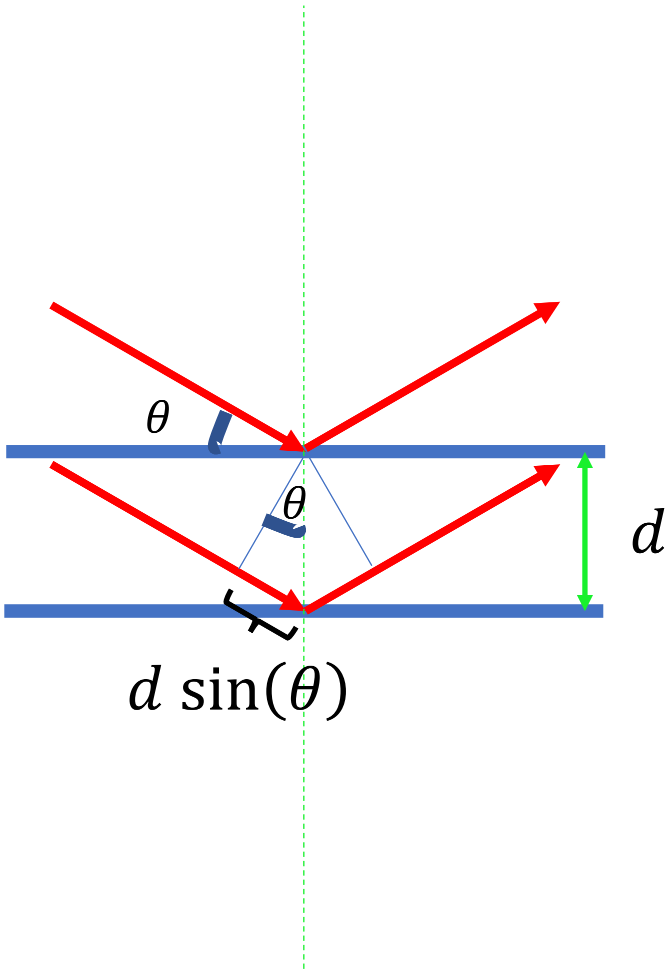
Reflection High Energy Electron Diffraction (RHEED) basics

Xiaoshan Xu

2017/07/26

Cullity, B. D. (1956). *Elements of X-ray diffraction.* Reading, Mass.: Addison-Wesley Pub. Co.
Wang, Z. L. (2011). *Reflection Electron Microscopy and Spectroscopy for Surface Analysis*. Cambridge, GBR: Cambridge University Press.

Diffraction of crystal planes: **real** space



The difference between the two beam path is:

$$2d\,\sin(\theta)$$

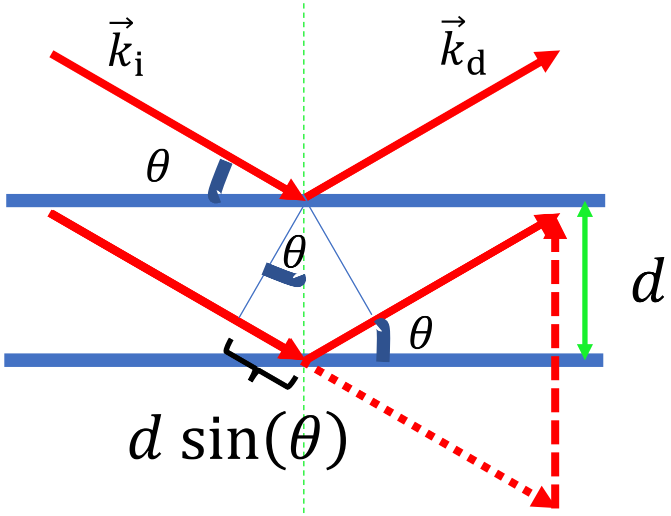
Bragg's law: The diffraction has maximum when

$$2d\,\sin(\theta) = n\lambda$$

Diffraction of crystal planes: **wave vectors**

$\vec{k}_\mathrm{i}, \vec{k}_\mathrm{d}$: wave vectors of the incident and diffracted beams.

$$|\vec{k}_\mathrm{i}| = |\vec{k}_\mathrm{d}| = \frac{2\pi}{\lambda}$$



$$|\vec{k}_\mathrm{d} - \vec{k}_\mathrm{i}| = 2|\vec{k_i}|\sin(\theta)$$

$$\sin(\theta) = \frac{|\vec{k}_\mathrm{d} - \vec{k}_\mathrm{i}|}{2|\vec{k_i}|}$$

Rewrite **Bragg's** law:

$$2d\sin(\theta) = n\lambda$$

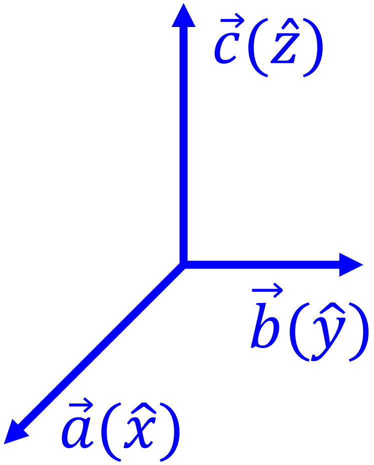
$$2d\,\frac{|\vec{k}_\mathrm{d} - \vec{k}_\mathrm{i}|}{2|\vec{k_i}|} = n\lambda$$

$$|\vec{k_i}|\lambda = 2\pi$$

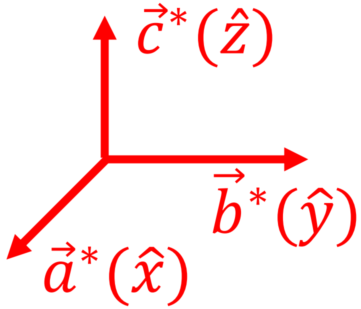
$$|\vec{k}_\mathrm{d} - \vec{k}_\mathrm{i}| = n\frac{2\pi}{d}$$

What's $\frac{2\pi}{d}$?

$$\left|\vec{k}_{\mathrm{d}} - \vec{k}_{\mathrm{i}}\right| = n\frac{2\pi}{d}$$



Real space

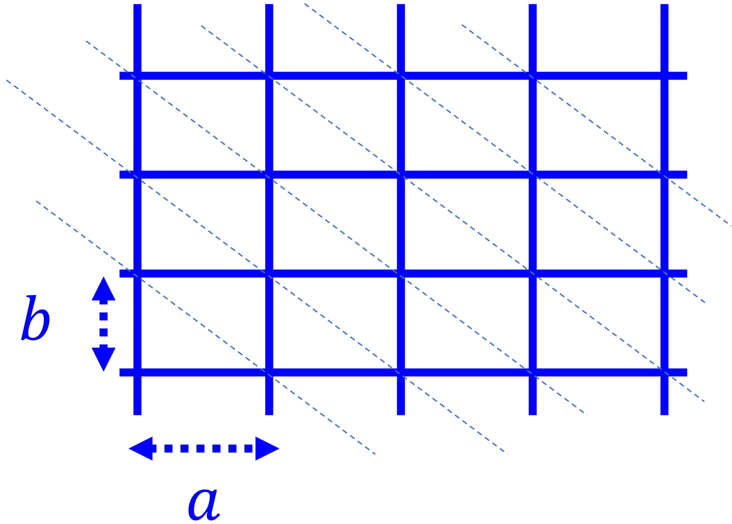


Reciprocal space

$$\vec{a}^* = 2\pi\frac{\vec{b}\times\vec{c}}{(\vec{a}\times\vec{b})\cdot\vec{c}} = \frac{2\pi}{a}\ \hat{x}$$

$$\vec{b}^* = 2\pi\frac{\vec{c}\times\vec{a}}{(\vec{a}\times\vec{b})\cdot\vec{c}} = \frac{2\pi}{b}\ \hat{y}$$

$$\vec{c}^* = 2\pi\frac{\vec{a}\times\vec{b}}{(\vec{a}\times\vec{b})\cdot\vec{c}} = \frac{2\pi}{c}\ \hat{z}$$



$$\vec{b}^* = 2\pi \frac{\vec{c} \times \vec{a}}{(\vec{a} \times \vec{b}) \cdot \vec{c}} = \frac{2\pi}{b}\ \hat{y}$$

$$\vec{a}^* = 2\pi \frac{\vec{b} \times \vec{c}}{(\vec{a} \times \vec{b}) \cdot \vec{c}} = \frac{2\pi}{a}\ \hat{x}$$

Example: the (110) plane

$$d_{(110)} = \frac{ab}{\sqrt{a^2 + b^2}}$$

$$\frac{2\pi}{d_{(110)}} = 2\pi \frac{\sqrt{a^2 + b^2}}{ab}$$

The reciprocal vector (110)

$$|\vec{G}_{(110)}| = \left|\vec{a}^* + \vec{b}^*\right|$$

$$= \left|\frac{2\pi}{a}\ \hat{x} + \frac{2\pi}{b}\ \hat{y}\right|$$

$$= 2\pi \frac{\sqrt{a^2 + b^2}}{ab}$$

$$\left|\vec{k}_\mathrm{d} - \vec{k}_\mathrm{i}\right| = n\frac{2\pi}{d}$$

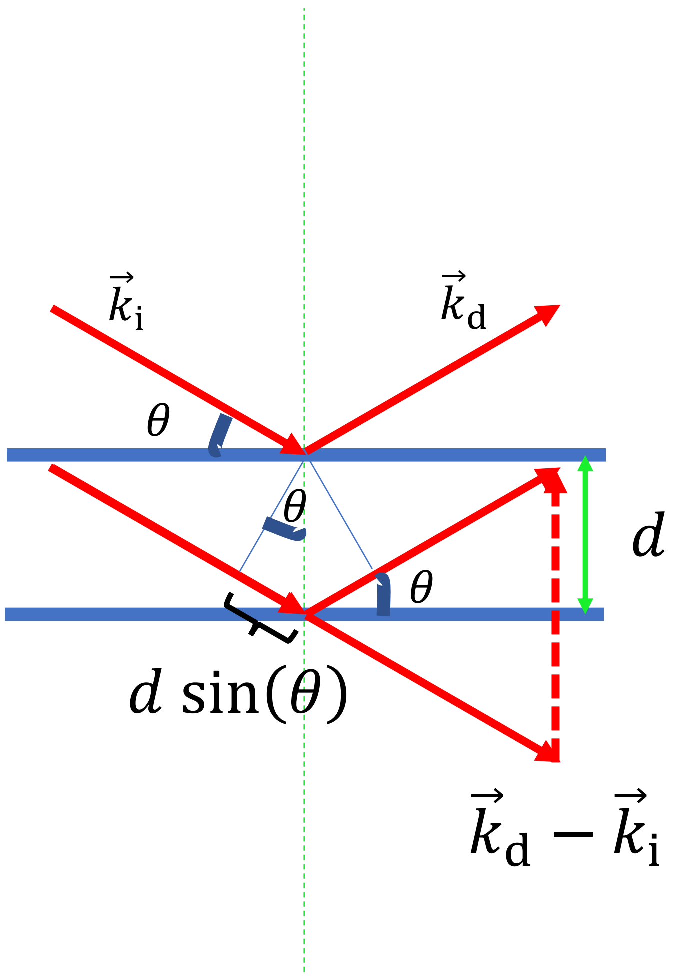
$$\frac{2\pi}{d_{(110)}} = |\vec{G}_{(110)}|$$

$$\left|\vec{k}_\mathrm{d} - \vec{k}_\mathrm{i}\right| = \left|\vec{G}\right|$$

$$\vec{G} = h\vec{a}^* + k\vec{b}^* + l\vec{c}^*$$

h, k, l are integers (Miller indices).

$$\left|\vec{k}_{\mathrm{d}} - \vec{k}_{\mathrm{i}}\right| = \left|\vec{G}\right|$$

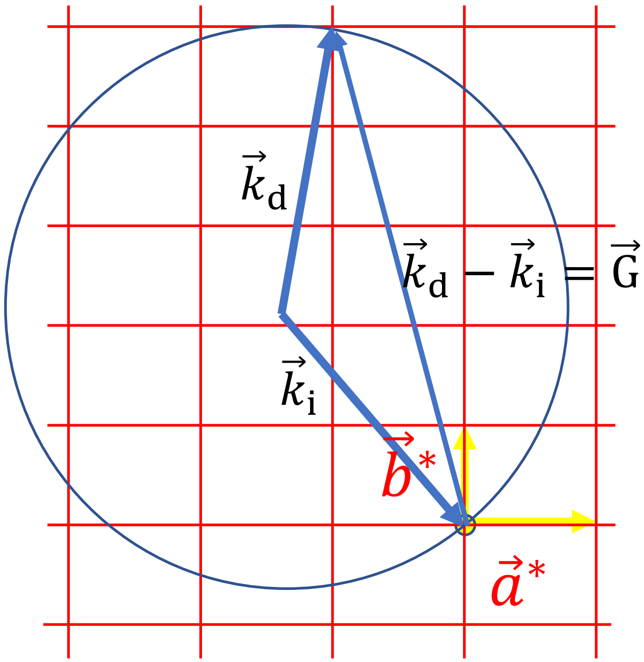


$\vec{k}_{\mathrm{d}} - \vec{k}_{\mathrm{i}}$ is along the normal of crystal plane. So,

$$\vec{k}_{\mathrm{d}} - \vec{k}_{\mathrm{i}} = \vec{G}$$
$$2d\sin(\theta) = n\lambda$$

$$\vec{G} = h\vec{a}^{*} + k\vec{b}^{*} + l\vec{c}^{*}$$
h, k, l are integers (Miller indices).

Crystal diffraction in **reciprocal space**



A cross section of the ***Ewald sphere*** in 3D reciprocal space.

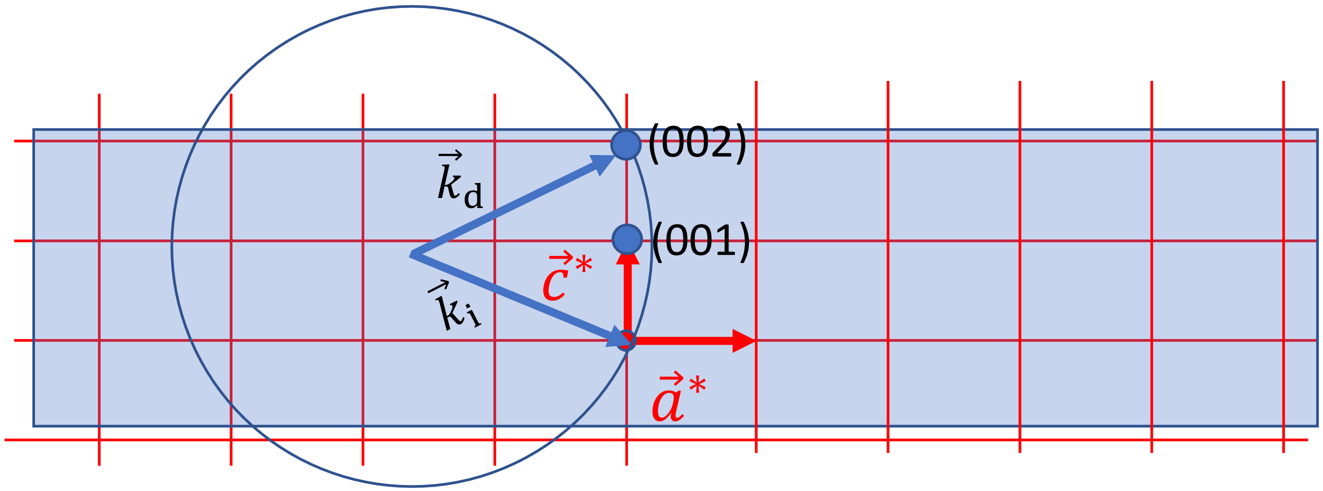
$$2d\sin(\theta) = n\lambda$$

$$\vec{k}_\mathrm{d} - \vec{k}_\mathrm{i} = \vec{G}$$

In reciprocal space,

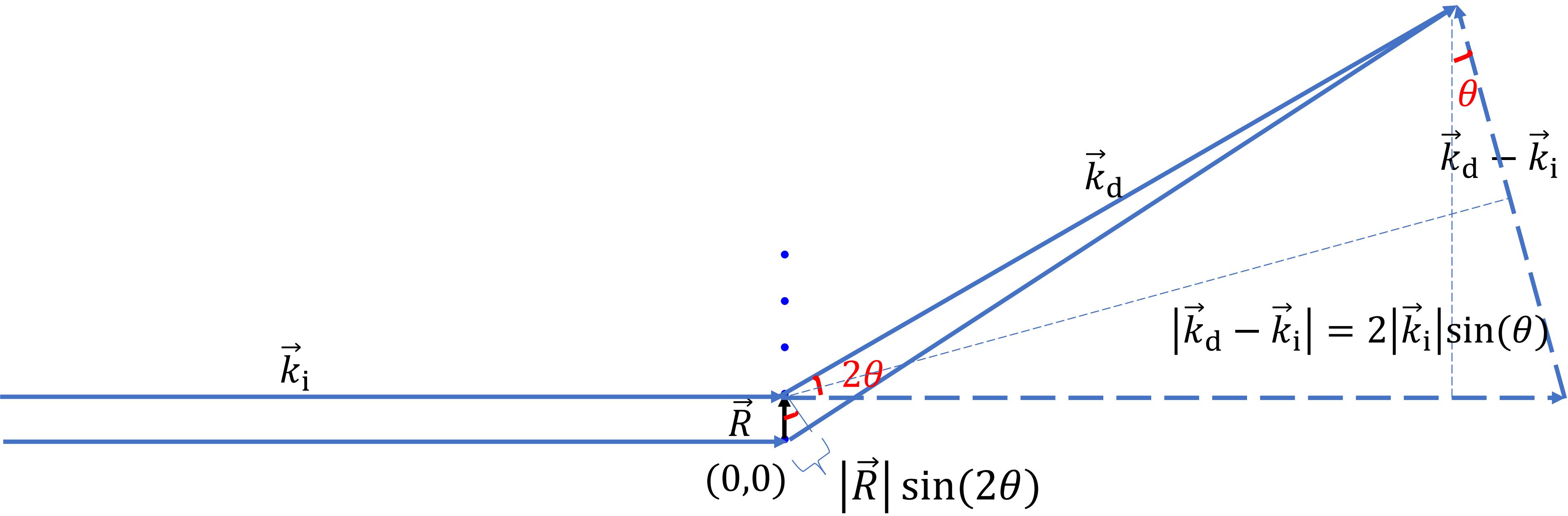
1) Draw a circle, using the origin of $\vec{k}_i$ as the center and $|\vec{k}_i|$ as the radius.
2) Draw $\vec{k}_i$ on the circle
3) Move the circle so that the tip of $\vec{k}_i$ is at the origin the reciprocal space.
4) The reciprocal lattice points that fall on the perimeter of the circle correspond to diffraction conditions.

Example, (00L) diffraction



$$\vec{k}_{\rm d} - \vec{k}_{\rm i} = (00l)$$

Contribution from **individual atoms** to diffraction



Phase change:

$$2\pi\frac{\left|\vec{R}\right|\sin(2\theta)}{\lambda} = \frac{2\pi}{\lambda}\left|\vec{R}\right|2\sin(\theta)\cos(\theta)$$

$$= \left|\vec{k}_\mathrm{i}\right|\left|\vec{R}\right|2\sin(\theta)\cos(\theta)$$

$$= \left|\vec{k}_\mathrm{d} - \vec{k}_\mathrm{i}\right|\left|\vec{R}\right|\cos(\theta)$$

$$= \left(\vec{k}_\mathrm{d} - \vec{k}_\mathrm{i}\right)\cdot\vec{R}$$

$$\vec{R} = u\vec{a} + v\vec{b} + w\,\vec{c}$$

u, v, w are integers

$$\left|\vec{k}_\mathrm{d} - \vec{k}_\mathrm{i}\right| = 2\left|\vec{k}_\mathrm{i}\right|\sin(\theta)$$

Every atom contributes an amplitude proportional to:

$$\exp\left[-i\left(\vec{k}_\mathrm{d} - \vec{k}_\mathrm{i}\right)\cdot\vec{R}\right]$$

Crystal diffraction and **Fourier transform**

Every atom contributes an amplitude proportional to:

$$\exp\left[-i\left(\vec{k}_{\mathrm{d}}-\vec{k}_{\mathrm{i}}\right)\cdot\vec{R}\right]$$

Diffraction intensity:

$$I(\vec{k}_{\mathrm{d}}-\vec{k}_{\mathrm{i}})=\left|\sum_{\vec{R}}\exp\left[-i\left(\vec{k}_{\mathrm{d}}-\vec{k}_{\mathrm{i}}\right)\cdot\vec{R}\right]\right|^{2}$$

$\vec{R}=u\vec{a}+v\vec{b}+w\vec{c}$ are the position of atoms.

This is actually a Fourier transform of the lattice from real space into the reciprocal space.

Rewrite: $I(\vec{k}_\mathrm{d} - \vec{k}_\mathrm{i}) = \sum_i \exp\left[-i\left(\vec{k}_\mathrm{d} - \vec{k}_\mathrm{i}\right) \cdot \vec{R}\right]$

$\vec{R} = u\vec{a} + v\vec{b} + w\,\vec{c}$
u, v, w are integers

$$I\left(\vec{k}\right) = \left|\sum_{\vec{R}} \exp\left[-i\vec{k} \cdot \vec{R}\right]\right|^2$$

$$= \left|\sum_{u,v,w} \exp[-i(k_1 \cdot ua + k_2 \cdot vb + k_3 \cdot wc)]\right|^2$$

$$\vec{k} \equiv \vec{k}_\mathrm{d} - \vec{k}_\mathrm{i}$$
$$\equiv k_1\vec{a}^* + k_2\vec{b}^* + k_3\,\vec{c}^*$$

$$= \left|\sum_u \exp[-i(k_1 \cdot ua)] \sum_v \exp[-i(k_2 \cdot vb)] \sum_w \exp[-i(k_3 \cdot wc)]\right|^2$$

Let's look at one of the sum:

$$\sum_u \exp[-i(k_1 \cdot ua)]$$

$= N$ if $k_1 \cdot ua = \mathrm{n}2\pi$

$= 0$ otherwise

So, after the transform:

$$k_1 = ha^*$$
$$k_2 = kb^*$$
$$k_3 = lc^*$$

This is the reciprocal lattice

Fourier transform of lattice of **different dimensions**

$$I(\vec{k}) = \left|\sum_u \exp[-i(k_1 \cdot ua)] \sum_v \exp[-i(k_2 \cdot vb)] \sum_w \exp[-i(k_3 \cdot wc)]\right|^2$$

If the lattice is ***two*** *dimensional (e.g. in the a-b plane):*

$$\vec{R} = u\vec{a} + v\vec{b}$$

$u, v,$ are integers to sum over

$w = 0$

After the transform:

$$k_1 = ha^*$$
$$k_2 = kb^*$$
$$k_3 \text{ is arbitrary}$$

If the lattice is ***one*** *dimensional (e.g. along the a axis):*

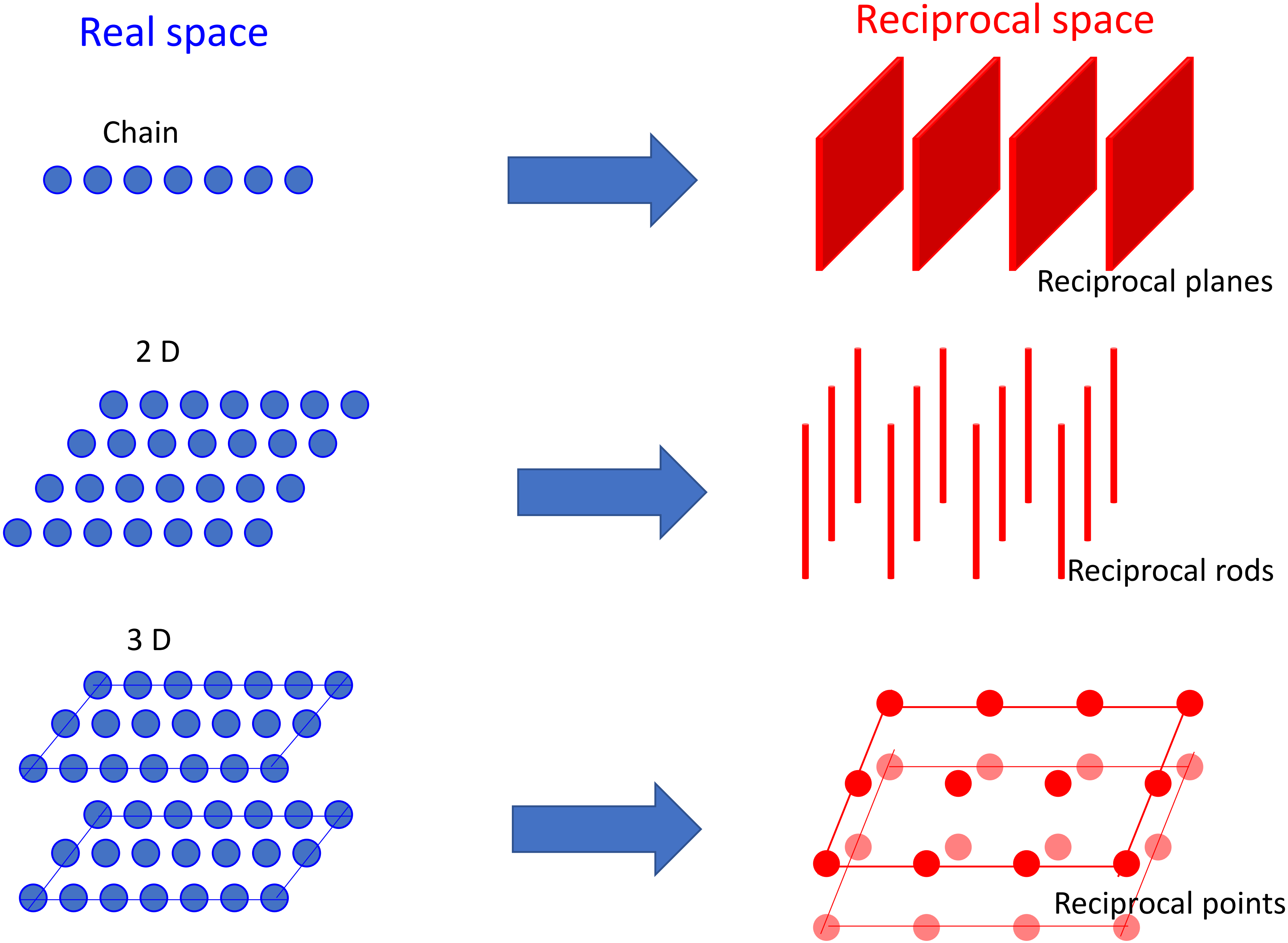
$$\vec{R} = u\vec{a}$$

$u,$ are integers to sum over

$v, w = 0$

After the transform:

$$k_1 = ha^*$$
$$k_2, k_3 \text{ are arbitrary}$$

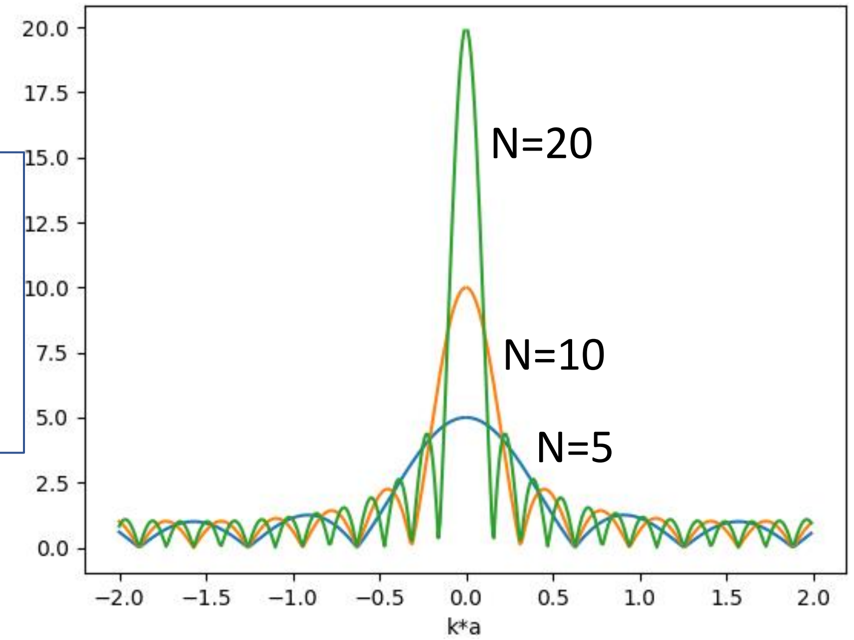

Real space
Reciprocal space
Chain
Reciprocal planes
2 D
Reciprocal rods
3 D
Reciprocal points

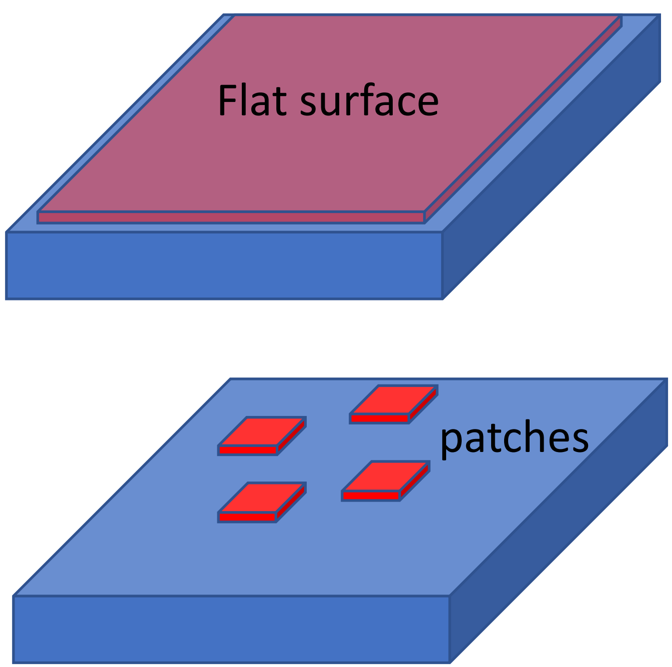
Real crystal, **finite size**

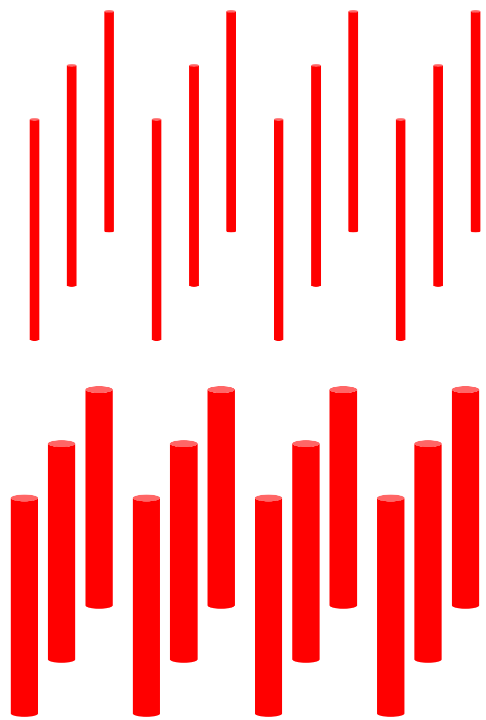
Let's look at the sum again:

$$\left|\sum_{u=1..N} \exp[-i(k_x \cdot ua)]\right|^2 = \frac{\sin^2(Nk_x a)}{\sin^2(k_x a)}$$

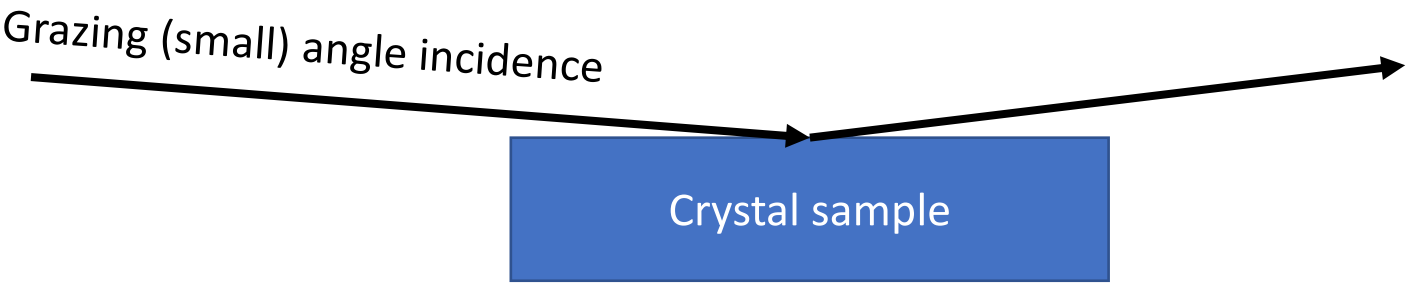
For intensity: $\Delta k \propto \frac{1}{N^2}$

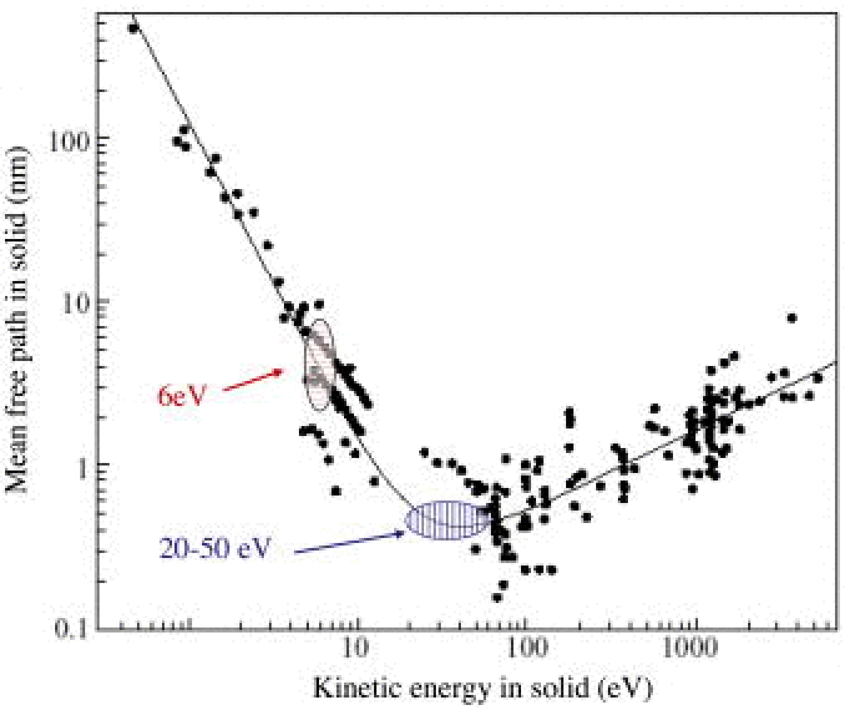


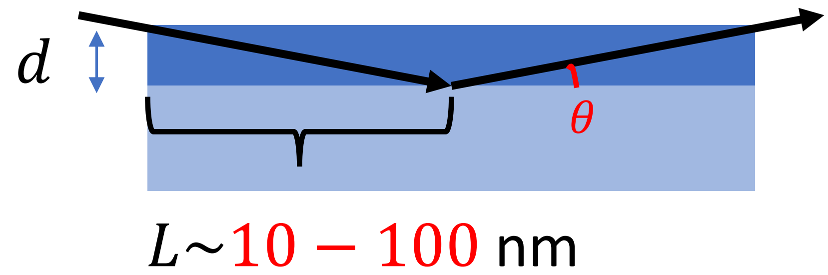




Reflection high energy diffraction (RHEED) **geometry**







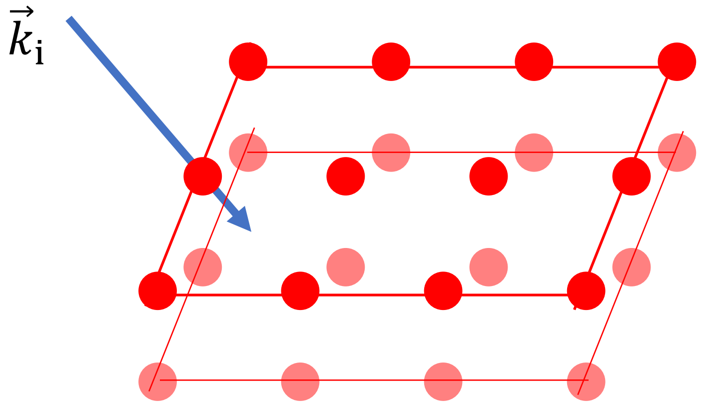
Penetration depth for 30 keV electron is $L \sim 10 - 100$ nm.

$d \approx L \tan(\theta) = 1.7$ nm

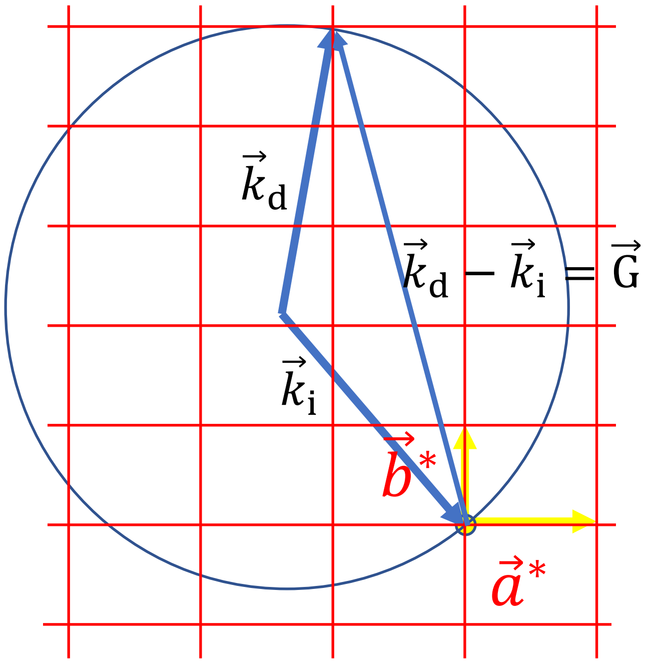
Assuming: $L = 100\ nm, \theta = 1\ degree$

RHEED probes the **surface (2 D lattice)**.

Ewald sphere and reciprocal **points**



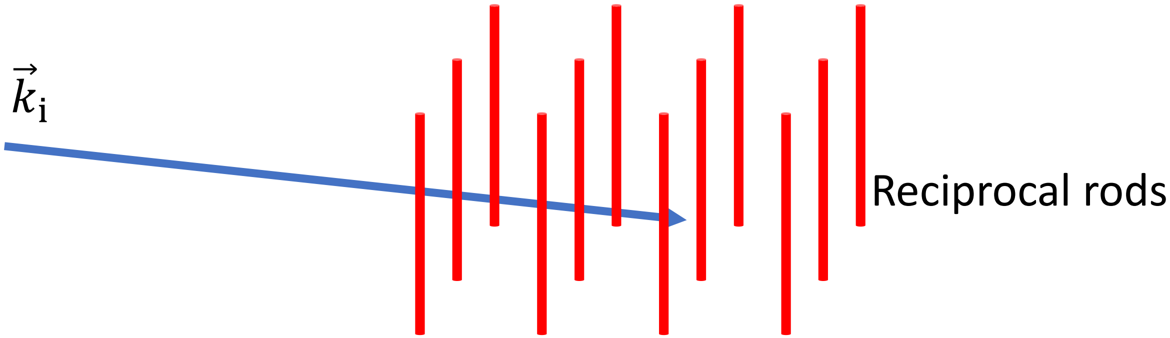
Reciprocal points



Cross section of ***Ewald sphere*** in 3D reciprocal space.

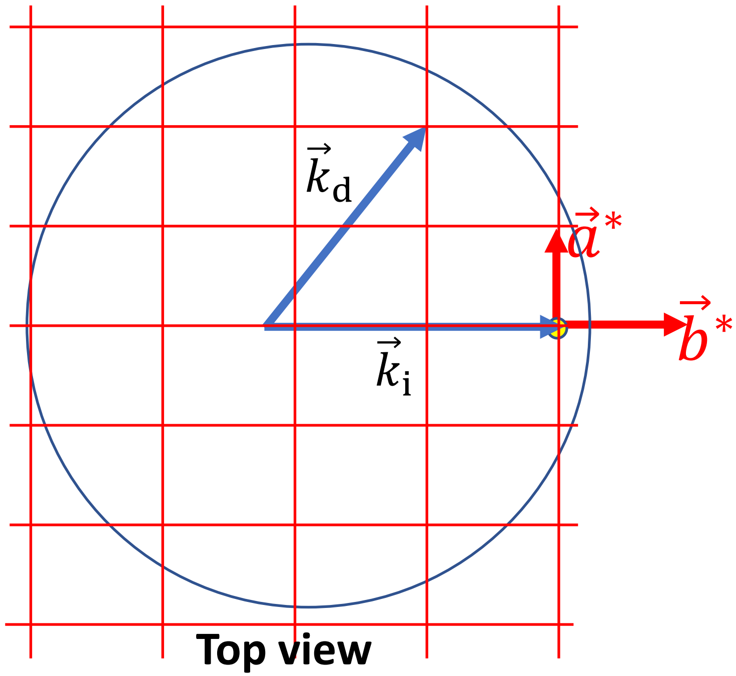
For 3 D real space, the reciprocal space consists of reciprocal points.
Only when the reciprocal points fall on the ***Ewald sphere***, diffraction occurs.

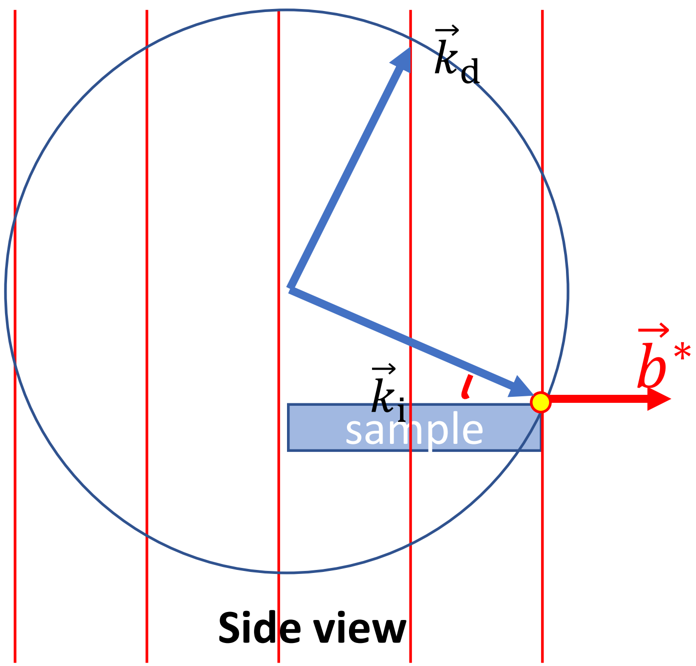
Ewald sphere and reciprocal **rods**

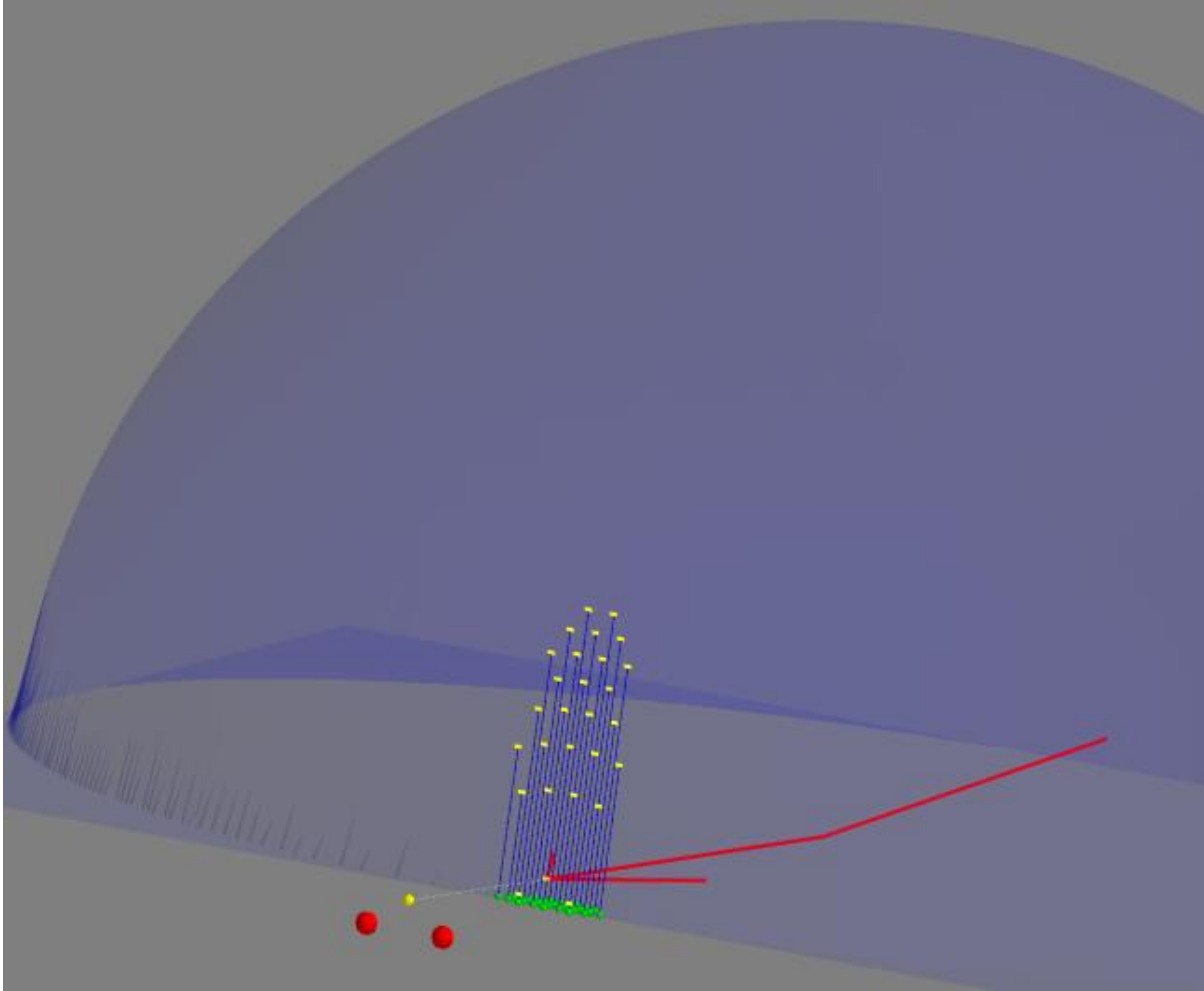


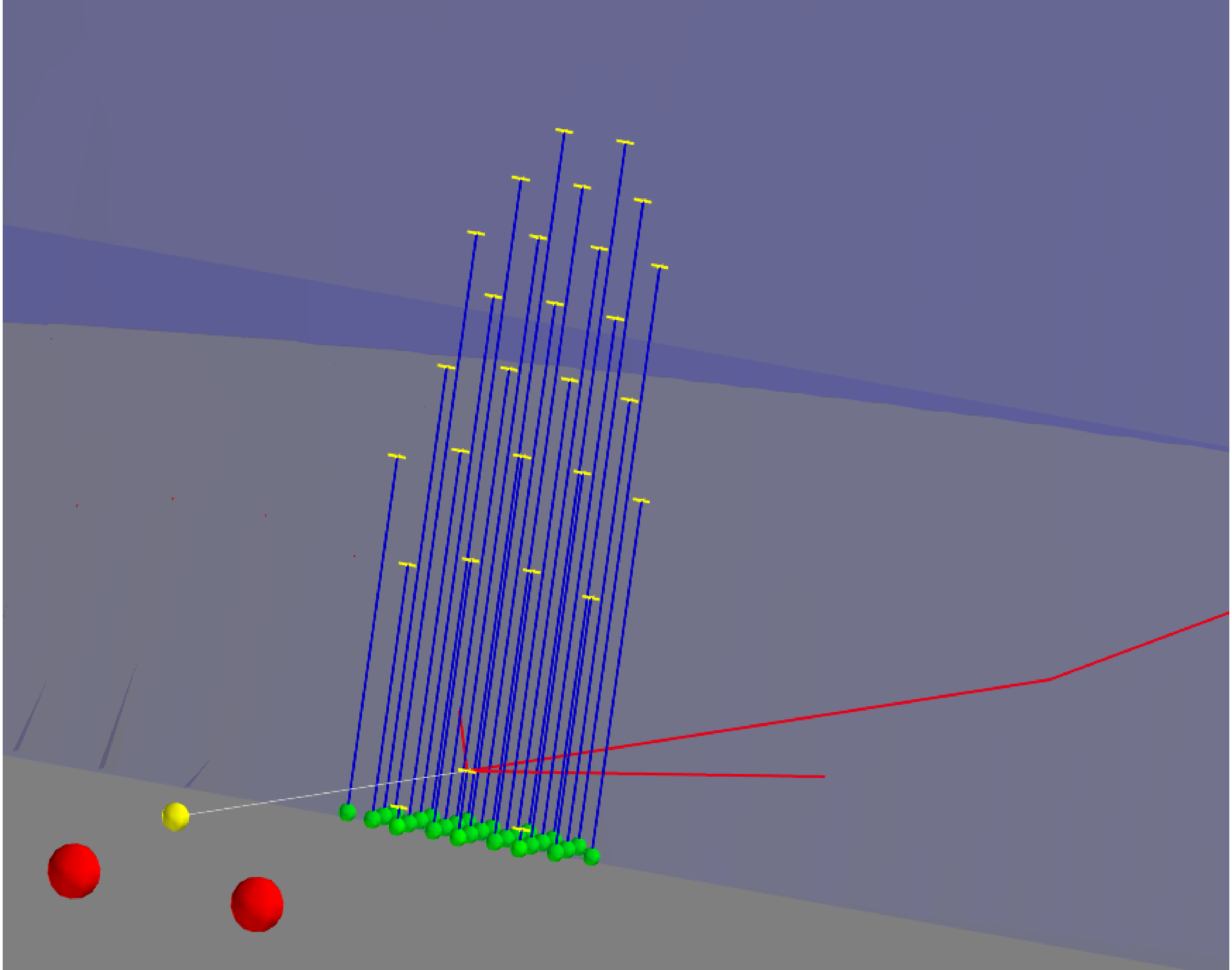
Top and side view of the ***Ewald sphere*** in RHEED

For 2D real space, **every** reciprocal rods can intersect with the ***Ewald sphere***, causing diffraction.

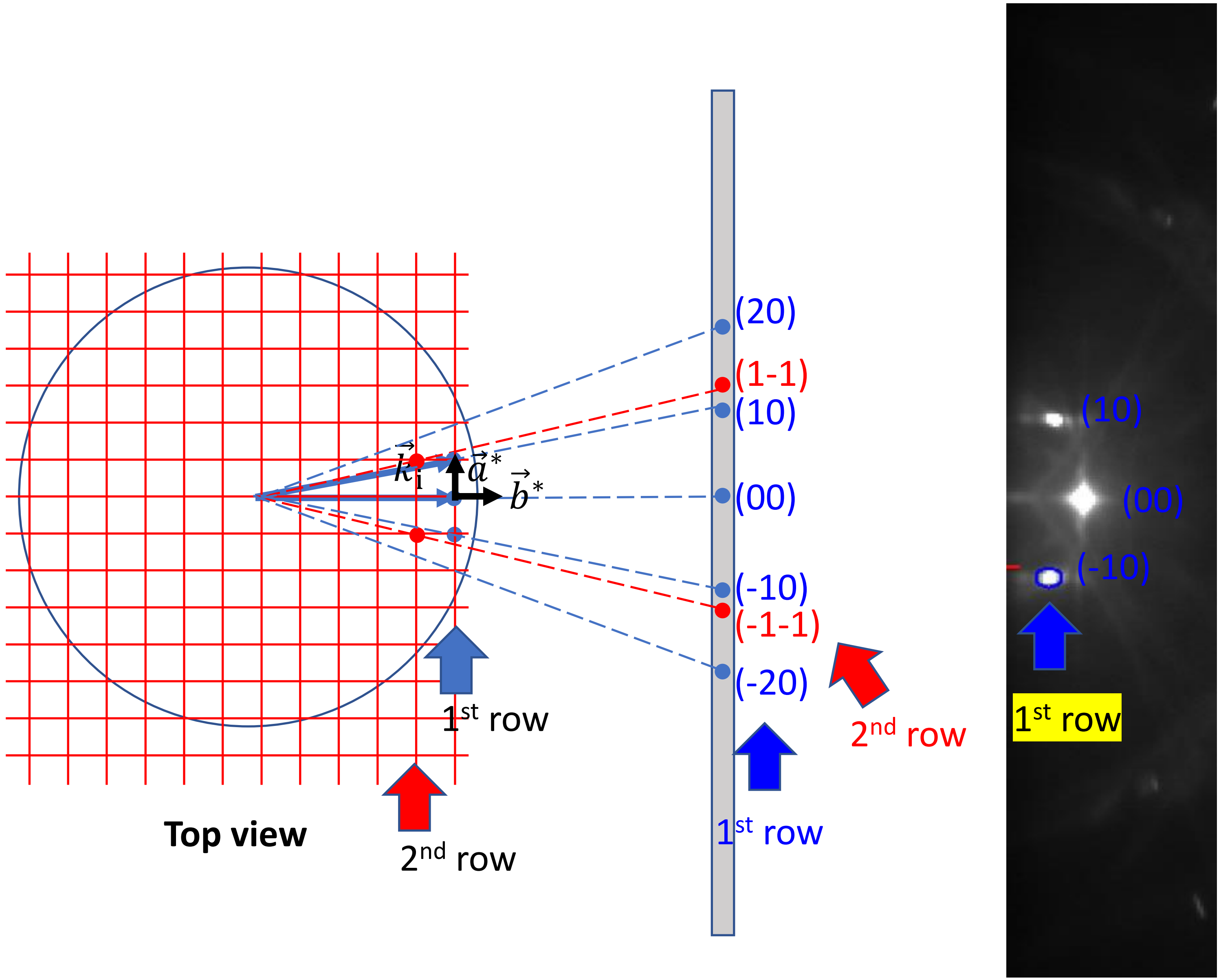






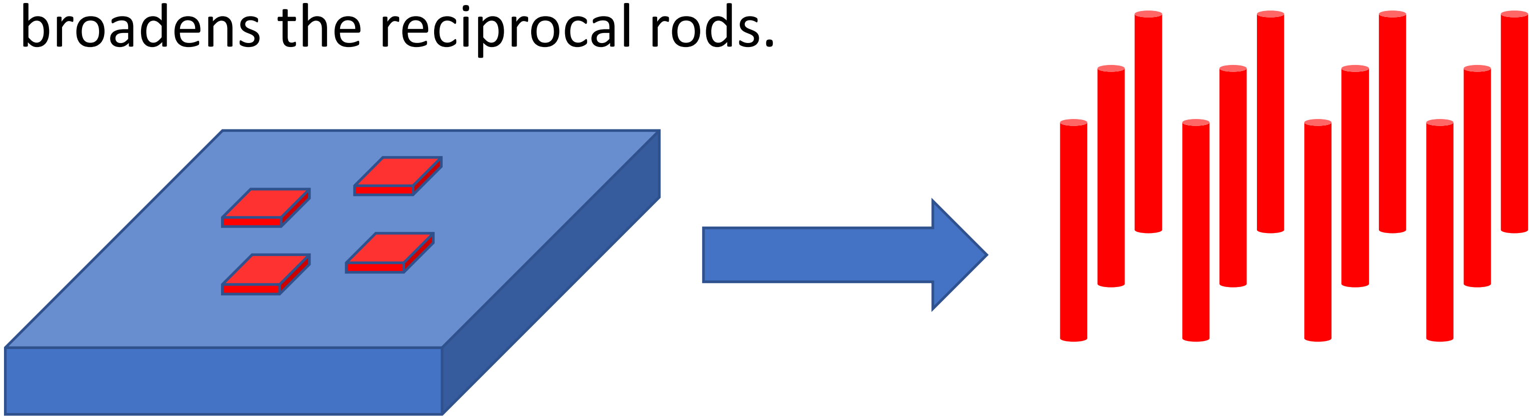


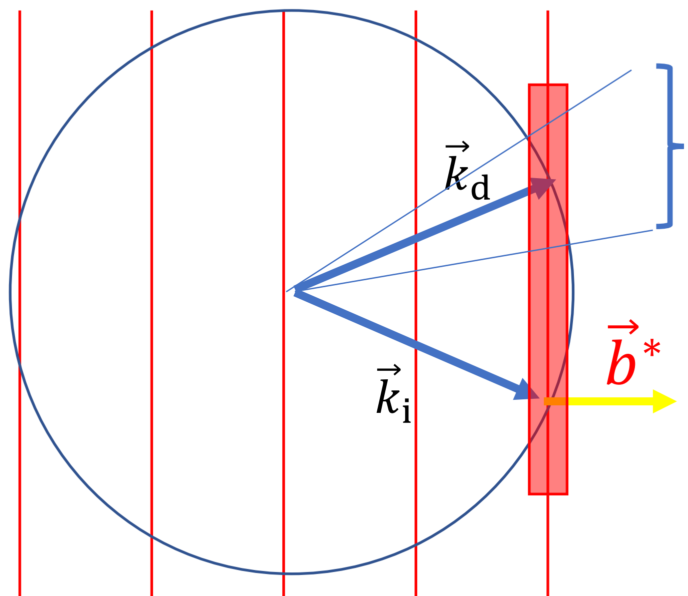
Diffraction pattern and reciprocal space



Why diffraction streaks?

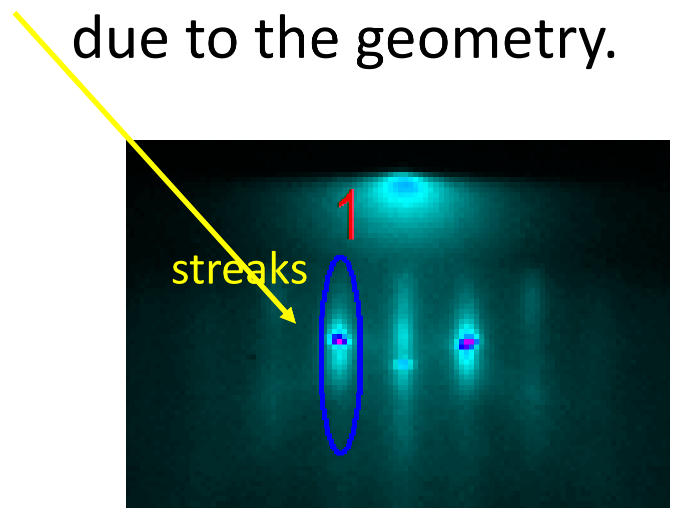
Patches on the surface broadens the reciprocal rods.



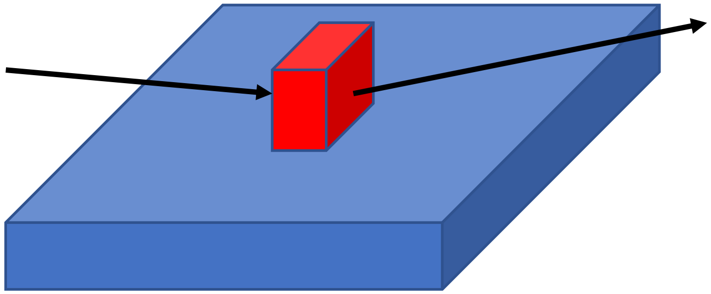


Side view

Broadening amplified in the vertical direction due to the geometry.

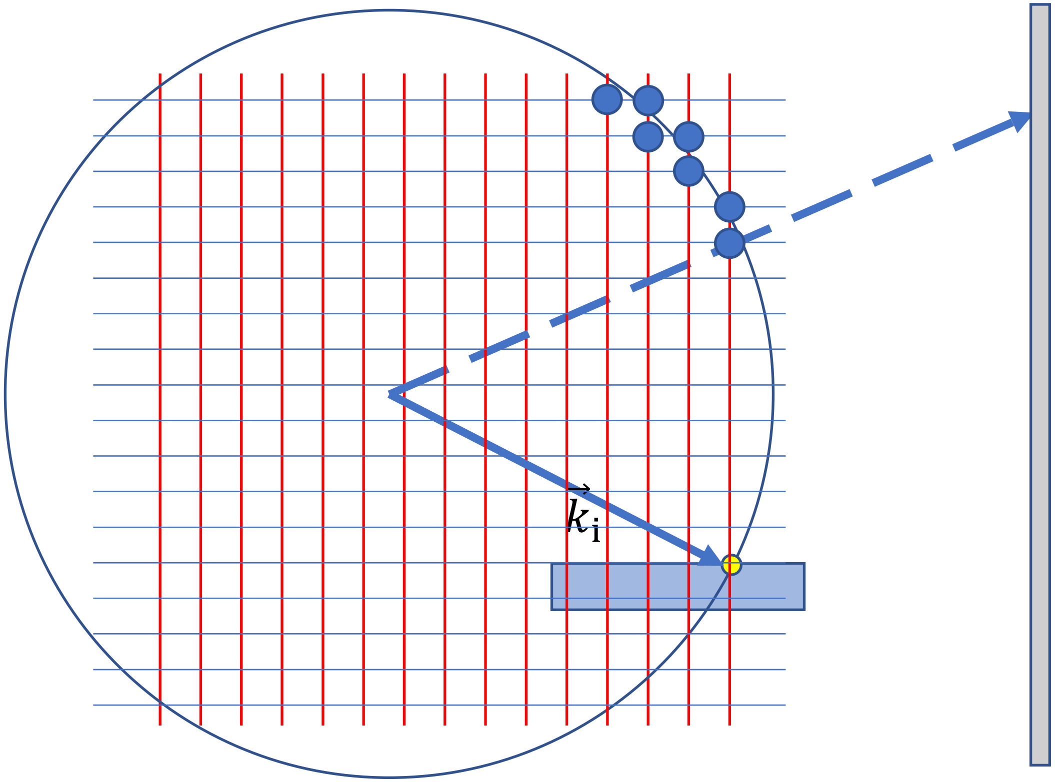


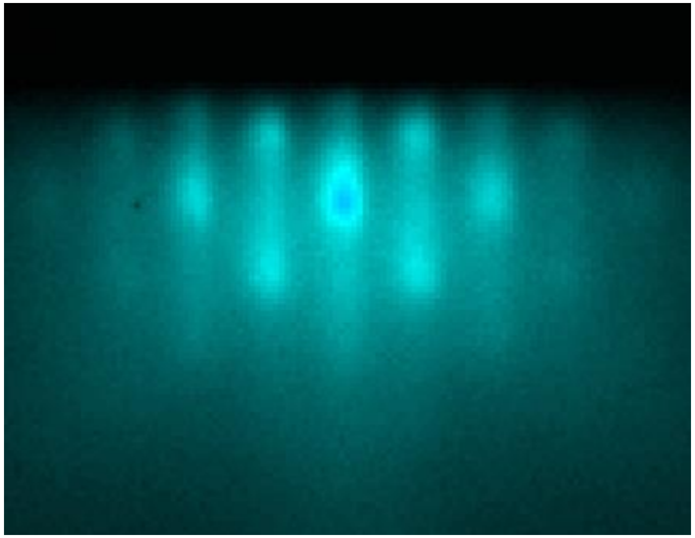
What about islands?



- The horizontal dimension is not too large (<100 nm).
- The vertical dimension is not too small (> 5 nm).

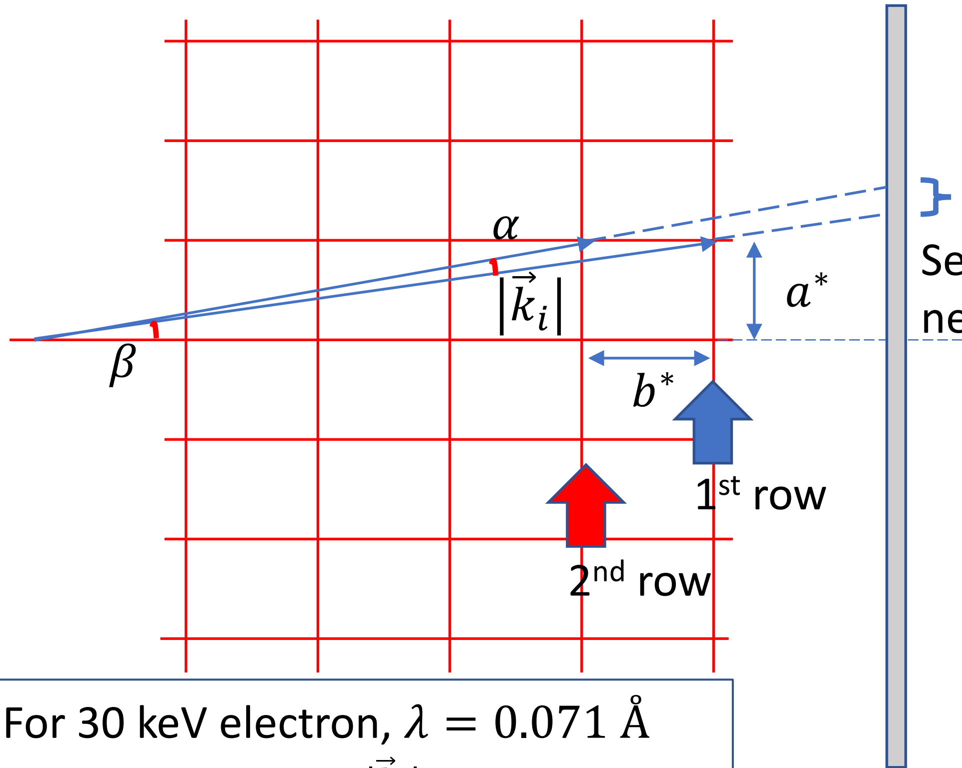
Side view

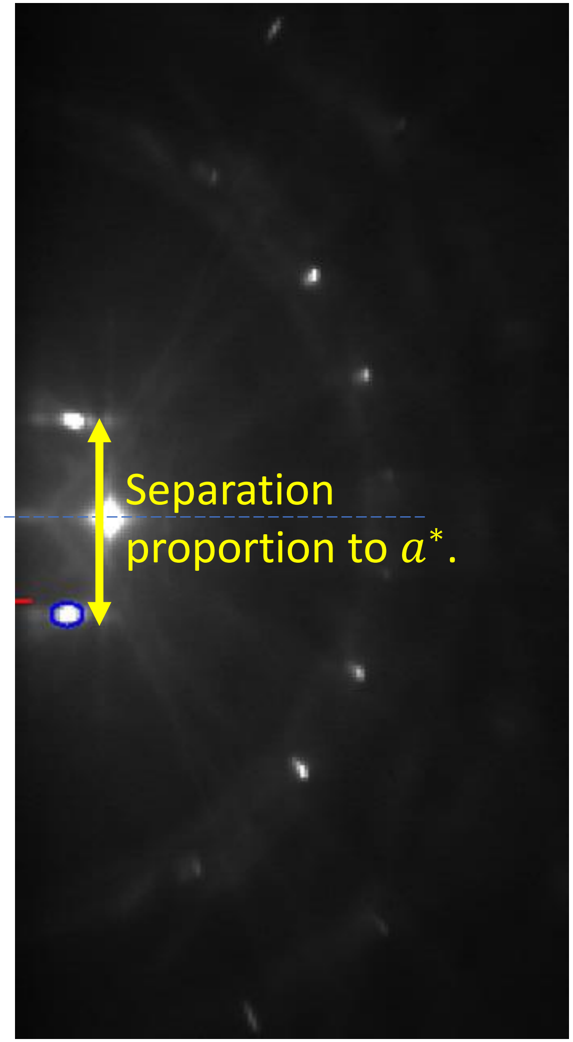




Surface structure analysis

$$\beta = \arcsin\left(\frac{a^*}{|\vec{k}_i|}\right), \alpha = \arctan\left[\frac{a^*}{\cos(\beta)|\vec{k}_i| - b^*}\right] - \beta$$





For 30 keV electron, $\lambda = 0.071$ Å

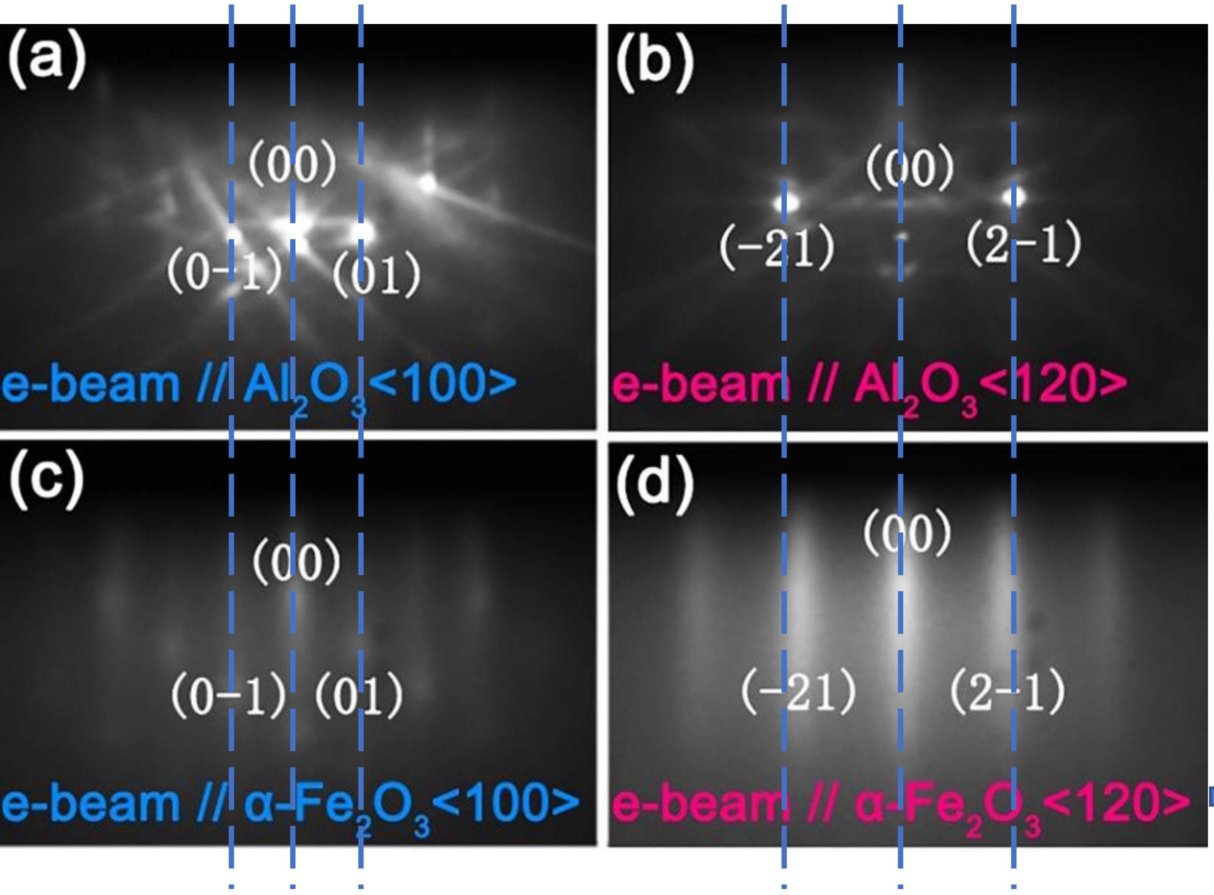
If we take $a \approx 4$ Å, $\frac{|\vec{k}_\mathrm{i}|}{\mathrm{a}^*} = 56$

$\beta \approx 0.018$ rad

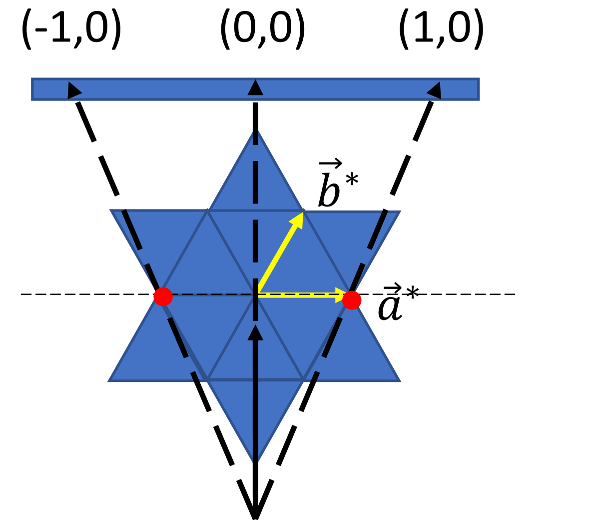
$\alpha \approx 0.00032$ rad

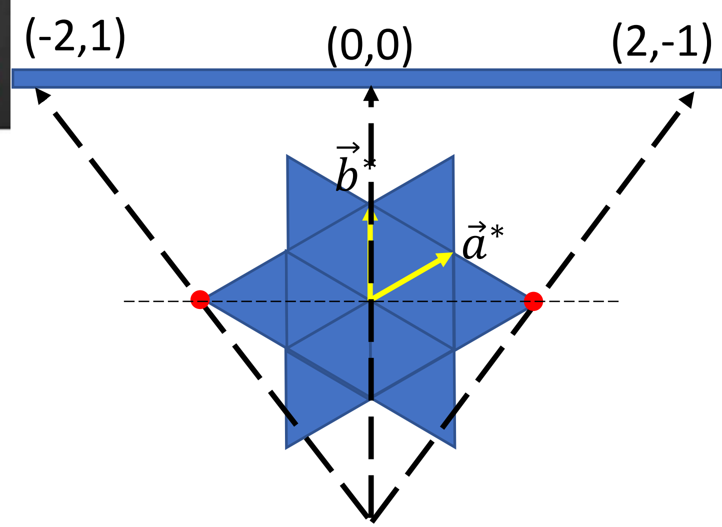
$$\frac{\alpha}{\beta} = 0.018$$

Surface structure analysis



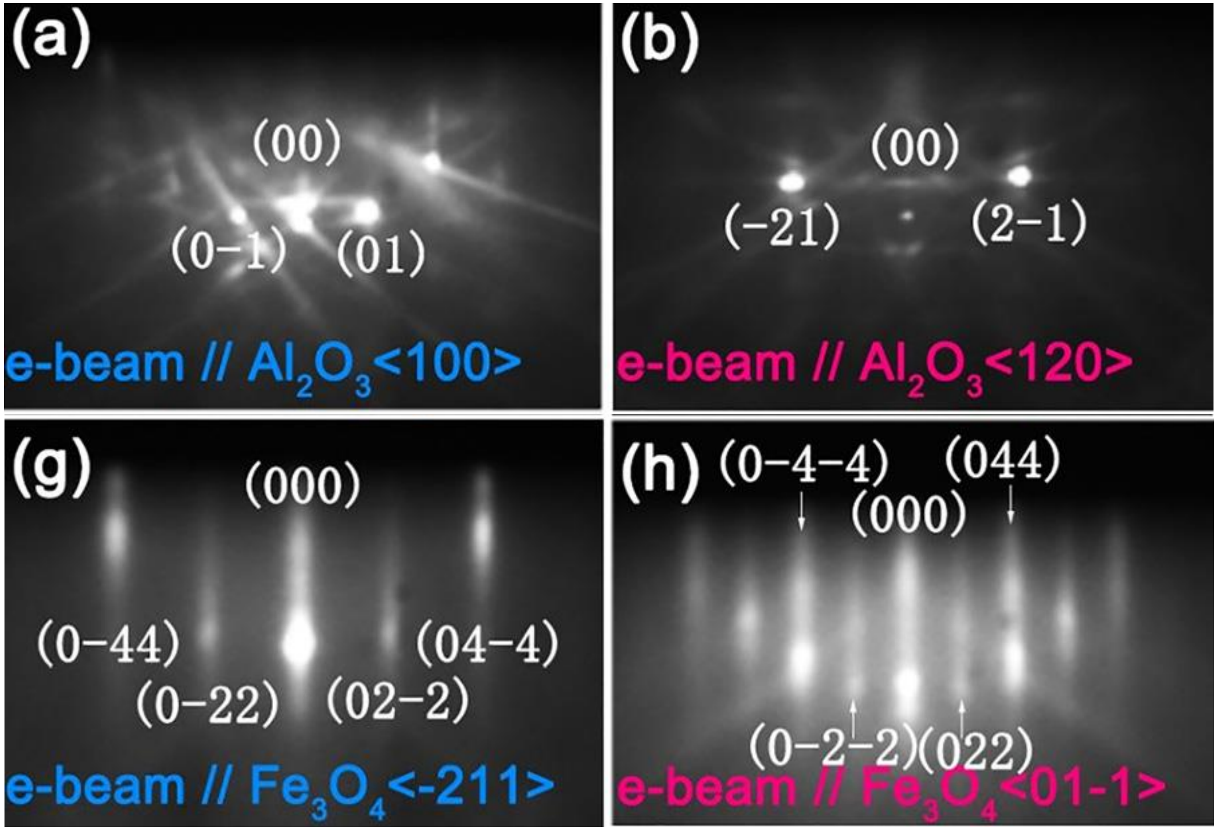
Al_2O_3 triangular lattice

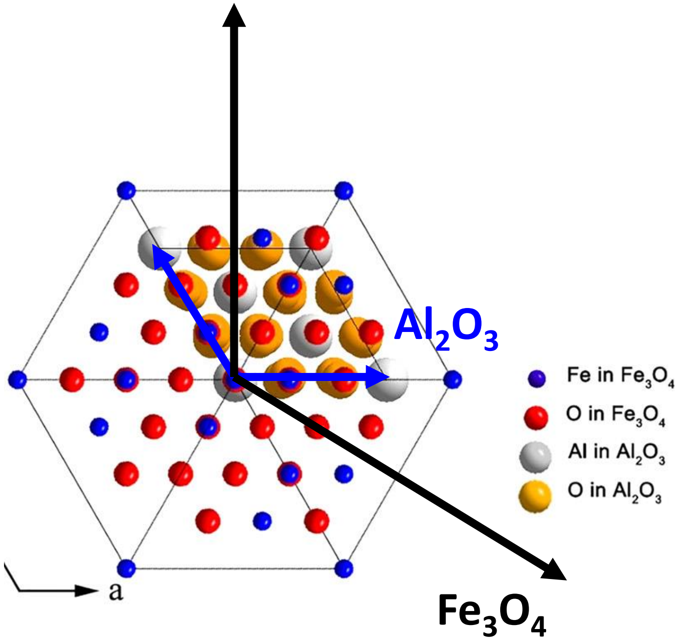




Al_2O_3 and Fe_2O_3 have similar triangular lattice.
The lattice constant of Al_2O_3 is slightly smaller, which is consistent with the observation.

Epitaxial relation analysis





Al_2O_3 (001) and Fe_3O_4 (111) both have triangular lattice.

From the RHEED pattern, the basis of the two lattices are rotate by 30 (or 90) degree.

Conclusion

- Basic kinematic diffraction theory is reviewed.
- Two dimensional diffraction geometry for RHEED is discussed
- Analysis of surface morphology, structure, and epitaxial relation is introduced.