EPFL

Diffraction and Imaging part V

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Diffraction and imaging V program

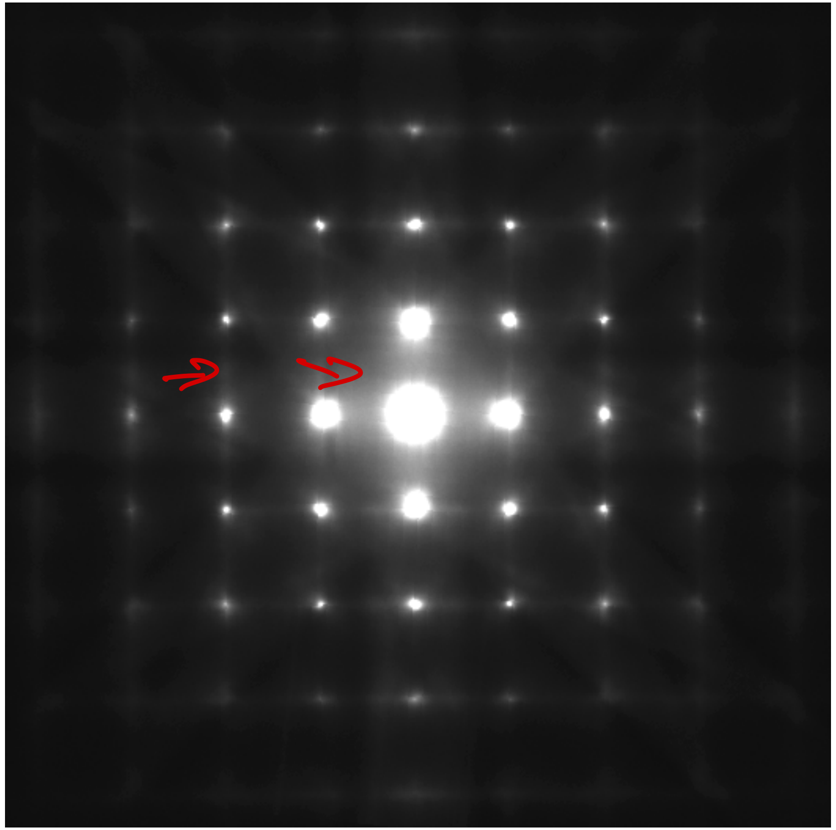
- Lecture on:
  - Kikuchi line formation
  - Analysis of crystal defects with diffraction and diffraction contrast:
    - Strong beam imaging of 1-D defects (dislocations)
    - Weak beam imaging of dislocations
    - 2-D defects: crystal twinning in diffraction mode, stacking faults
- Demo at microscope: imaging of dislocations in strong beam and weak beam

Kikuchi diffraction

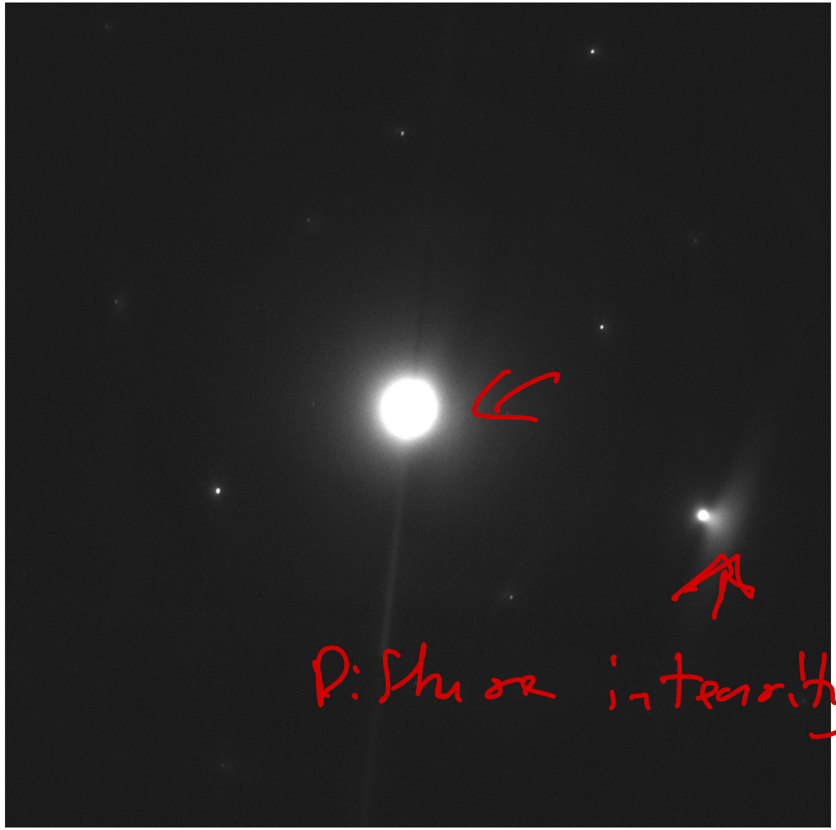
Diffuse intensity in diffraction

- Observe diffuse intensity between the sharp diffracted beams of selected area DPs
- Diffuse intensity must derive from *incoherent scattering*

Si on [0 0 1] ZA

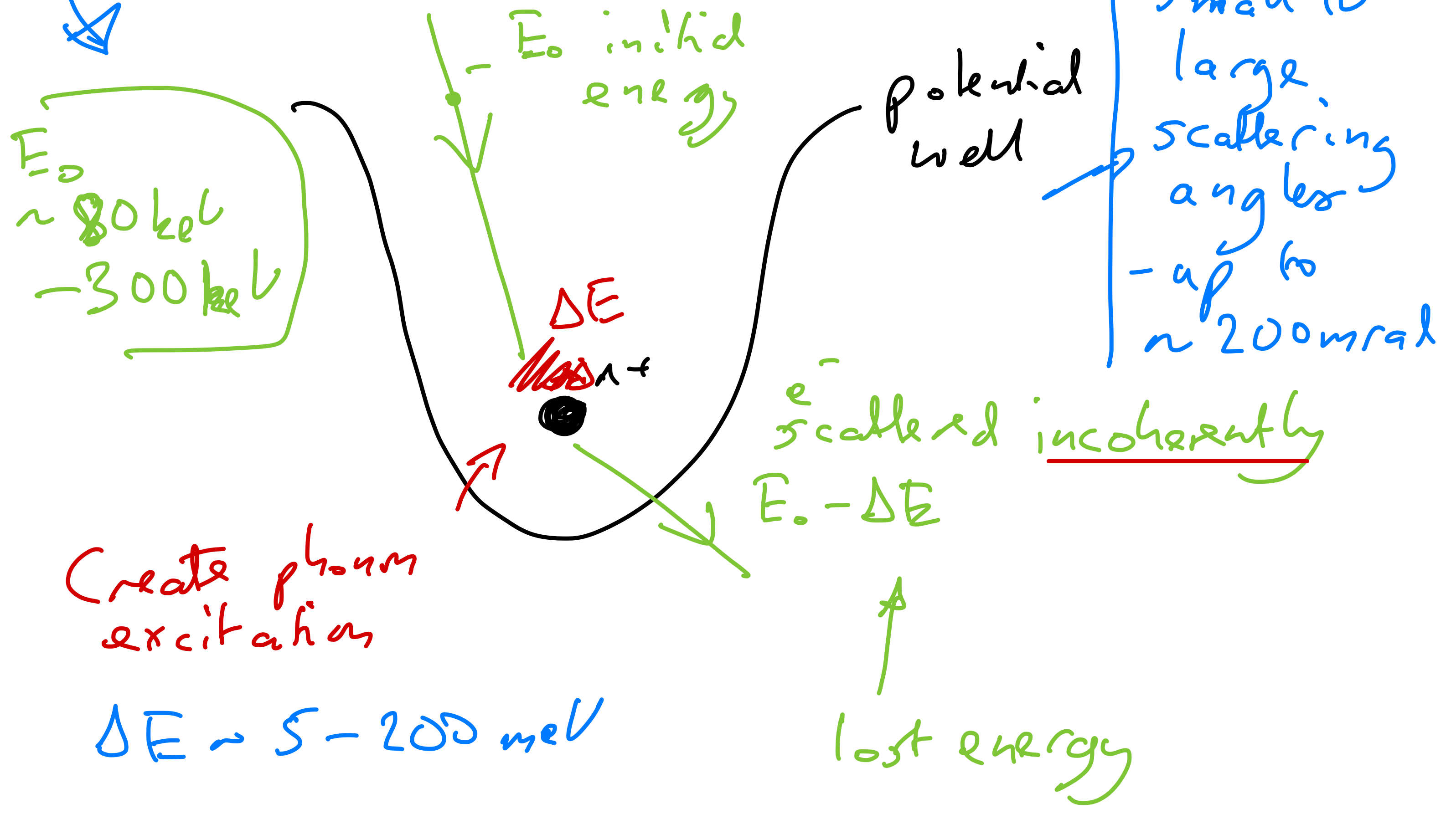


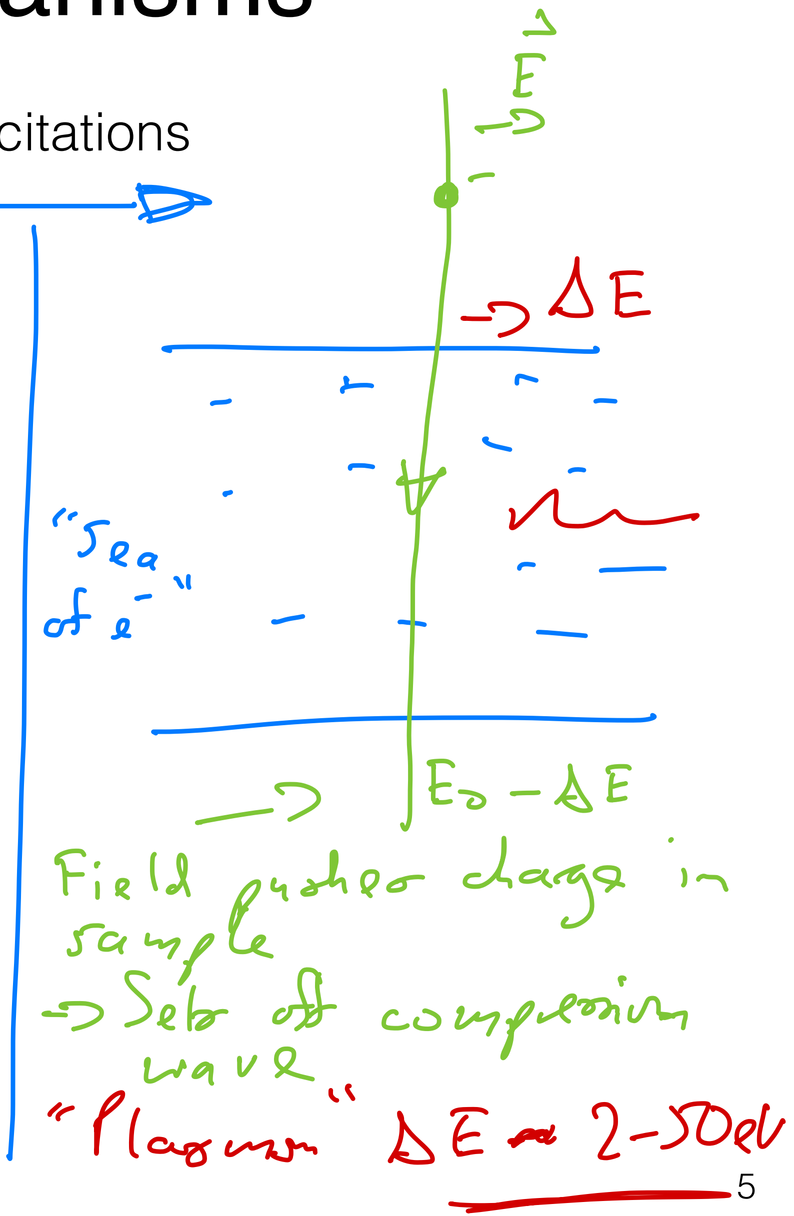
Si near a 2-beam condition



Incoherent scattering mechanisms

- “Thermal diffuse scattering” i.e. scattering from phonon excitations
- Scattering from plasmon excitations





Kikuchi diffraction

Plasmon scattering:
Very low scattering angles
(< 5 mrad)

- Parallel incident beam geometry: formation of bright *excess* and dark *deficient* lines in a selected area diffraction pattern
- Position of the Kikuchi line pairs of very sensitive to specimen orientation
- Can use to identify excitation vector; in particular ***s*** = 0 when diffracted beam coincides exactly with excess Kikuchi line (and direct beam with deficient Kikuchi line)

“Multi-beam” condition; general case

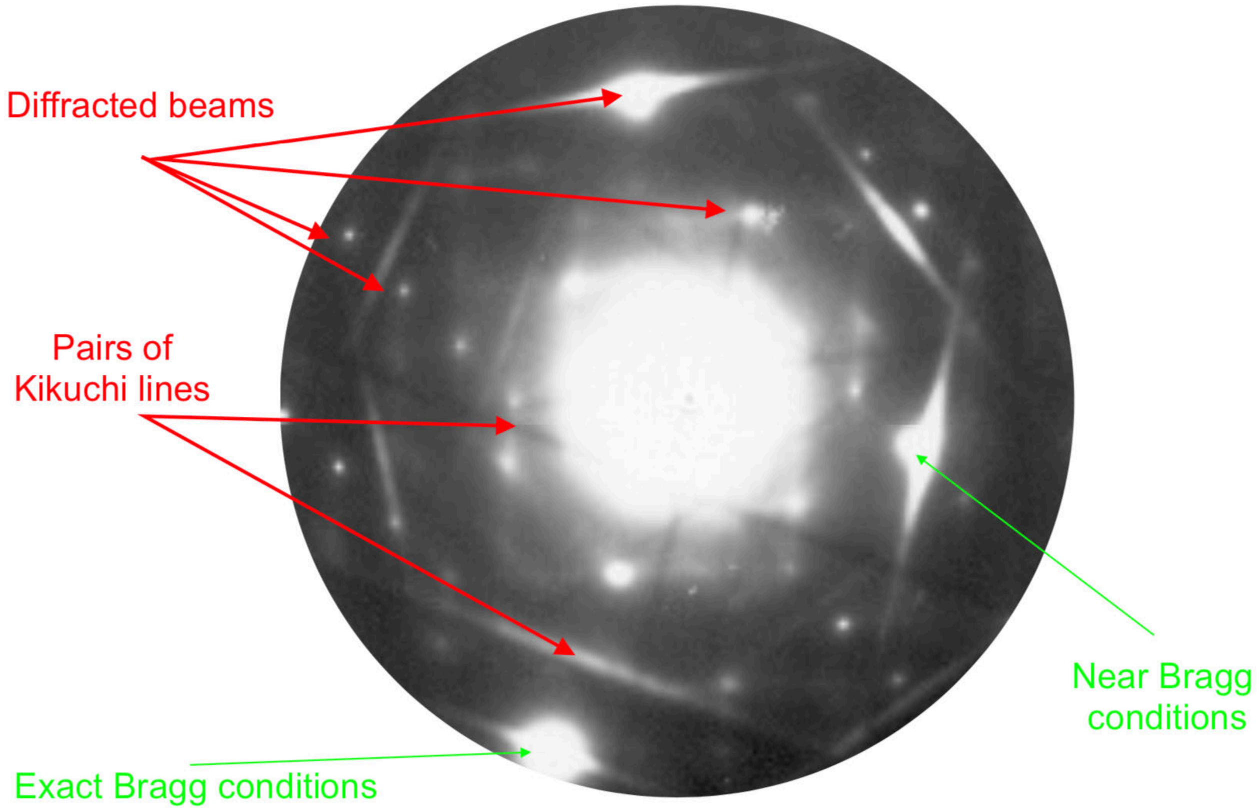
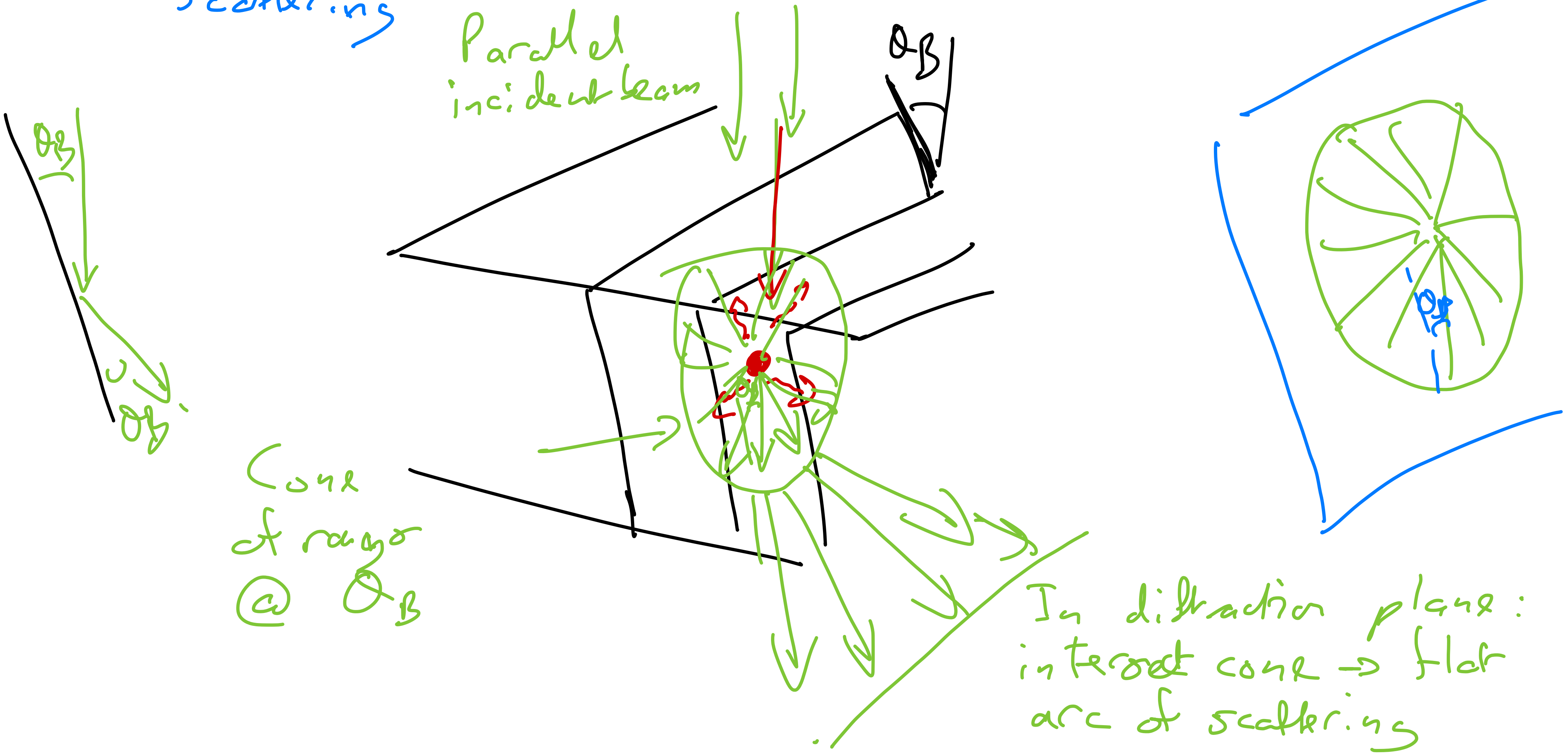


Figure from J.-P. Morniroli

Kikuchi scattering mechanism

Incoherent scattering followed by coherent elastic scattering



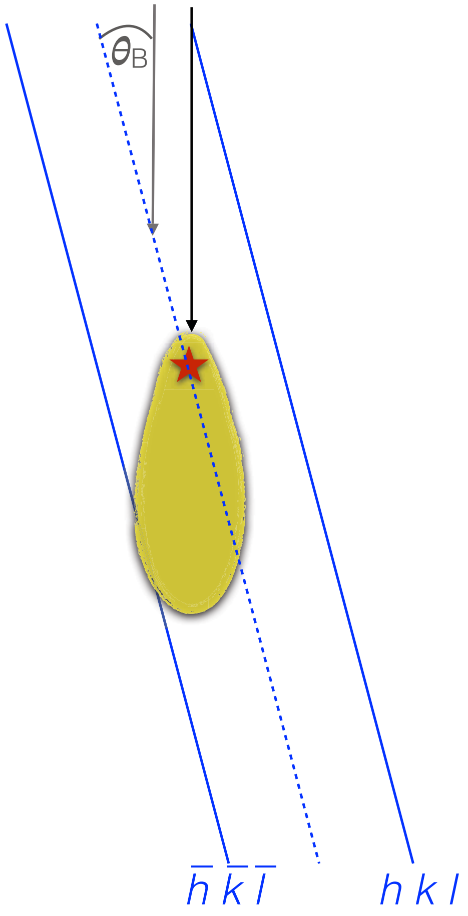
Kikuchi scattering mechanism

- Formation of bright and dark lines (“excess” and “deficient” lines) by combination of **incoherent scattering*** followed by elastic scattering, in parallel incident beam geometry (e.g. SADP)
- Incoherent scattering: no preferential scattering vectors, but is generally in *forwards* direction
- Cones of incoherently-scattered electrons then elastically scattered, creating arcs in the diffraction plane. Because angles are small the arcs look straight.
- Resulting lines are *very similar* to the excess and deficient lines of CBED. They are equally sensitive to specimen orientation, and we use them e.g. to set a 2-beam condition. However the specimen must be thick (for sufficient incoherent scattering) and flat (to have sharp lines) to see them well.

**Origin of incoherent scattering: “thermal diffuse scattering” i.e. phonon scattering (quasi-elastic); plasmon excitations (inelastic)*

Kikuchi formation

- 2-beam Bragg scattering condition
- Treat problem in real space
- Yellow volume represents intensity distribution of incoherent scattering event; mainly forward scattered
- Origin of incoherent scattering event represented by: ★



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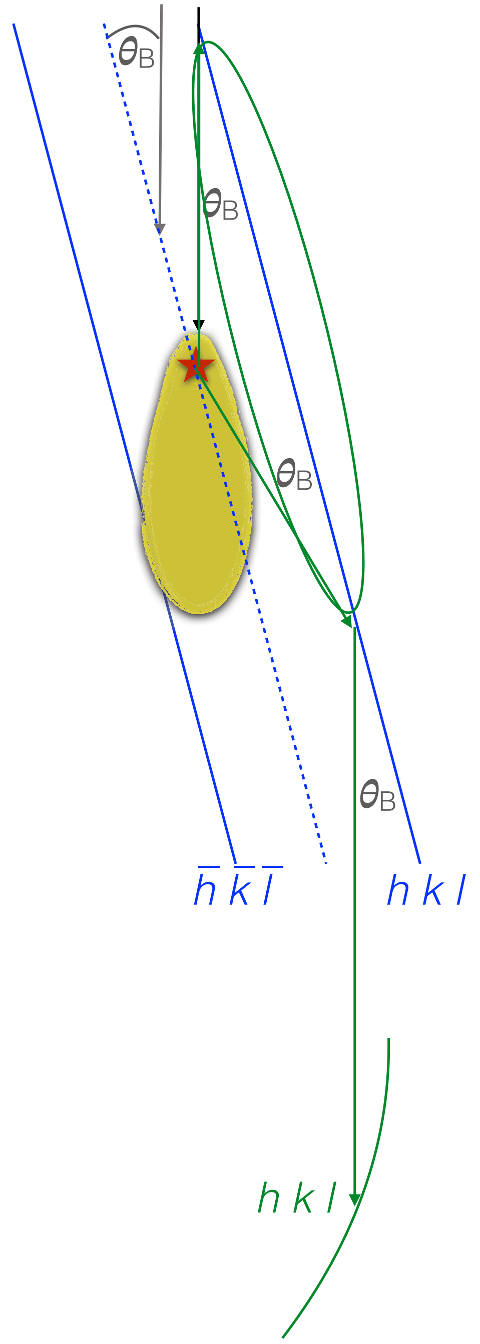

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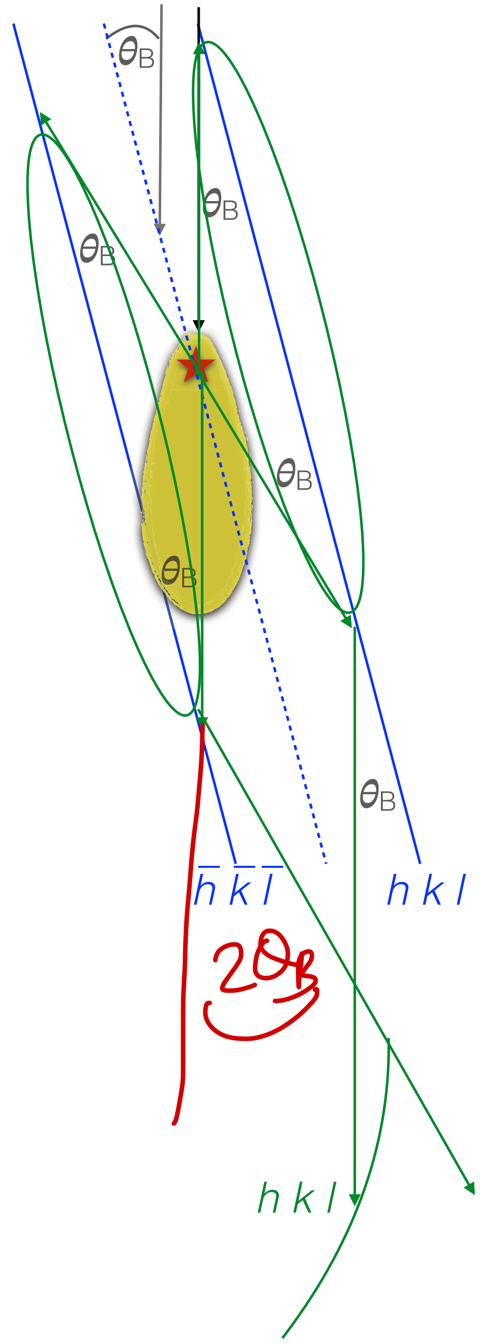

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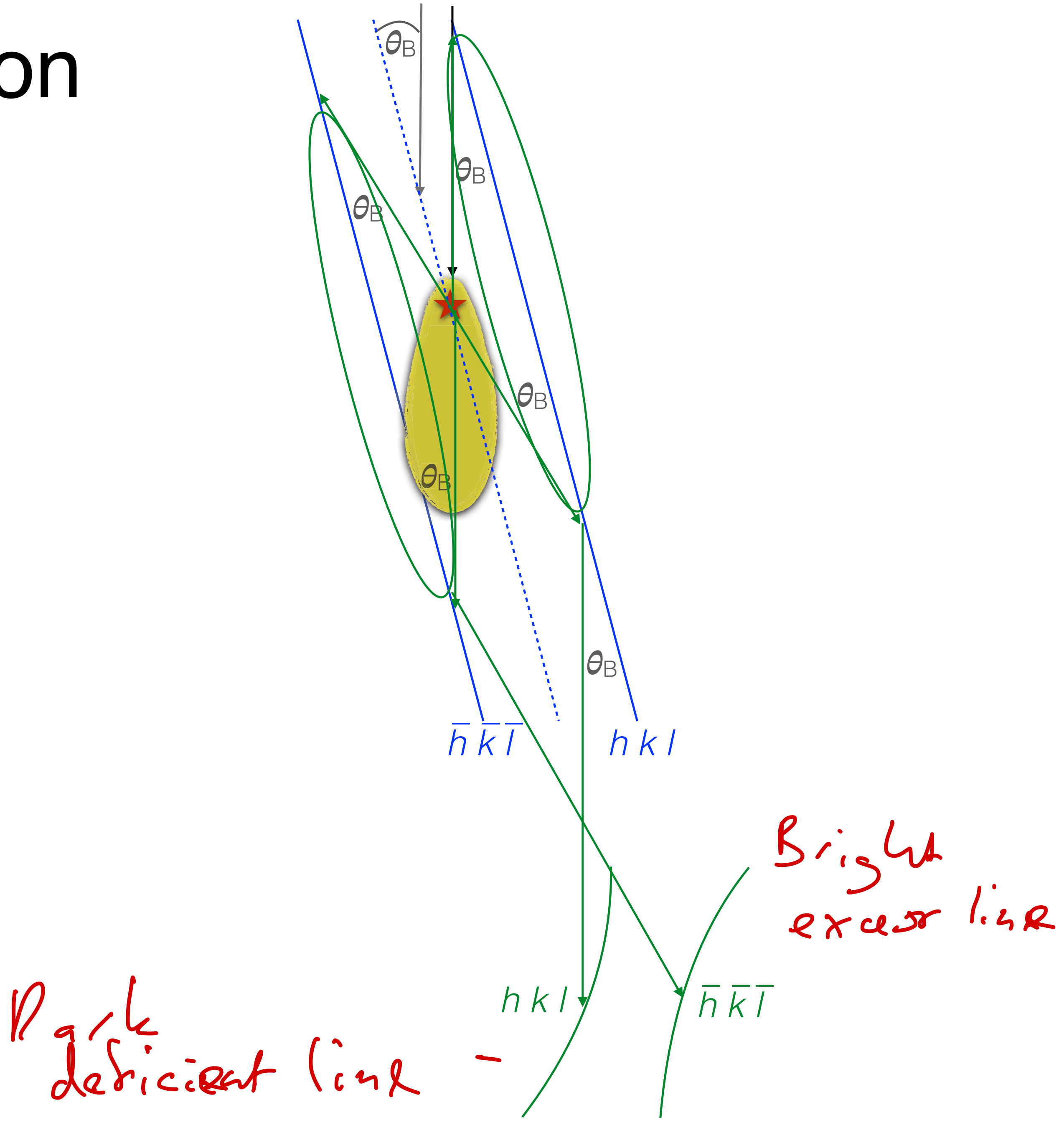

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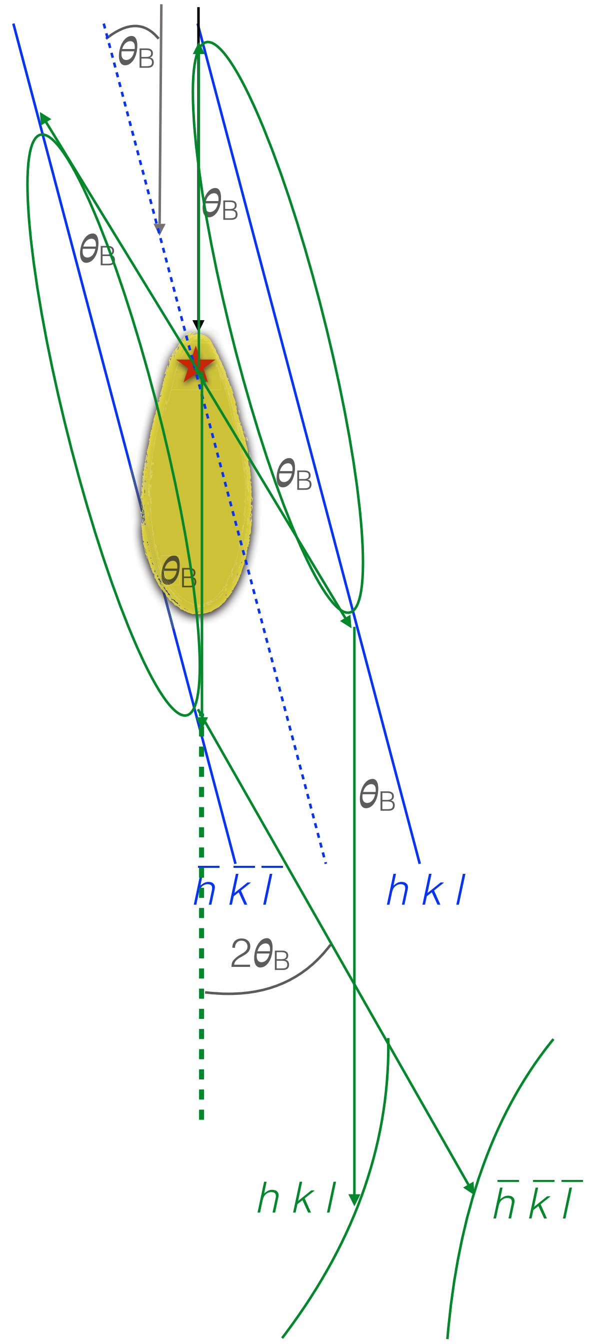
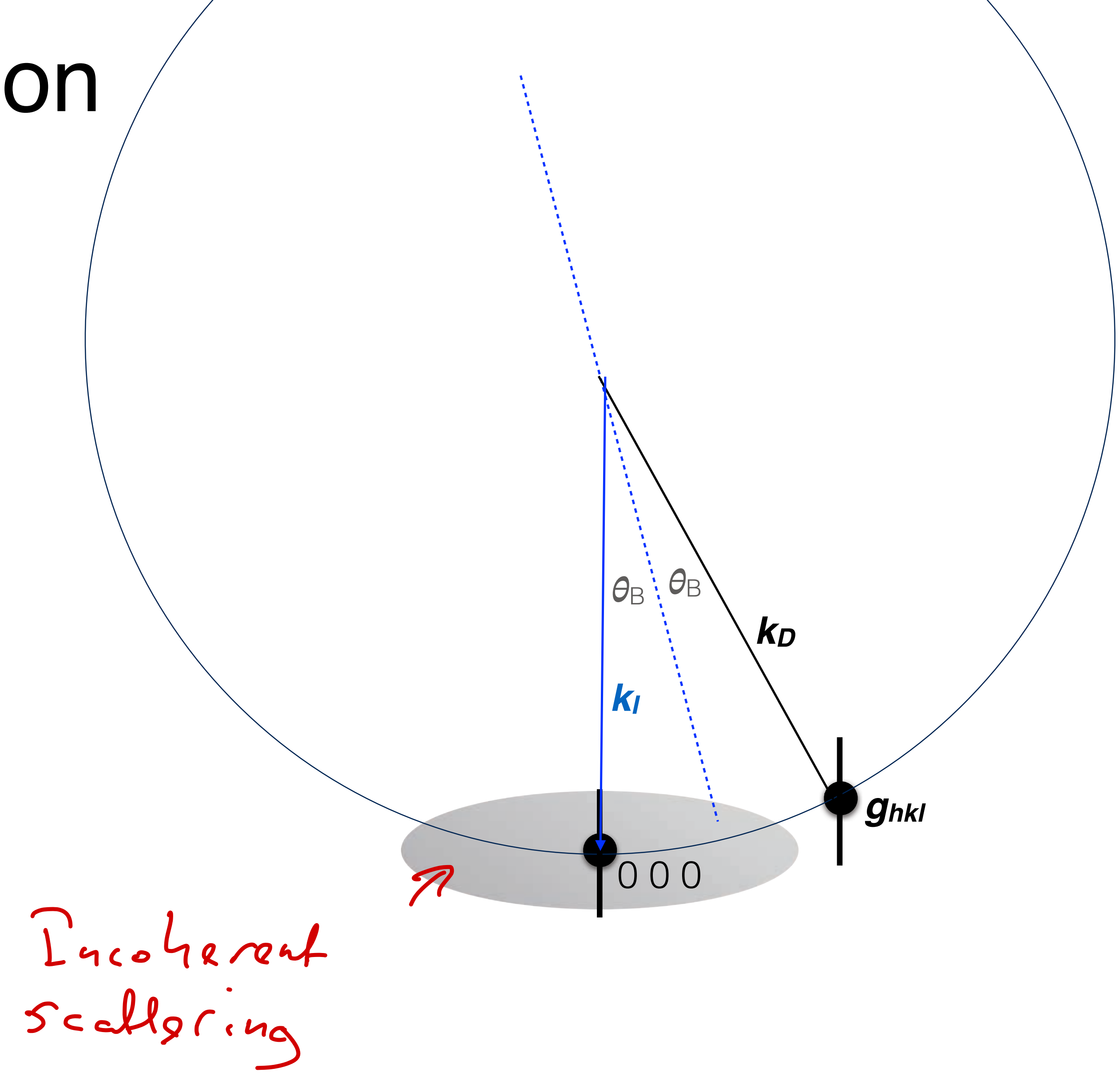


Kikuchi formation

- 2-beam Bragg scattering condition
- Treat problem in reciprocal space with Ewald sphere construction
- Forwards incoherent scattering gives diffuse intensity at low scattering angles around (0 0 0) direct beam spot
- Exact Bragg condition: Kikuchi lines coincide with direct and diffracted beam



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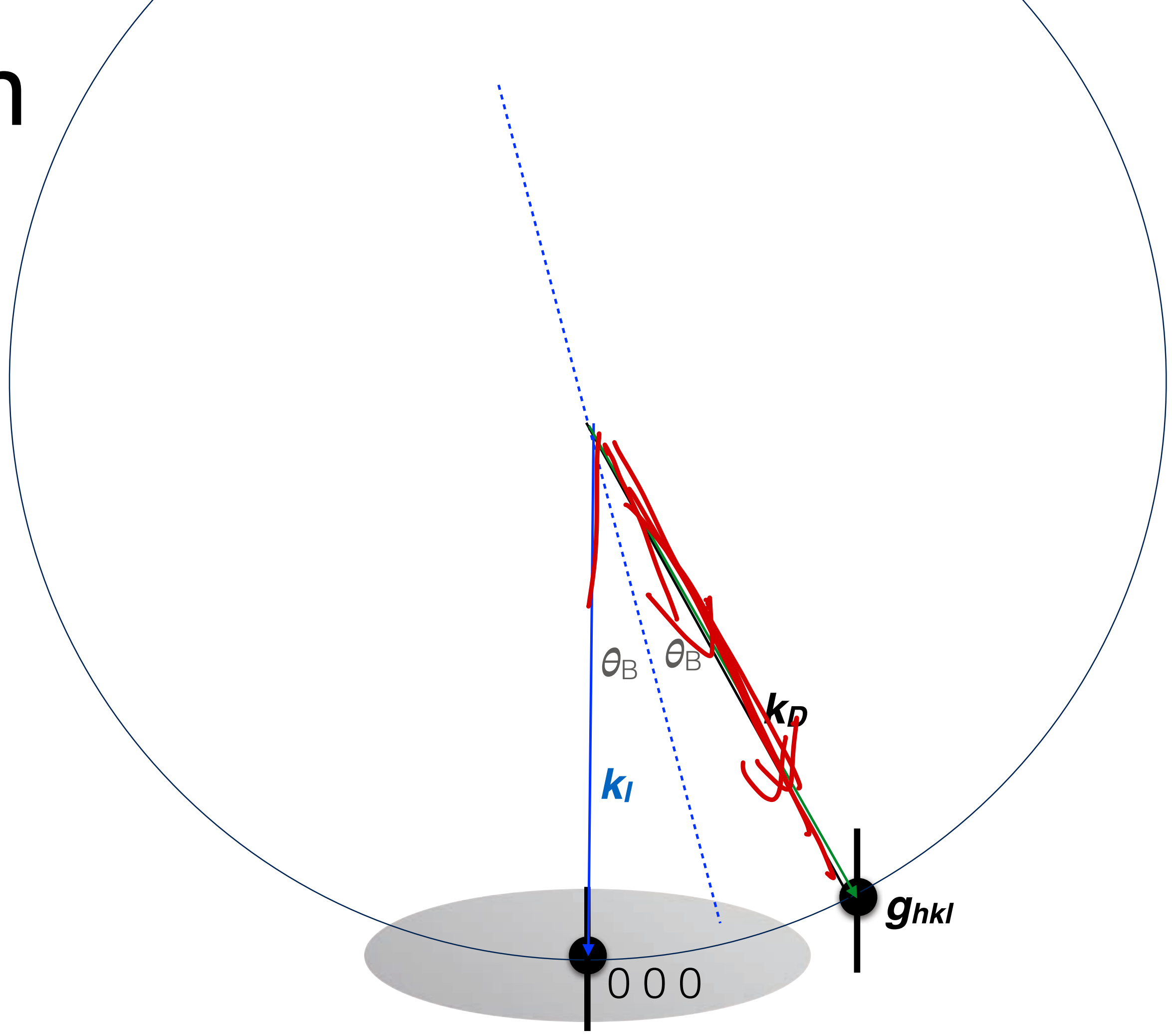

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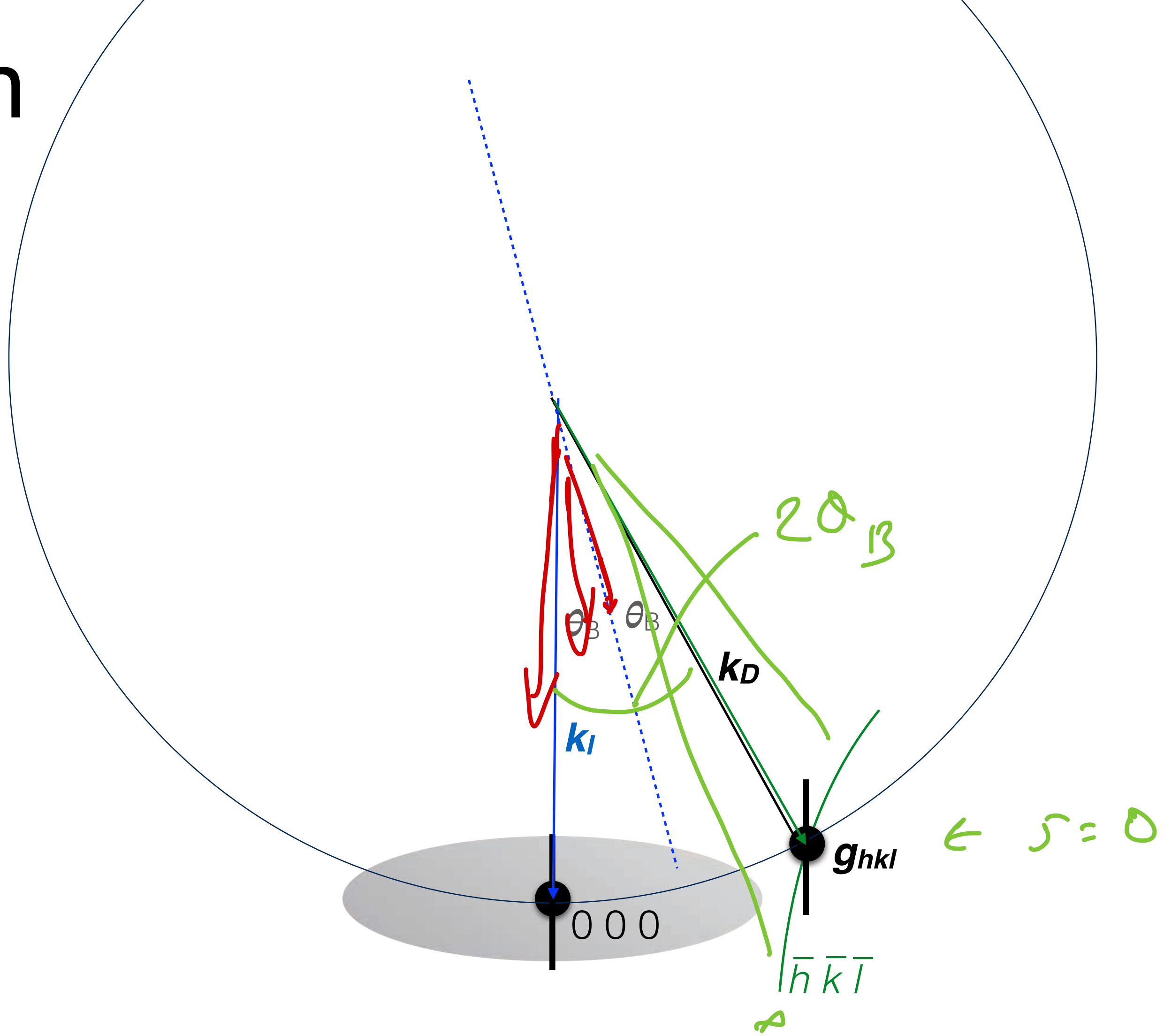

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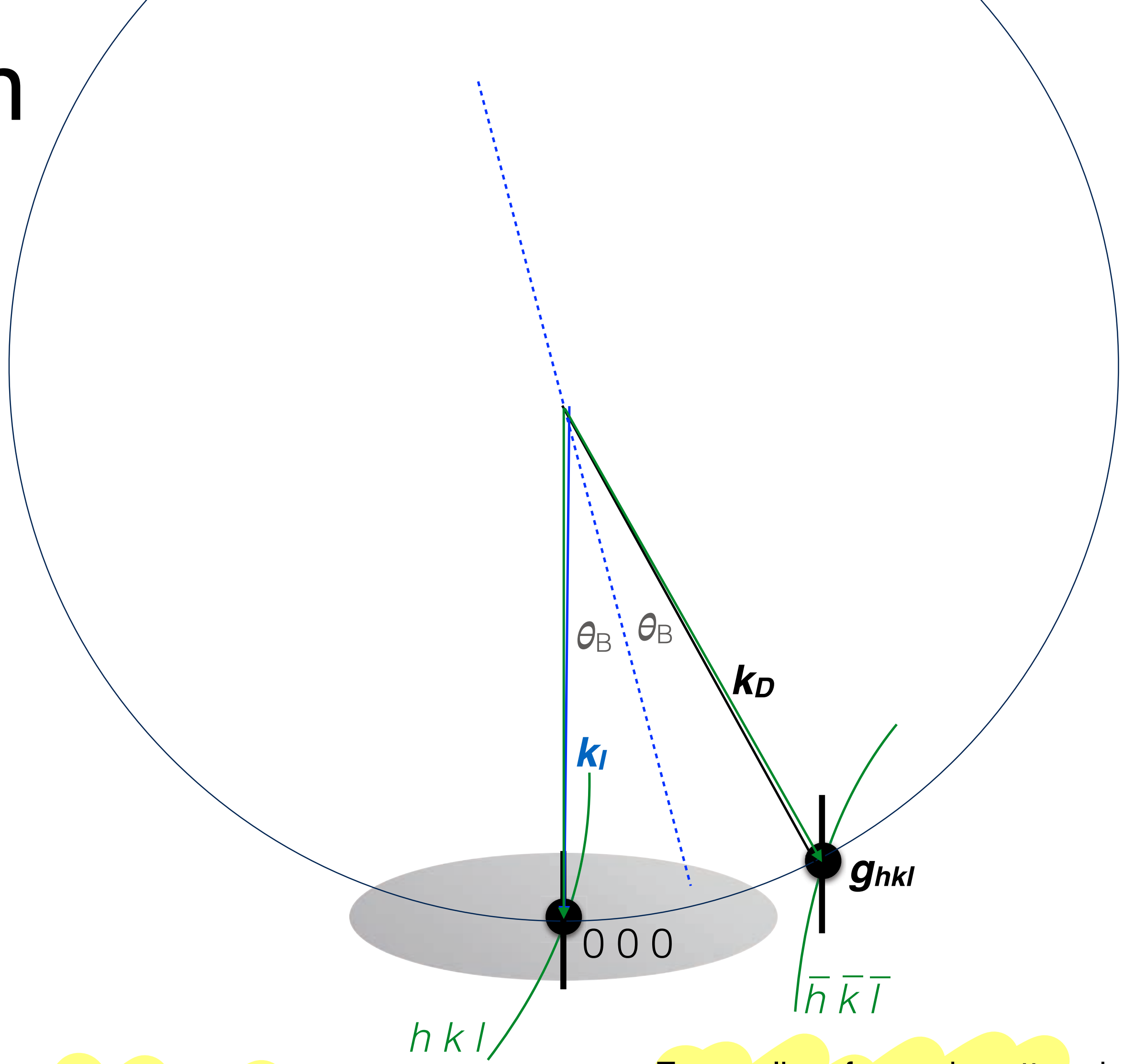
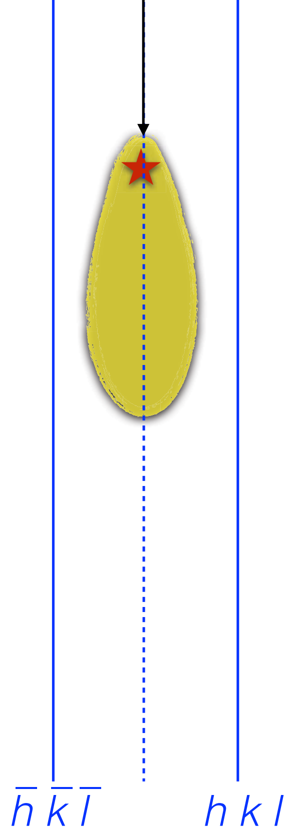


Deficient line: forward-scattered incoherent intensity removed from Kikuchi line ⟹ dark line on brighter background

Excess line: forward-scattered incoherent intensity redistributed into Kikuchi line ⟹ bright line on dark background

Kikuchi formation

- Zone axis or systematic row condition
- Treat problem in real space
- Yellow volume represents intensity distribution of incoherent scattering event; mainly forward scattered
- Origin of incoherent scattering event: ★



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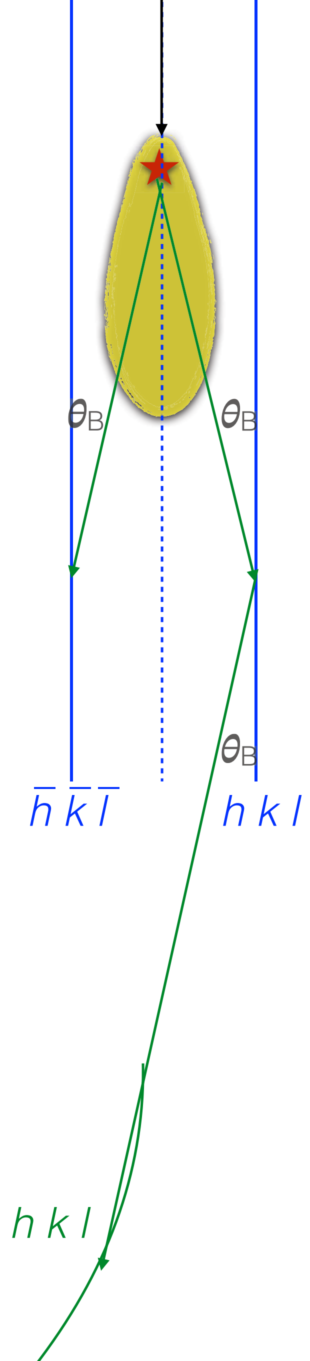

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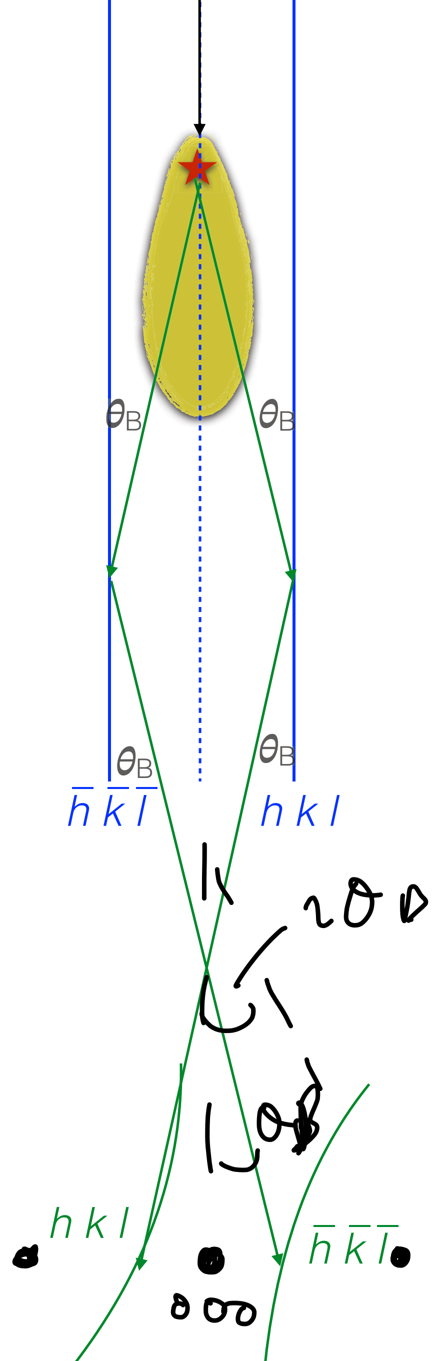
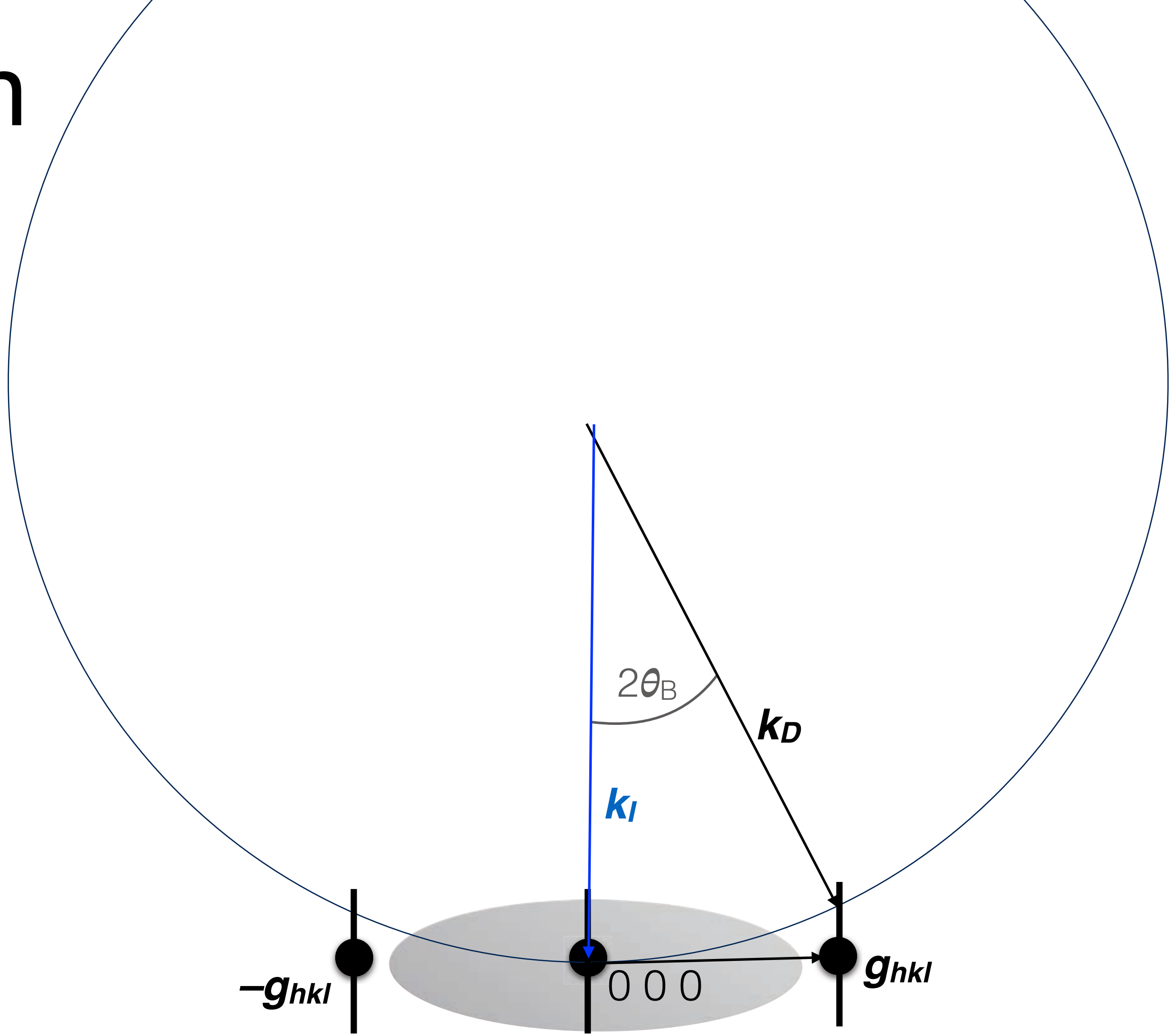


Kikuchi formation

- Zone axis or systematic row condition
- Treat problem in reciprocal space
- By geometry, Kikuchi lines halfway between diffraction spots ***g*** and ***–g***



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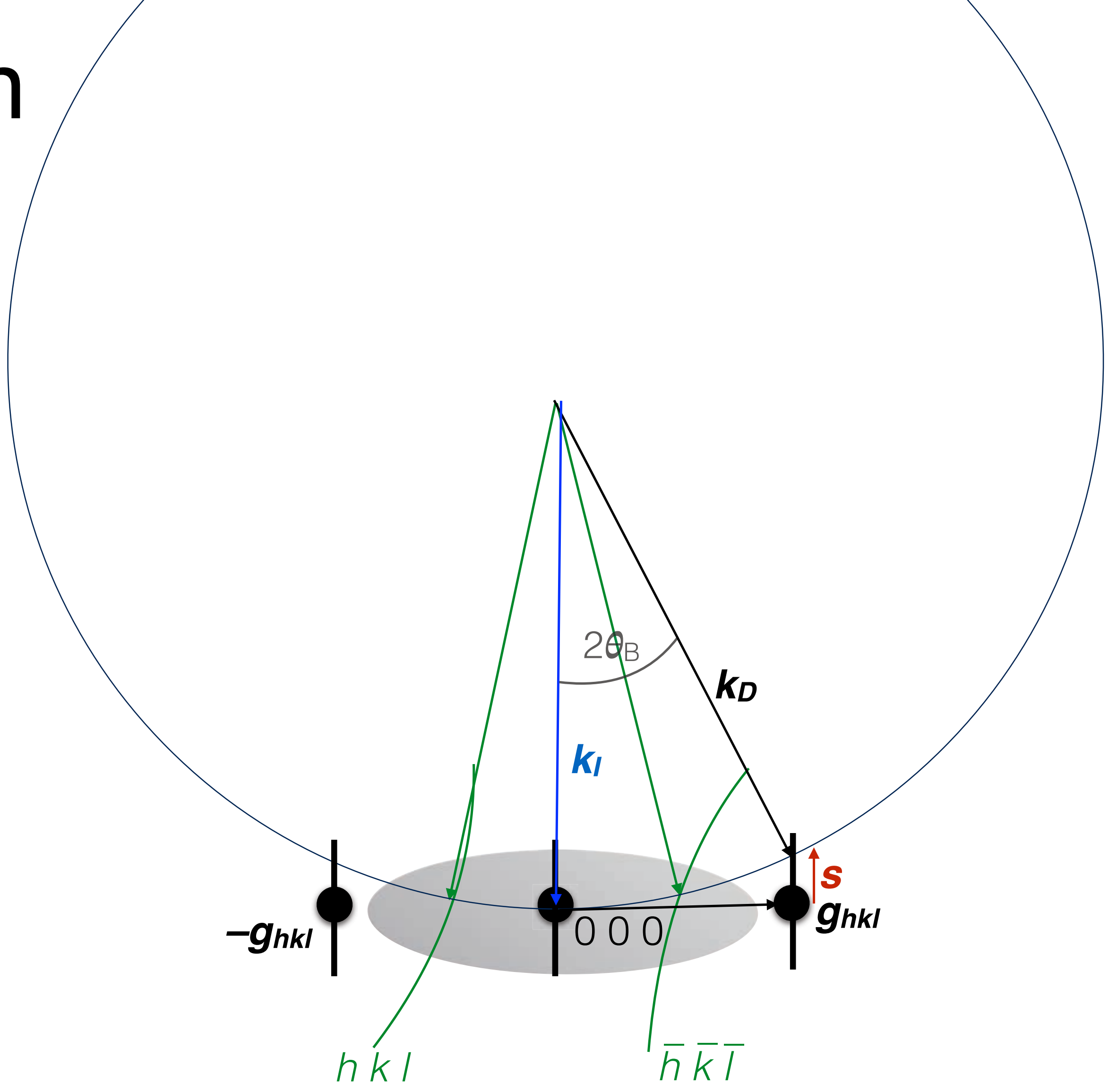
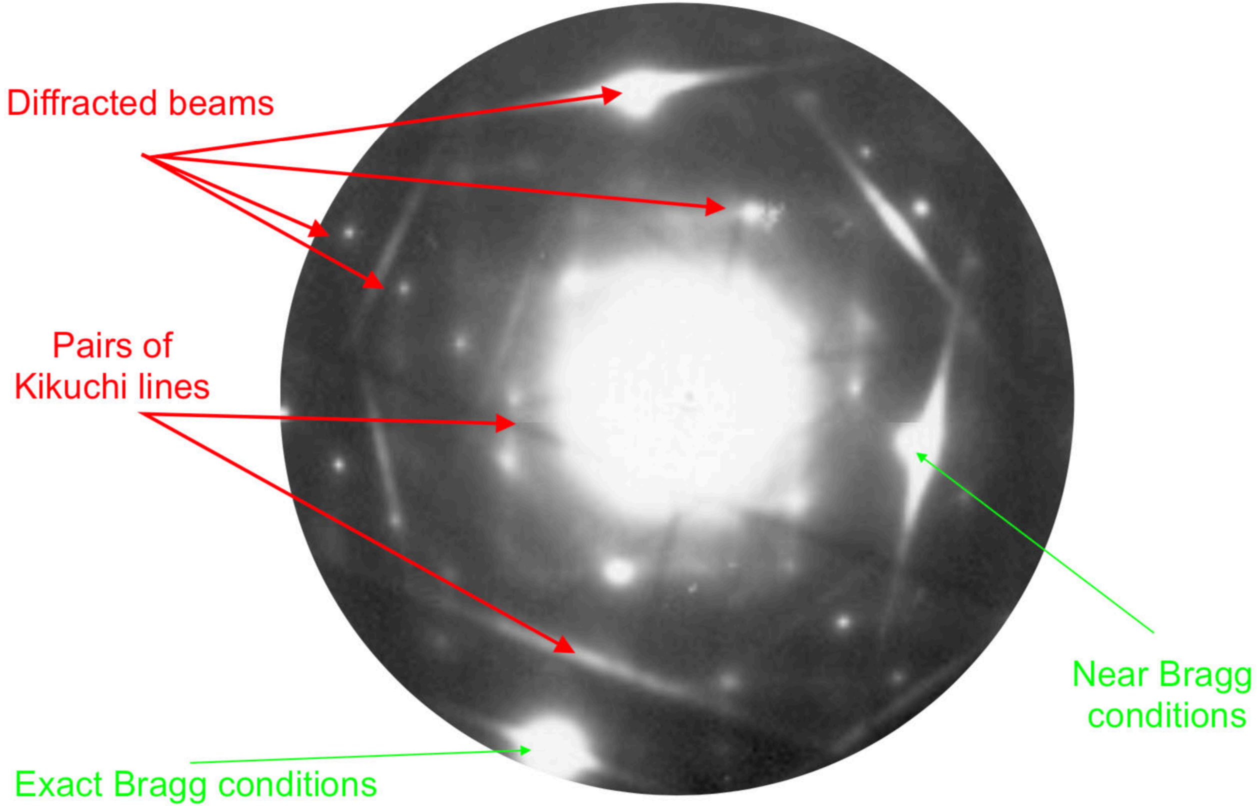


Symmetrical relationship so both Kikuchi lines should have same intensity

Kikuchi diffraction

- Geometrically Kikuchi lines are very similar to excess and deficient lines in CBED
- However seen in parallel beam geometry. Good lines require: a thick sample for sufficient incoherent scattering; a flat sample for sharp lines
- *Q*uiz: what happens to width of Kikuchi line pairs as (*h k l*) indices become bigger?

"Multi-beam" condition; general case



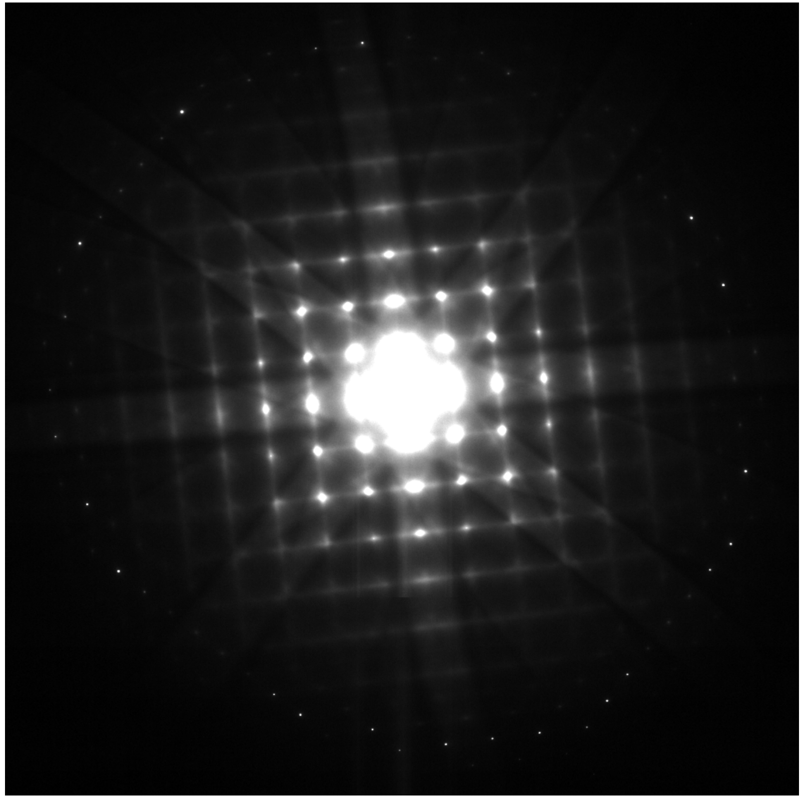
Width ↑ because d_{hkl} ↓ and θ_B ↑ – "Width" = $2\theta_B$

Figure from J.-P. Morniroli

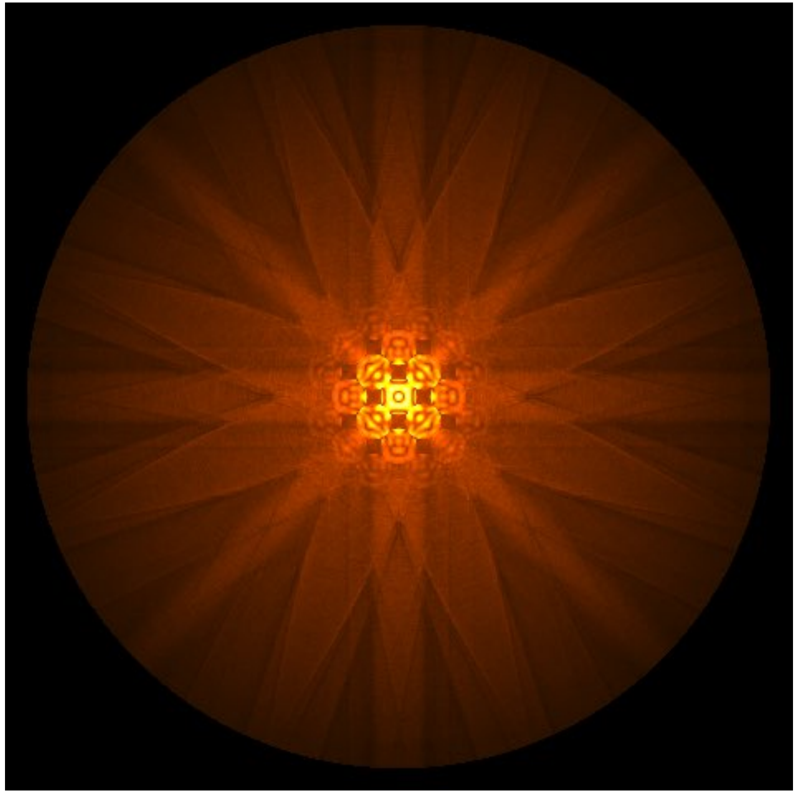
Diffuse scattering and zone axis patterns

- In zone axis SADP and CBED patterns of thick samples, the combination of diffuse incoherent scattering followed by coherent elastic scattering gives rise to “Kikuchi lines” which cannot be accounted for from pure coherent elastic scattering.

Si [0 0 1] SADP – experimental



$SrTiO_3$ [0 0 1] CBED – simulation



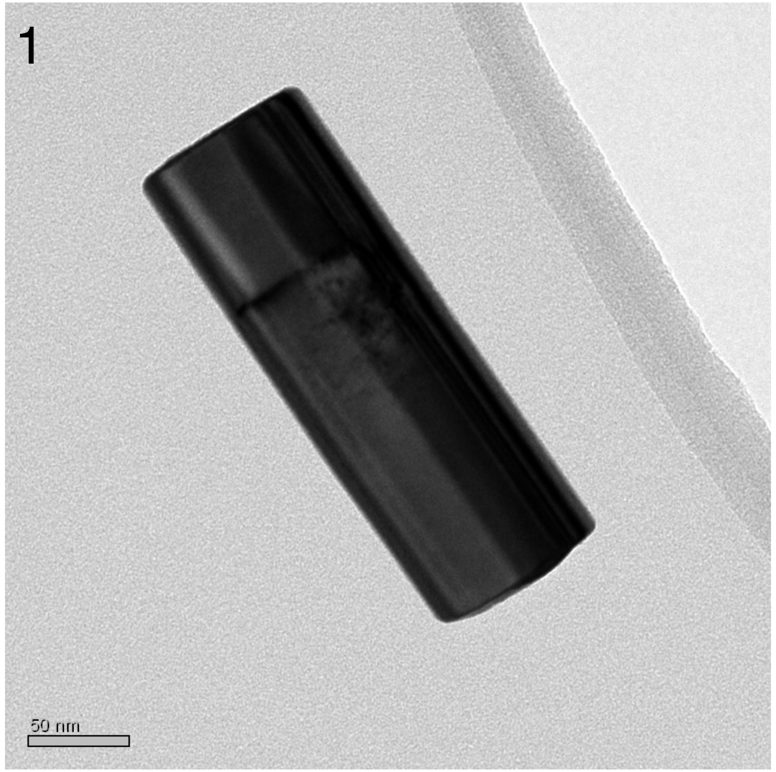
TEM analysis of crystal defects

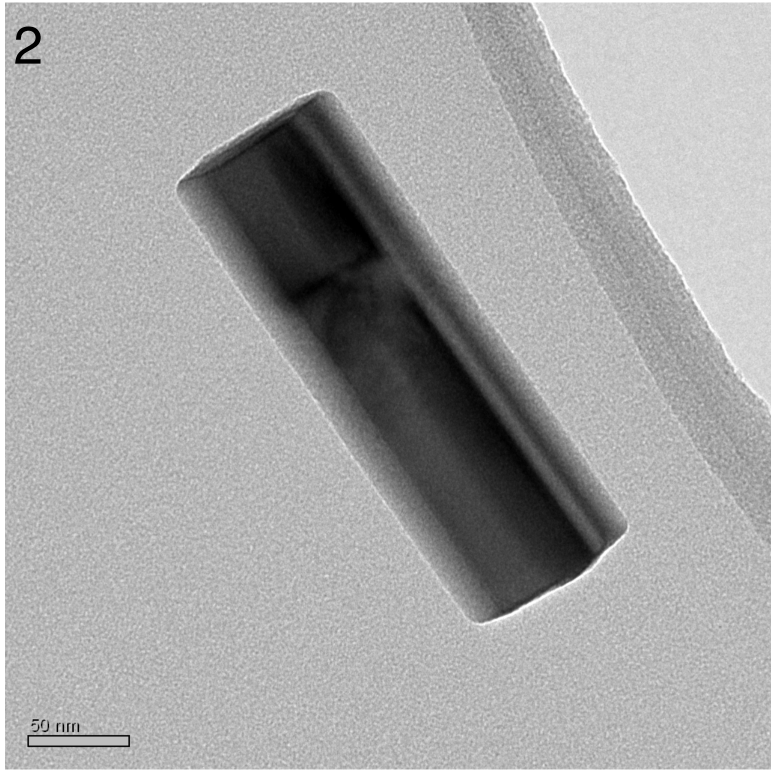
Diffraction contrast analysis

- Diffraction combined with diffraction contrast imaging allows a precise, nano-scale analysis of 2-D and 1-D crystal defects. (0-D point defects are a different matter.)
- When we do diffraction contrast imaging we select the direct beam or a particular diffracted beam with the objective aperture to form the TEM image. By doing this, *we make a spatial map of the intensity of the chosen beam*.
- Any local structural change that modifies the intensity of the beam gives a corresponding contrast change in the TEM image. For dislocations this can be from the strain field of the dislocation.
- Note that a small objective aperture will be the resolution limiter of the optical system (see phase contrast lectures)

Diffraction contrast quiz

- Which of these BF images of a GaN nanowire was taken in zone axis condition?





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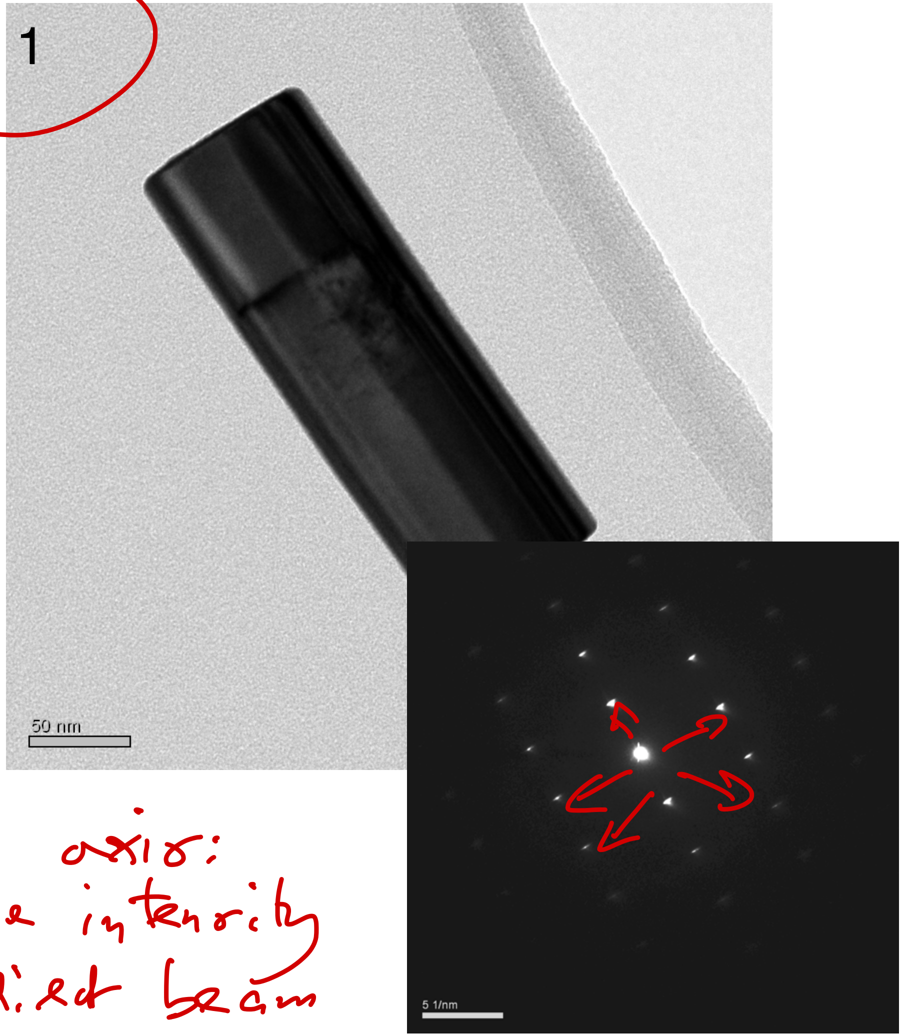
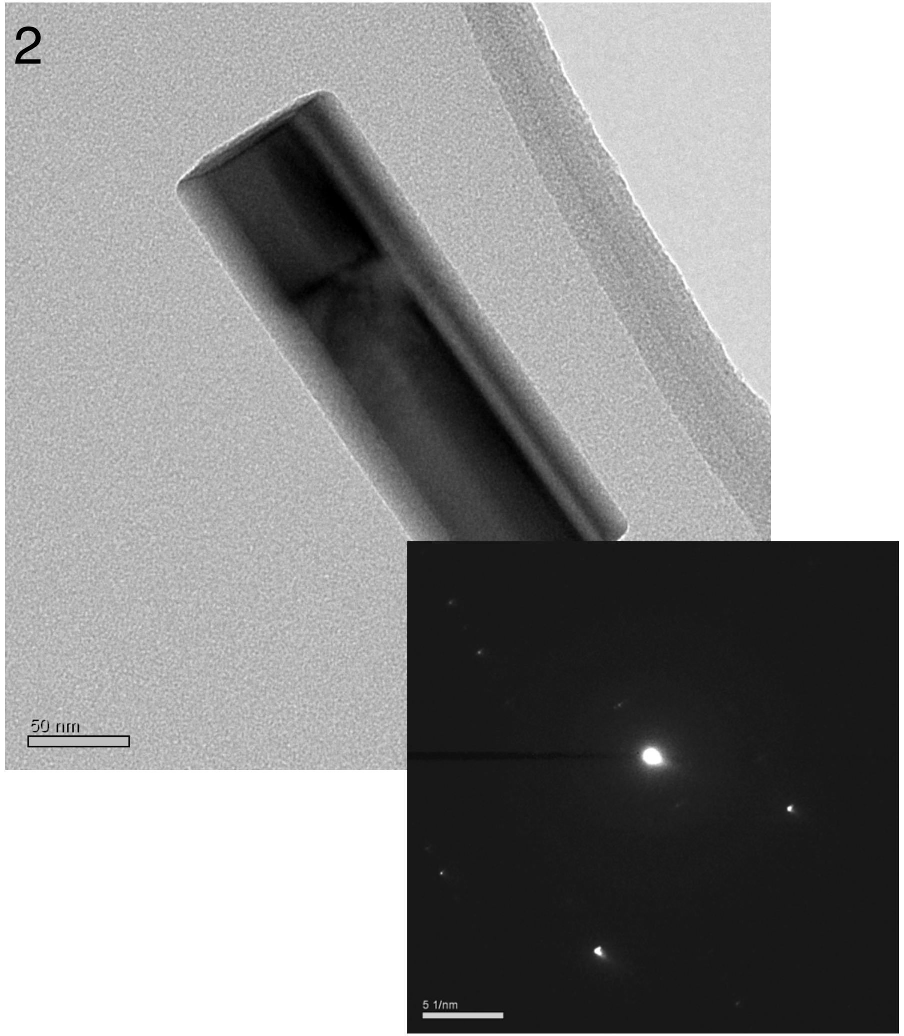


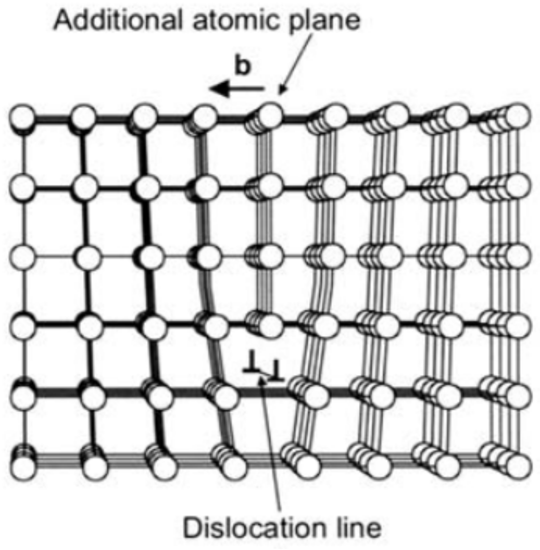


zone axis: remove intensity from direct beam

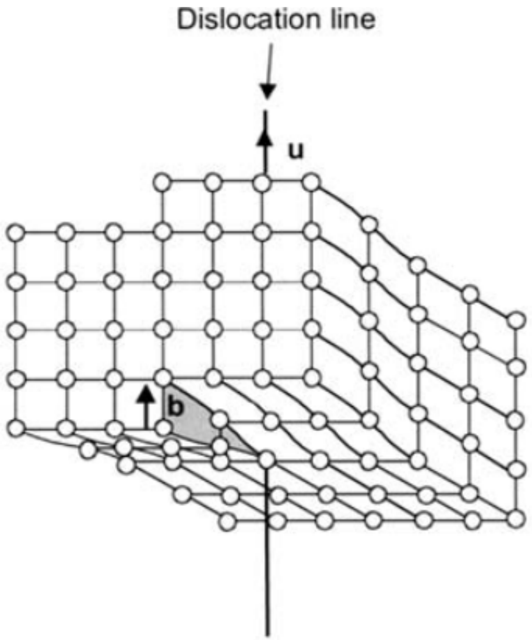
1-D crystal defects

1-D crystal defects: dislocations

Edge dislocation:
Burgers vector ***b*** perpendicular to dislocation line ***u***



Screw dislocation:
Burgers vector ***b*** parallel to dislocation line ***u***



- Note dislocations are often *mixed* in nature (both edge and screw components)

Figures from J.-P. Morniroli LACBED book

Definition of Burgers vector

Edge dislocation:
$\vec{b} \perp \vec{u}$

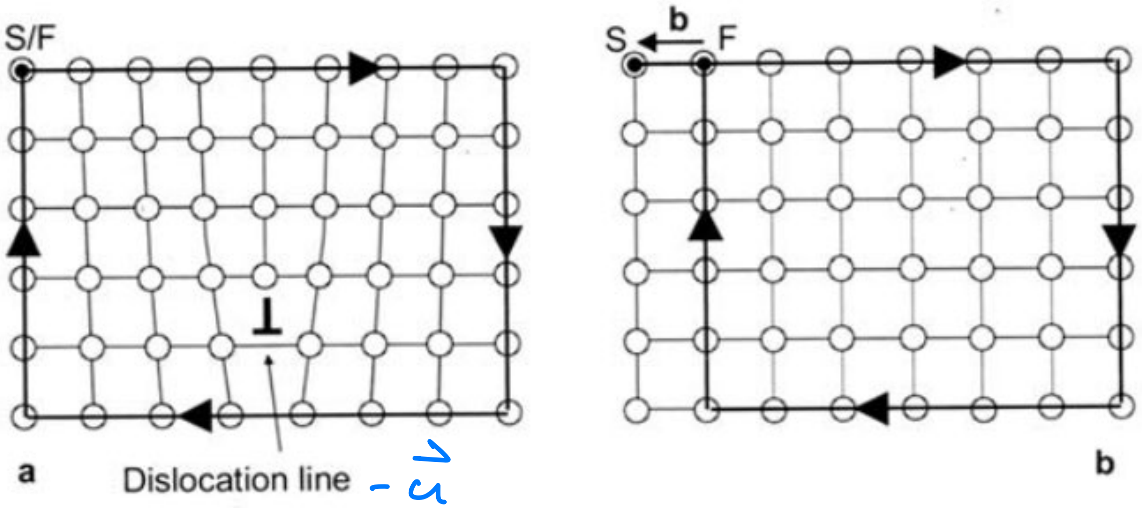


Figure XI.4 - Burgers circuit around an edge dislocation.
a - A closed right-handed circuit is drawn around the dislocation line.
b - The same circuit is drawn in a perfect crystal. The F →S gap characterizes the Burgers vector **b**.
On this diagram, the vector **u** points into the page.

- For dislocation analysis want to:
  - ➡ image dislocation
  - ➡ characterise both ***b*** and ***u***

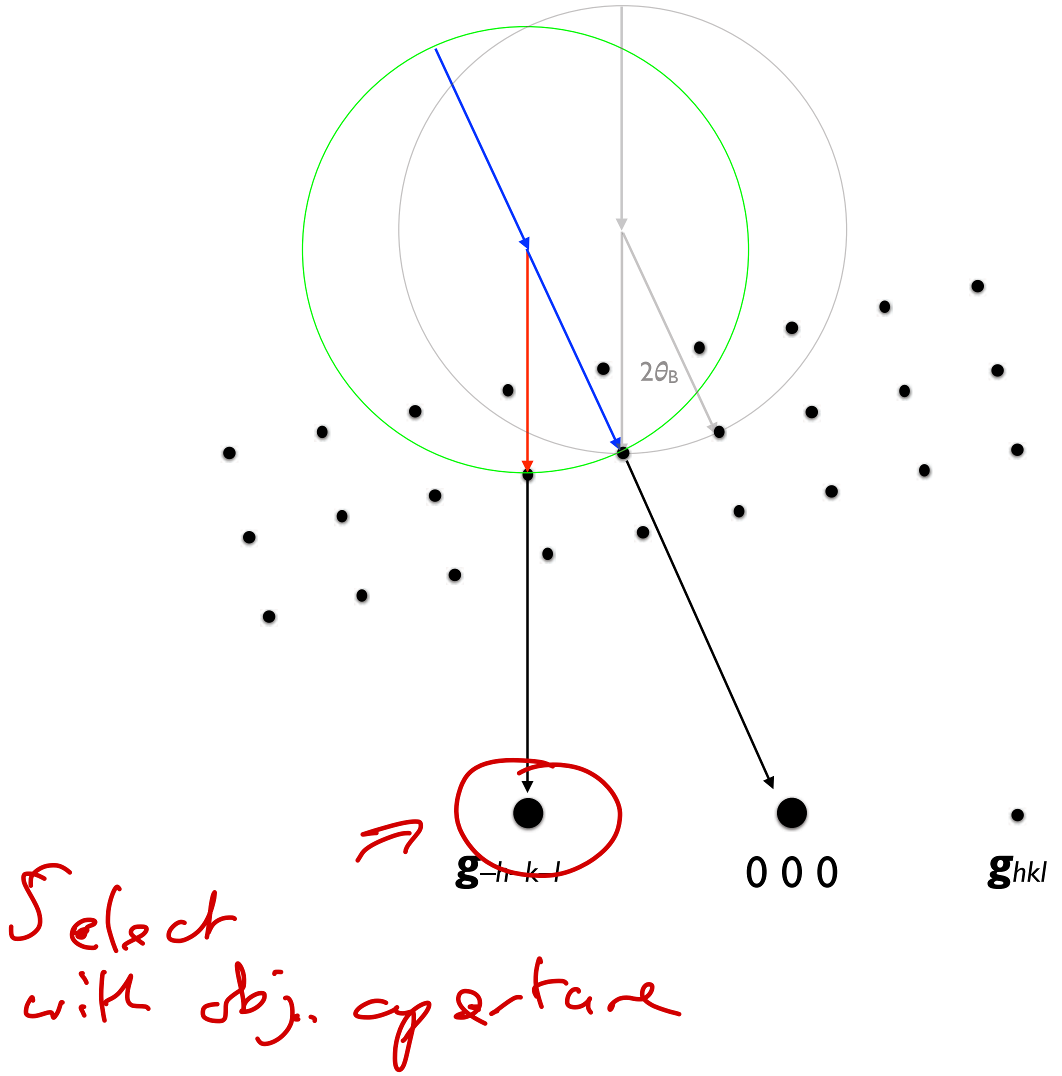
Figure from J.-P. Morniroli LACBED book

Centred aperture dark-field: *strong* beam

- Corresponds to tilting of Ewald sphere by $2\theta_B$,

  excite $\boldsymbol{g}_{-h\,-k\,-l}$;
  0 0 0 takes place of $\boldsymbol{g}_{h\,k\,l}$ in SADP

- "Strong beam" imaging condition with excitation error $s = 0$



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Principle of dislocation imaging

- Bending of crystal plane around the dislocation core locally changes its diffraction condition
- This produces a contrast in the image ⟹ ***g.b*** analysis for Burgers vector

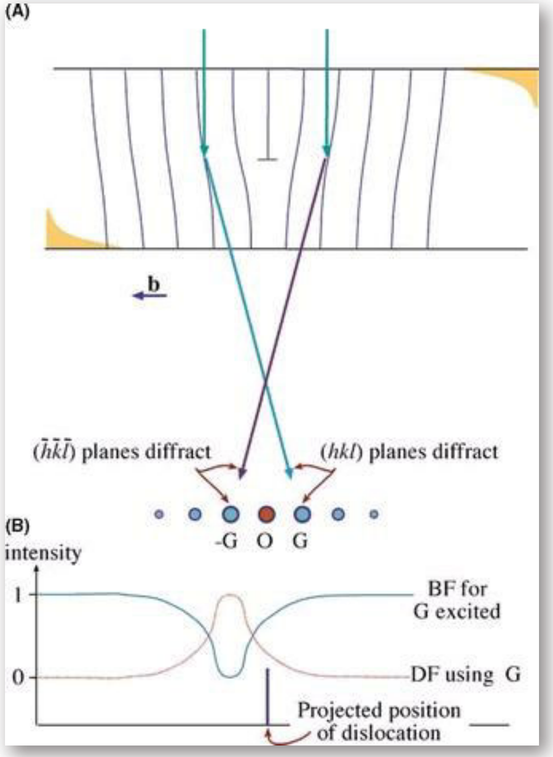
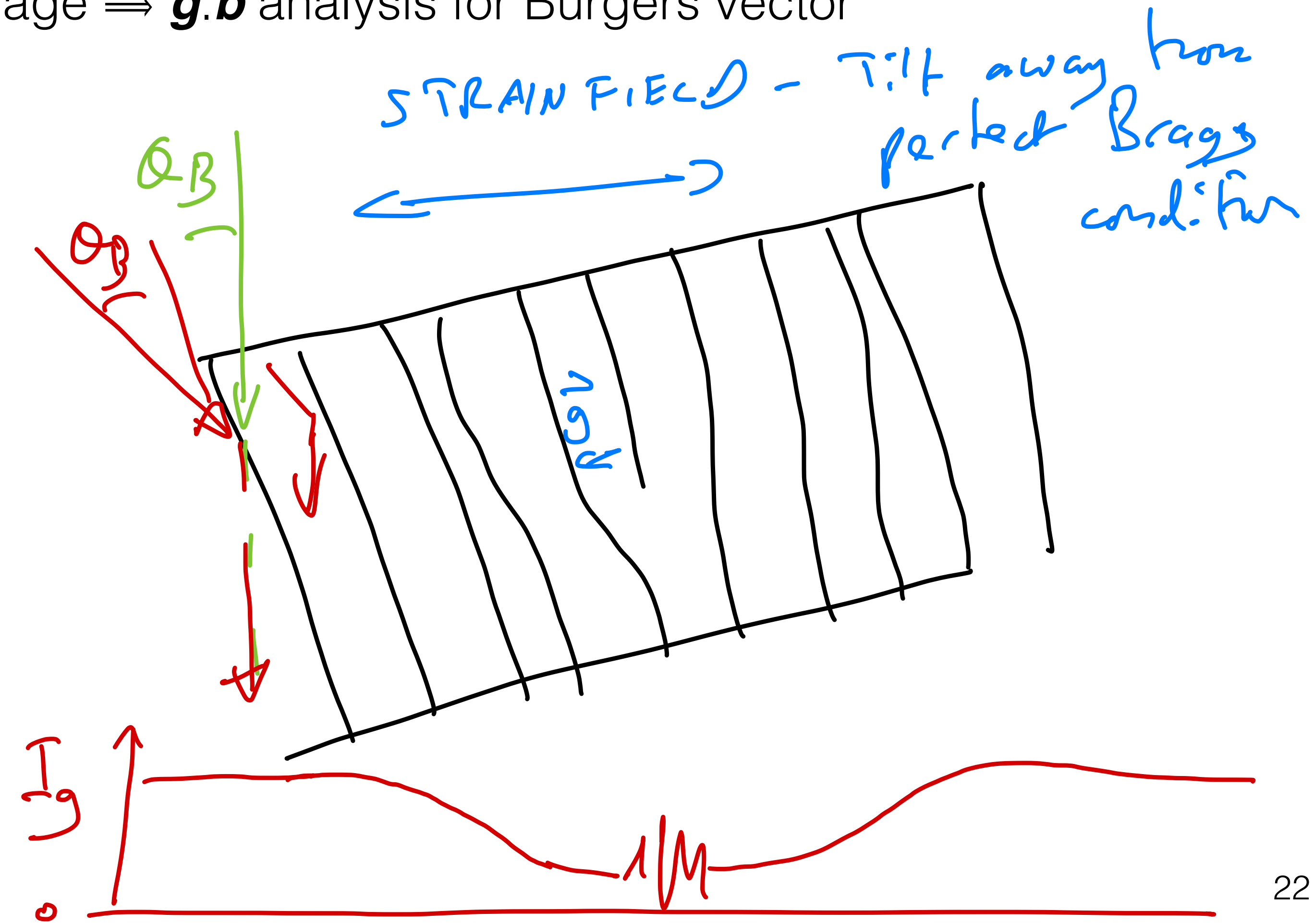


Diagram from Williams & Carter *Transmission Electron Microscopy*



g.**b** analysis

- On a basic level, planes parallel to ***b*** are not distorted by the dislocation
  ⟹ these planes show no change in contrast

- This condition corresponds to ***g***.***b*** = 0 – the *invisibility criterion*

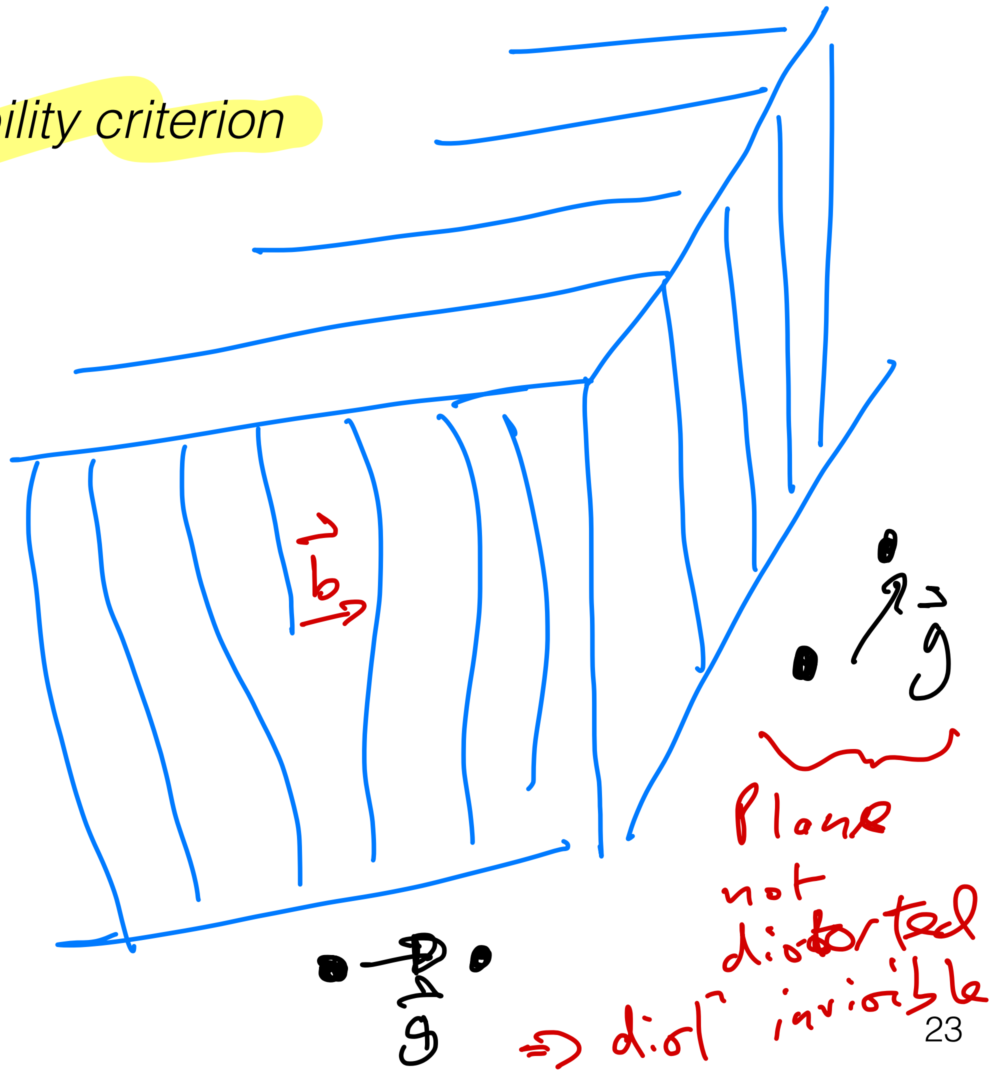
Defects in TEM
R. Schäublin

g.b analysis

g perpendicular to b → dislocation invisible, g.b = 0

b → 3 unknowns → 3 equations

In practice: Minimum 4 different g, non-coplanar



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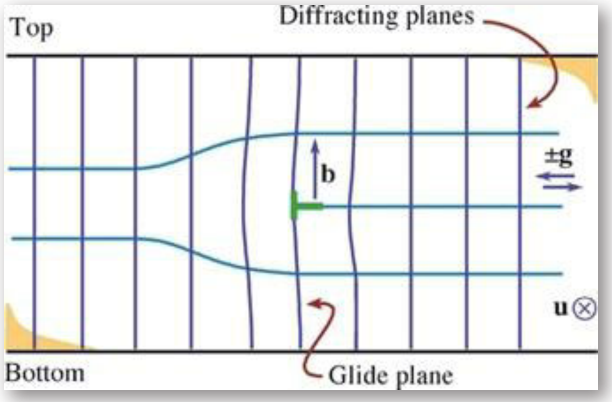
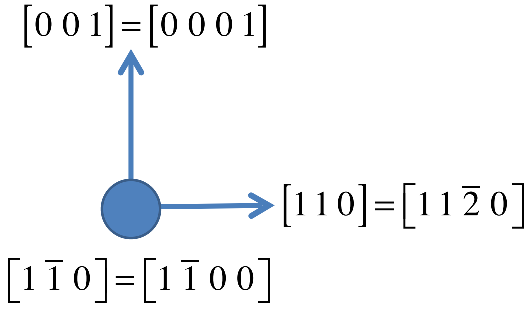
In practice: Minimum 4 different g, non-coplanar

- Note: for edge dislocation the glide plane parallel to ***b*** can be buckled
  ⟹ still gives some contrast even for ***g***.***b*** = 0.
  Plane perpendicular to ***u*** and parallel to ***b*** gives no contrast: ***g***.(***b*** x ***u***) = 0

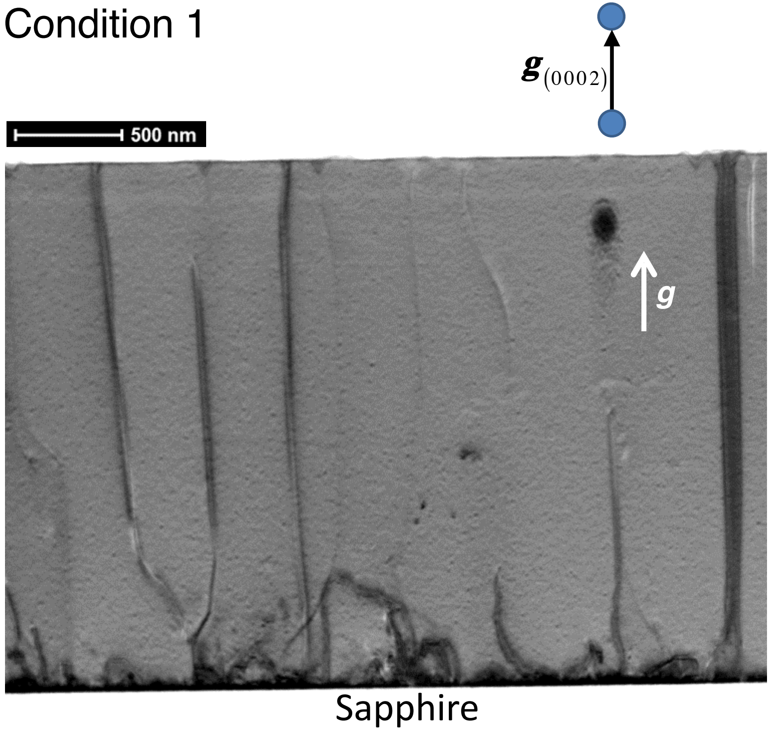


Strong beam dislocation imaging

- Example: threading dislocations of GaN grown on sapphire substrate



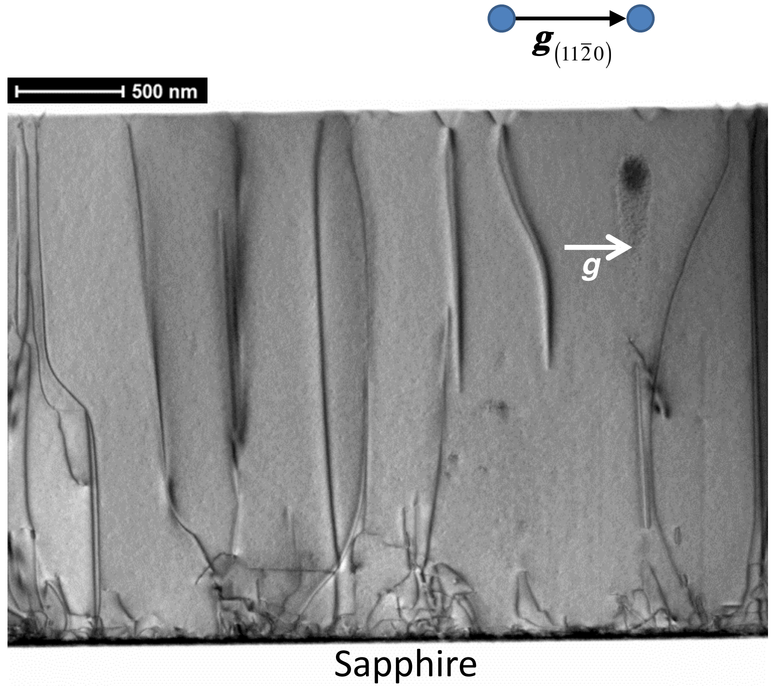
Condition 1



Visible:
Screw $[0\,0\,0\,1]$ and Screw $[0\,0\,0\,\bar{1}]$
Mixed $1/3[2\,\bar{1}\,\bar{1}\,3],[\bar{1}\,\bar{1}\,2\,3],[\bar{1}\,2\,\bar{1}\,3]$

Invisible:
Edge $1/3[2\,\bar{1}\,\bar{1}\,0],[\bar{1}\,\bar{1}\,2\,0],[\bar{1}\,2\,\bar{1}\,0]$

Condition 2



Visible:
Edge $1/3[2\,\bar{1}\,\bar{1}\,0],[\bar{1}\,\bar{1}\,2\,0],[\bar{1}\,2\,\bar{1}\,0]$
Mixed $1/3[2\,\bar{1}\,\bar{1}\,3],[\bar{1}\,\bar{1}\,2\,3],[\bar{1}\,2\,\bar{1}\,3]$

Invisible:
Screw $[0\,0\,0\,1]$ and Screw $[0\,0\,0\,\bar{1}]$

Data from:
Dr Emad Oveisi, CIME

Weak beam imaging: $s \gg 0$

- Remember basic 2-beam dynamical scattering intensities:

$I_g \propto \sin^2(\pi t s)$ ("kinematical")

$$I_g(t) = \frac{1}{1+\xi_g^2 s^2}\sin^2\left(\pi t\sqrt{\frac{1}{\xi_g^2}+s^2}\right) \qquad I_0(t) = 1 - I_g(t)$$

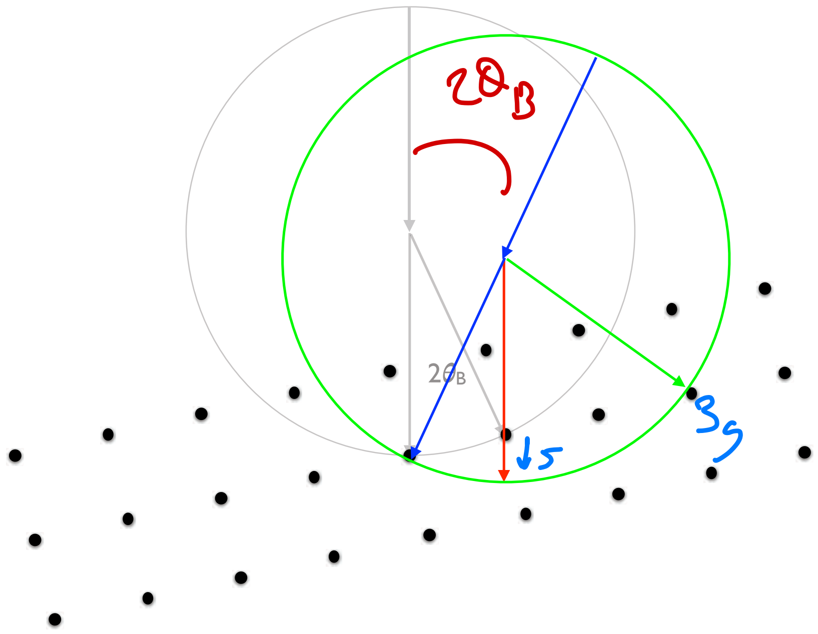
- Imaging with $s = 0$ gives contrast which is highly "dynamical": I_g is very sensitive to proximity to the Bragg condition

- Dislocation images are also not sharp, because you measure intensity variation across the whole strain field, and contrast can be complicated (not just simple bright/dark)

- Can solve these issues by using ***weak beam imaging*** where the excitation error is *large* and *positive*, i.e. $s \gg 0$, e.g. $s \approx 0.2$ nm^{-1}

Weak beam imaging: $s \gg 0$

- Typical imaging conditions are: $\boldsymbol{g}$(3$\boldsymbol{g}$) or 2$\boldsymbol{g}$(5$\boldsymbol{g}$)
- Example: $\boldsymbol{g}$(3$\boldsymbol{g}$)
    - ➡ 1. Tilt sample to excite $\boldsymbol{g}$
    - ➡ 2. Tilt incident beam/Ewald sphere by +2θ_B to excite 3$\boldsymbol{g}$ reflection



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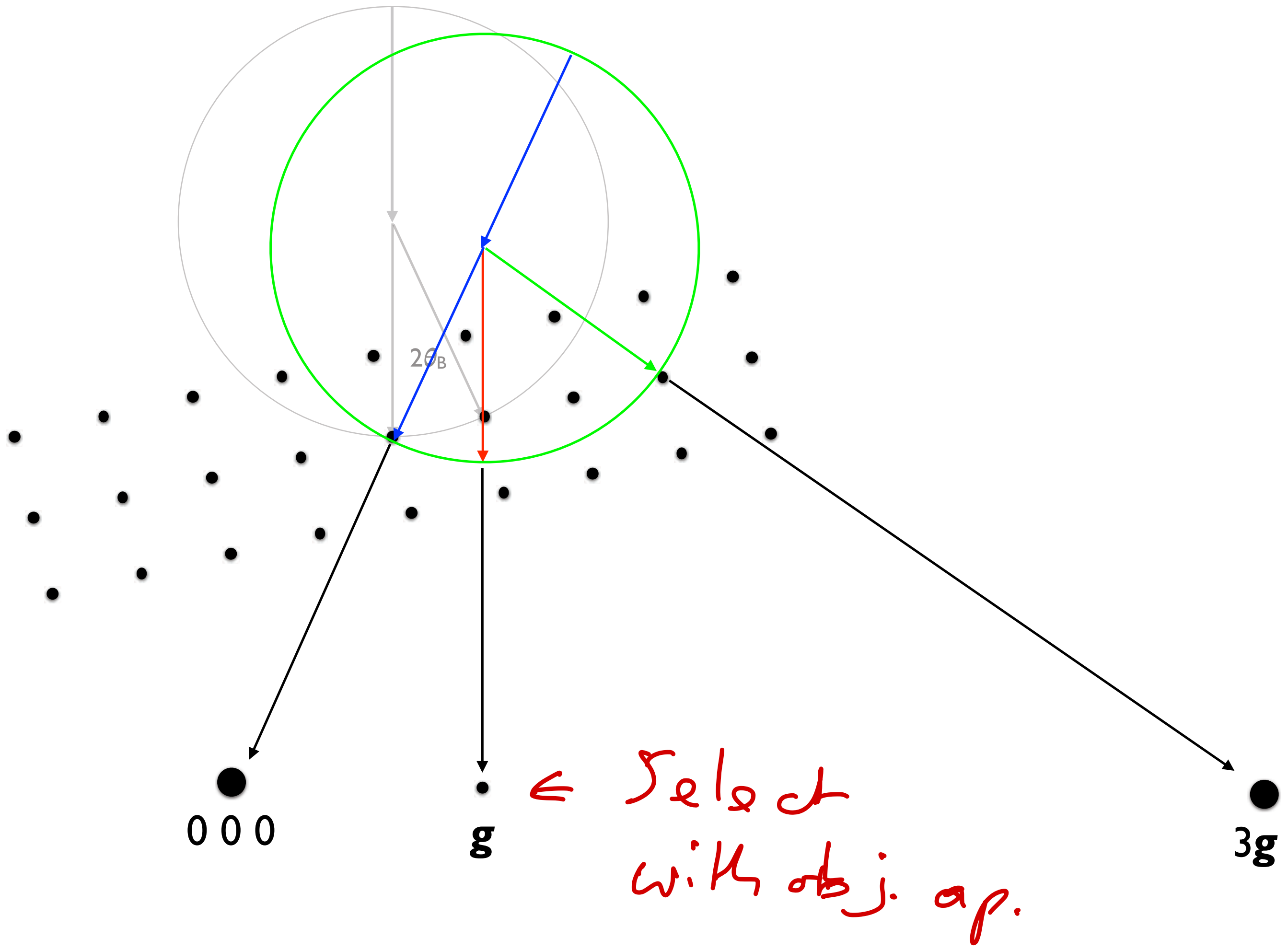
  - ➡ 2. Tilt incident beam/Ewald sphere by $+2\theta_B$ to excite $3\boldsymbol{g}$ reflection
  - ➡ 3. Image with reflection $\boldsymbol{g}$ which has large s and $\boldsymbol{k}_D$ parallel to the optic axis



Weak beam dislocation imaging

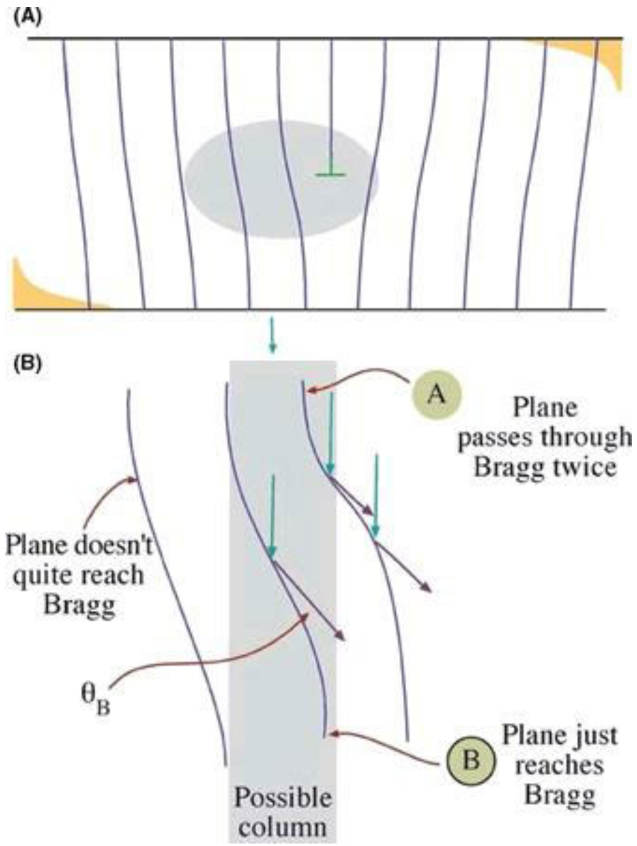
- The bright line corresponds to where planes are tilted towards satisfying the Bragg condition for diffraction vector ***g***, which only happens close to the dislocation core

- The intensity peak is always displaced to one side of the dislocation core

- Hirsch's kinematical approximation for screw dislocations finds half-width of dislocation given by:

$$\Delta x = \frac{1}{\pi s_g}$$

- s_g = 0.2 nm^{-1} $\Longrightarrow$ $\Delta x \approx$ 1.6 nm
(c.f. typically Δx > 10 nm for strong beam)



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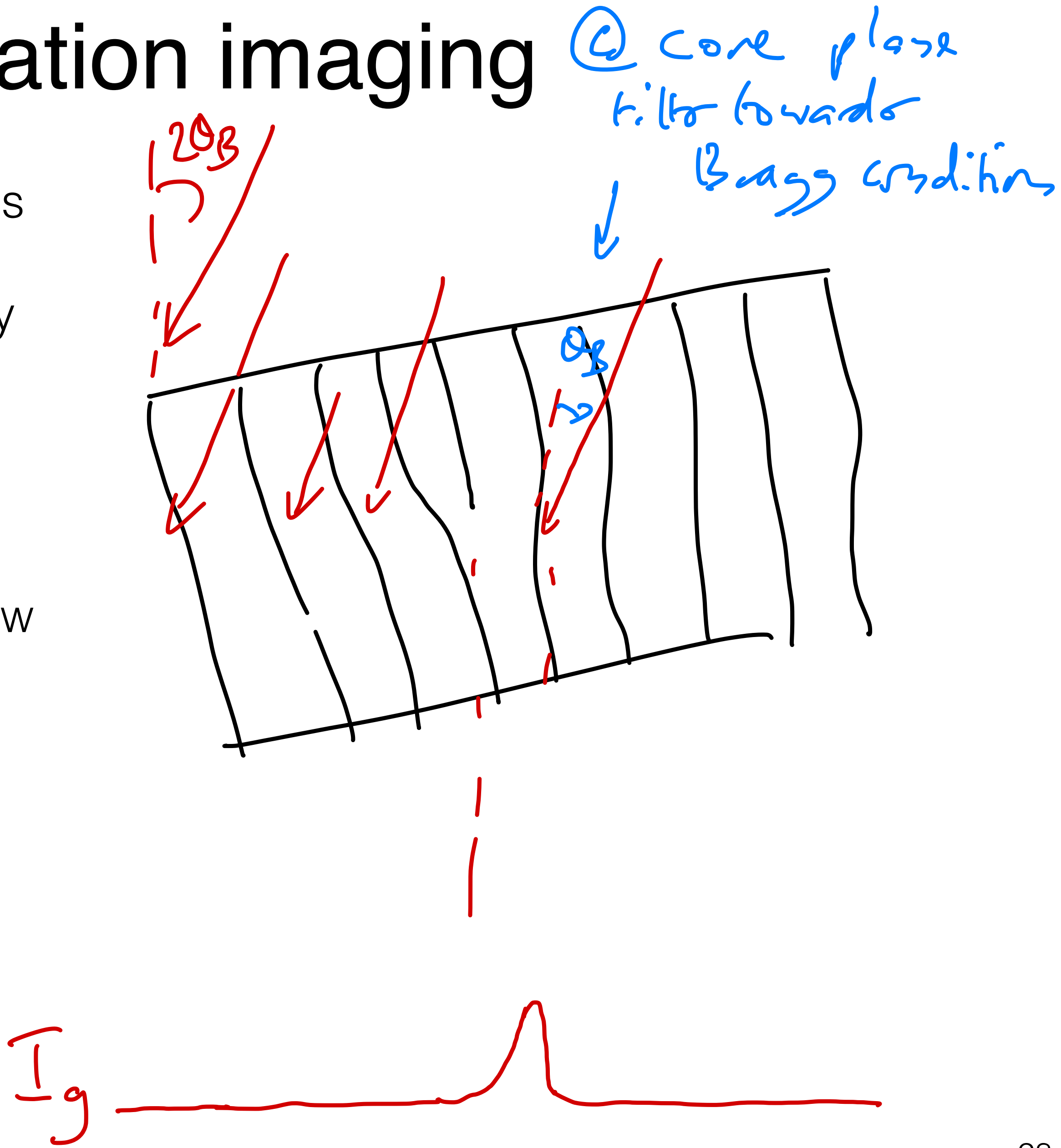
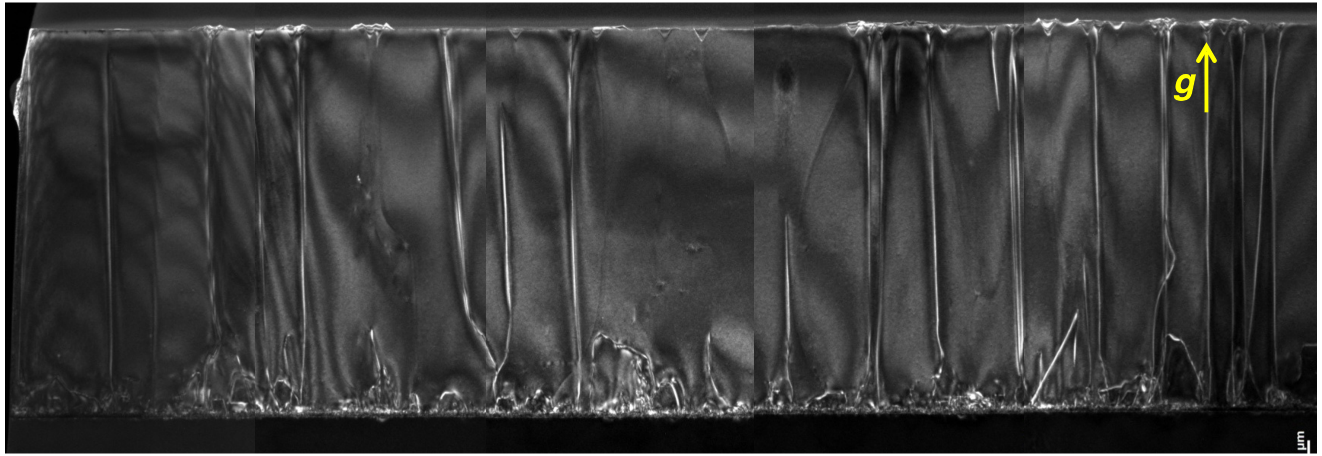
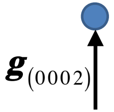


Weak beam dislocation imaging

- Example: threading dislocations of GaN grown on sapphire substrate

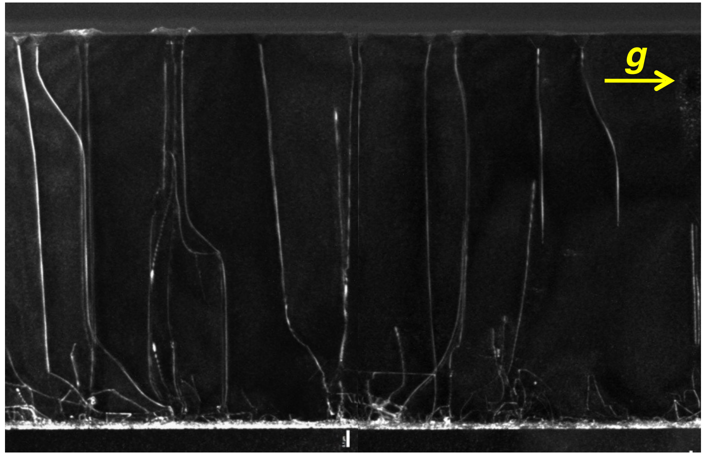


0.5 µm

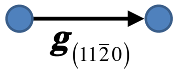


Visible:
Screw $[0\,0\,0\,1]$ and Screw $[0\,0\,0\,\bar{1}]$
Mixed $1/3[2\,\bar{1}\,\bar{1}\,3],[\bar{1}\,\bar{1}\,2\,3],[\bar{1}\,2\,\bar{1}\,3]$

Invisible:
Edge $1/3[2\,\bar{1}\,\bar{1}\,0],[\bar{1}\,\bar{1}\,2\,0],[\bar{1}\,2\,\bar{1}\,0]$



0.5 µm



Visible:
Edge $1/3[2\,\bar{1}\,\bar{1}\,0],[\bar{1}\,\bar{1}\,2\,0],[\bar{1}\,2\,\bar{1}\,0]$
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Invisible:
Screw $[0\,0\,0\,1]$ and Screw $[0\,0\,0\,\bar{1}]$

Data from:
Dr Emad Oveisi, CIME

2-D crystal defects: twinning and stacking faults

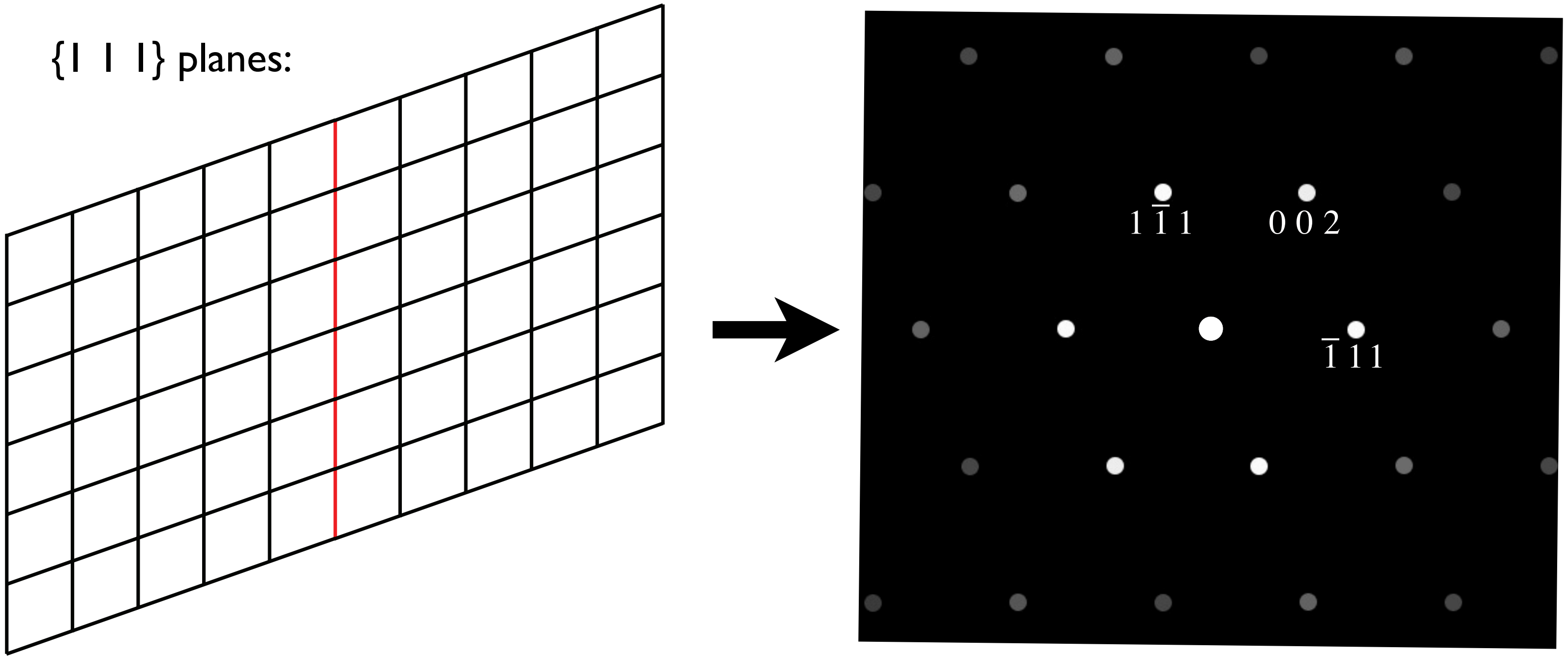
Crystal twinning

- Example: FCC twins
- Stacking of close packed {1 1 1} planes reversed at twin boundary:

A B C A B C A B C A B C

→ A B C A B C B A C B A C

- View on [1 1 0] zone axis:



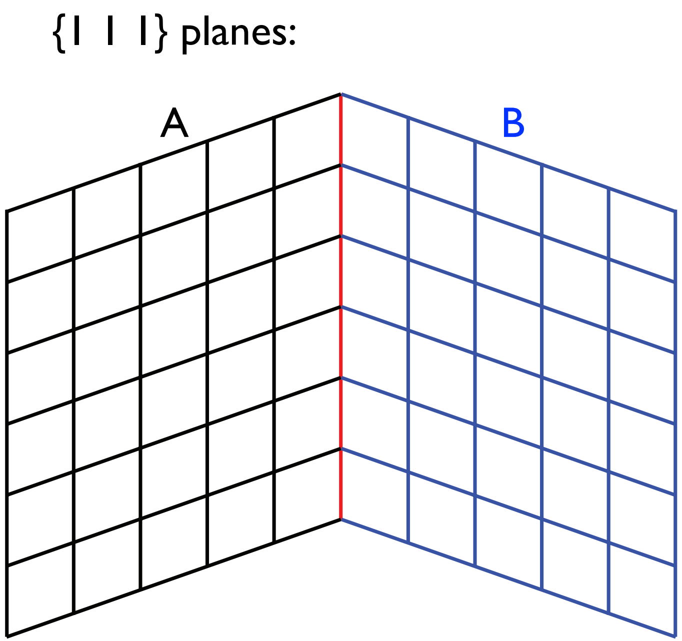
Crystal twinning → Pair of spots/shared spots

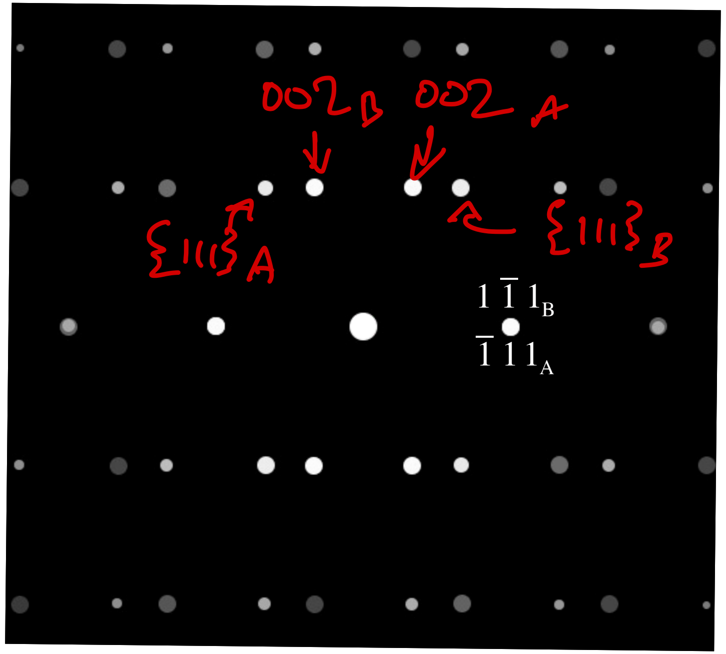
- Example: FCC twins
- Stacking of close packed {1 1 1} planes reversed at twin boundary:

A B C A B C A B C A B C

→ A B C A B C B A C B A C

- View on [1 1 0] zone axis:

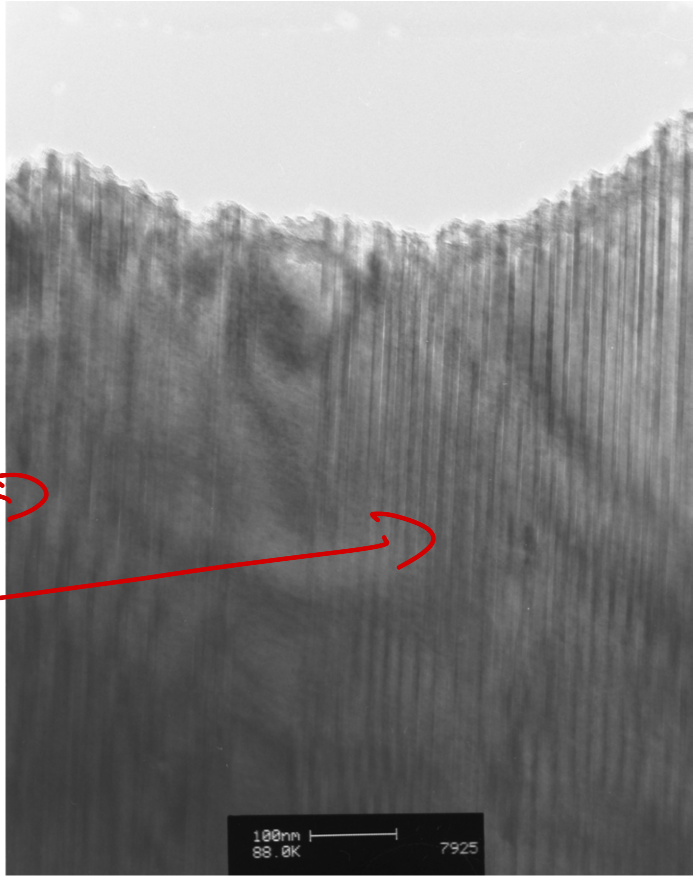


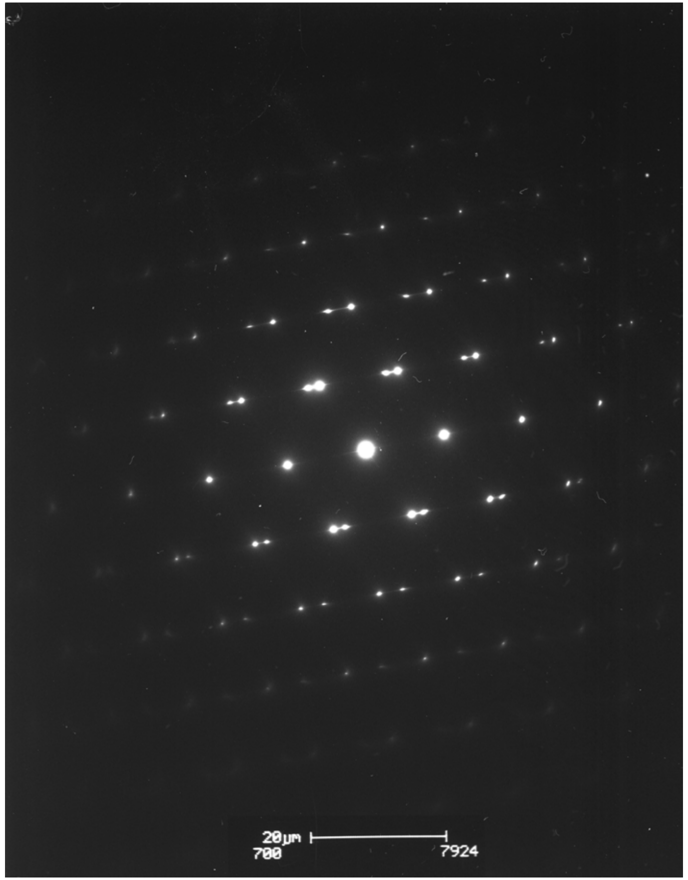


Crystal twinning

- Example: Co-Ni-Al shape memory FCC twins observed on [1 1 0] zone axis
- {1 1 1} close-packed twin plane diffraction spots overlap in SADP

Twin boundaries

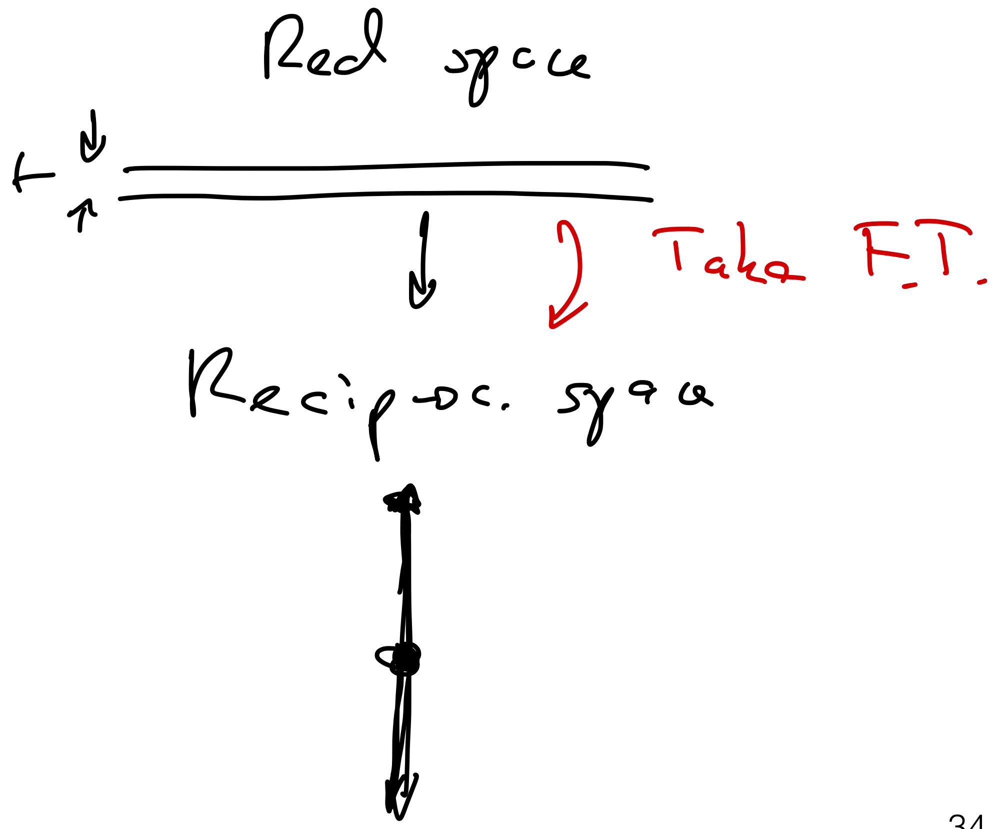




Data from:
Dr Barbora Bartová, CIME

Stacking faults

- Stacking fault: error in crystal stacking sequence
- Example: intrinsic basal plane stacking faults in hexagonal Wurtzite:
  AaBbAaBbAaBbCcBbCcBbCc
- This creates unit cell thick layer of cubic zinc blende structure interrupting the hexagonal stacking
- *How will this affect a diffraction pattern that includes basal plane diffraction spots?*



Stacking faults

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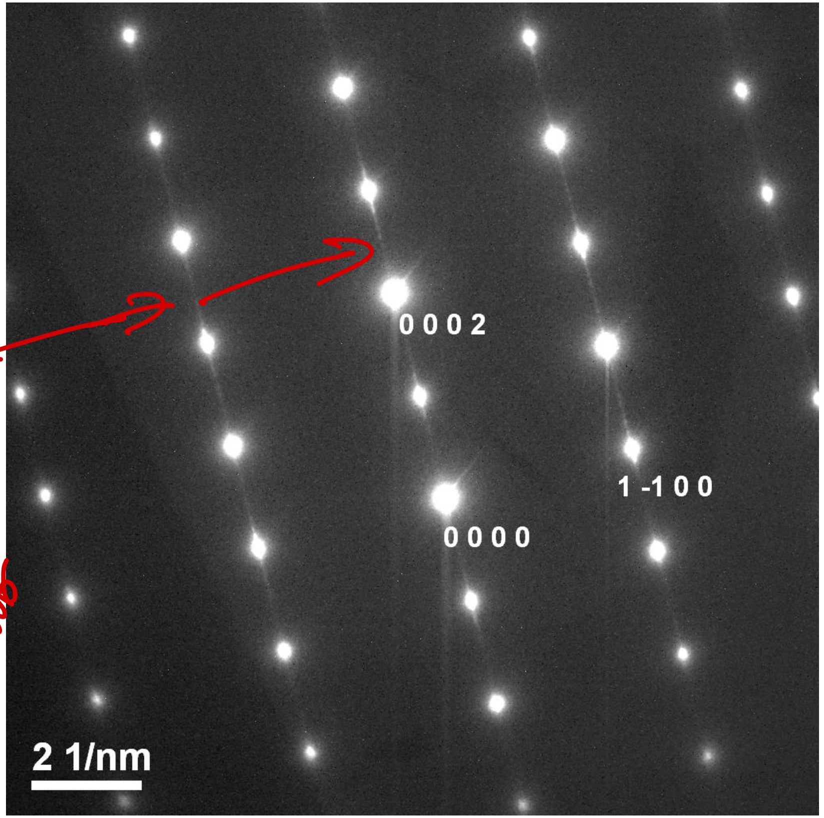
ZnO grain in thin film:
SADP on $[1 1 \bar{2} 0]$ zone axis



Smearing of intensity from stacking faults

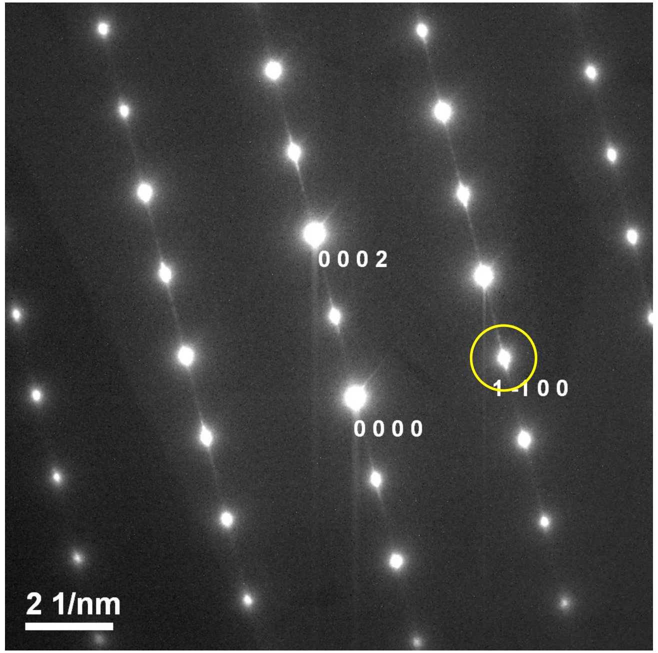
Stacking faults: imaging

- Similarly to Burgers vector analysis, stacking faults with displacement vector ***R*** are invisible for ***g***.***R*** = 0

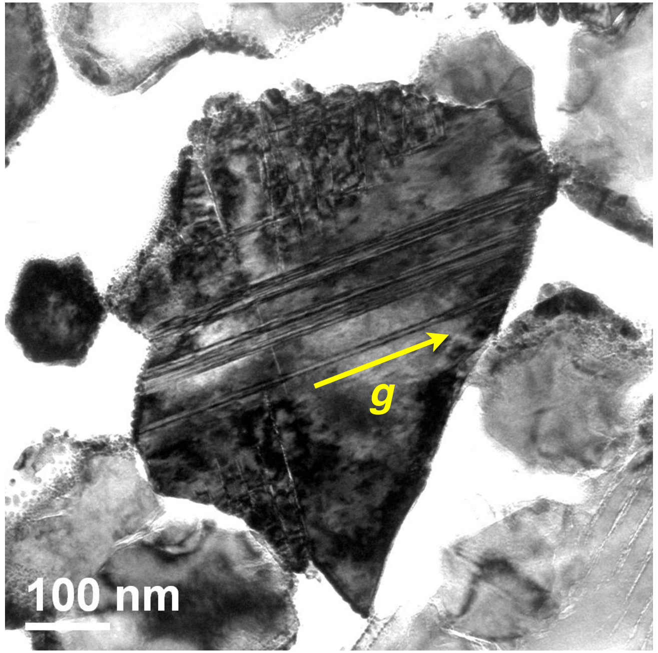
$\vec{g} \parallel \vec{R} \Rightarrow$ S.F. visible

- Example: analysis of basal plane stacking faults in ZnO grain:

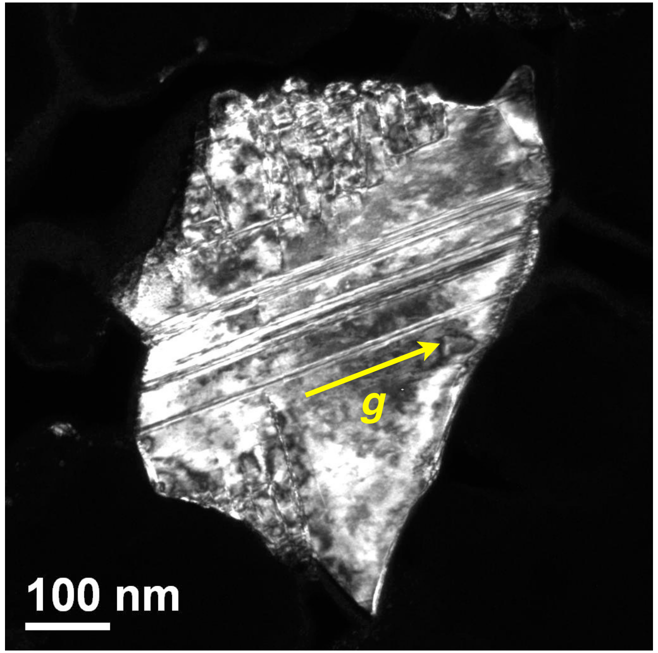
SADP on $[1 1 \bar{2} 0]$ zone axis



Bright-field: $\boldsymbol{g} = (1 \bar{1} 0 0)$



Dark-field: $\boldsymbol{g} = (1 \bar{1} 0 0)$



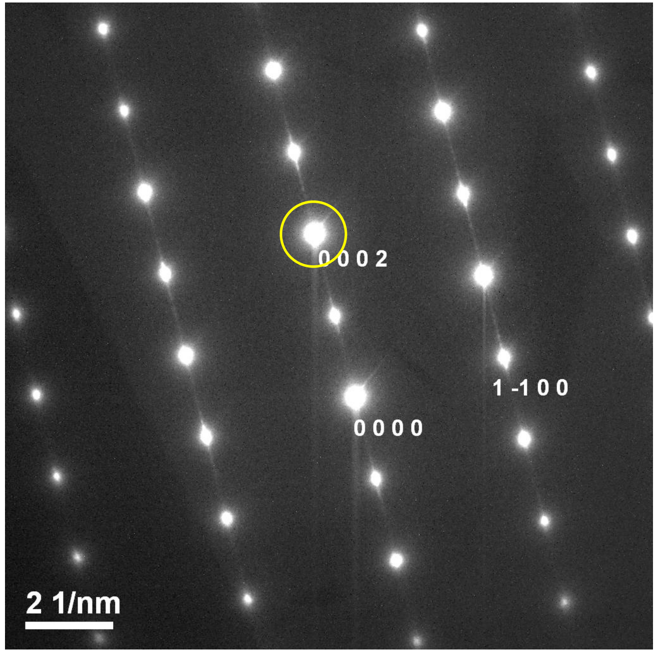
- ***g*** parallel to ***R***: stacking faults visible

Stacking faults: imaging

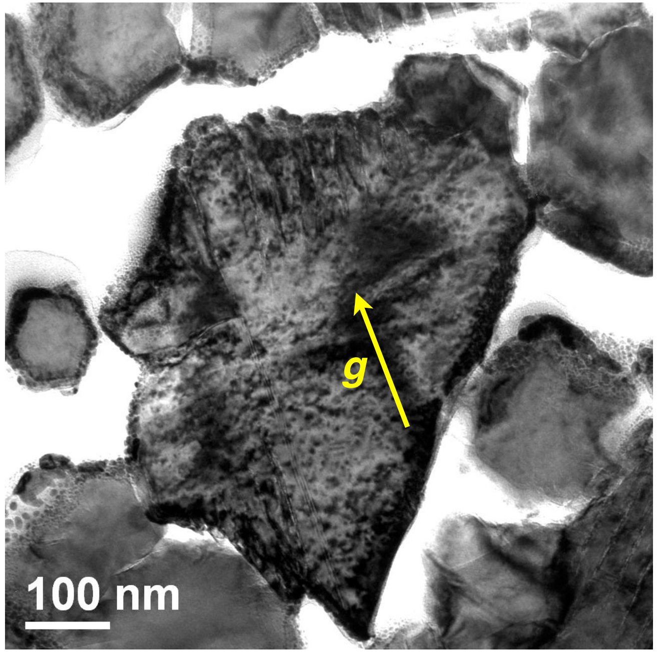
- Similarly to Burgers vector analysis, stacking faults with displacement vector ***R*** are invisible for ***g***.***R*** = 0

- Example: analysis of basal plane stacking faults in ZnO grain:

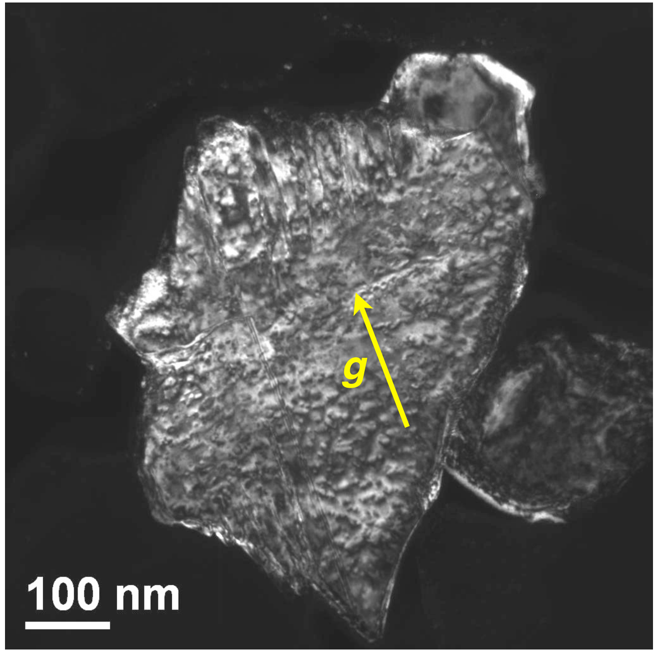
SADP on $[1 1 \bar{2} 0]$ zone axis



Bright-field: $\boldsymbol{g} = (0\,0\,0\,2)$

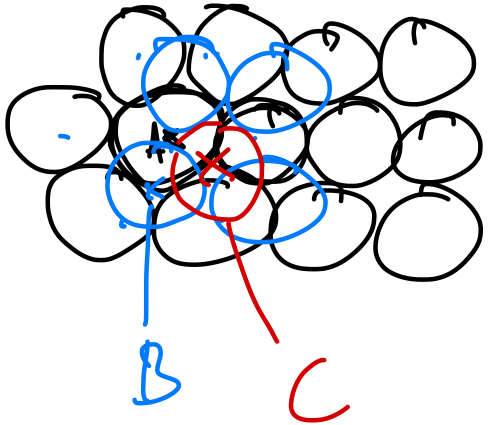


Dark-field: $\boldsymbol{g} = (0\,0\,0\,2)$



- ***g*** perpendicular to ***R***: stacking faults invisible

Close packed material



A

FCC:

{111} – CCP

↓

ABCABC

Hexagonal close packed

ABABAB