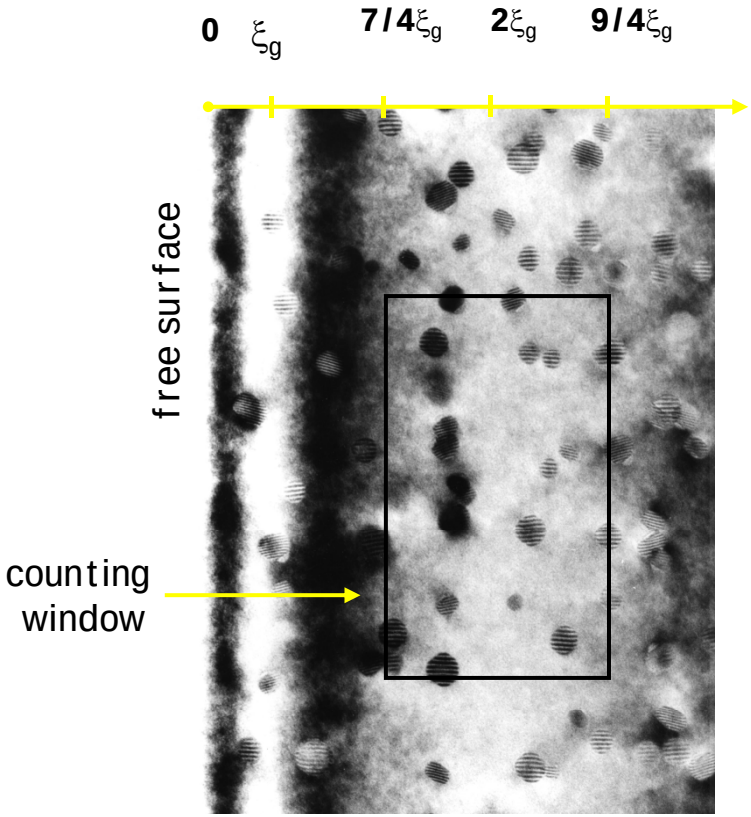


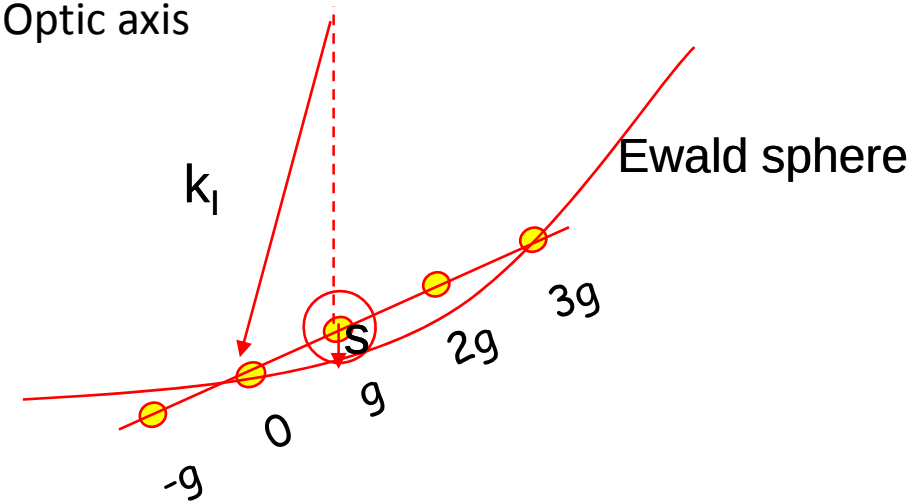
Image in Bright Field two beams of As precipitates in GaAs with thickness fringes

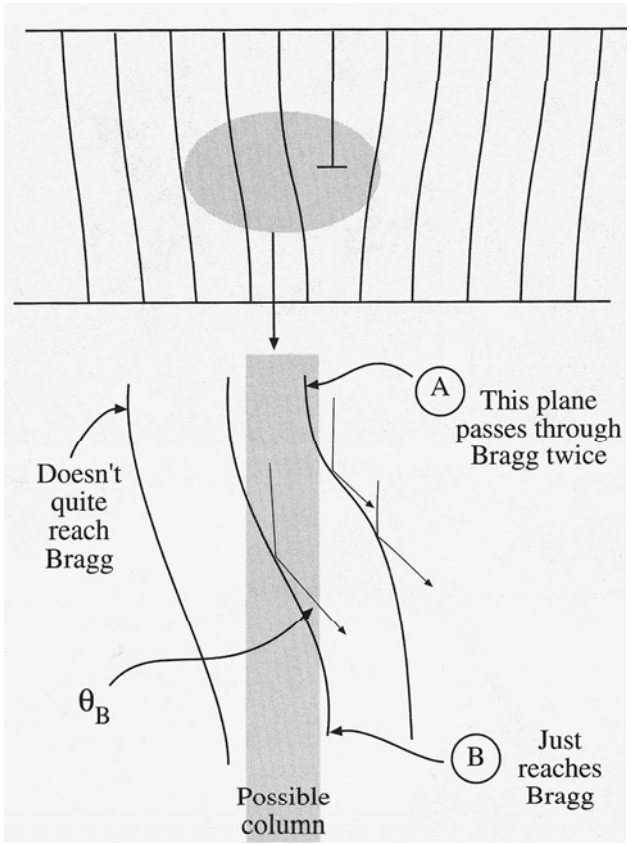
GaAs, V=200 kV, g= 220, ξ_g = 96 nm

Weak Beam Dark Field (WBDF) imaging

The only way to come close to a dislocation core

WBDF with (g,3g)
Optic axis

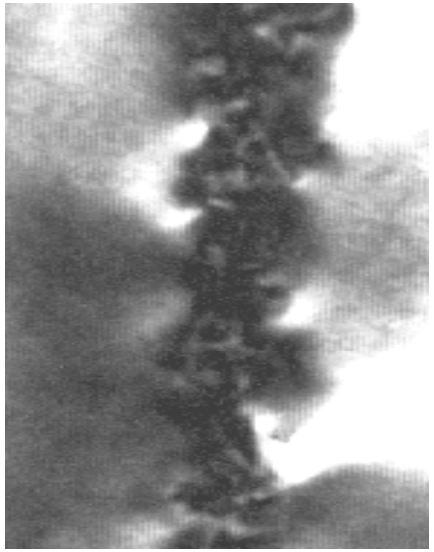




Use g_{hkl} far from the Bragg position:
=> lattice planes from the specimen rotated away from the Bragg condition
=> only the « distorded » zones close to the core of the defect diffract electrons (bent back into Bragg conditions) and appear bright on a dark background

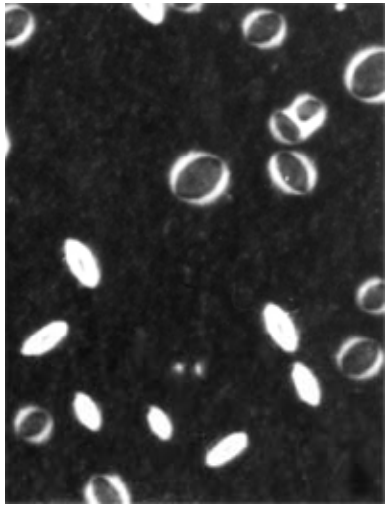
Weak Beam Dark Field (WBDF) imaging

BF, 2 beams



Dislocation loops

WBDF (g,2g) with g=400



☹ overlap of deformation fields surrounding the defects
=> size measurement and identification impossible

☺ dislocation loops get sharper
=> size, density measurement + identification (b, habit plane)

Identifying a defect by WBDF

1) A dislocation loops is totally determined by

- Vector normal to habit plane (u)
- Burgers vector, b (direction and amplitude of displacement field)
- Type (vacancy or interstitial)

2) Its contrast depend on g.b and g.b^u (if both = 0 then loop is not visible)

3) Burgers vector determination

- b direction. Find g so that loop is no more or faintly visible => g.b=0
- b amplitude. Study contrast rules when g.b=0 /

4) Habit plane

- Find how to put it vertical and horizontal (stereographic projection)

5) Type

- Tricky. Study inside outside contrast as function of (g.b)s
- Better use HREM !