Data Processing for Neutron Total Scattering

– From Theory to Practice

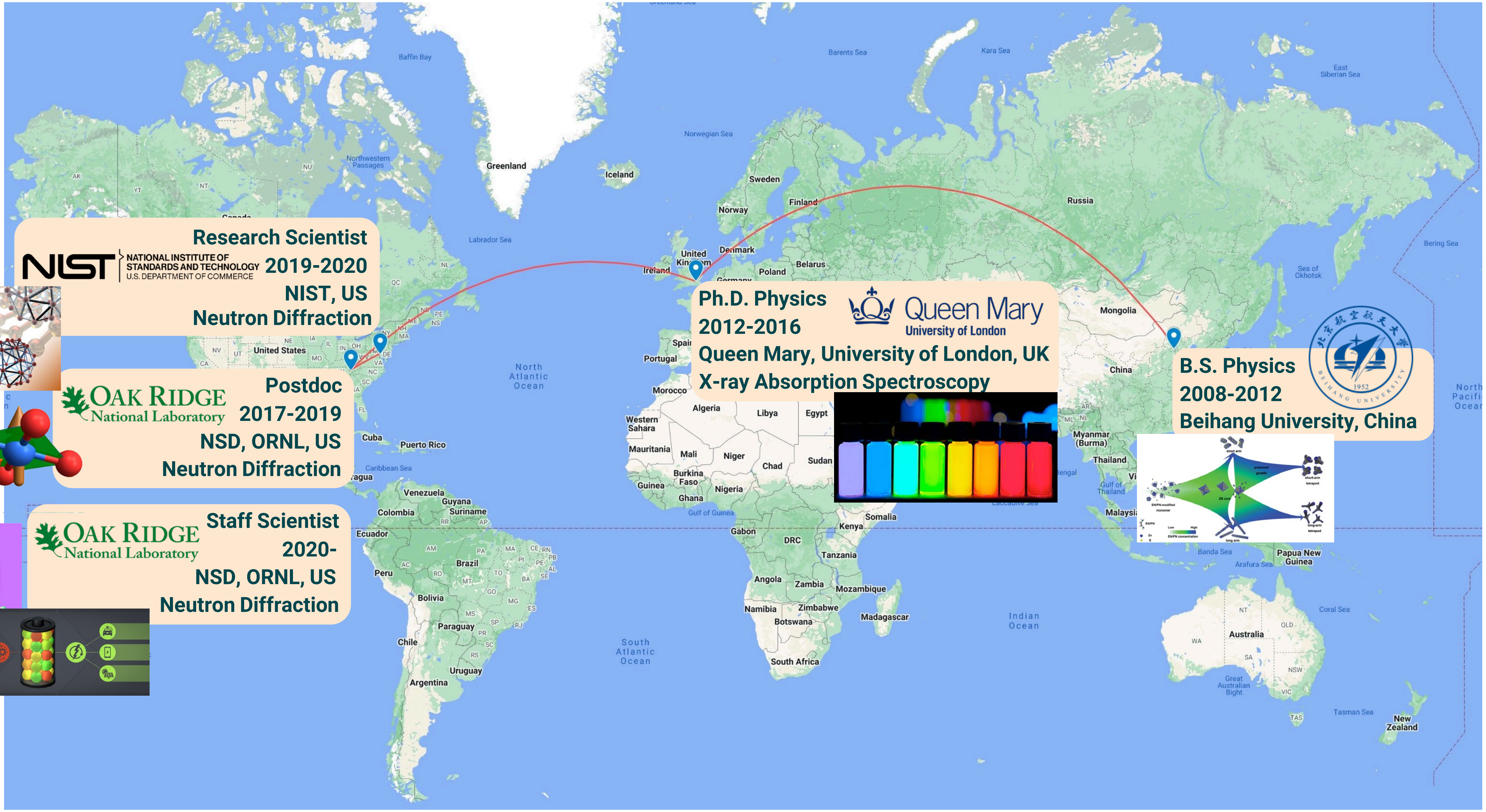
Dr. Yuanpeng Zhang

Neutron Scattering Division

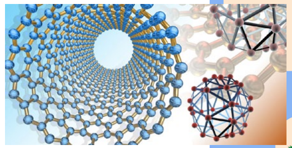
Apr-03-2025

Guest Lecture for MSE 405/572
Invited by Prof. Katharine Page

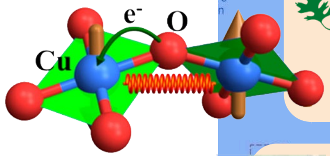
My Career Path



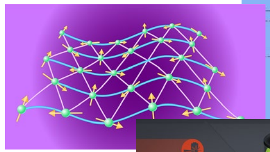
Research Scientist
NIST | NATIONAL INSTITUTE OF STANDARDS AND TECHNOLOGY
U.S. DEPARTMENT OF COMMERCE
2019-2020
NIST, US
Neutron Diffraction



Postdoc
OAK RIDGE National Laboratory
2017-2019
NSD, ORNL, US
Neutron Diffraction



Staff Scientist
OAK RIDGE National Laboratory
2020-
NSD, ORNL, US
Neutron Diffraction



Ph.D. Physics
2012-2016
Queen Mary University of London
Queen Mary, University of London, UK
X-ray Absorption Spectroscopy

B.S. Physics
2008-2012
Beihang University, China

OAK RIDGE National Laboratory

My Profile

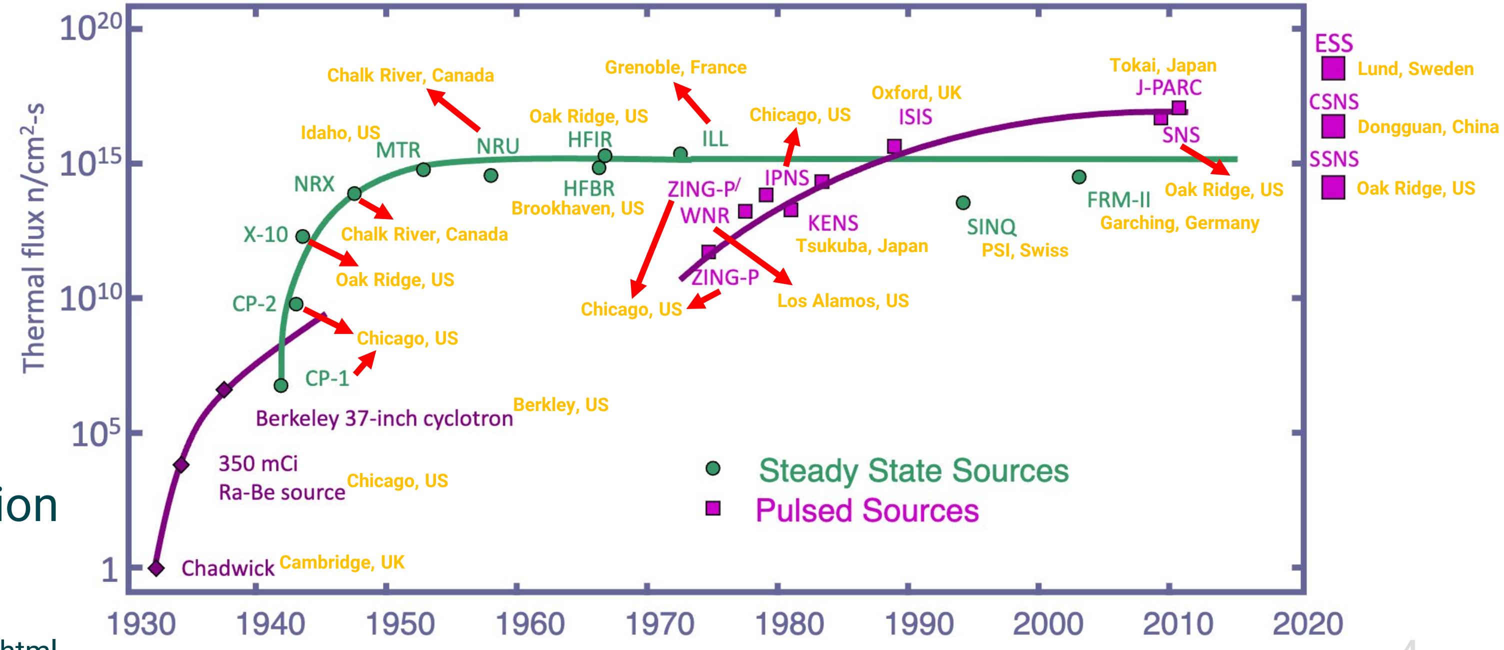
- ORNL Profile, https://www.ornl.gov/staff-profile/yuanpeng-zhang
- My homepage, https://me.iris-home.net
- My blog, https://iris2020.net
- My GitHub profile, https://github.com/Kvieta1990
- Email: zhangy3@ornl.gov, zyroc1990@gmail.com

Neutrons

J. Chadwick
(1891-1974)

- One of the elementary particles
- Discovered in 1932 by J. Chadwick
- Utilized for scattering experiments, starting in 1940s, by C. Shull, E. Wollan, etc.
- Reactor based source for experiments operational in 1950s
- Spallation based source operational in 1970s

C. Shull (left) and E. Wollan (right)



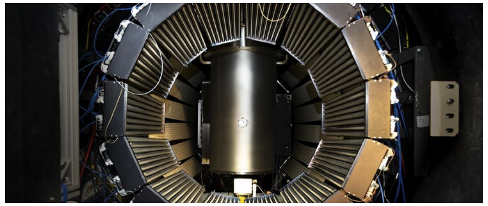
Neutron Sources Evolution

[1] K. Skold, et al. Neutron Scattering, 1986.
[2] https://powder.ornl.gov/general_aspects/nsources.html

Neutron Powder Diffraction Instruments at ORNL

Spallation

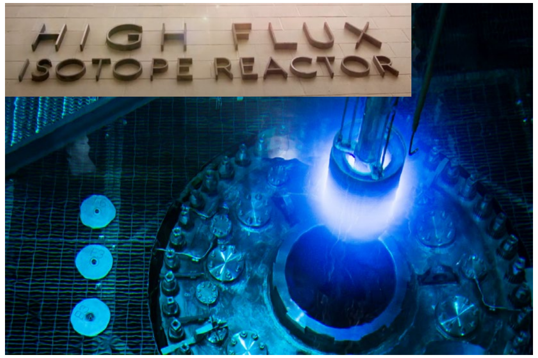
SNS
SPALLATION NEUTRON SOURCE



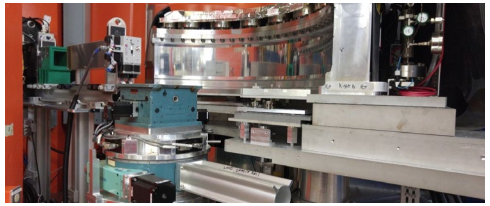
NOMAD

POWGEN

Reactor



HB-2A



HB-2C

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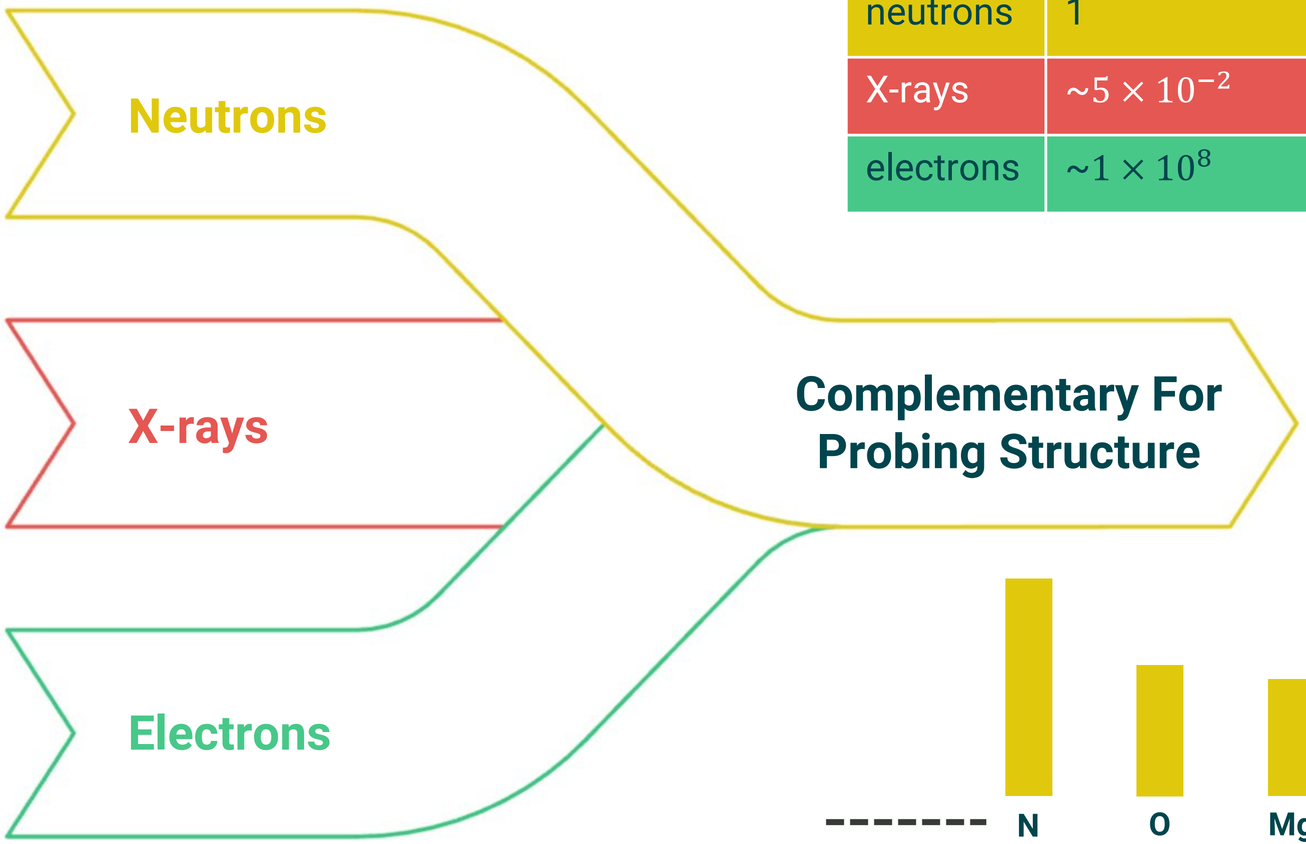
https://neutrons.ornl.gov/sns
https://neutrons.ornl.gov/hfir

X-rays, Neutrons and Electrons are Complementary

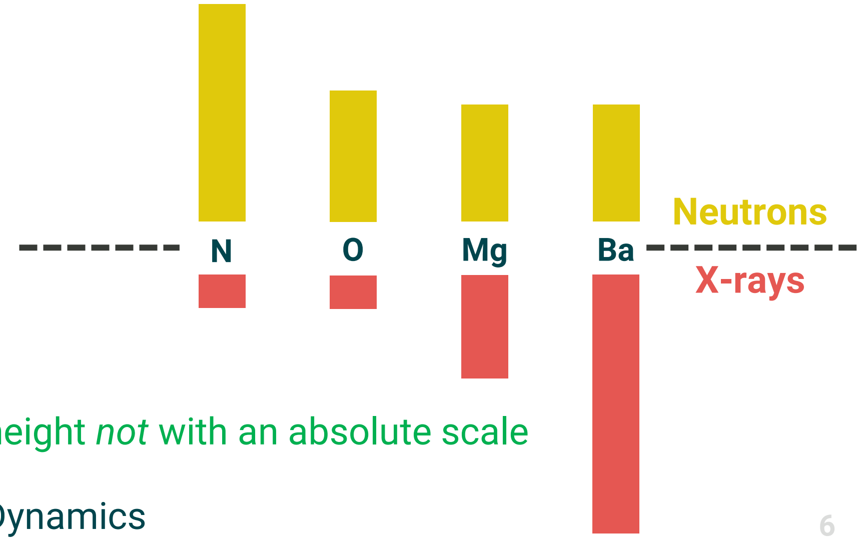
- No intrinsic dampening vs. Q
- Carry magnetic moment

- Flux high
- Strong interaction

- Flux high
- Super strong interaction



	Cross-section [barn/particle]	Flux $[s^{-1}cm^{-2}BW^{-1}]$
neutrons	1	10^7
X-rays	$\sim 5 \times 10^{-2}$	10^{12}
electrons	$\sim 1 \times 10^{8}$	10^{10}



Bar height *not* with an absolute scale

Outline

- ❖ Brief recovery of basic crystallography
- ❖ From Bragg diffraction to Total Scattering
- ❖ Link experiment to theory
- ❖ Ingredients of Data Processing

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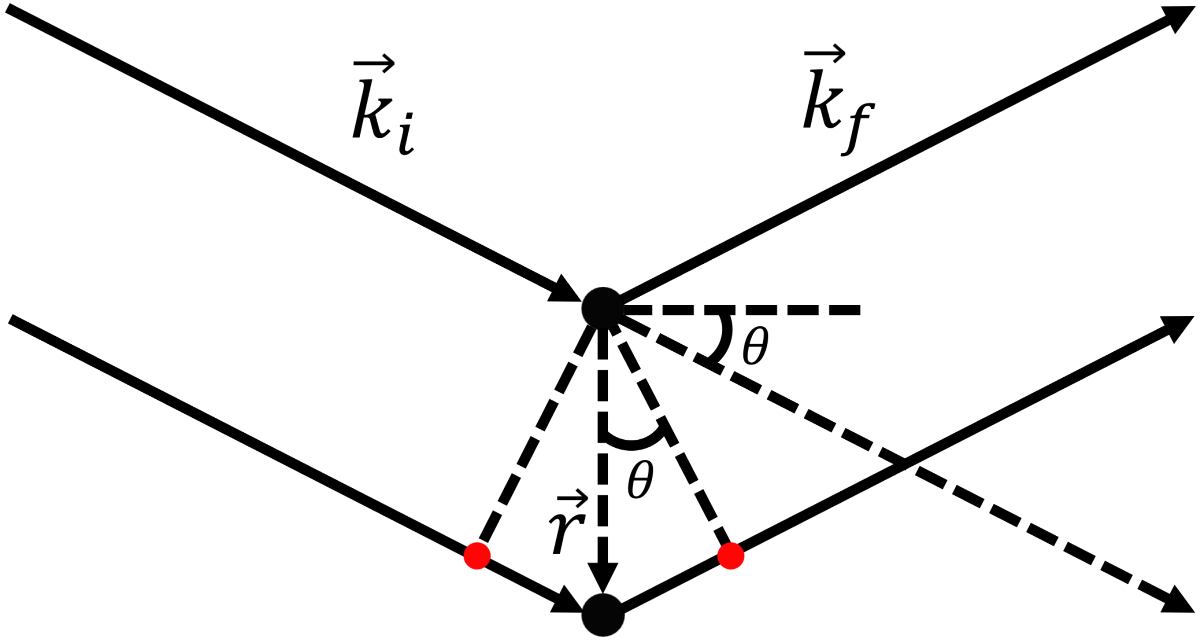
Brief recovery of basic crystallography

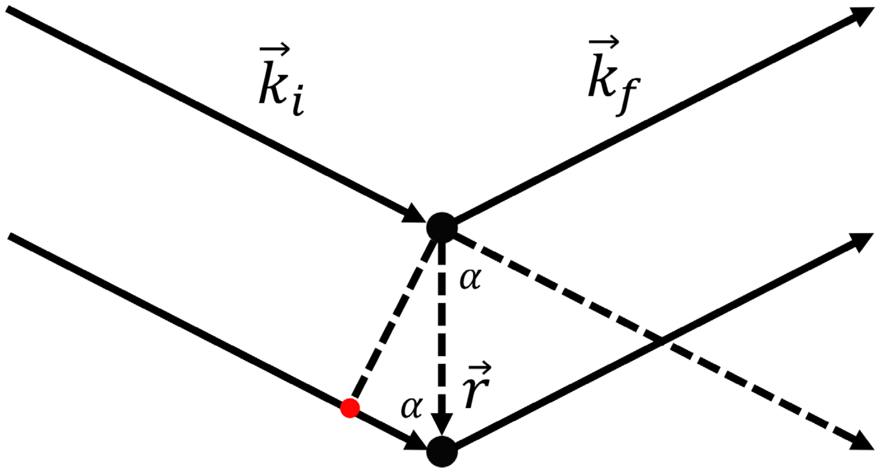
$$k = \frac{2\pi}{\lambda}$$

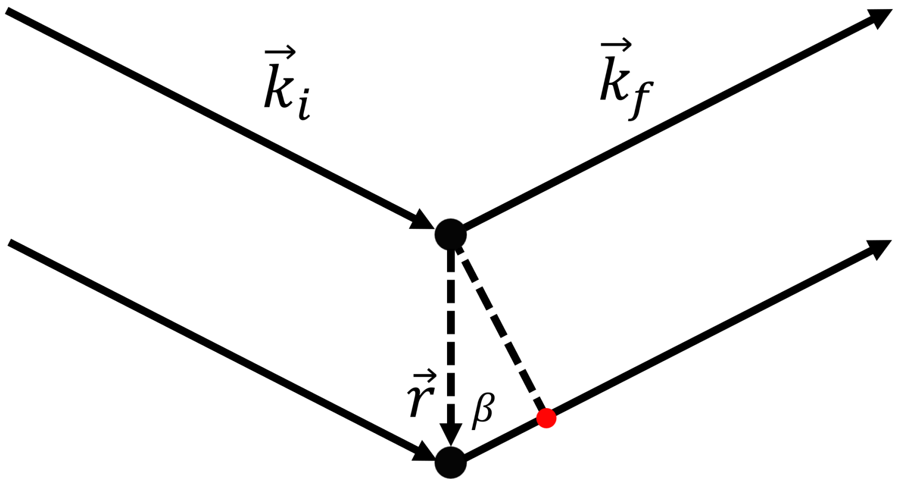


$$\frac{2|\vec{r}|sin\theta}{\lambda} \times 2\pi = 2n\pi \Rightarrow 2|\vec{r}|sin\theta = n\lambda$$

Brief recovery of basic crystallography

$$k = \frac{2\pi}{\lambda}$$



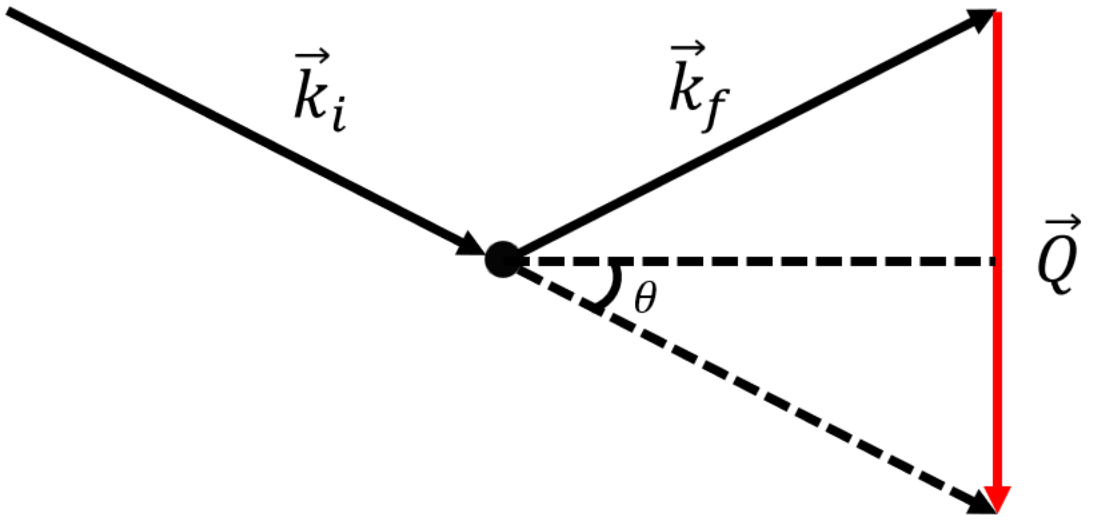


$$\frac{|\vec{r}|cos\alpha}{\lambda} \times 2\pi = \frac{2\pi}{\lambda}|\vec{r}|cos\alpha = \vec{k}_i \cdot \vec{r}$$

$$\frac{|\vec{r}|cos\beta}{\lambda} \times 2\pi = -\frac{2\pi}{\lambda}|\vec{r}|\cos(180-\beta) = -\vec{k}_f \cdot \vec{r}$$

$$\left(\vec{k}_i - \vec{k}_f\right) \cdot \vec{r}$$

Brief recovery of basic crystallography



$\Rightarrow \vec{Q} = \vec{k}_i - \vec{k}_f \Rightarrow$ Momentum Transfer

$\Rightarrow 1 + e^{i\vec{Q}\cdot\vec{r}} \Rightarrow$ Modulation over the plain wave, giving rise to diffraction

$$\Rightarrow F(\vec{Q}) = \sum_{n=0}^{n=N} e^{i\vec{Q}\cdot\vec{r}}$$

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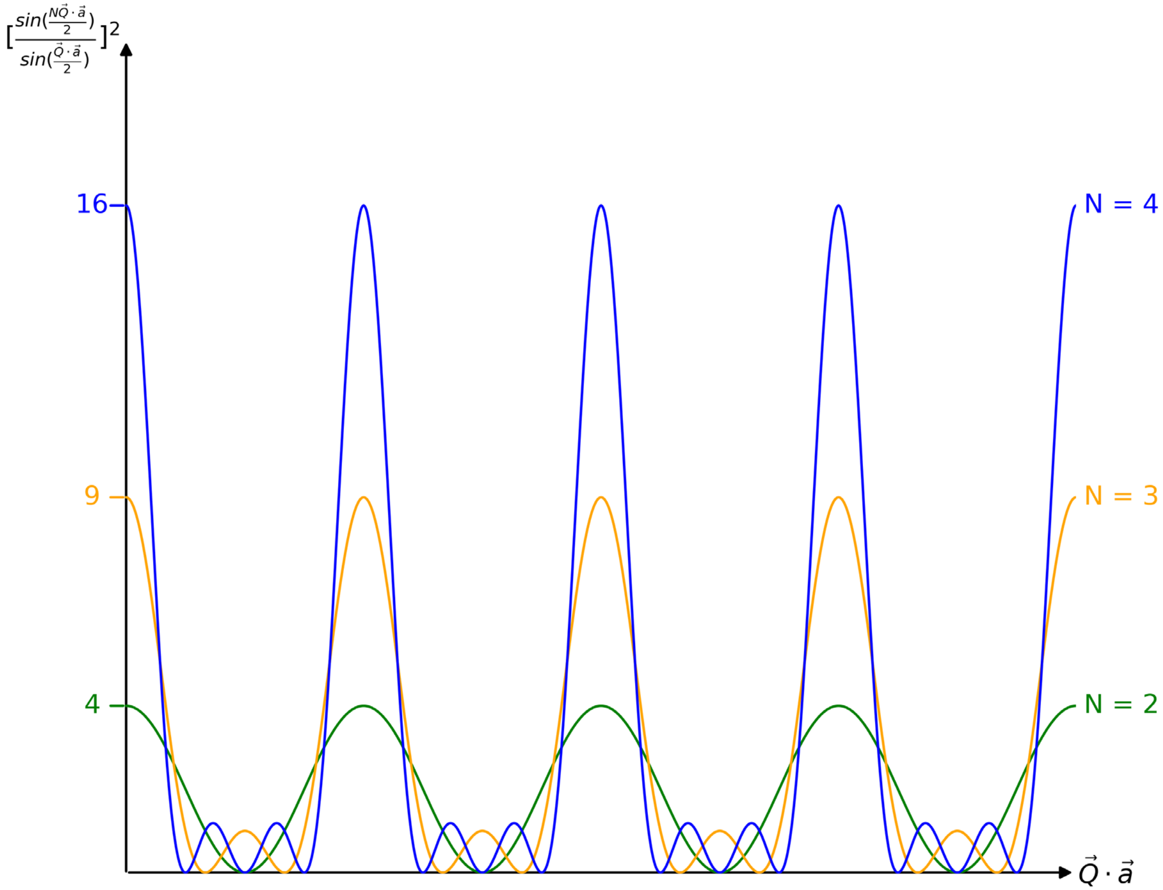
- ❖ Ingredients of Data Processing

From Bragg diffraction to Total Scattering – A simple 1D example

$$I(\vec{Q}) = |F(\vec{Q})|^2 = \left|\sum_{n=0}^{n=N} \exp(i\vec{Q}\cdot\vec{r}_n)\right|^2$$

$$= \left|\sum_{n=0}^{n=N} \exp[i\vec{Q}\cdot(\vec{r}_0 + n\vec{a})]\right|^2$$

$$= \left[\frac{\sin(\frac{N\vec{Q}\cdot\vec{a}}{2})}{\sin(\frac{\vec{Q}\cdot\vec{a}}{2})}\right]^2$$

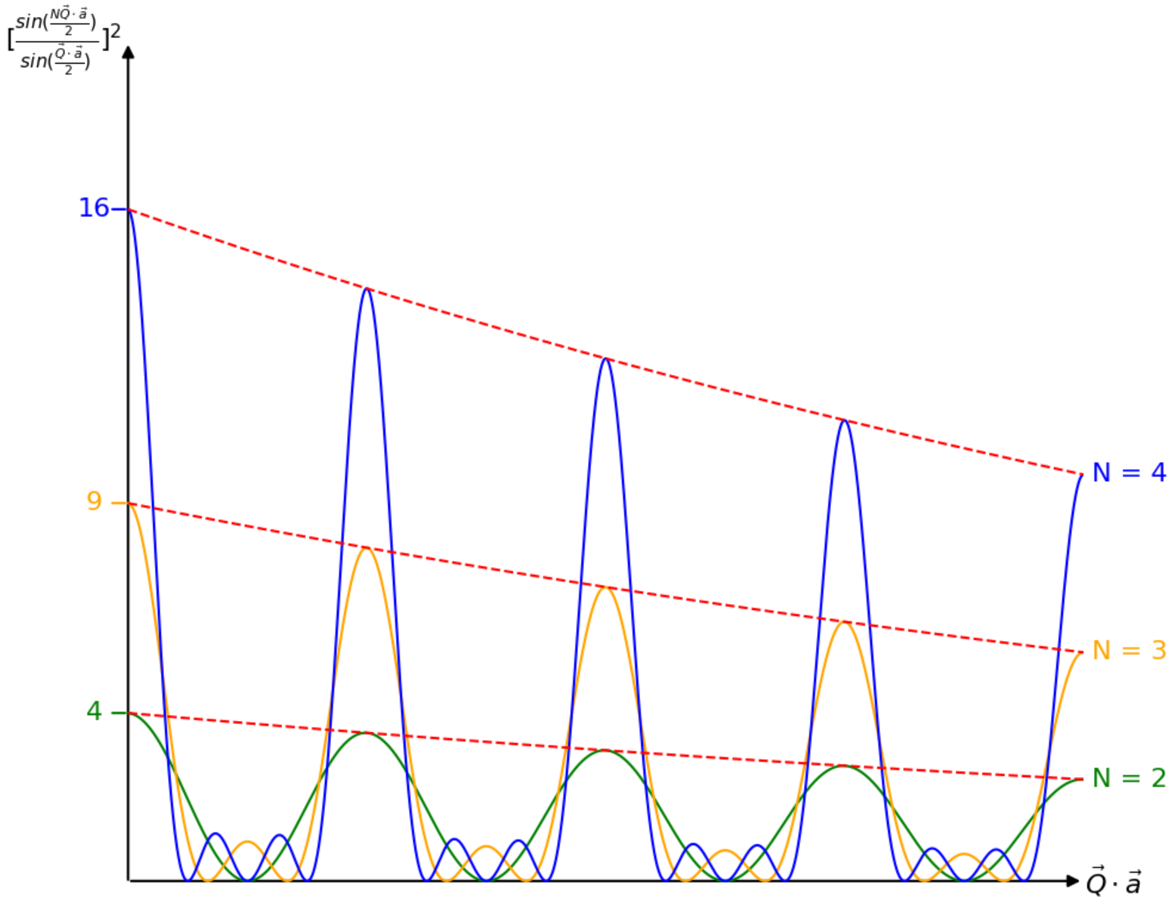


From Bragg diffraction to Total Scattering – A simple 1D example

$$I(\vec{Q}) = \left| \sum_{n=0}^{n=N} \exp[i\vec{Q} \cdot (\vec{r}_0 + n\vec{a} + \vec{u}_n)] \right|^2$$

$\Downarrow$ Average out $\vec{u}_n$

$$= \left[\frac{\sin(\frac{N\vec{Q} \cdot \vec{a}}{2})}{\sin(\frac{\vec{Q} \cdot \vec{a}}{2})} \right]^2 \exp[-\frac{1}{3} Q^2 \langle u^2 \rangle]$$



https://yr.iris-home.net/dataproc

From Bragg diffraction to Total Scattering – A simple 1D example

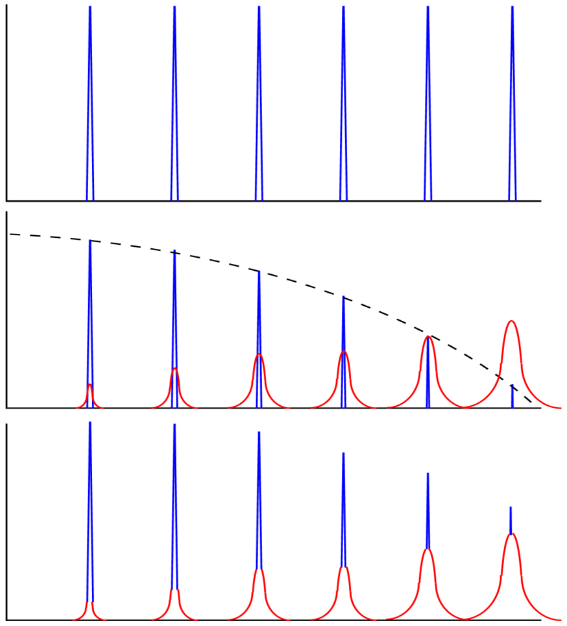
By averaging out the distortions, we lose the variation of local distortions from one place to another

$$I(\vec{Q}) = \left| \sum_{n=0}^{n=N} \exp[i\vec{Q} \cdot (\vec{r}_0 + n\vec{a} + \vec{u}_n)] \right|^2$$

$\Downarrow$ Expand explicitly

$$I_{0,1}(\vec{Q}) + \cdots = 2 + 2\cos[\vec{Q} \cdot \vec{a} + \vec{Q} \cdot (\vec{u}_1 - \vec{u}_0)] + \cdots$$

origin of diffuse scattering



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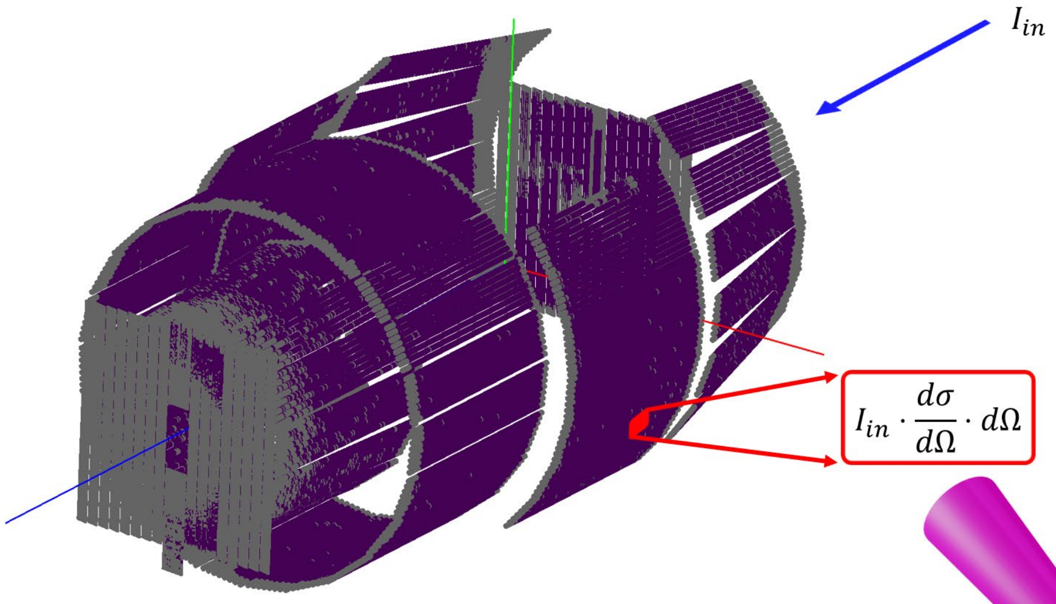
Link experiment to theory

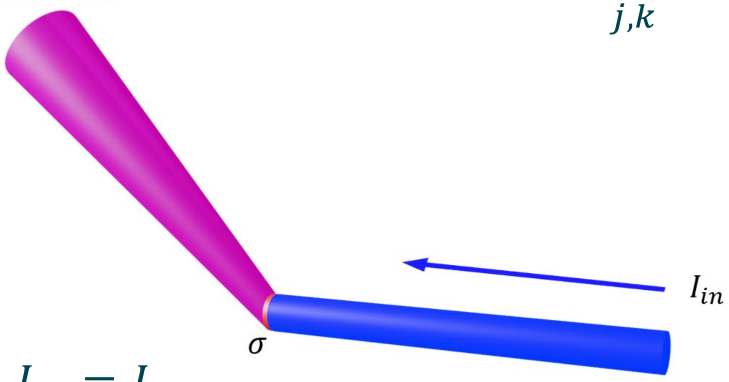


NOMAD Diffractometer
Instrument View

$$d\sigma = \frac{W_{\vec{k}_i \to \vec{k}_f}}{\text{incident flux}}$$

$$\Downarrow$$

$$\frac{d\sigma}{d\Omega} = \sum_{j,k} b_j b_k \exp[-i\vec{Q} \cdot (\vec{R}_j - \vec{R}_k)]$$



$$\sigma \cdot I_{in} = I_{scattered}$$

[1] W. Marshall and S. W. Lovesey, *Theory of thermal neutron scattering*
[2] J. J. Sakurai, *Modern Quantum Mechanics*
[3] https://yr.iris-home.net/dataproc

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Link experiment to theory

Theory

$$d\sigma = \frac{W_{\vec{k}_i \to \vec{k}_f}}{\text{incident flux}}$$

$$\Downarrow$$

$$\frac{d\sigma}{d\Omega} = \sum_{j,k} b_j b_k \exp[-i\vec{Q} \cdot (\vec{R}_j - \vec{R}_k)]$$

Experiment

$$I_s = I_{in} \cdot \frac{d\sigma}{d\Omega} \cdot d\Omega \Rightarrow d\sigma = \frac{I_s}{I_{in}}$$

$$\Downarrow$$

$$\frac{d\sigma}{d\Omega} = \frac{1}{\Phi_x} \frac{1}{d\Omega} \frac{I_s^e}{I_{in}^e} = \Phi I_s^{norm}$$

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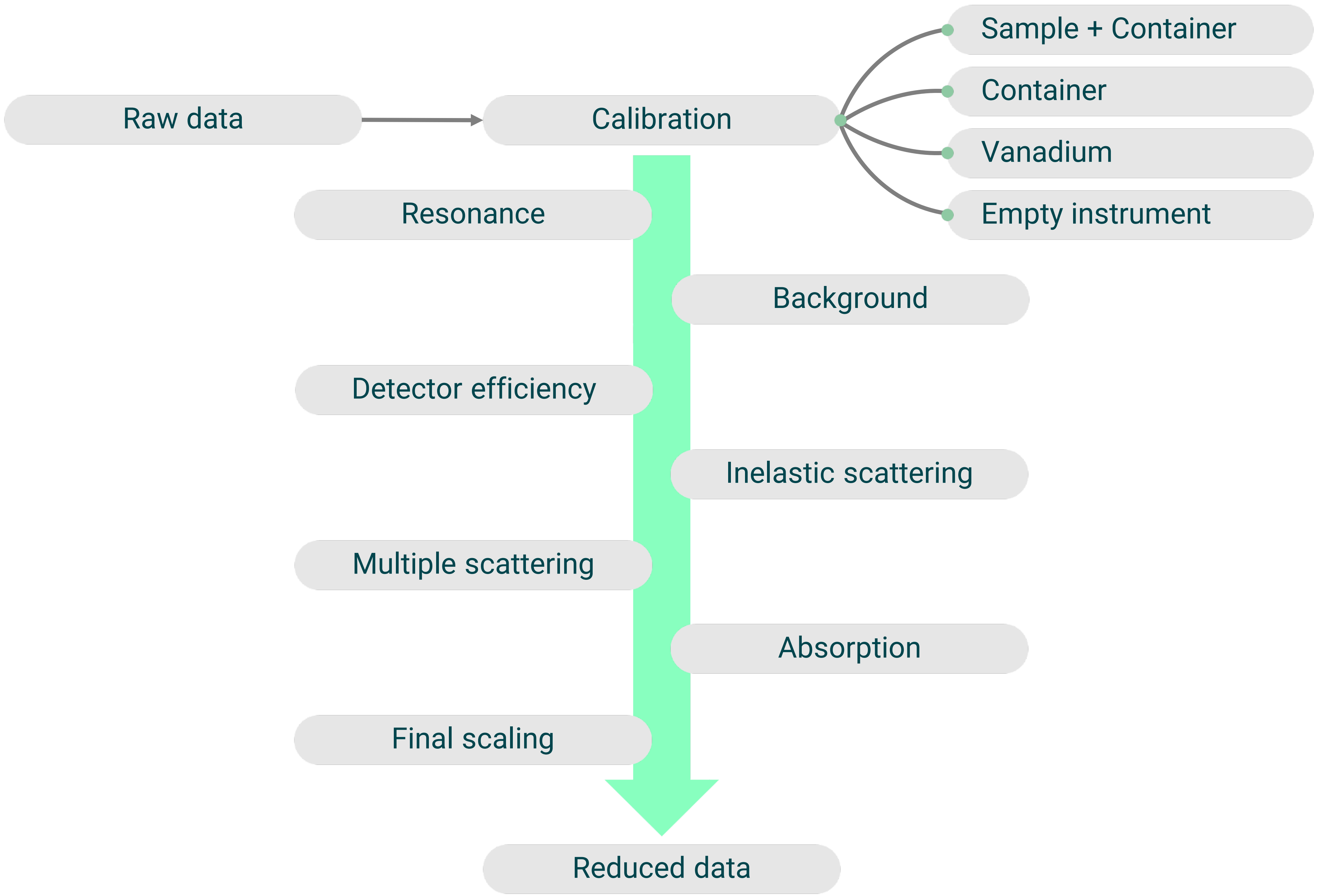
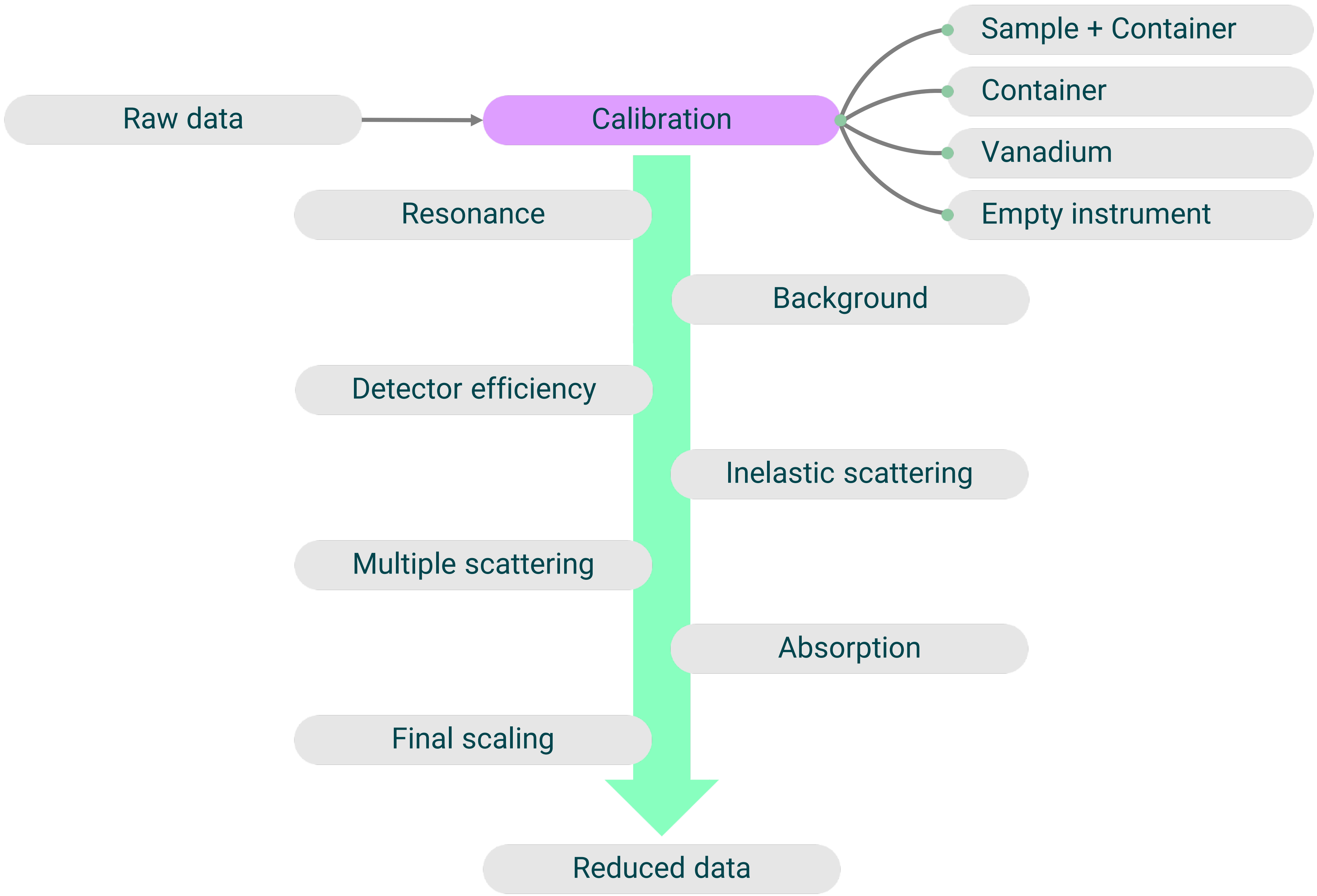
Ingredients of Data Processing

- Calibration
- Background subtraction & Normalization
- Absorption & Multiple Scattering
- Resonant absorption
- Inelastic scattering effect

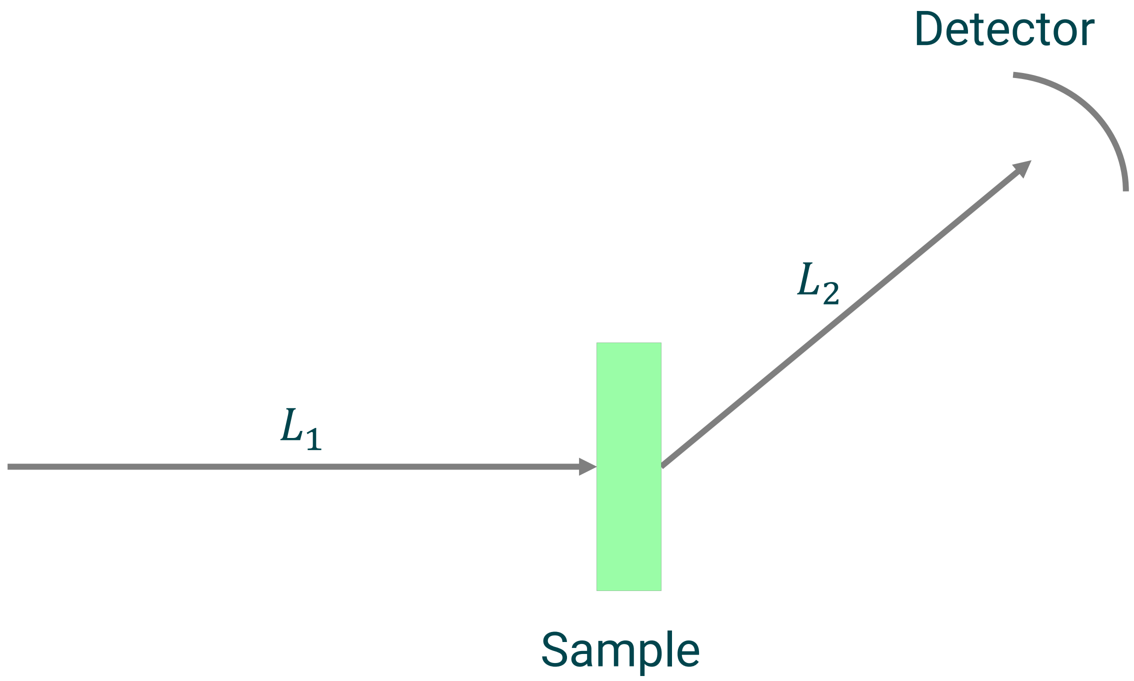
Ingredients of Data Processing



Ingredients of Data Processing



Ingredients of Data Processing – Calibration

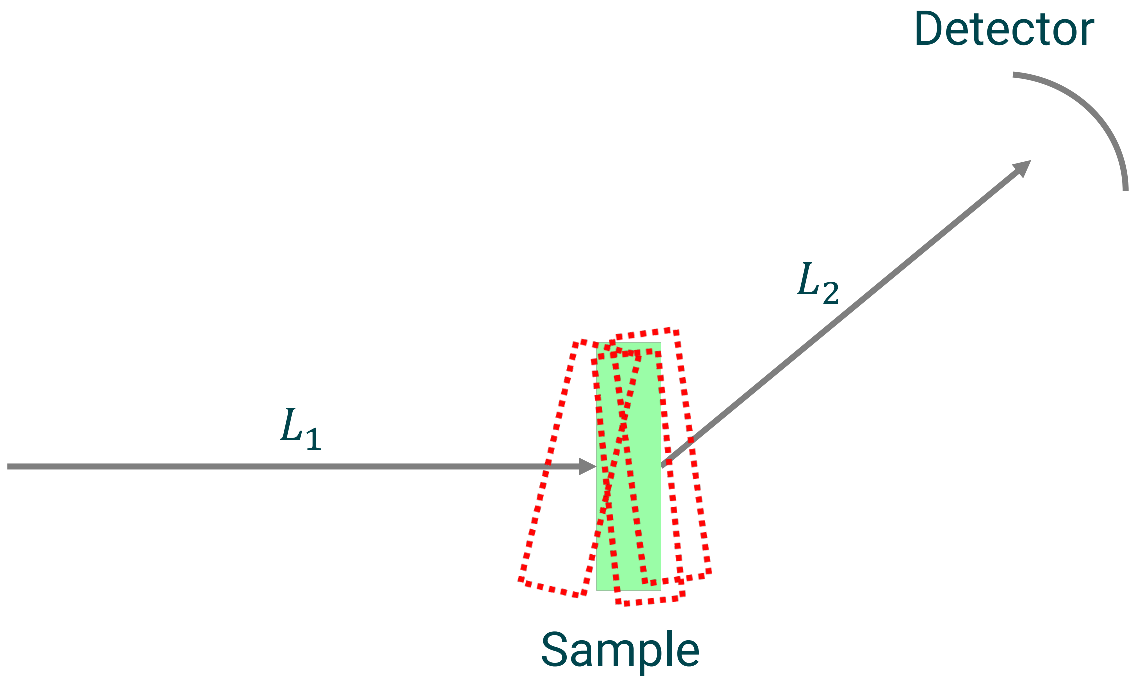


$$v = \frac{L_1 + L_2}{TOF}$$

$$\lambda = \frac{h}{p} = \frac{h}{mv} = \frac{h}{m} \cdot \frac{TOF}{L_1 + L_2}$$

$$2dsin\theta = \lambda$$

Ingredients of Data Processing – Calibration



‘unknown’

$$v = \frac{L_1 + L_2}{TOF}$$

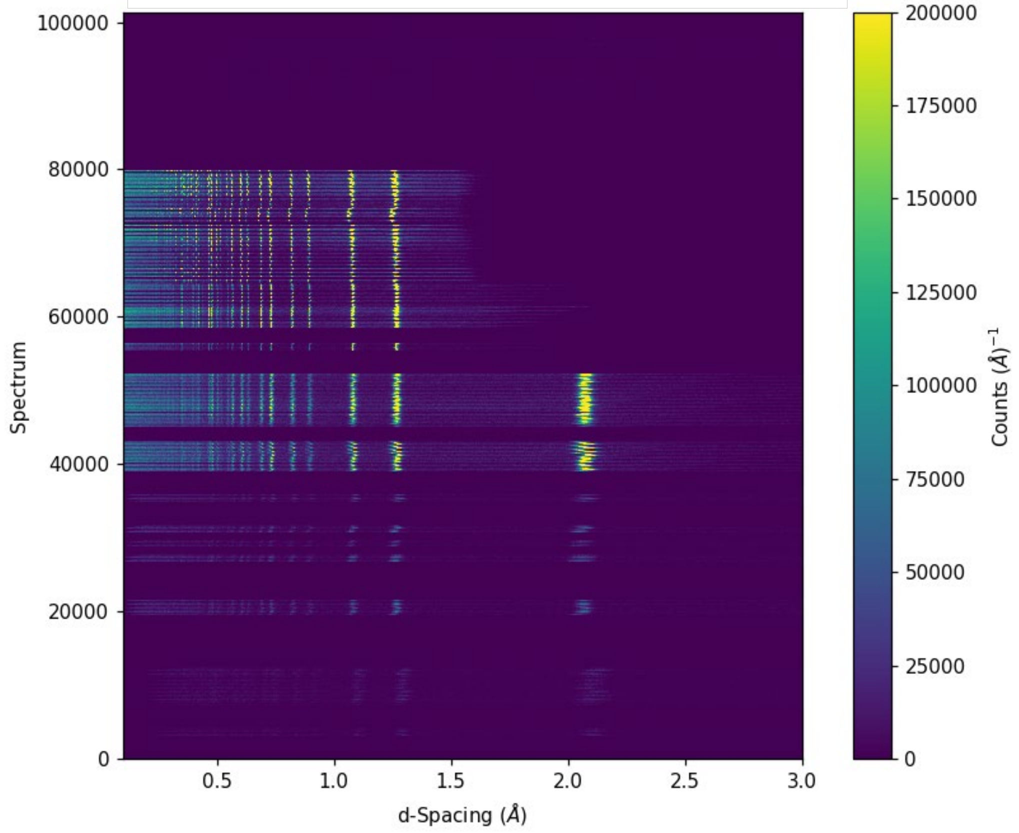
$$\lambda = \frac{h}{p} = \frac{h}{mv} = \frac{h}{m} \cdot \frac{TOF}{L_1 + L_2}$$

$$2d sin\theta = \lambda$$

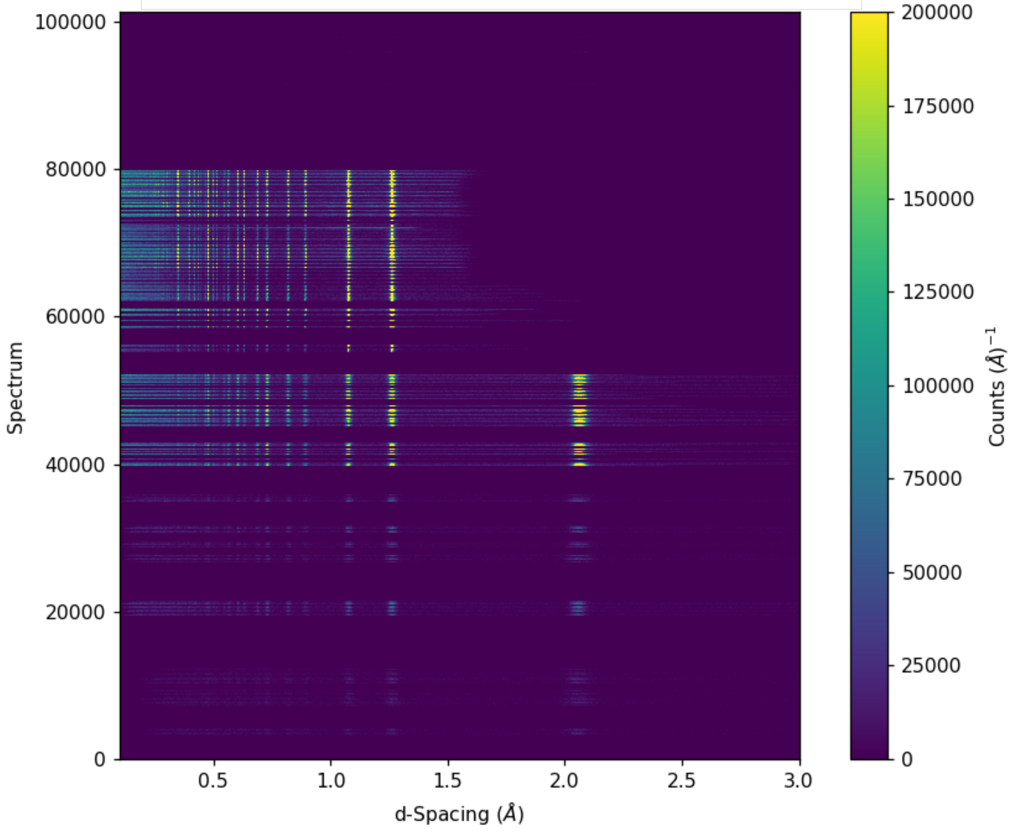
‘unknown’ ‘unknown’

Ingredients of Data Processing – Calibration

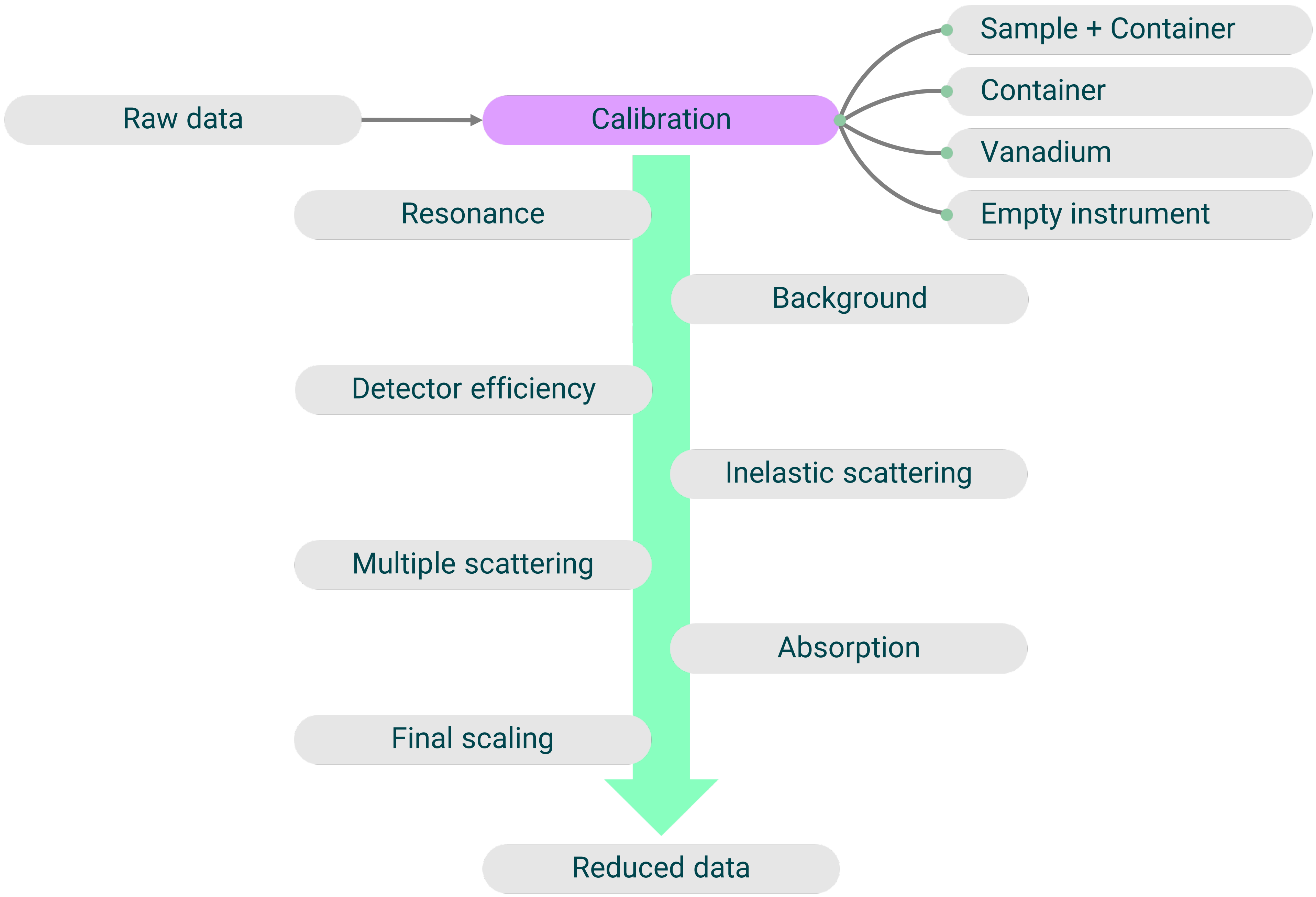
Engineering alignment



Calibrated alignment



Ingredients of Data Processing – Bkg & Normalization



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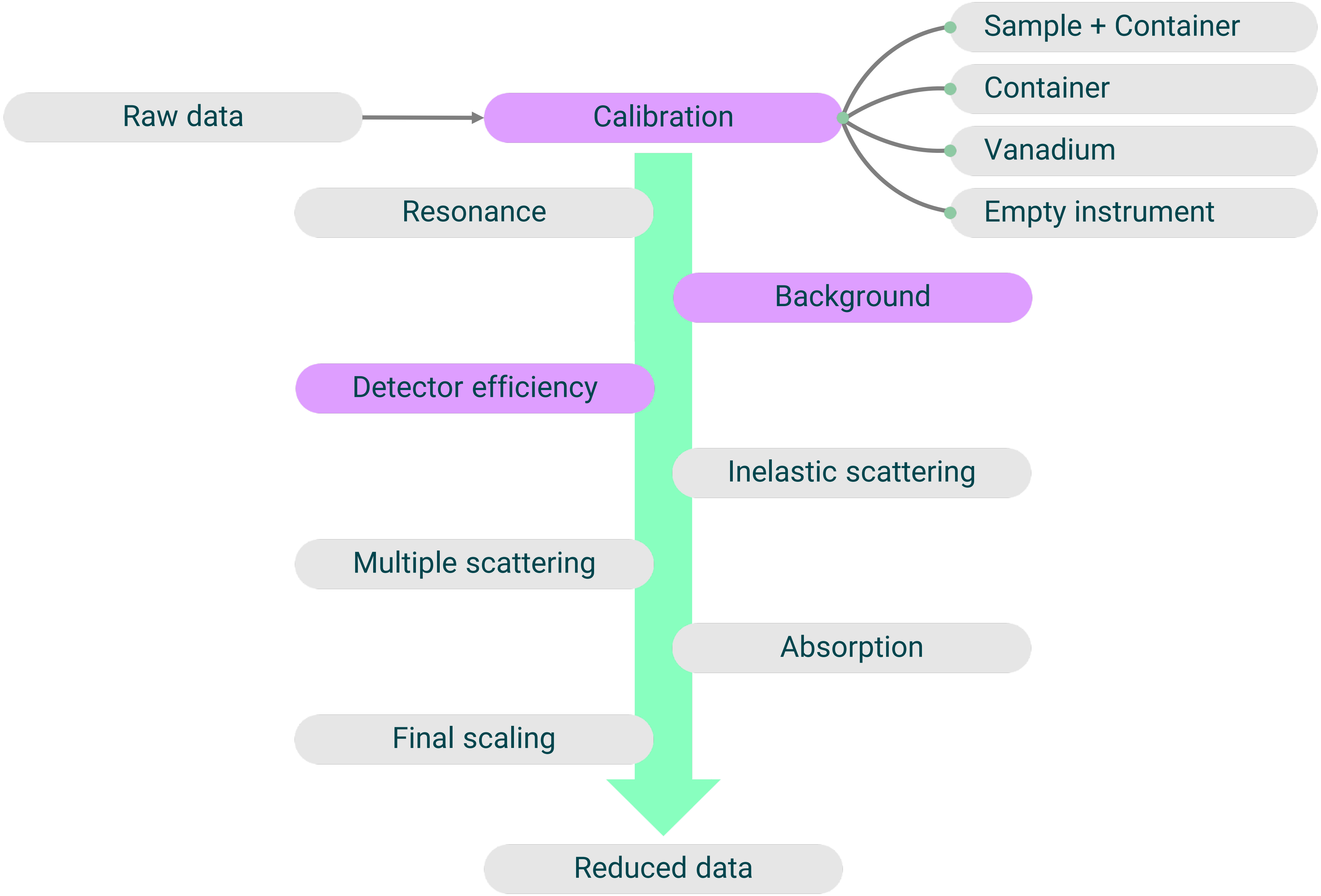
Ingredients of Data Processing – Bkg & Normalization



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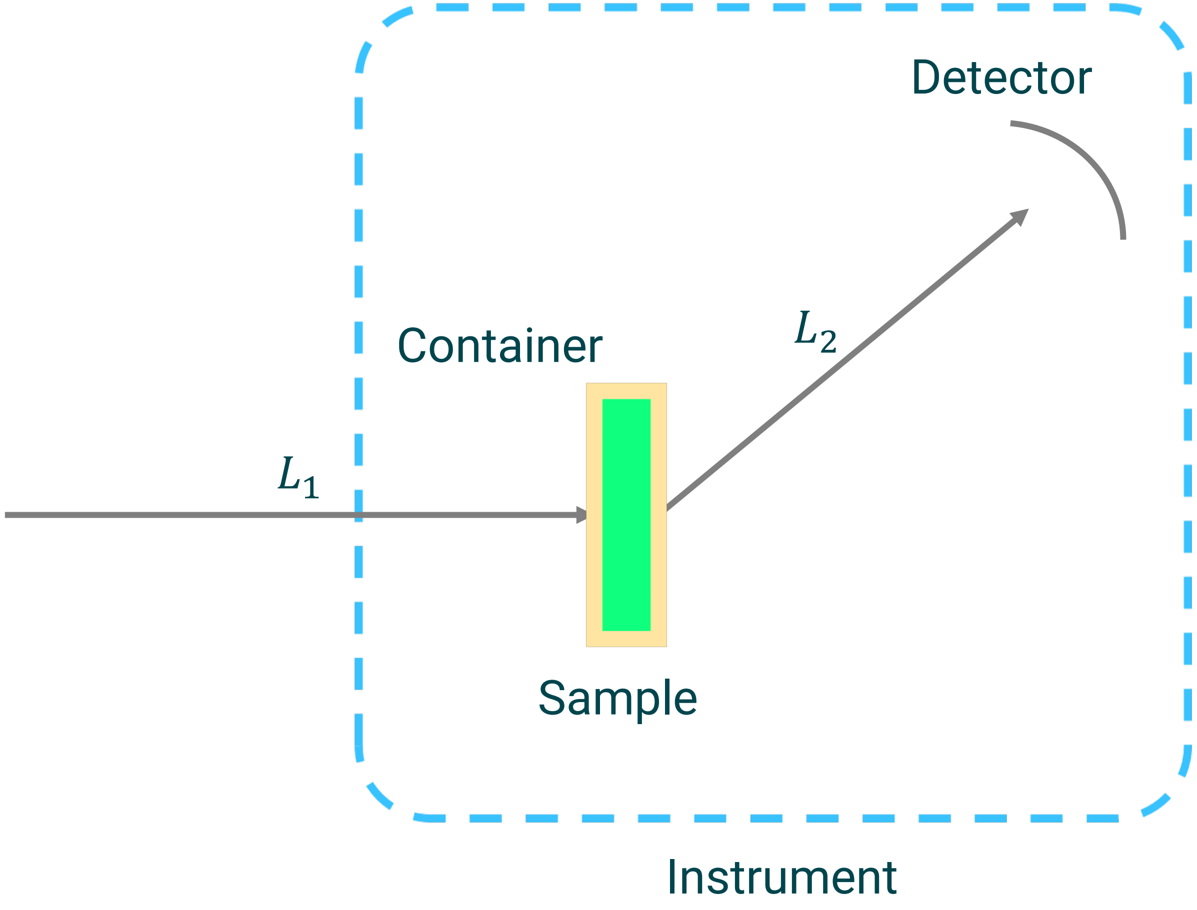
Ingredients of Data Processing – Bkg & Normalization



$\Rightarrow$ Background – Container & instrument

$\Rightarrow I_s^E - I_e^E \rightarrow$ Sample & container

$\Rightarrow I_c^E - I_e^E \rightarrow$ Container

$\Rightarrow I_V^E - I_e^E \rightarrow$ Vanadium

$\Rightarrow$ Detector efficiency, etc.

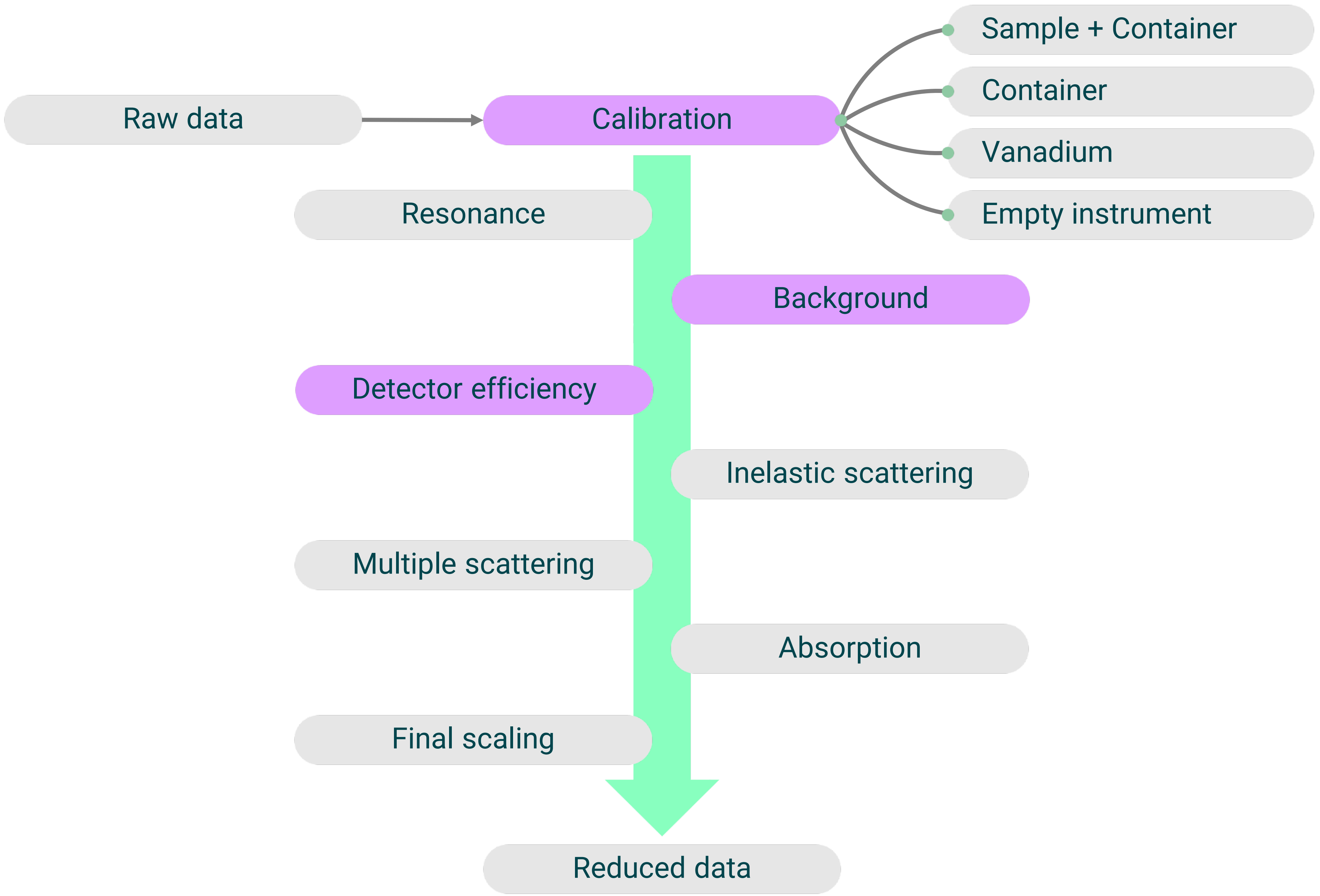
$$\Rightarrow \Phi = \frac{I_V^E - I_e^E}{N_V \langle b_{tot}^2 \rangle}$$

$\Rightarrow$ Putting all together,

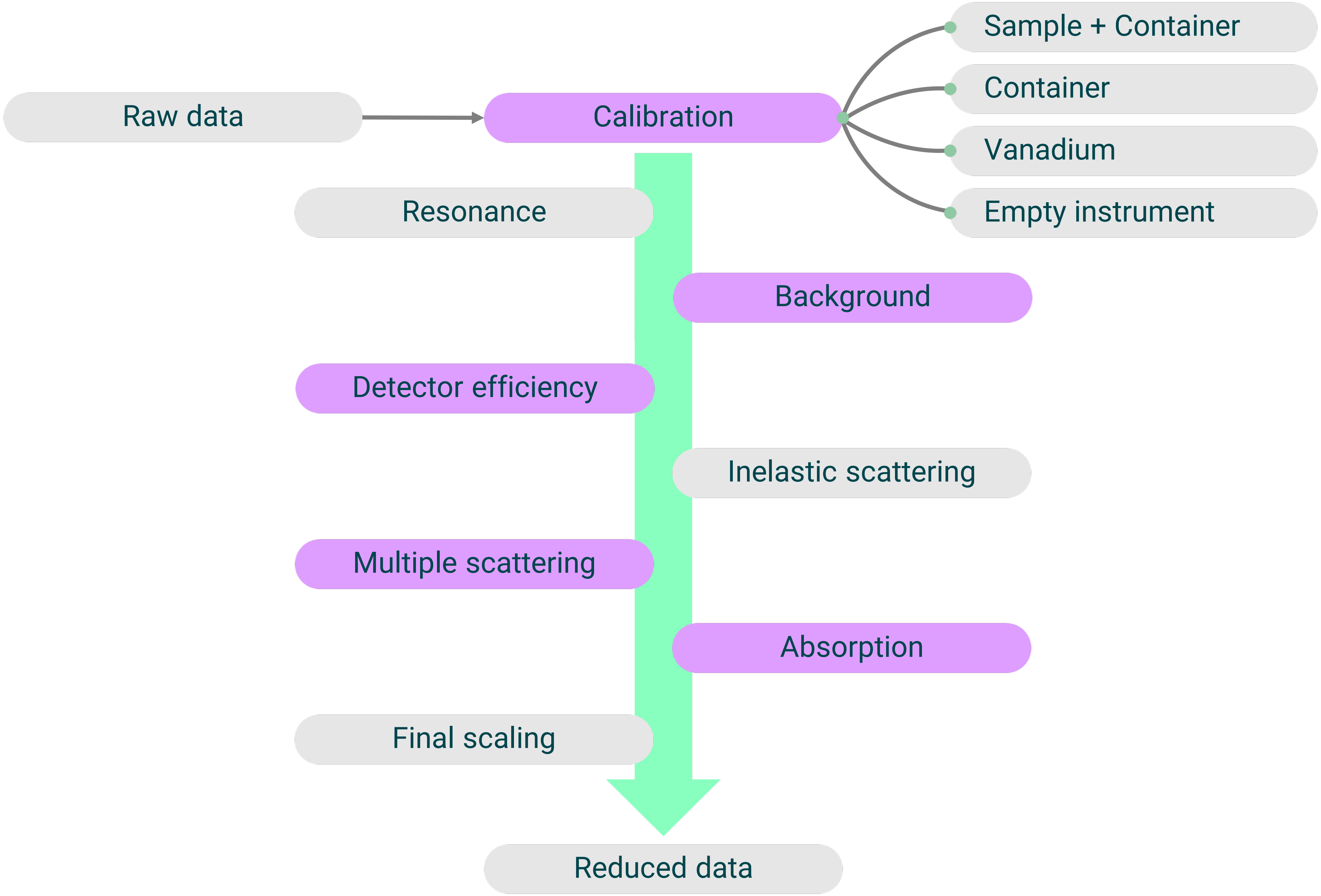
$$\Rightarrow \frac{d\sigma^S}{d\Omega} = \frac{(I_S^E - I_e^E) - (I_C^E - I_e^E)}{\Phi}$$

P. F. Peterson, *et al.* *J. Appl. Cryst*. (2021). **54**, 317–332.

Ingredients of Data Processing – Abs & MS

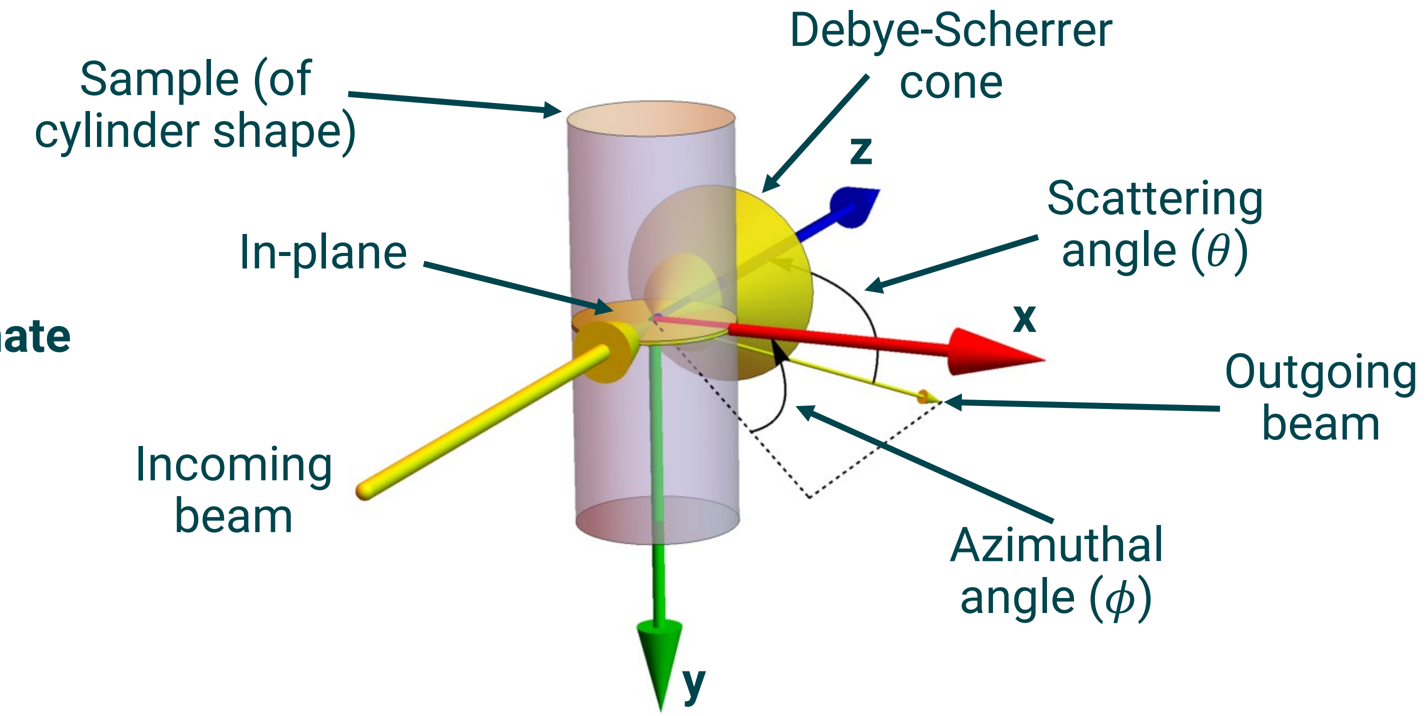


Ingredients of Data Processing – Abs & MS



Ingredients of Data Processing – Abs & MS

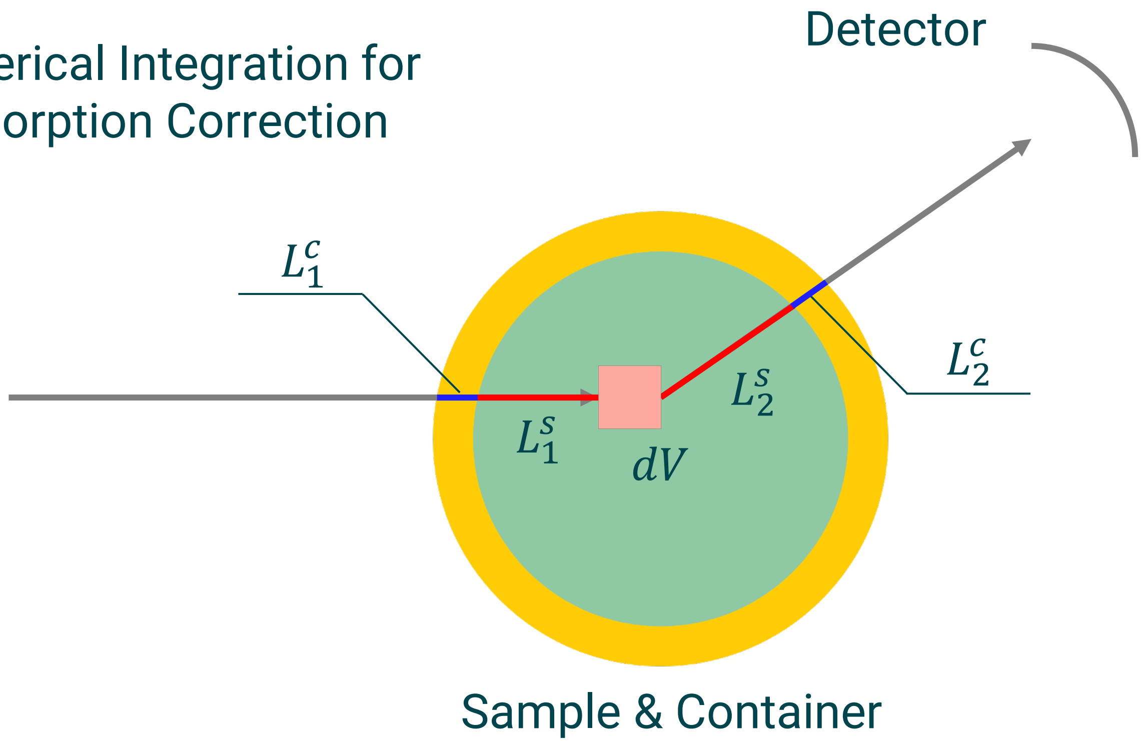
Scattering local coordinate



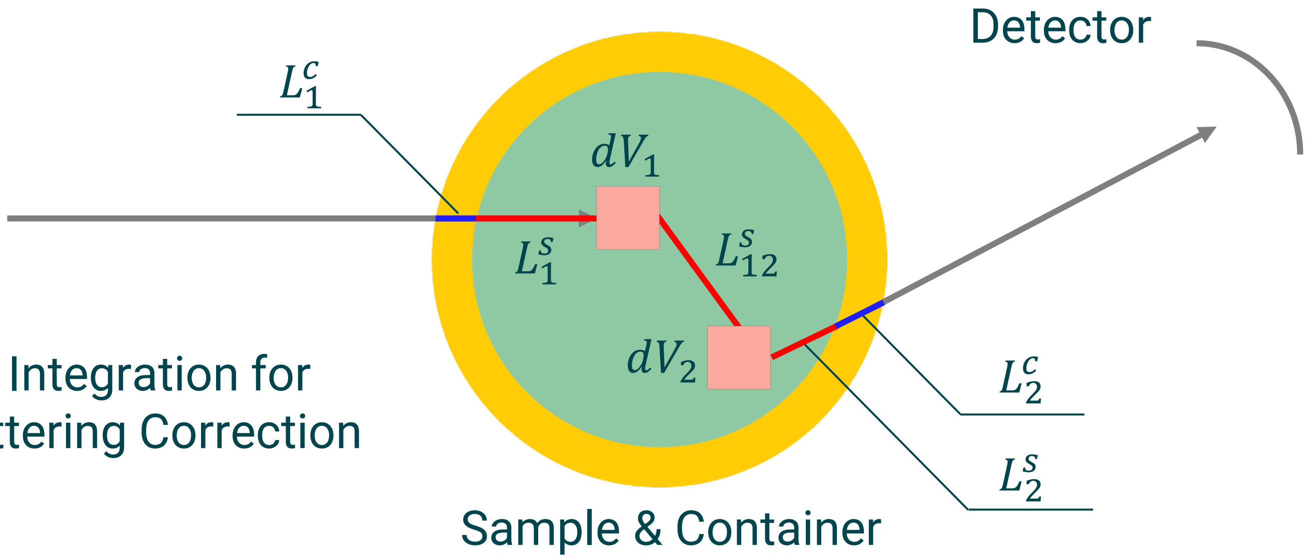
 https://powder.ornl.gov/total_scattering/data_reduction/mts_abs_ms.html

Ingredients of Data Processing – Abs & MS

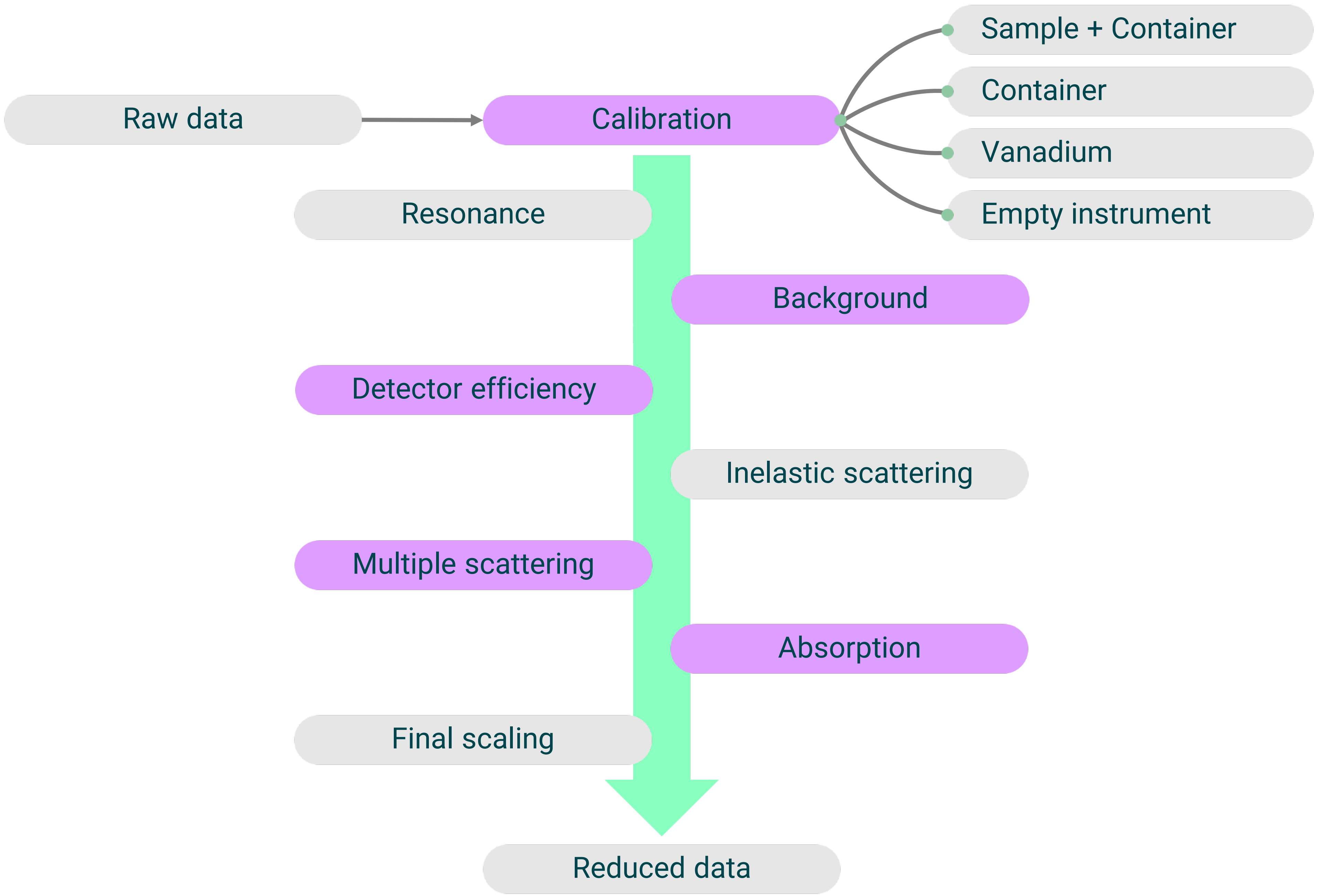
Numerical Integration for
Absorption Correction



Numerical Integration for
Multiple Scattering Correction



Ingredients of Data Processing – Resonance Absorption



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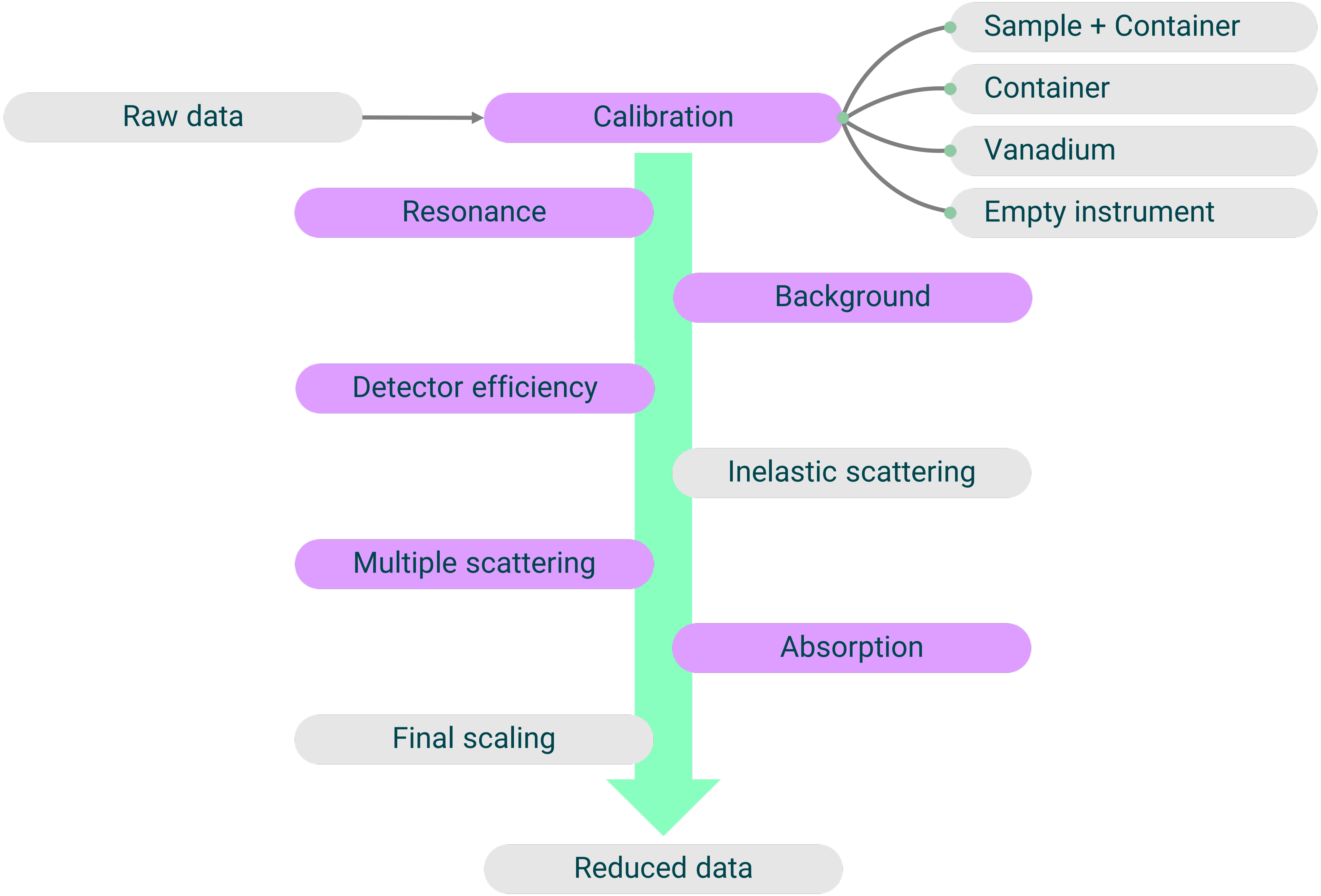
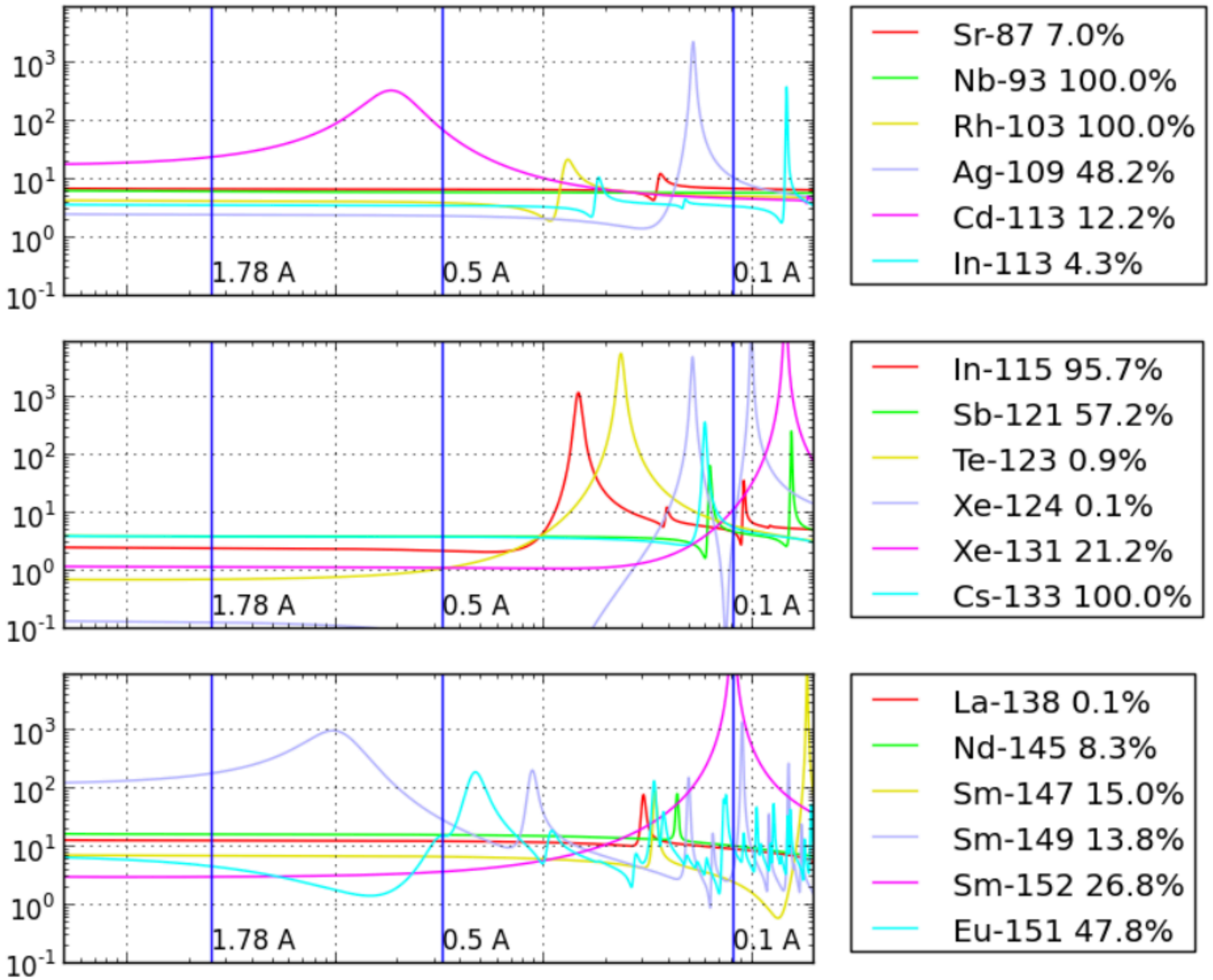
Ingredients of Data Processing – Resonance Absorption



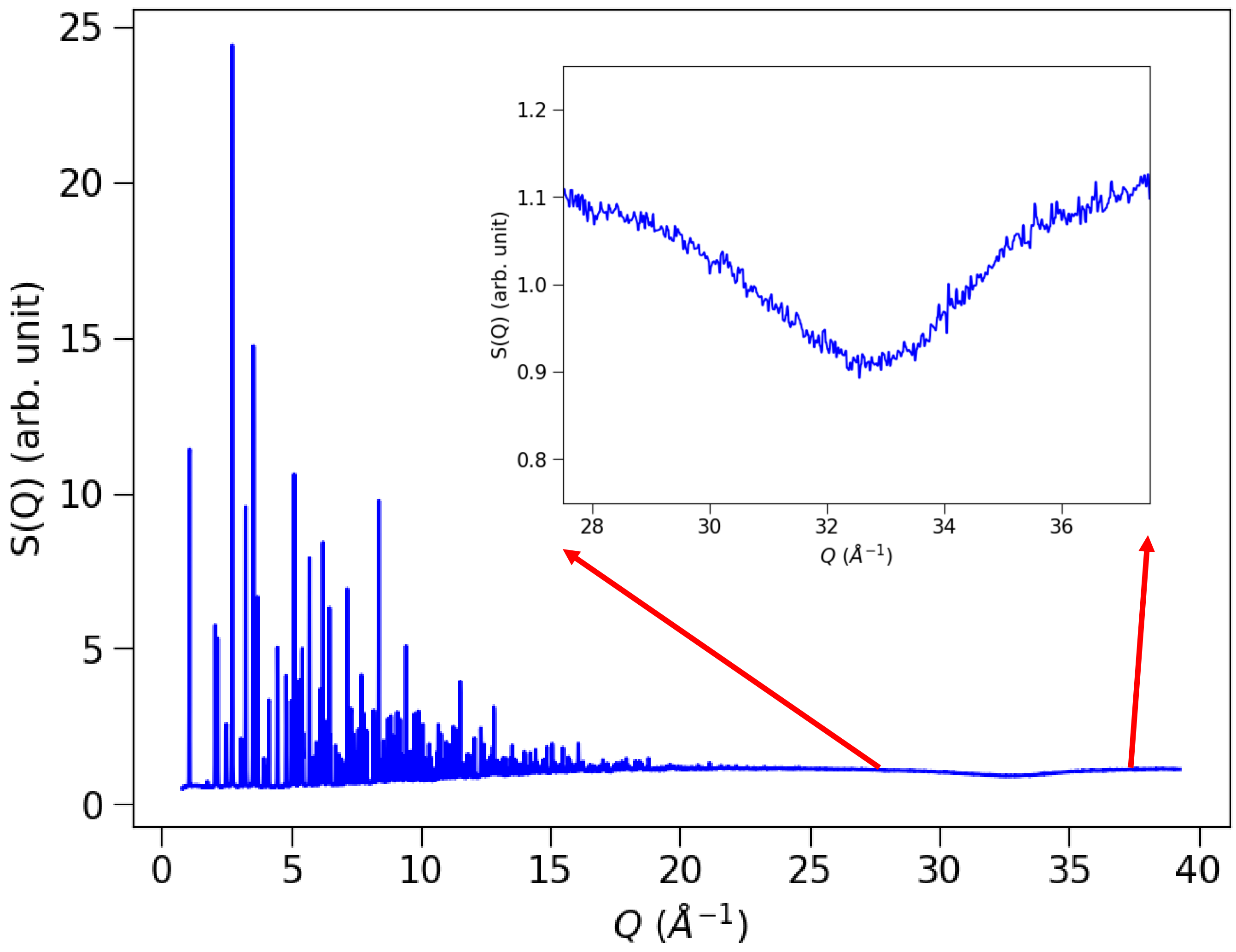
Ingredients of Data Processing – Resonance Absorption



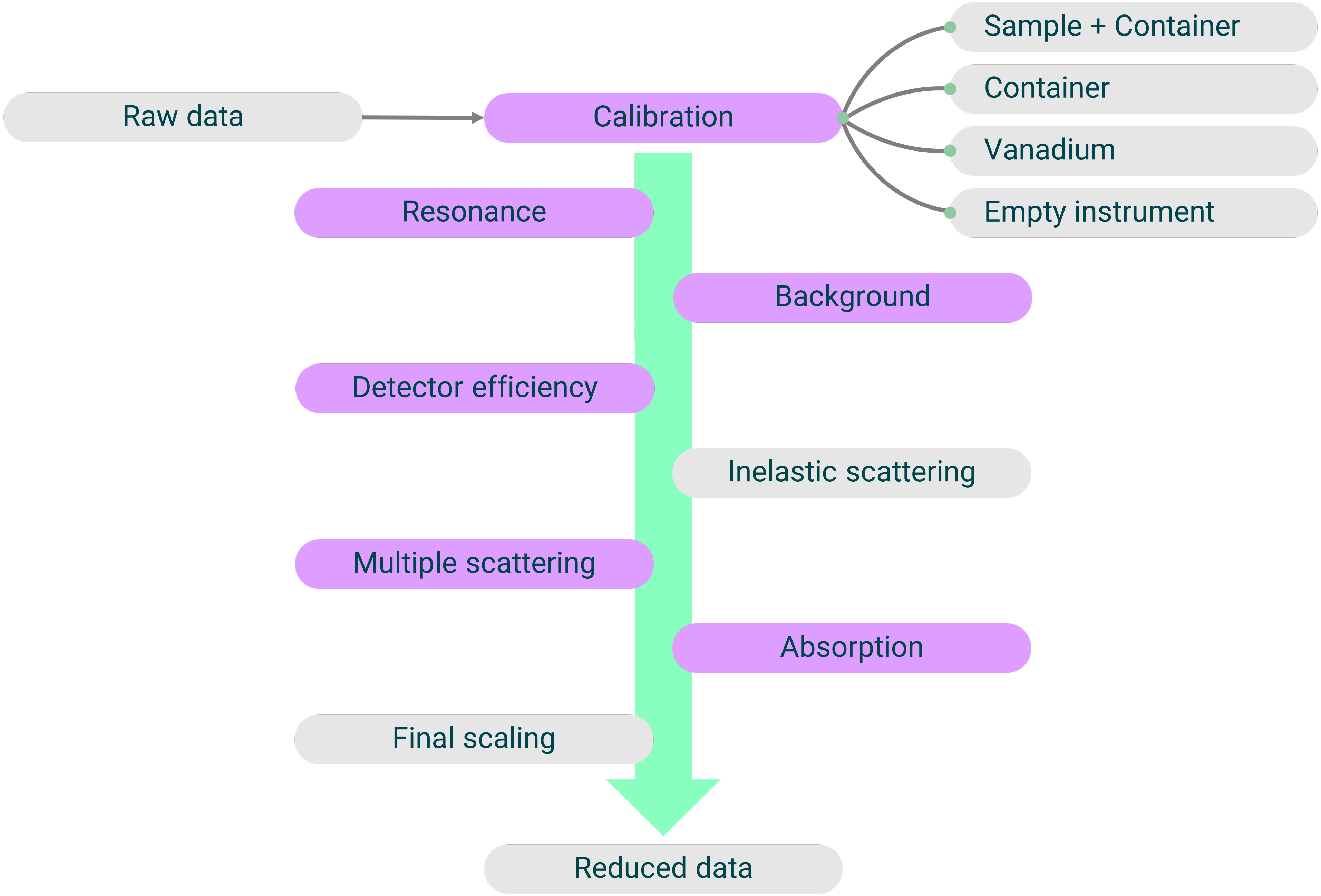
⋮

https://www.ncnr.nist.gov/resources/activation/resonance.html

Ingredients of Data Processing – Resonance Absorption



Ingredients of Data Processing – Inelastic Scattering



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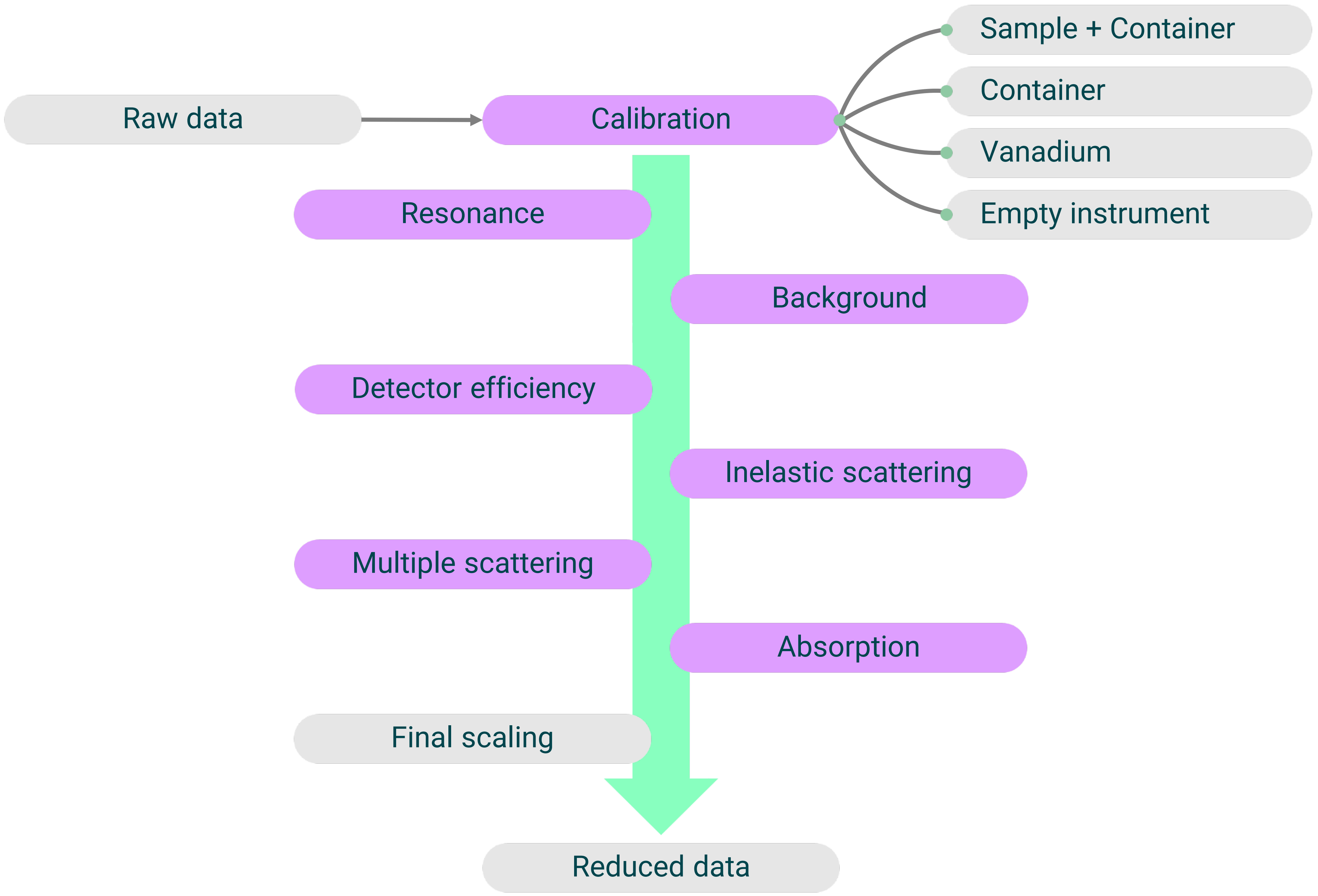
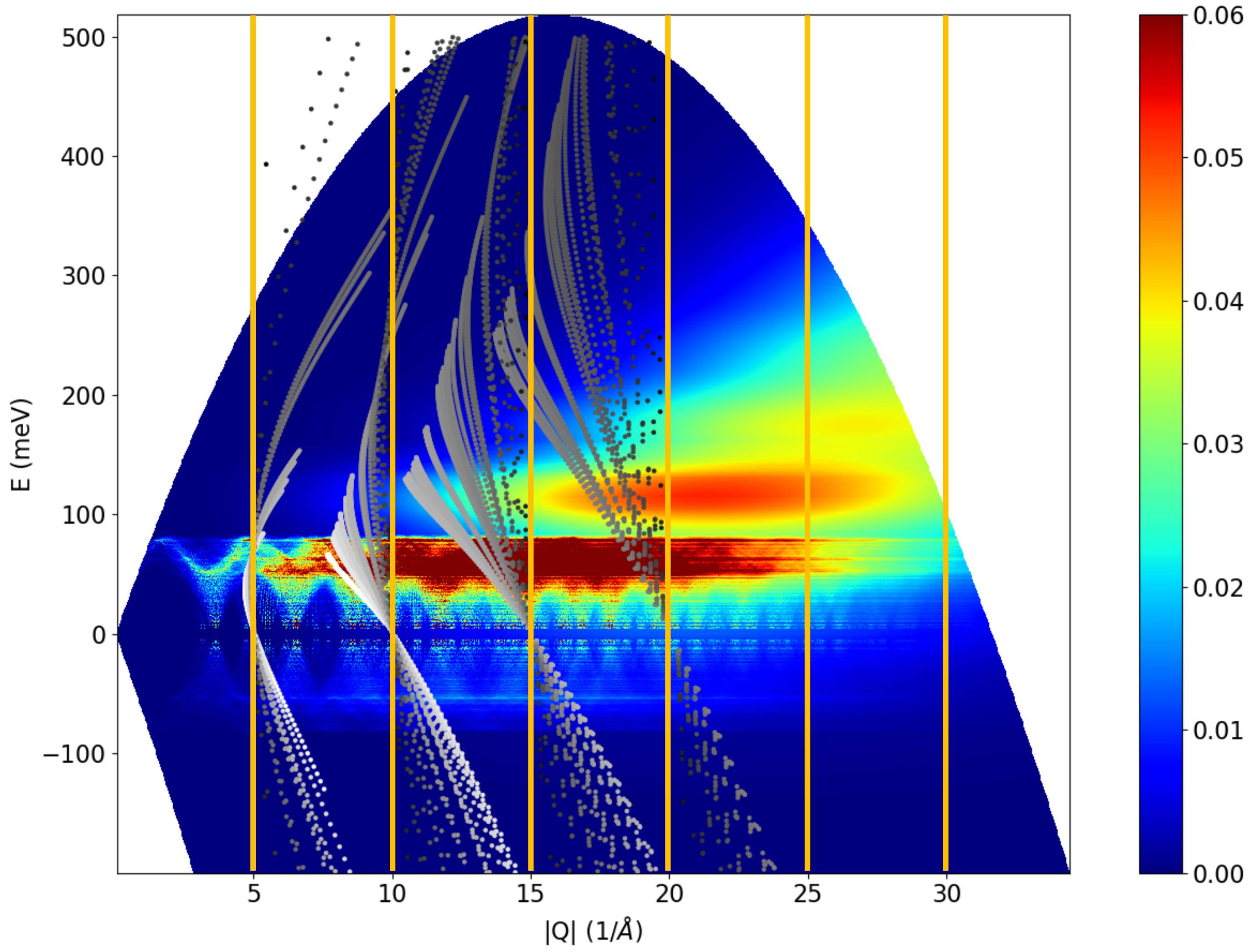
Ingredients of Data Processing – Inelastic Scattering



Ingredients of Data Processing – Inelastic Scattering



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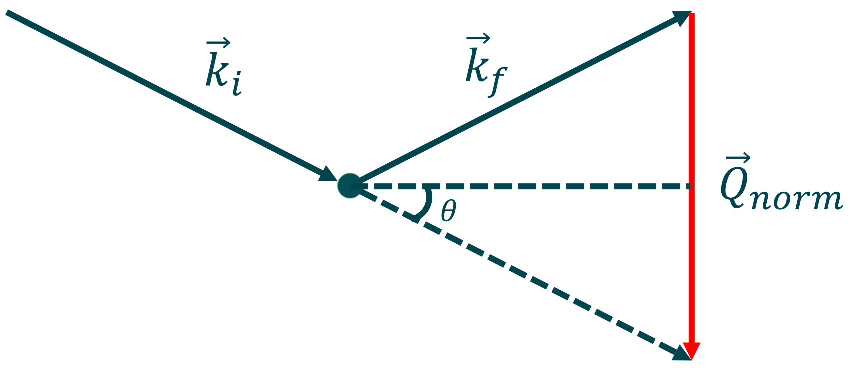
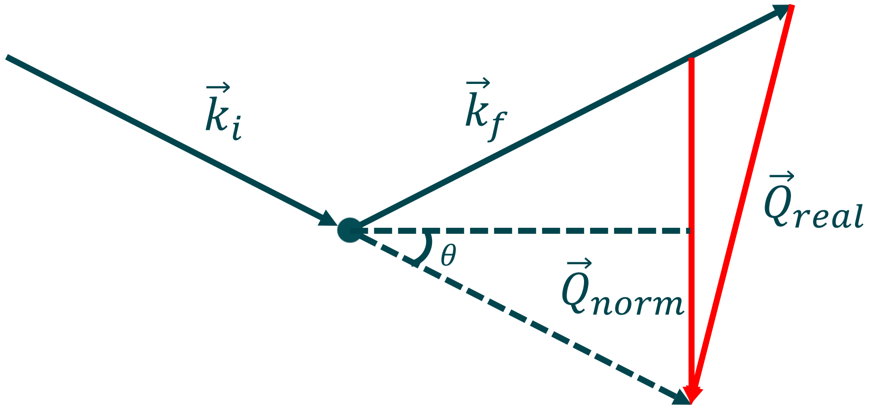
Ingredients of Data Processing – Inelastic Scattering



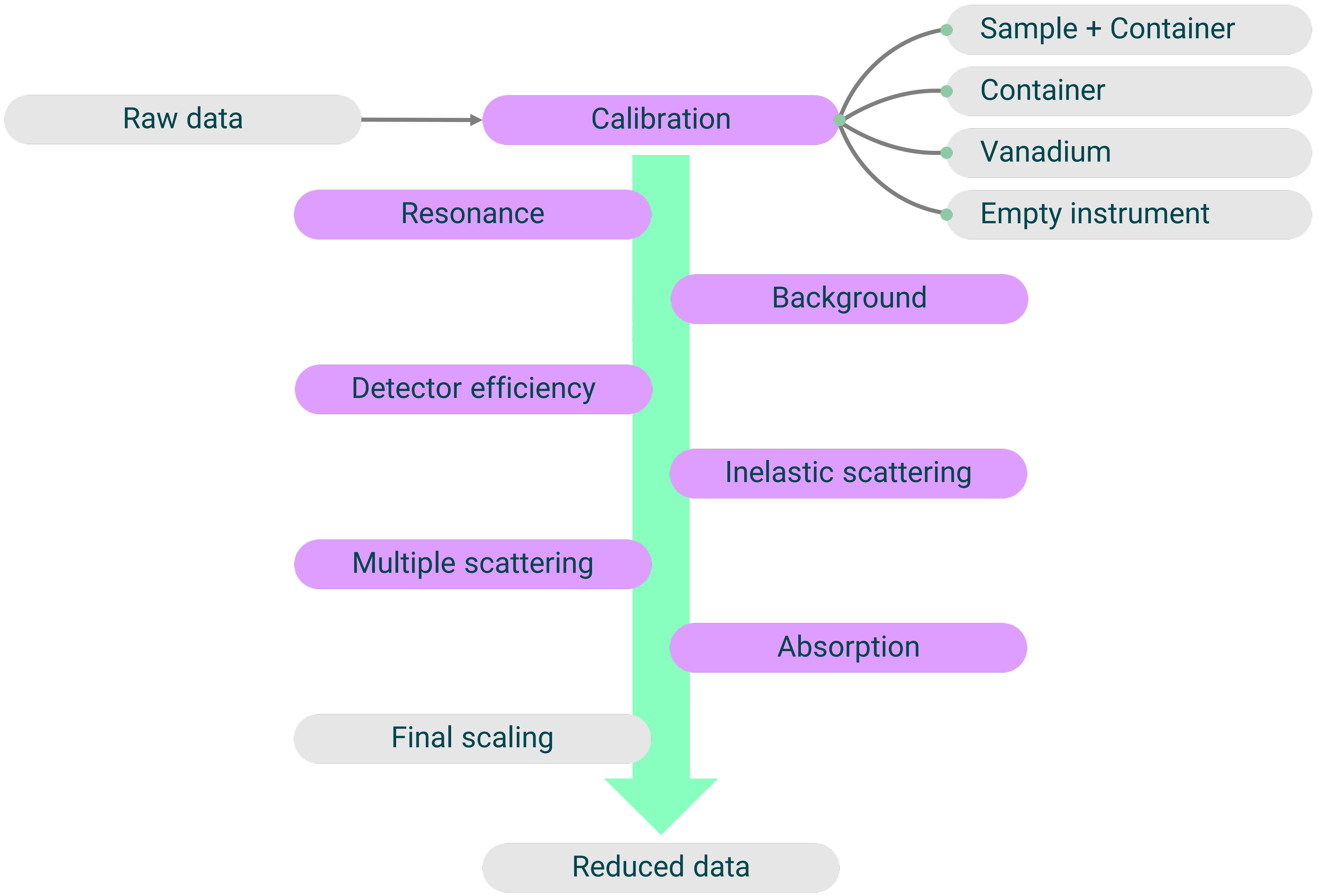


$$Q_{norm} = \frac{4\pi sin\theta}{\lambda} = 2k_i sin\theta$$

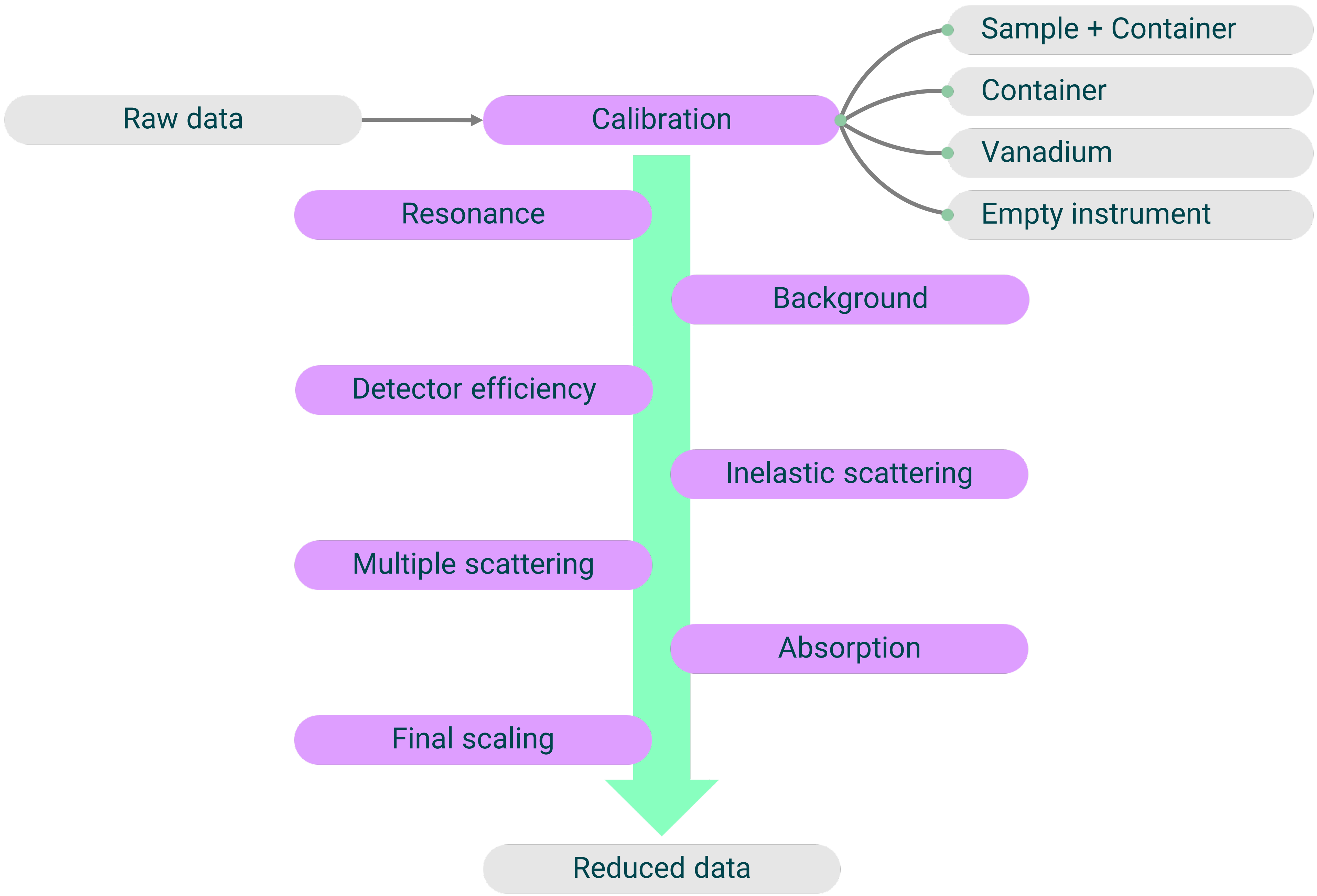
$$\frac{\hbar^2 k_f^2}{2m} - \frac{\hbar^2 k_i^2}{2m} = \Delta E$$

$$\vec{Q}_{real} = \vec{k}_i - \vec{k}_f \Rightarrow \left|\vec{Q}_{real}\right|^2 = \left|\vec{k}_i - \vec{k}_f\right|^2 = k_i^2 + k_f^2 - 2k_i k_f cos2\theta$$

Ingredients of Data Processing – Scaling



Ingredients of Data Processing – Scaling



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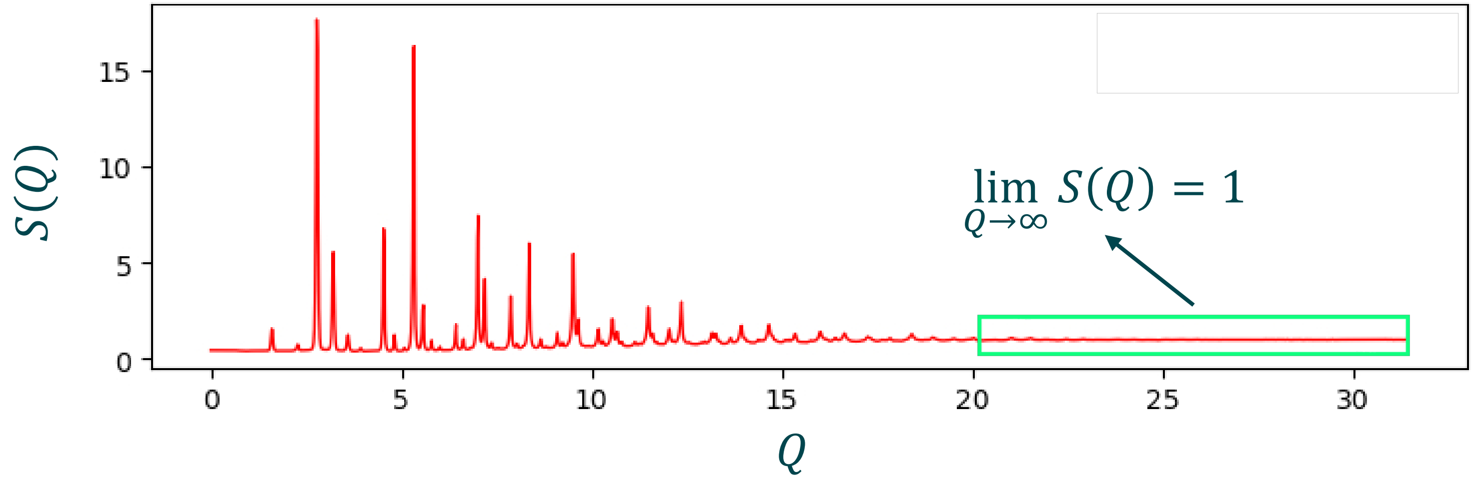
Ingredients of Data Processing – Scaling

$$\frac{d\sigma^s}{d\Omega} = \frac{1}{N_s}\sum_{j,k} b_j b_k e^{i\vec{Q}\cdot(\vec{r}_j-\vec{r}_k)} = \frac{1}{N_s}\sum_{j\neq k} b_j b_k e^{i\vec{Q}\cdot(\vec{r}_j-\vec{r}_k)} + \langle b^2\rangle = F(Q) + \langle b^2\rangle = I(Q)$$

Laue term

$$\frac{F(Q)}{\langle b\rangle^2} = \frac{I(Q)-\langle b^2\rangle}{\langle b\rangle^2} \stackrel{\text{def}}{=} S(Q) - 1 \Rightarrow S(Q) = \frac{I(Q)}{\langle b\rangle^2} + \boxed{\frac{\langle b\rangle^2 - \langle b^2\rangle}{\langle b\rangle^2}}$$



Final scaling,

$\Rightarrow$ scale & offset factors

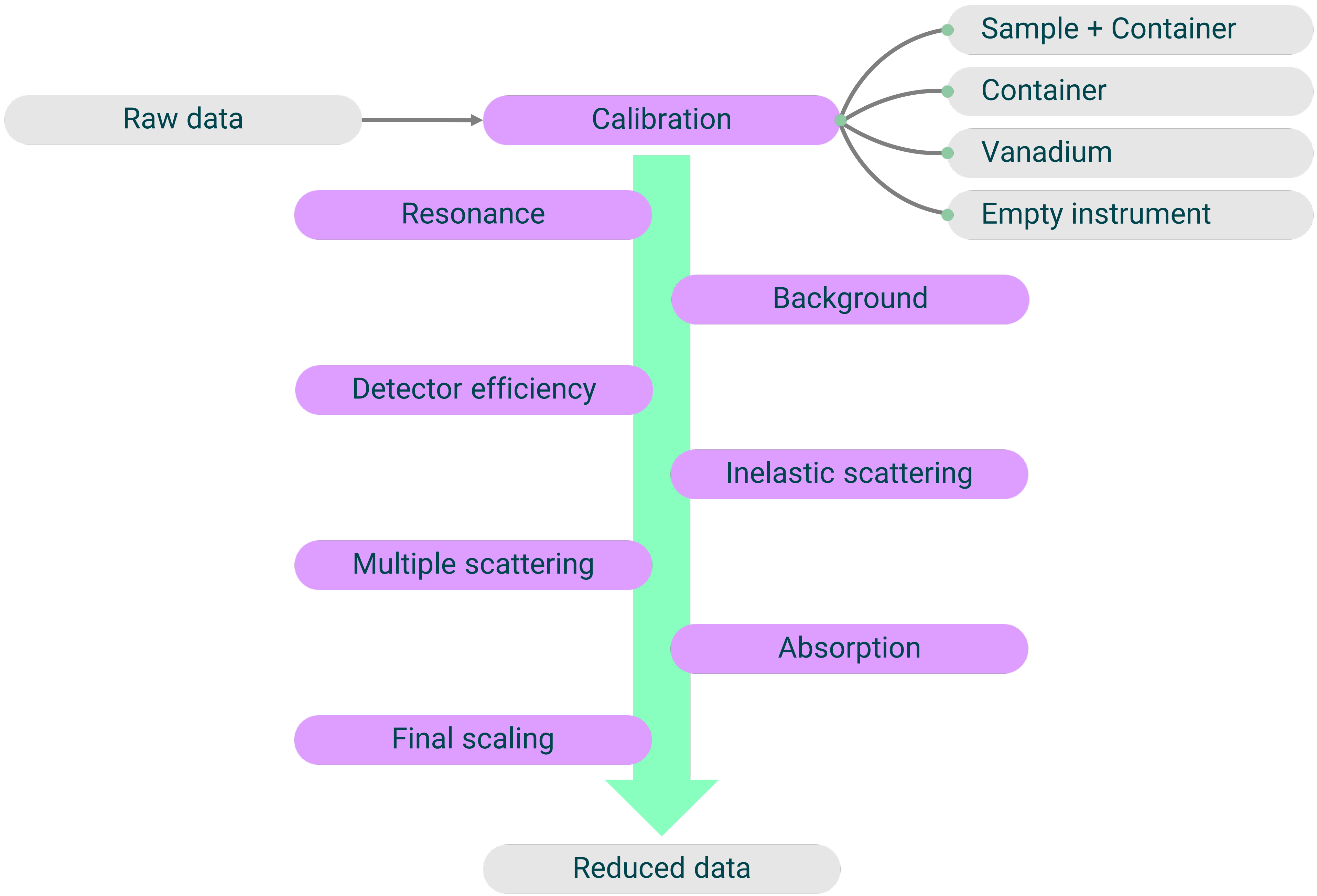
$\Rightarrow$ low-r behavior in real-space

$\Rightarrow$ unit-cell refinement

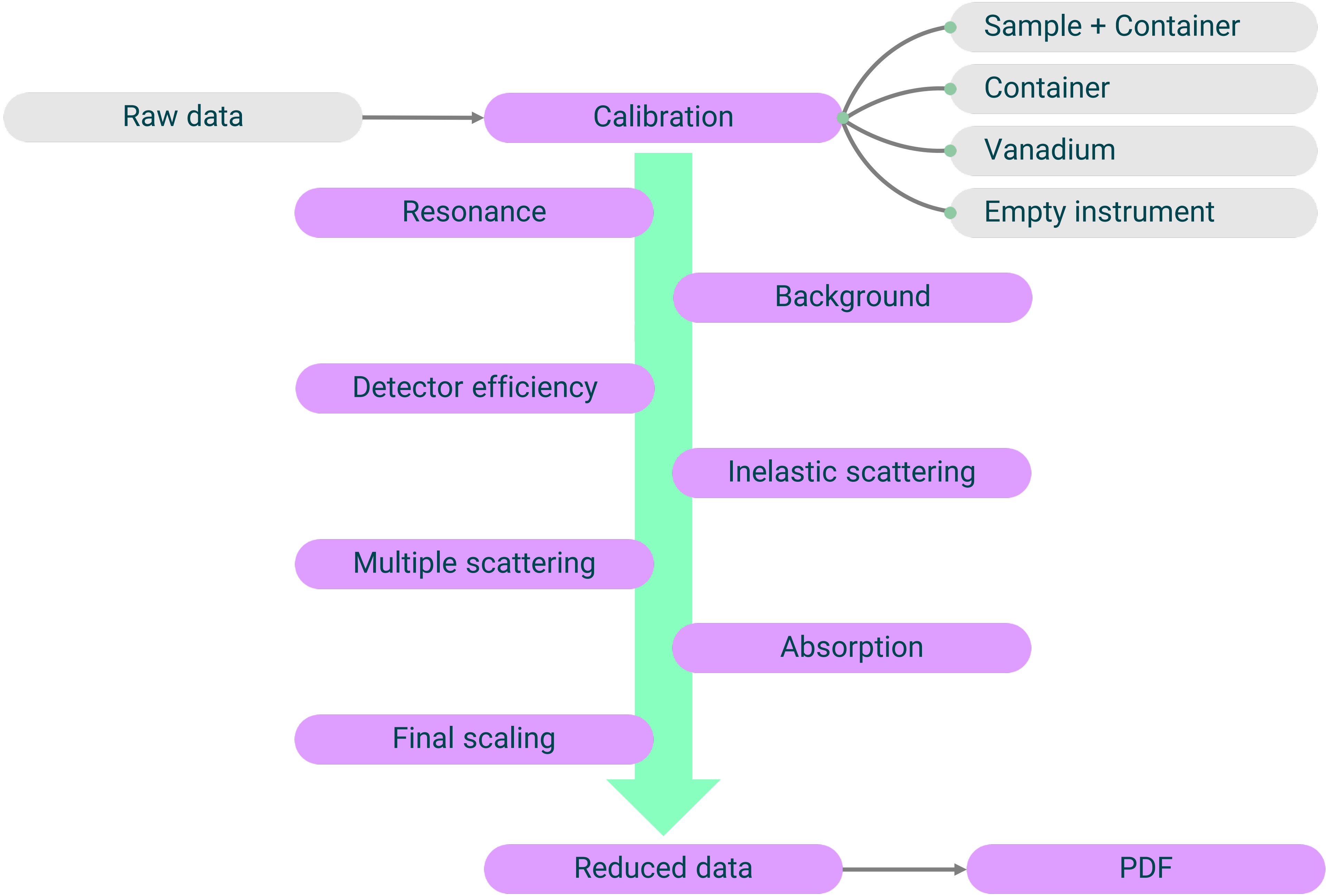
$\Rightarrow$ play with scale for supercell fitting

$\Rightarrow$ *only when have to!*

Ingredients of Data Processing – Fourier Transform



Ingredients of Data Processing – Fourier Transform



Ingredients of Data Processing – Fourier Transform

- Isotropic Orientational Average

$$\langle \exp[i\vec{Q}\cdot(\vec{r}_j-\vec{r}_k)]\rangle = \frac{sin(Qr_{jk})}{Qr_{jk}}$$

- Coordination Number

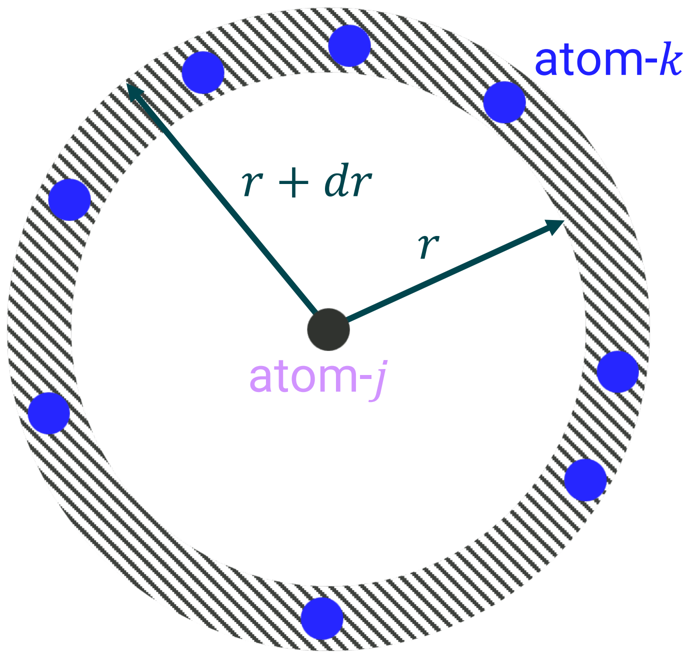
Def: $$g_{jk}(r) = \frac{n_{jk}/(4\pi r^2 dr)}{\rho_k}$$

$$n_{jk} = \rho_k \cdot 4\pi r^2 dr \cdot g_{jk}(r)$$

- Putting Together

$$F(Q) = \rho \sum_{jk} c_j c_k b_j b_k \cdot 4\pi \int_0^\infty r^2 g_{jk}(r) \frac{\sin(Qr)}{Qr} dr$$

Def: $$G(r) = \sum_{i,j=1}^{n} c_i c_j \bar{b}_i \bar{b}_j [g_{ij}(r) - 1]$$



M. Dove, *Structure and Dynamics: An Atomic View of Materials*, see Appendix-J

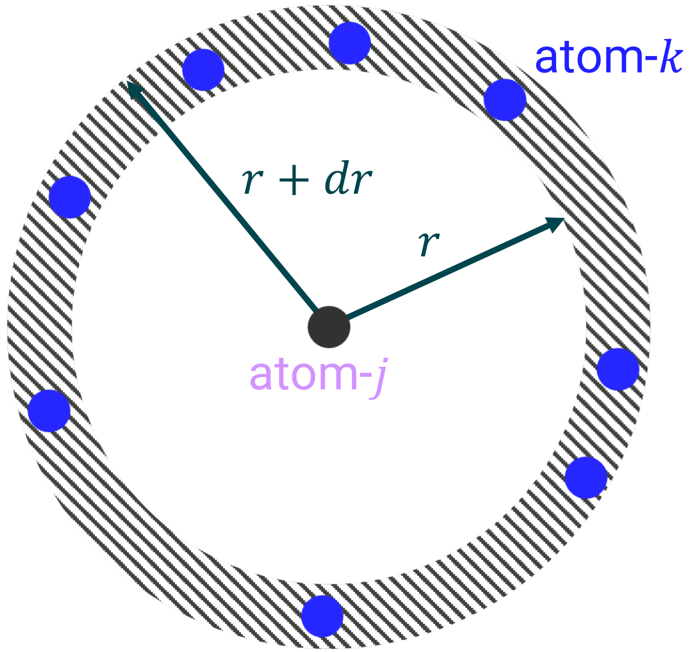
Ingredients of Data Processing – Fourier Transform

$$G(r) = \frac{1}{(2\pi)^3 \rho_0} \int_0^{\infty} 4\pi Q^2 F(Q) \frac{\sin Qr}{Qr} dQ$$

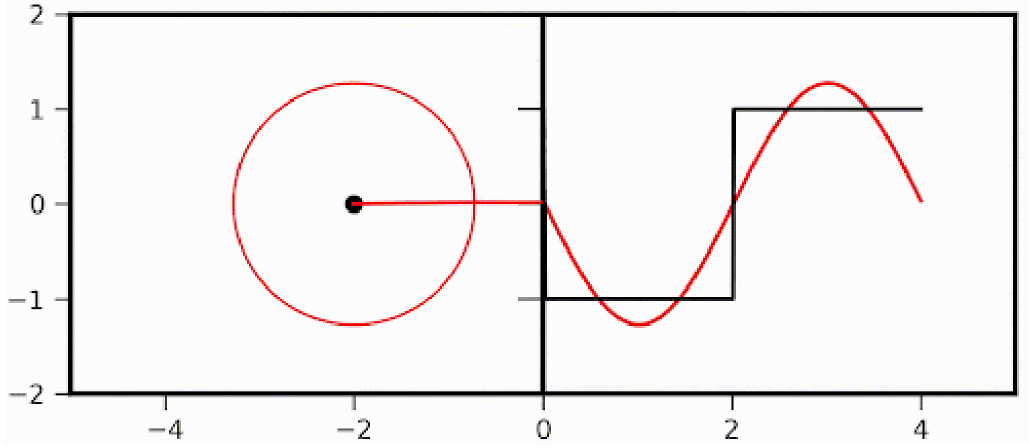
$$F(Q) = \rho_0 \int_0^{\infty} 4\pi r^2 G(r) \frac{\sin Qr}{Qr} dr$$

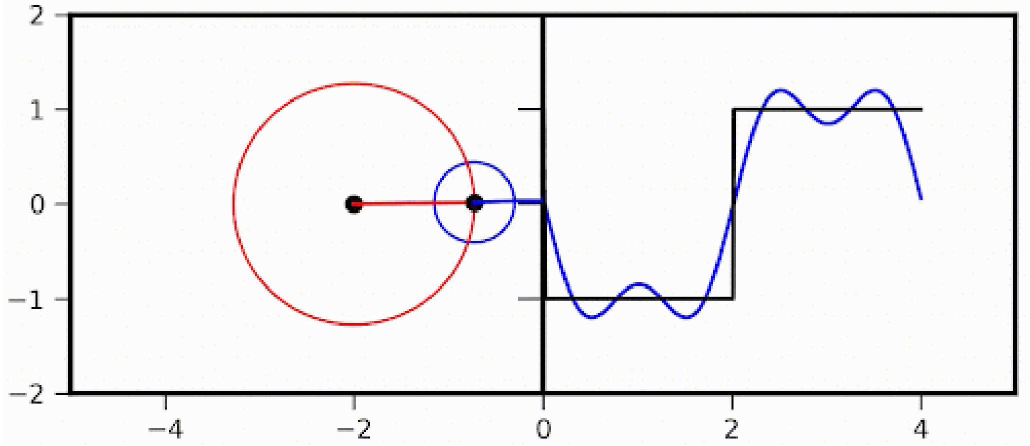
$$G(r) = \sum_{j,k=1}^{n} c_j c_k \bar{b}_j \bar{b}_k [g_{jk}(r) - 1]$$

$$g_{jk}(r) = \frac{n_{jk}/(4\pi r^2 dr)}{\rho_k} = \frac{\rho_k^{local}}{\rho_k}$$



Ingredients of Data Processing – Fourier Transform





https://iris2020.net/2020-06-28-fourier_transform/

Reading Materials & Tools

- M. Dove, *Structure and Dynamics: An Atomic View of Materials*
- W. Marshall and S. W. Lovesey, *Theory of thermal neutron scattering*
- J. J. Sakurai, *Modern Quantum Mechanics*
- D. Keen, *J. Appl. Cryst*. (2001), **34**, 172-177.
- P. Peterson, *et al*. *J. Appl. Cryst*. (2021), **54**, 317-332.
- Powder Diffraction Doc at ORNL, https://powder.ornl.gov
- Neutron Powder Diffraction Data Processing Handbook, https://yr.iris-home.net/dataproc
- ADDIE, https://addie.ornl.gov
- Powder Diffraction Forum: https://powder.ornl.gov/forum
- RMCProfile, https://rmcprofile.ornl.gov

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Ross Whitfield
Thomas Proffen
Victor Krayzman
Wenduo Zhou
Yang Long

Live Demo

- Neutron powder diffraction data processing workflow at SNS, ORNL

  - MantidTotalScattering, LiveRed & AutoRed workflow
    - `abs_pre_calc -c`
    - `abs_pre_calc NOM 28922`
    - `all_nom 28922 181278`, or `all_nom 32855 ‘203274-203280’`

  - ADDIE
    - Skip demo
    - Tutorial available here, https://powder.ornl.gov/total_scattering/data_reduction/dr_howto.html

- ADDIE web interface available at https://addie.ornl.gov

  - ScatteringInspector, CIFFinder, ConfigViewer, NXCalculator, SimulatePDF, SimulatePowder