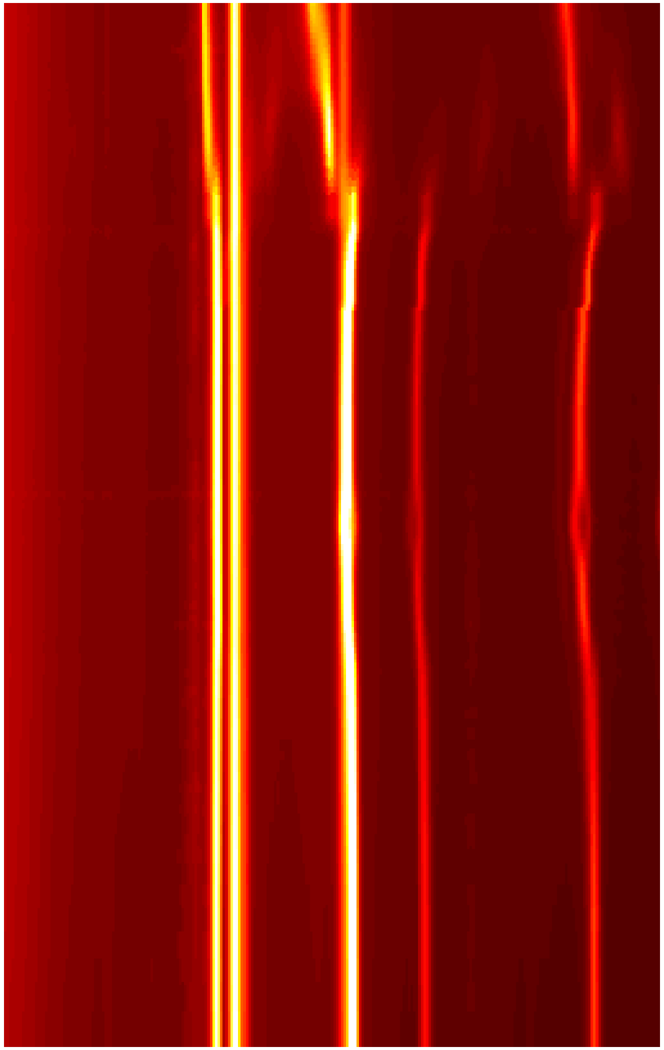
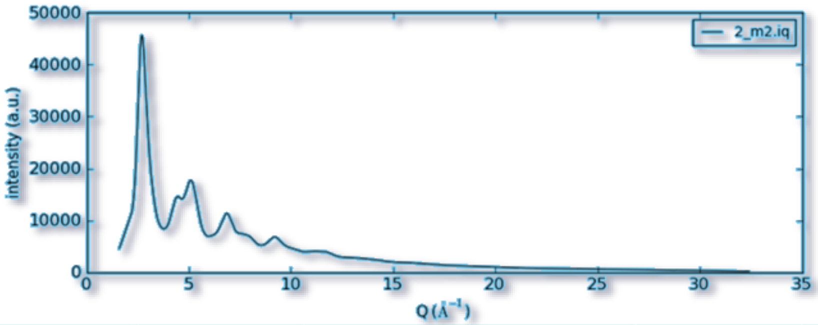
ESRF | The European Synchrotron



Synchrotron PDF Data Acquistion and Reduction

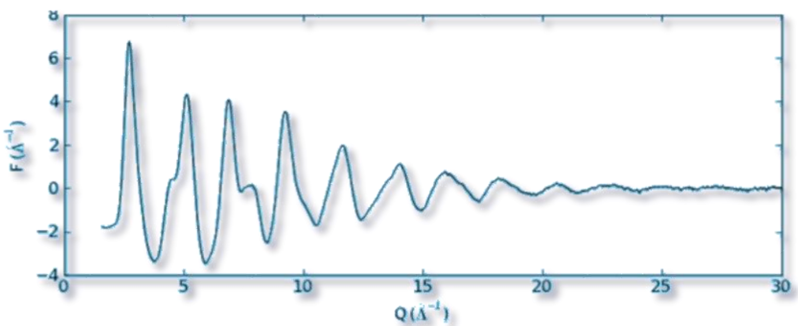
G. Vaughan, M. Brunelli, S. Checchia

GETTING THE PDF FROM THE DIFFRACTION DATA



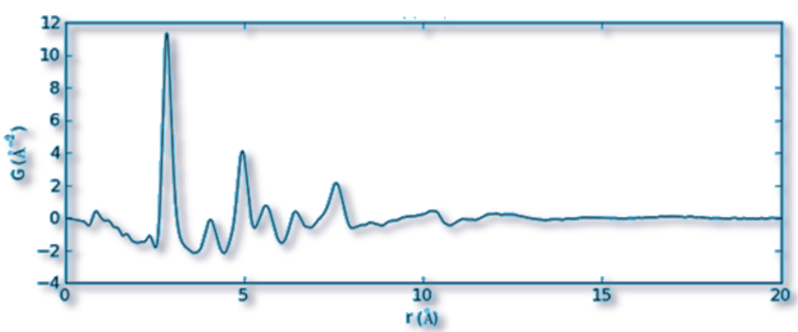
$$S(q) = \frac{I(q) - \langle f(q)^2 \rangle}{\langle f(q) \rangle^2} + 1$$

$$F(q) = q(S(q) - 1)$$



Debye Equation:

$$F(q) = \frac{1}{N\langle f(q) \rangle^2} \sum_{i \neq j} f_i(q)\, f_j(q) \frac{\sin q r_{ij}}{r_{ij}}$$



$$G(r) = \frac{2}{\pi} \int_{q_{min}}^{q_{max}} F(q) \sin qr \, dq$$

$$G(r) = \frac{1}{N\langle f \rangle^2} \sum_{i \neq j} f_i\, f_j \delta(r - r_{ij})$$

DATA ACQUISITION

Continuous Transform on finite data

- High q – implies high energy

Good statistics

- Particularly at high q; contrary to form factor behaviour

Low/well characterized background

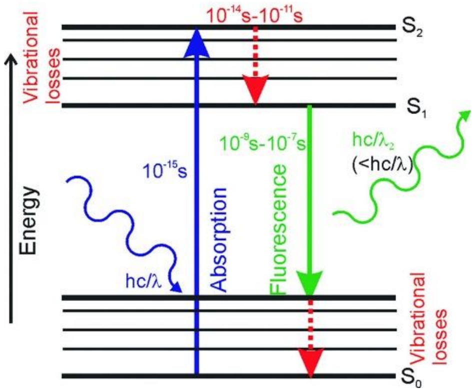
- Minimize inelastic scattering
  - avoid absorption edges (W, Pb, …)
  - using energy discrimination
- Clean background – minimize parasitic scattering
  - sample environment
  - tomographic methods

$$I = I_e + I_{ie} + I_p$$

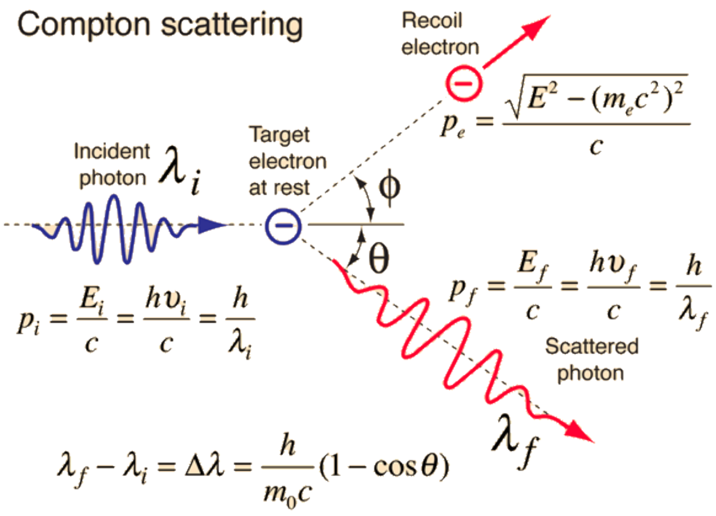
$$I = I_e + \left(I_{istruct} + I_{Comp} + I_{Fluo}\right) + I_p$$

Fluorescence comes from all absorption edges below the incident energy
Fluorescence can be 80% of the signal at high Q

Jablonksi diagram depicting simple 1 photon excitation fluorescence

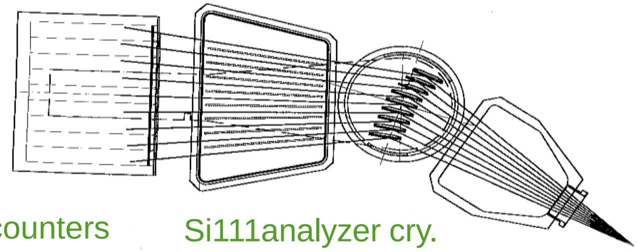


Compton scattering has a spatial and energy distribution

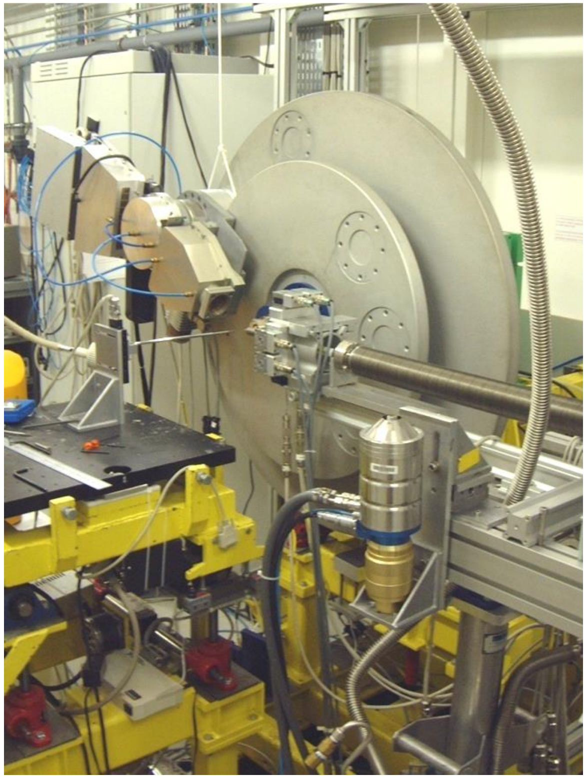


Scintilators and PMT

- Angle sensitive
- Energy discrimination
- Background elimination
- Good dynamic range
- Photon counting
- Very high angular resolution
- Accurate lattice parameters
- Slow

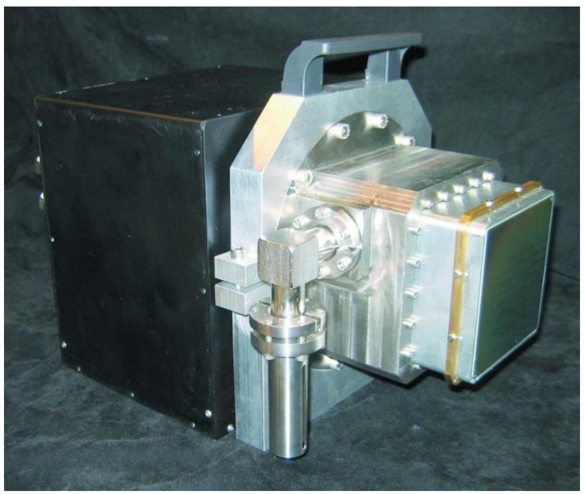


1-d counters Si111analyzer cry.



Mostly Developed for Medical Imaging

- Flat panel detectors
- CCD/CMOS cameras coupled to scintillators
- Pixel detectors



2D DETECTORS

	Advantages	Disadvantages
CCD/CMOS Cameras Phosphor coupled	Stable Background Stable Flat Field	High Background Limited Dynamic Range Large PSF Low Sensitivity Integrating
Flat Panel	High Sensitivity Stable Flat Field No PSF Cheap	Very High Background Variable Background Integrating
Pixel Detectors	High Dynamic Range High Sensitivity Photon Counting Zero Background Energy Discrimination Stable Flat Field No PSF	Price

EFFECT OF ELECTRONIC NOISE

$$I = (I_0 - D)R$$

$$I = (I_0 - D)\frac{\langle R \rangle}{R_i}$$

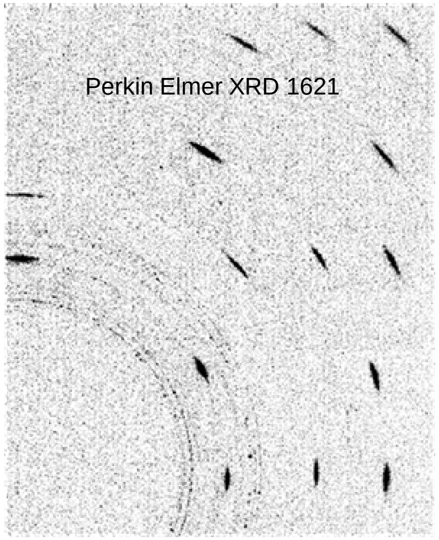
$$D = \frac{1}{N_D}\sum D_j = \frac{n_D}{N_D}\sum I_{0,j} = n_D I_0$$

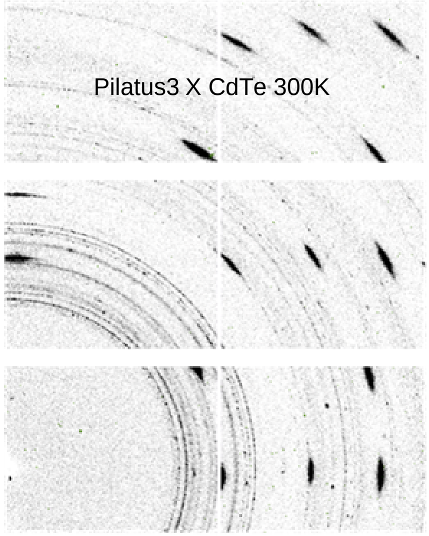
$$\sigma_I^2 \cong \sigma_{I_0}^2 \left[1 + \frac{n_D}{N_D} + \frac{(1 - n_D)^2}{n_R N_R} \right]$$

$$\begin{aligned} \sigma_{\langle R \rangle}^2 &= \left(\frac{1}{N}\right)^2 N \sigma_{R_i}^2 \\ &= \left(\frac{\sigma_{R_i}^2}{N}\right) \\ &\approx 0 \end{aligned}$$

COMPARISON OF FLAT PANEL AND PIXEL DETECTOR

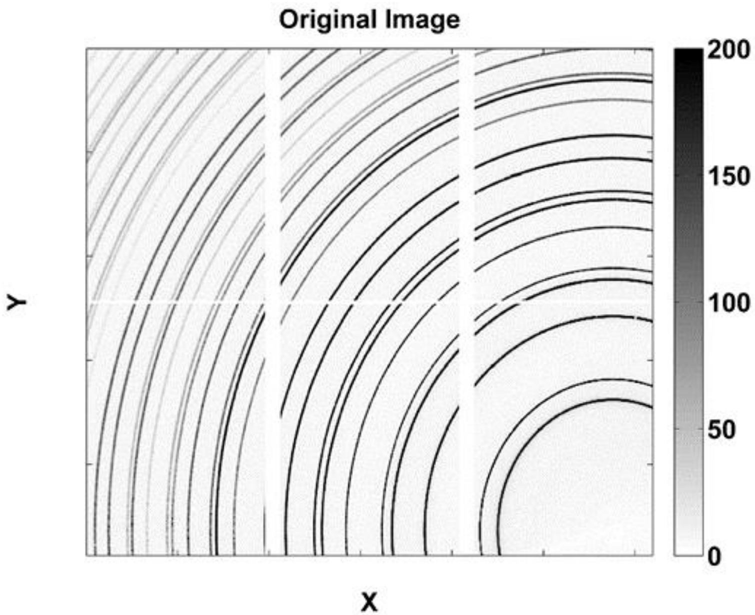
	Pilatus3 X CdTe 2M	Perkin Elmer XRD 1621
Detection technology	Hybrid photon counting	Flat panel
Sensor material	CdTe	CsI
Pixel size [μm^2]	172×172	200×200
Total number of pixels (H × V)	1475×1679	2024×2024
Maximum frame rate[Hz]	250 (500 with ROI)	15 (30 with 2×2binning)
Point Spread Function (FWHM)	1 pixel	2 pixels
Energy threshold [keV]	8-40	none
Maximum count rate [ph/s/pixel]	5×10^6	Integrating detector
Non linearity	<2% at 10^6 counts/s/pixel	
Counter depth	20 bit	16 bit
Dynamic range	20 bit	12.8 bit
Minimum exposure [ns]	200	33000000
Image lag	0	~1% after 100ms



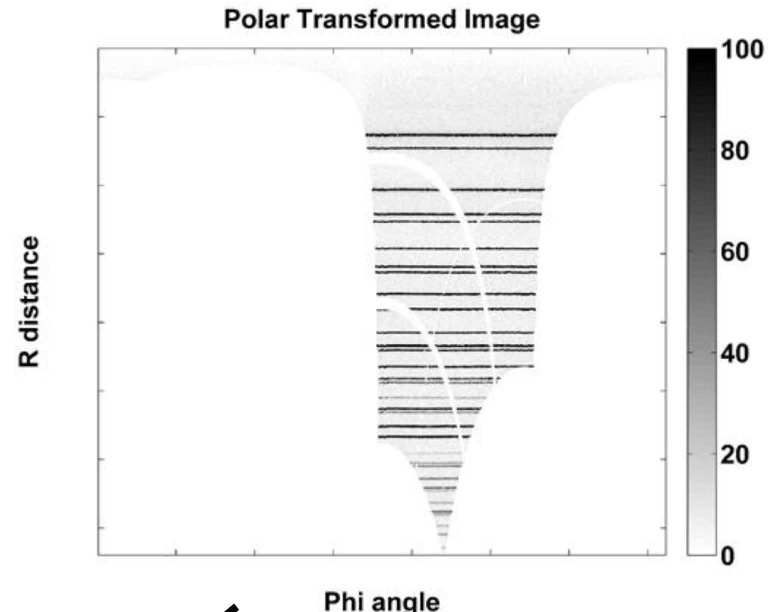


Superconducting filament, Ø50 µm, measured at 50 keV, with exposure time of 100ms with a Perkin Elmer XRD 1621 flat panel detector (left) and with the Dectris Pilatus3 X CdTe 300K prototype

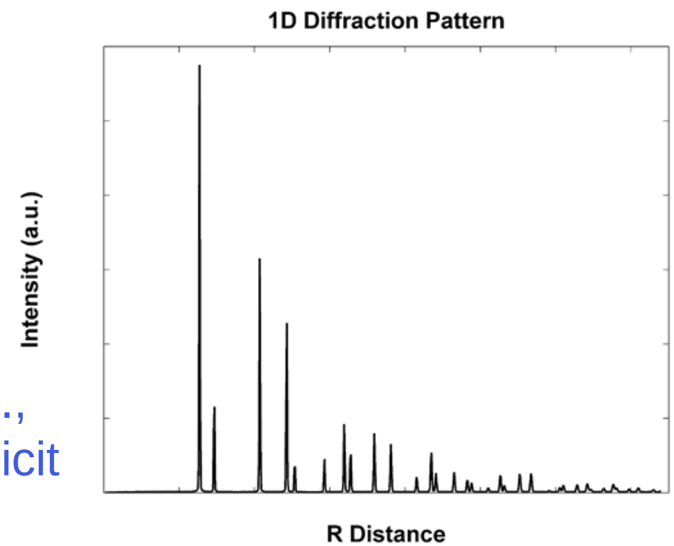
REDUCTION OF XRD DATA



Geometric corrections

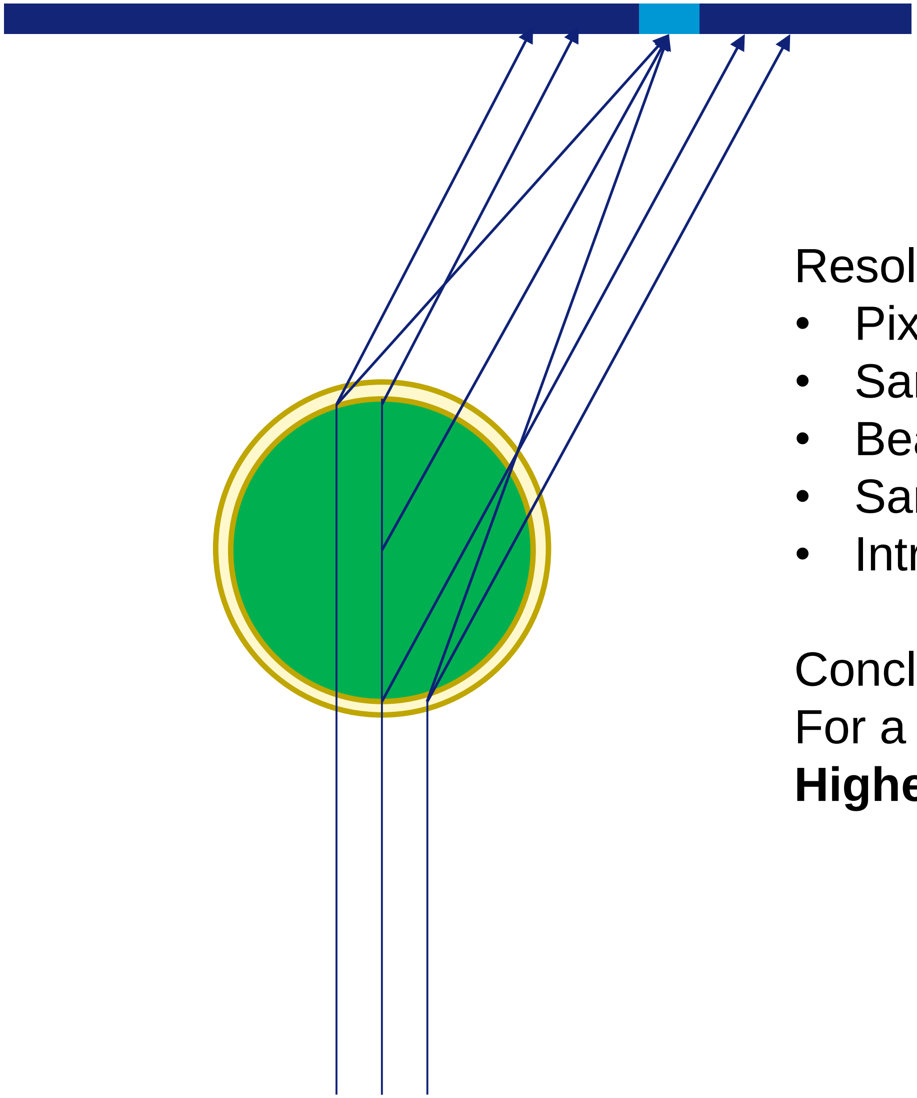


Outlier rejection, masking



Cylindrical symmetry, i.e., a perfect powder, is implicit

EFFECTS OF SAMPLE GEOMETRY - BROADENING

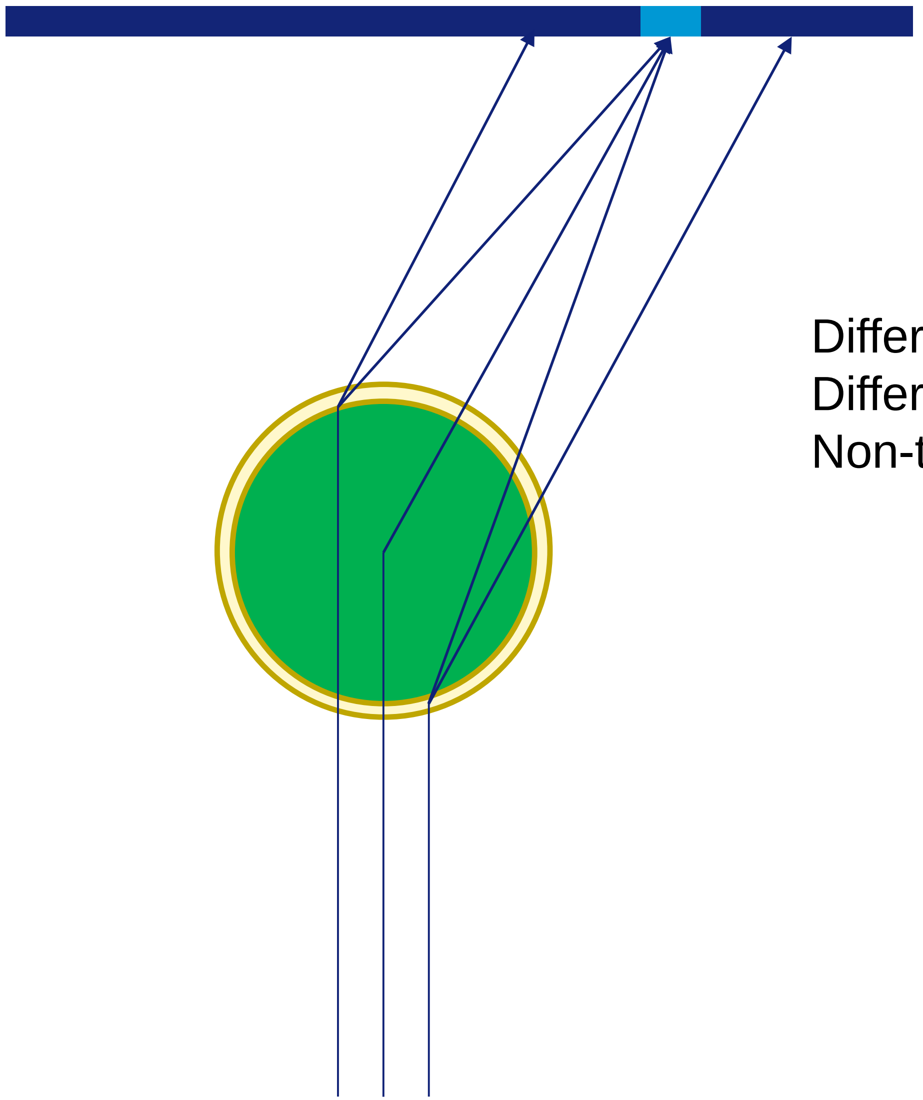


Resolution dependent on

- Pixel Size
- Sample – Detector Distance
- Beam Size
- Sample Size
- Intrinsic Broadening

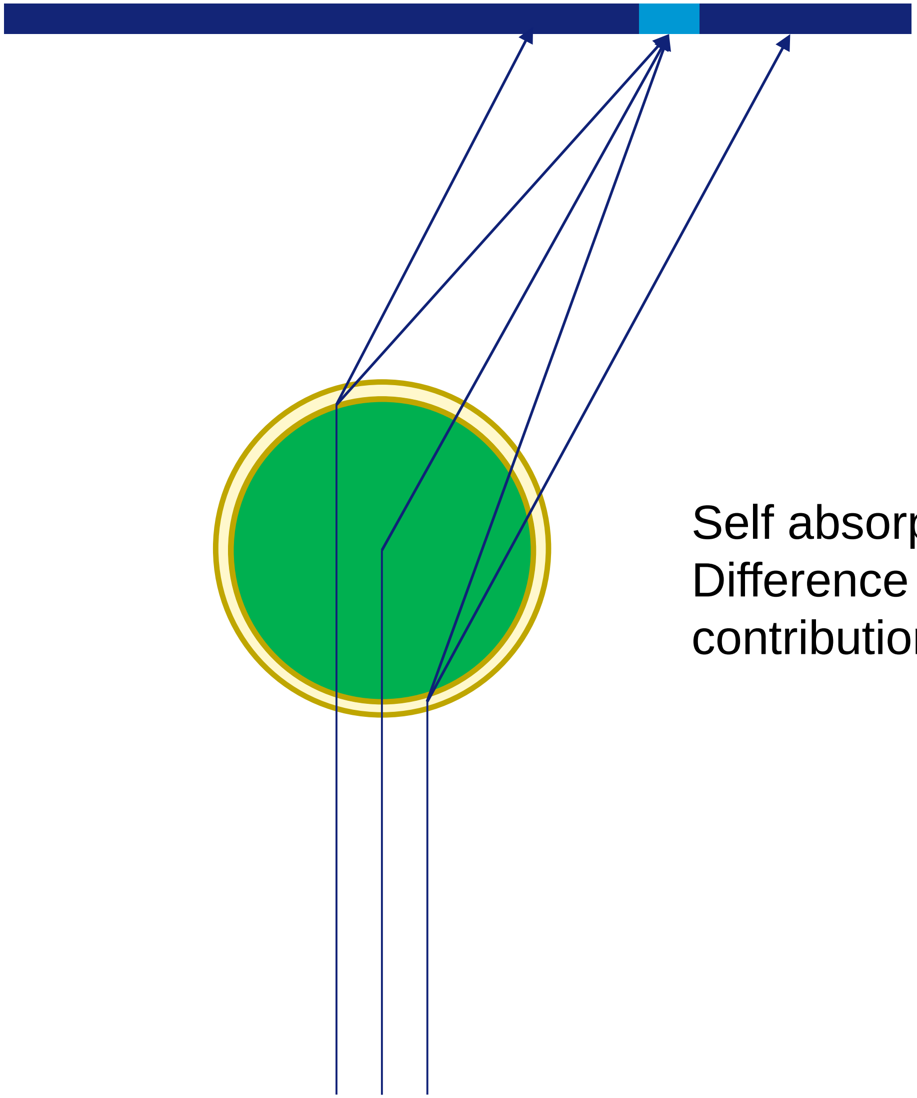
Conclusion:
For a given Q-range, best to use
Higher Energy/further distance

EFFECTS OF SAMPLE GEOMETRY – ABSORPTION



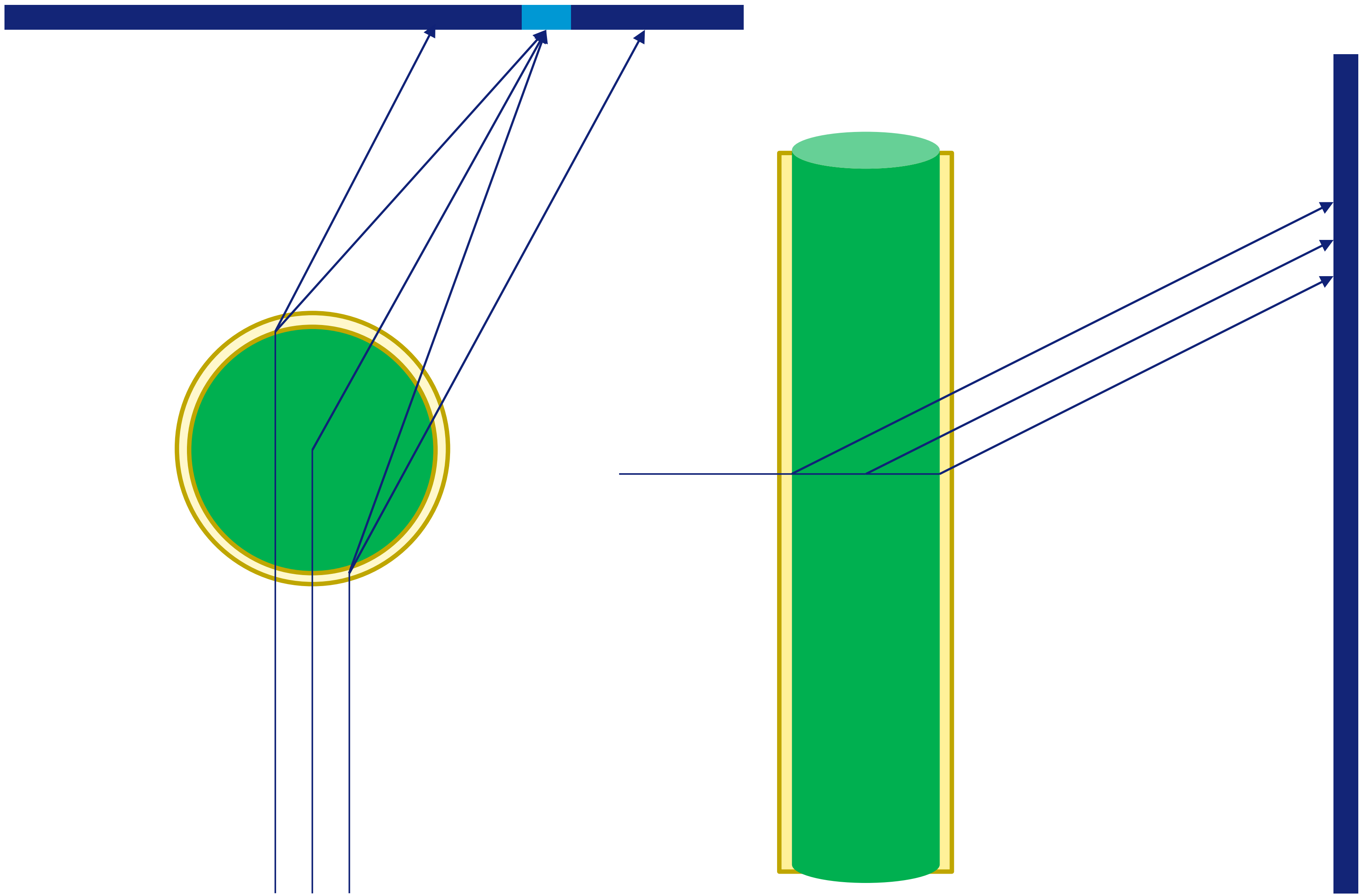
Different Rays have different pathlengths
Different angles have different signal
Non-trivial absorption correction

EFFECTS OF SAMPLE GEOMETRY – BACKGROUND SUBTRACTION



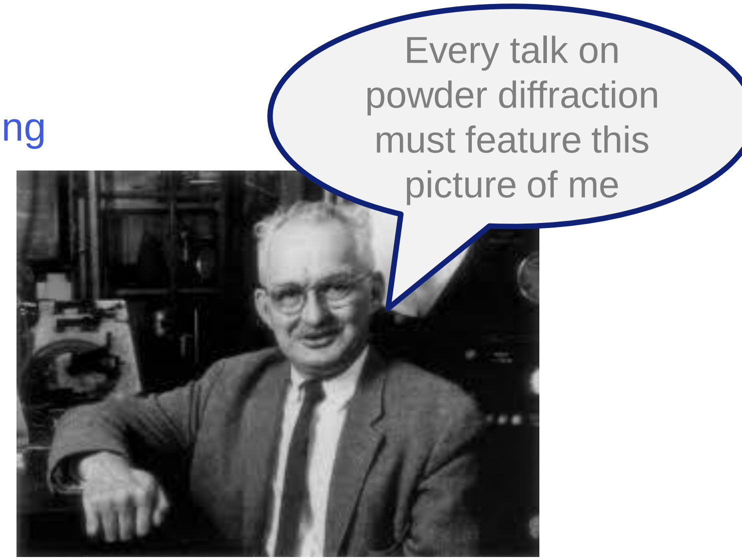
Self absorption affects background subtraction
Difference pattern will slightly oversubtract back contribution

EFFECTS OF SAMPLE GEOMETRY – 2D CASE

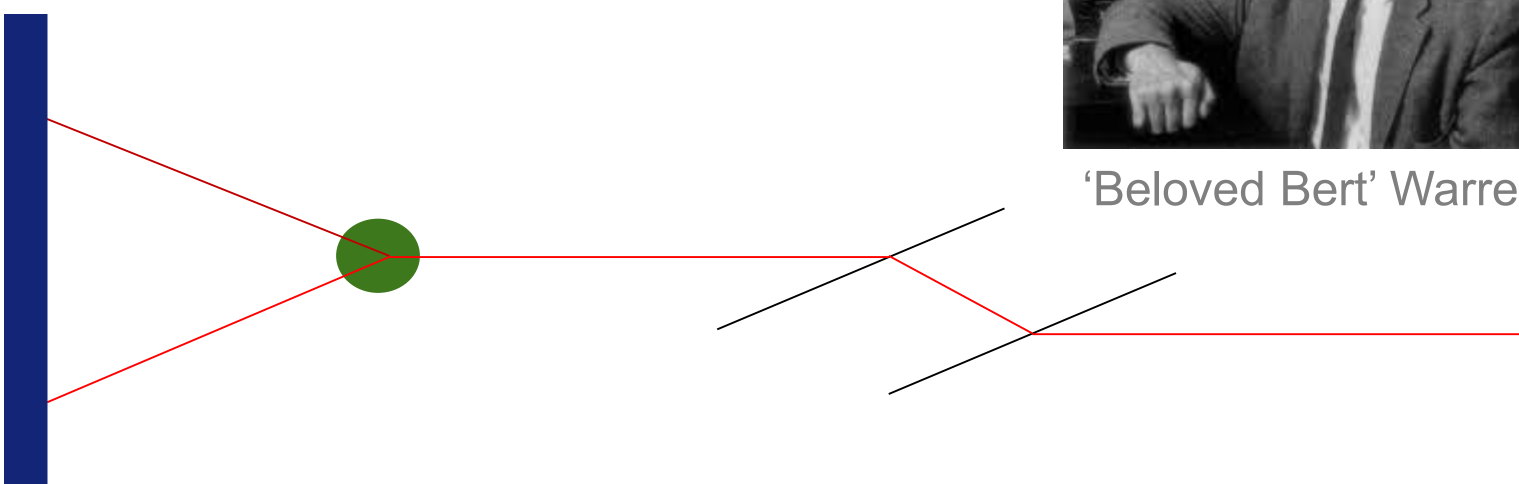


Polarization correction depends on

- **Scattering angle**
- **Azimuthal angle (synchrotron plane polarized)**
- **Optical and sample configuration**
  - Every scattering event affects the polarization
  - Right/left symmetry broken by sample scattering



'Beloved Bert' Warren



EFFECT OF SAMPLE GEOMETRY

Sample Geometry affects

- **Angular Resolution**
- **Absorption correction**
- **Background subtraction**

Convolution of (rapidly-varying) scattering pattern means that a proper treatment would require ray-tracing (algebraic reconstruction)

Achievable (in progress) but not in general plausible

- **Tomographic data collection**
- **Iterative computation**

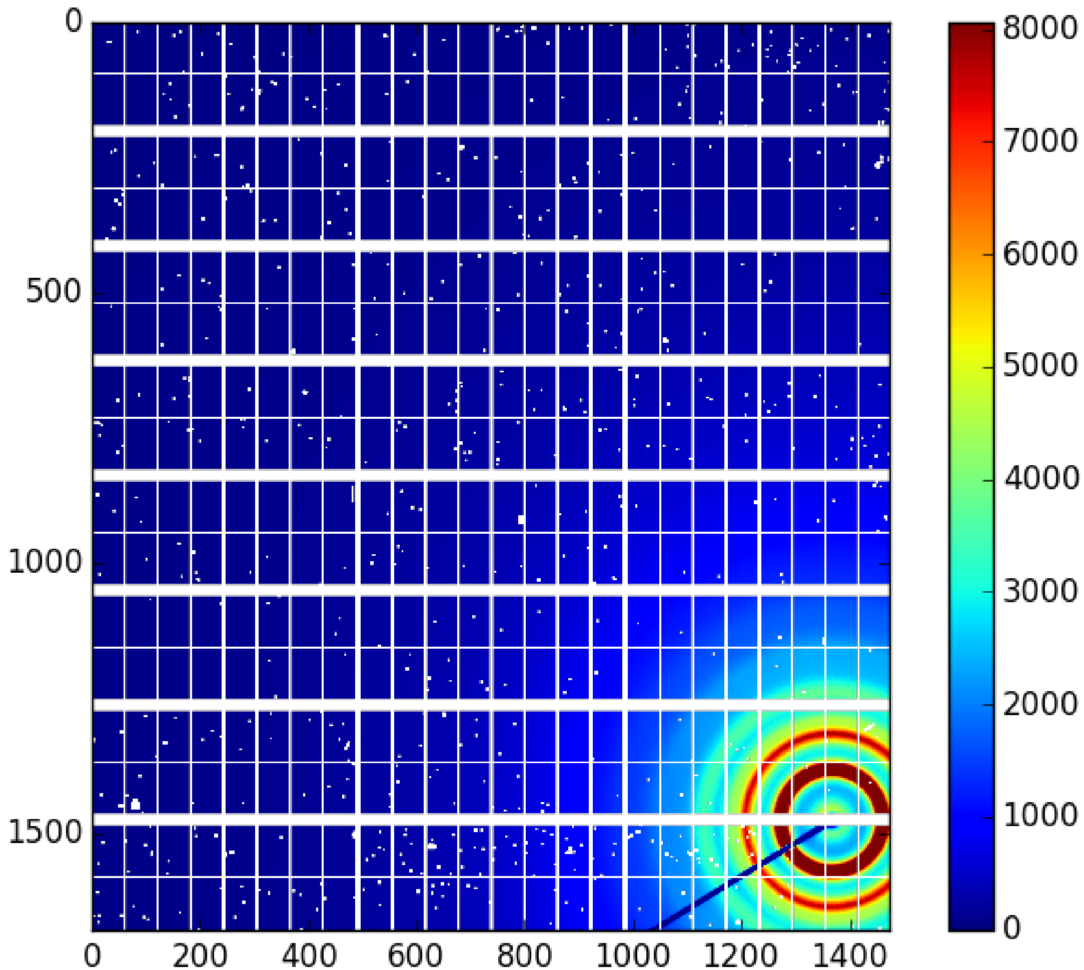
Precise polarization correction difficult to implement

- **This can be seen with noiseless detectors and good statistics at high Q**

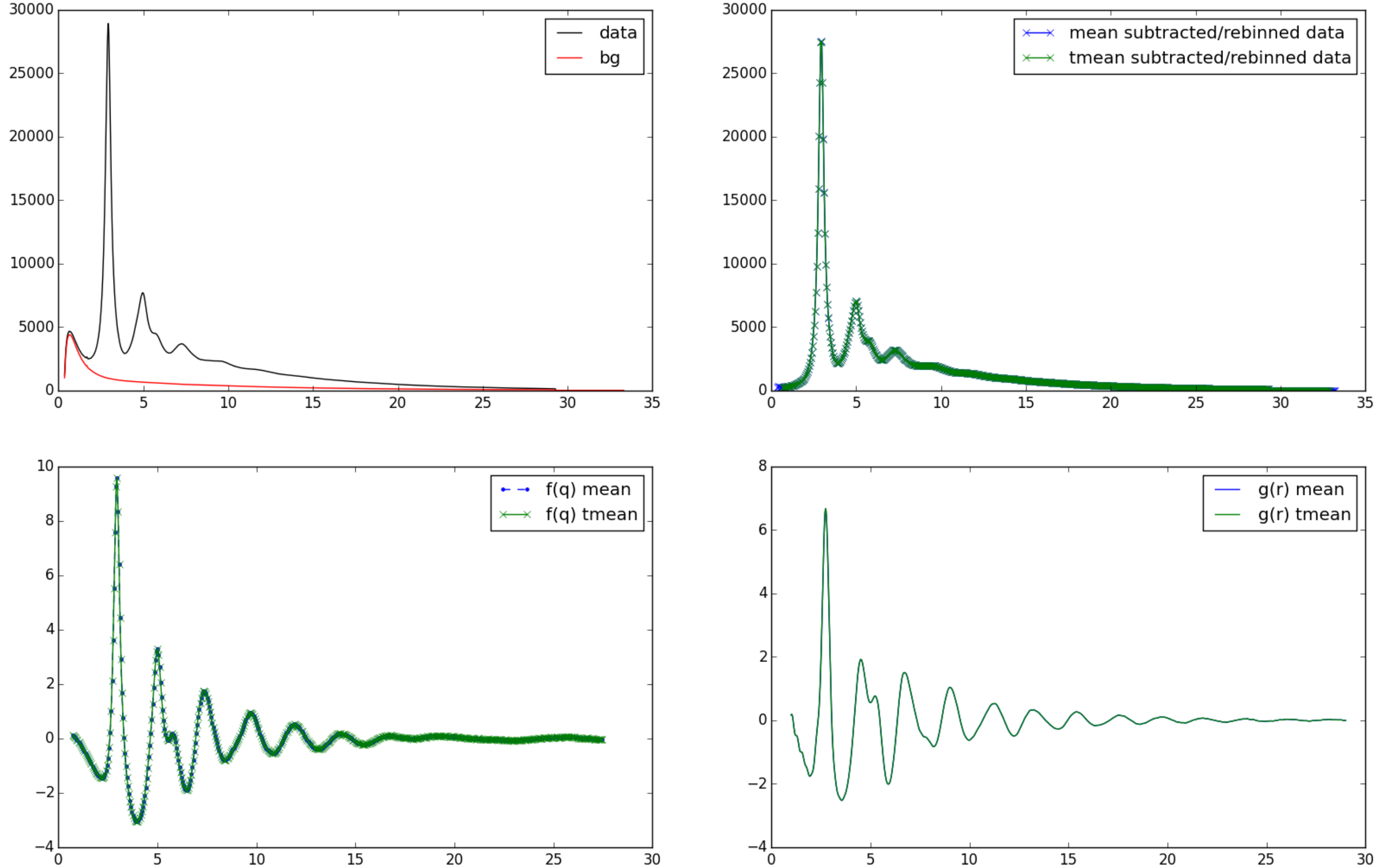
"Solution" ← i.e., work-around to hide the problems

- **Use either 90 or 360 azimuthal degrees**

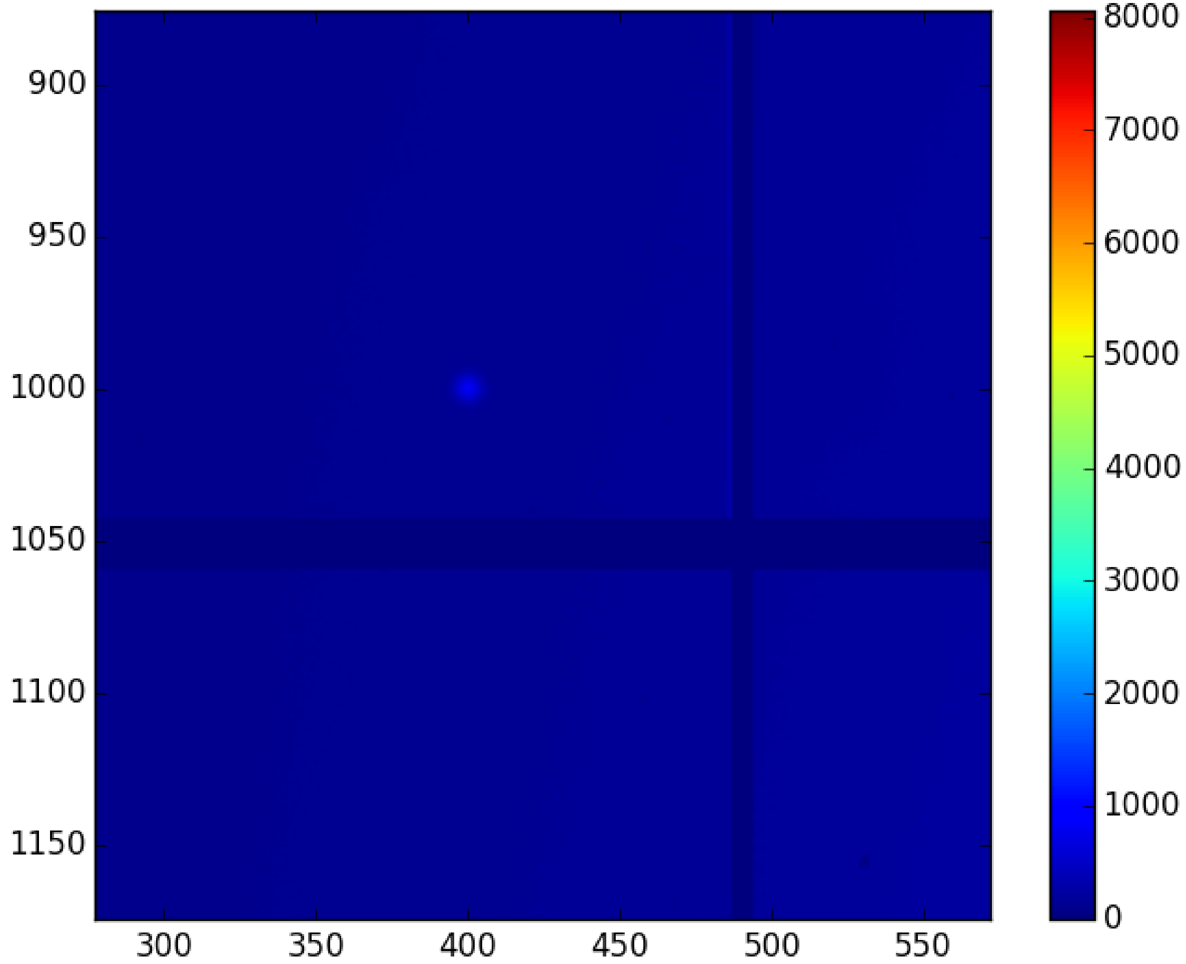
EXAMPLE OF A BMG - MASKING



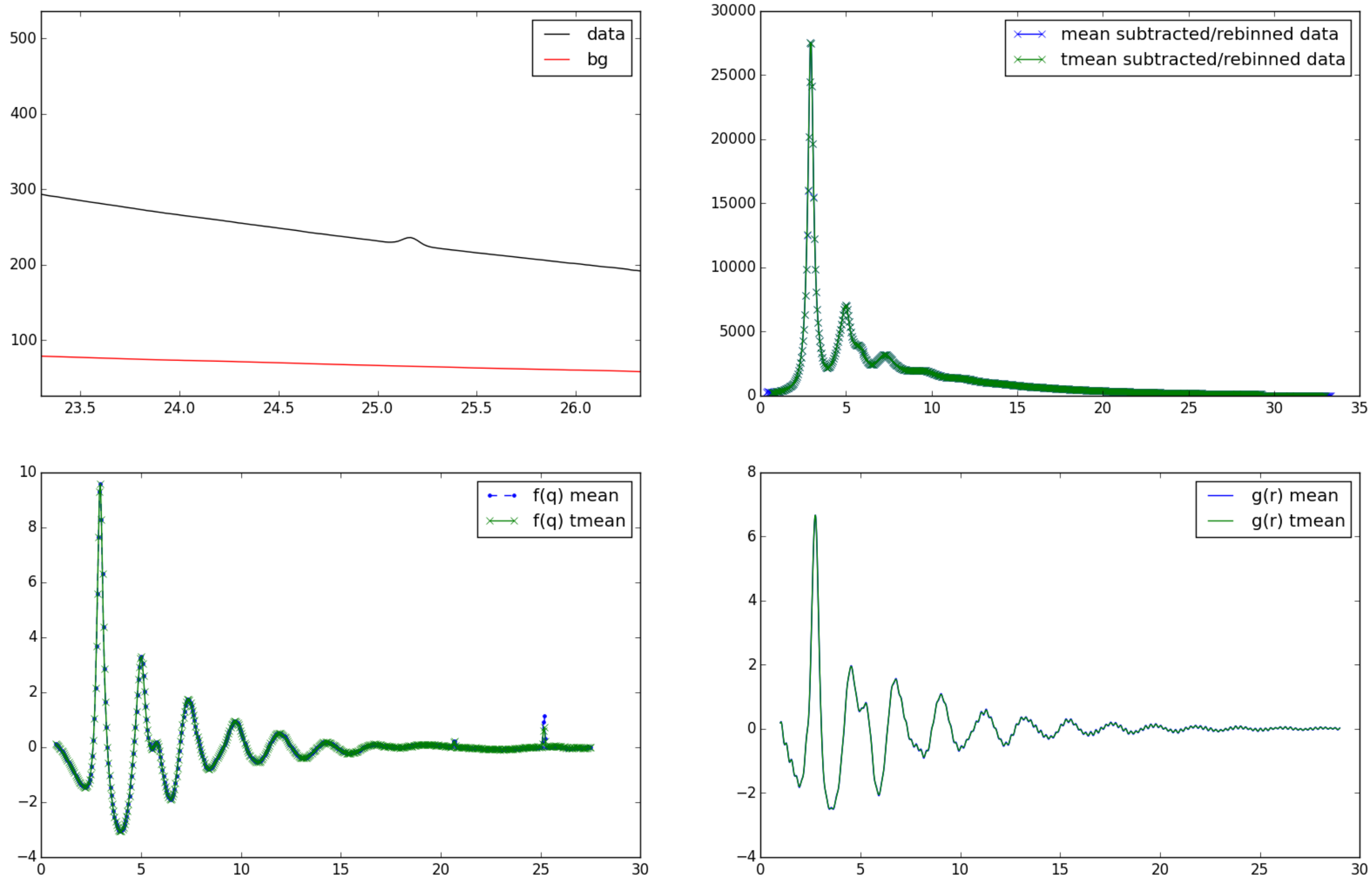
BMG


data
bg
mean subtracted/rebinned data
tmean subtracted/rebinned data
f(q) mean
f(q) tmean
g(r) mean
g(r) tmean

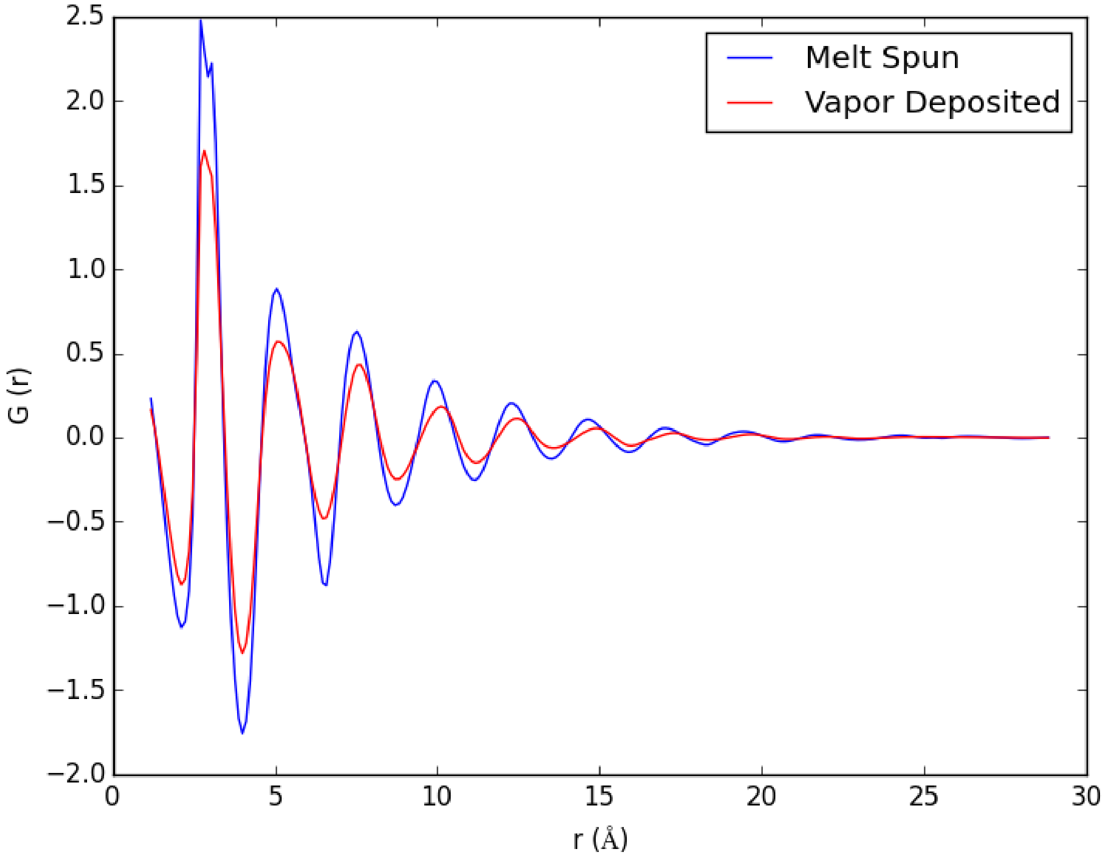
EFFECT OF NOISE ON G(R)



EFFECT OF NOISE ON G(R)



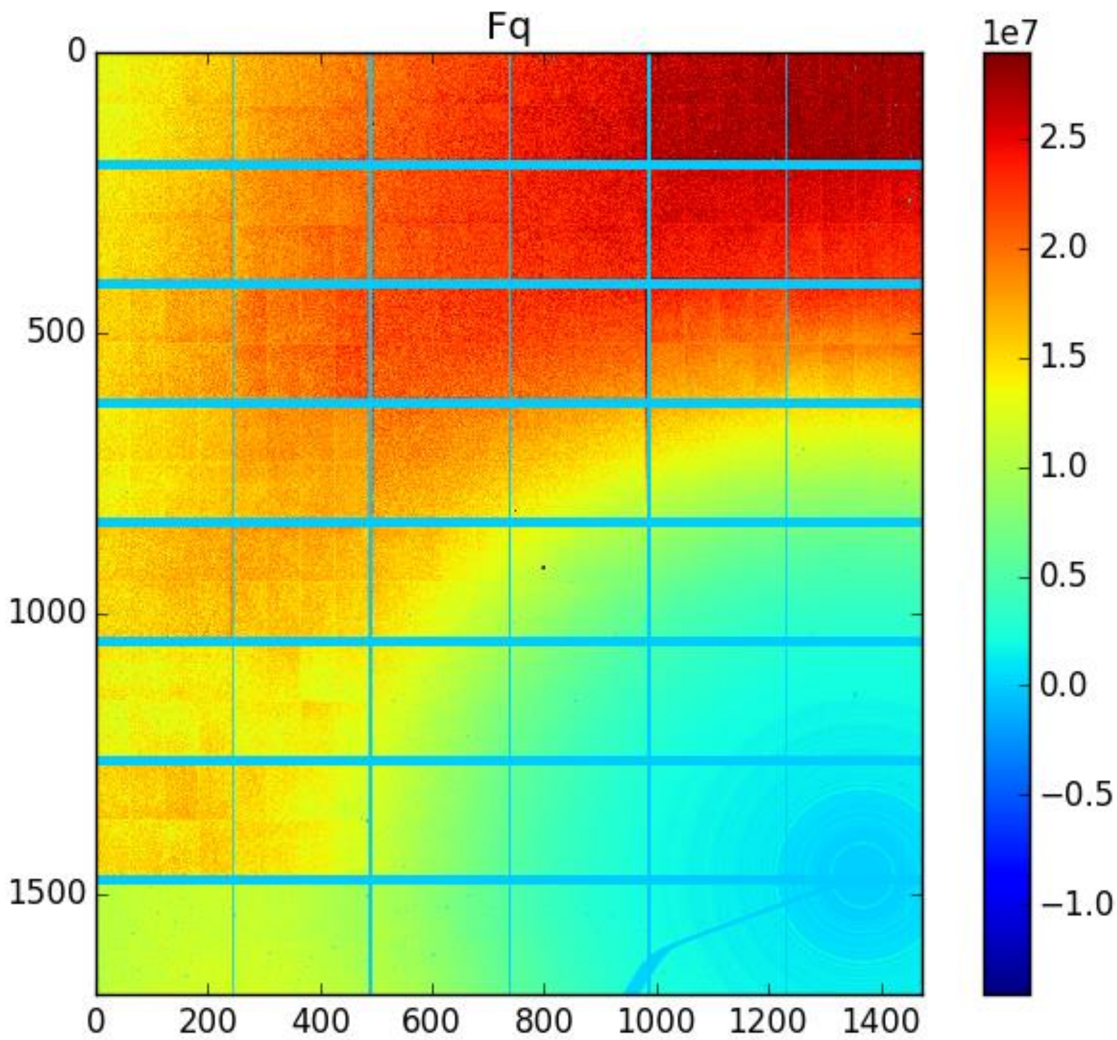
PDF OF ULTRATHIN GLASSES



The G(r) obtained from very thin (ca. 2 micron) thick films of BMG show the microstructural origin of the ultrastability of the CVP glasses

Luo et al, Nature Comm. ***9****, 1389 (2018)*

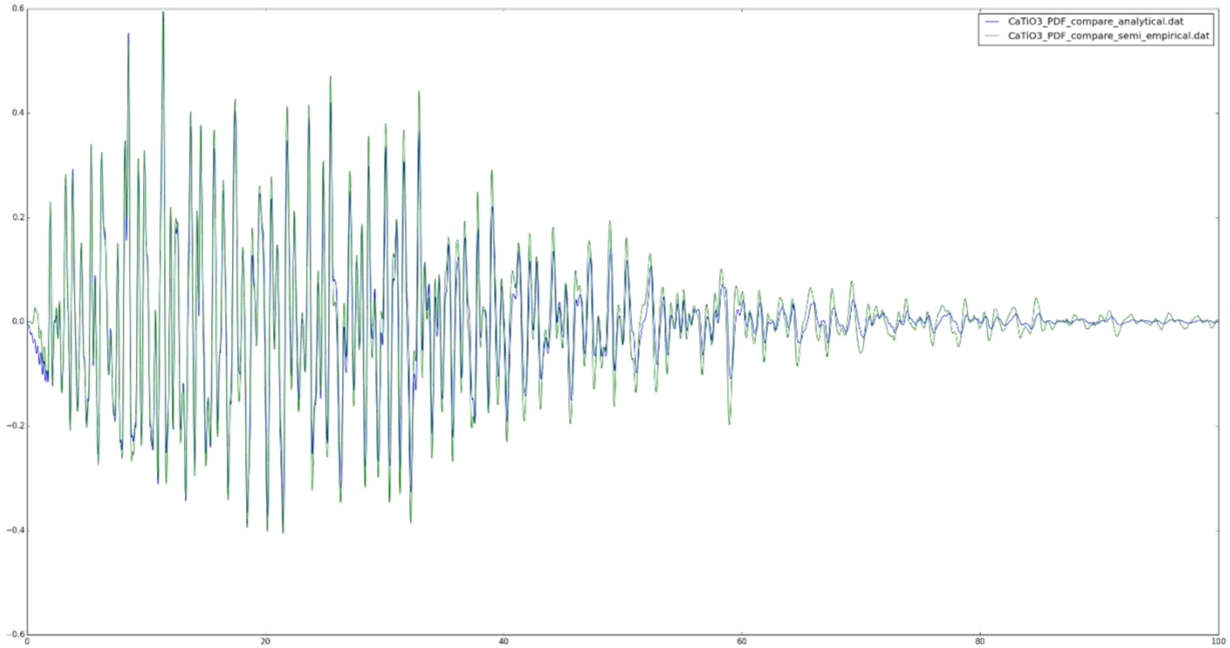
THE DREADFUL F(Q) ISSUE



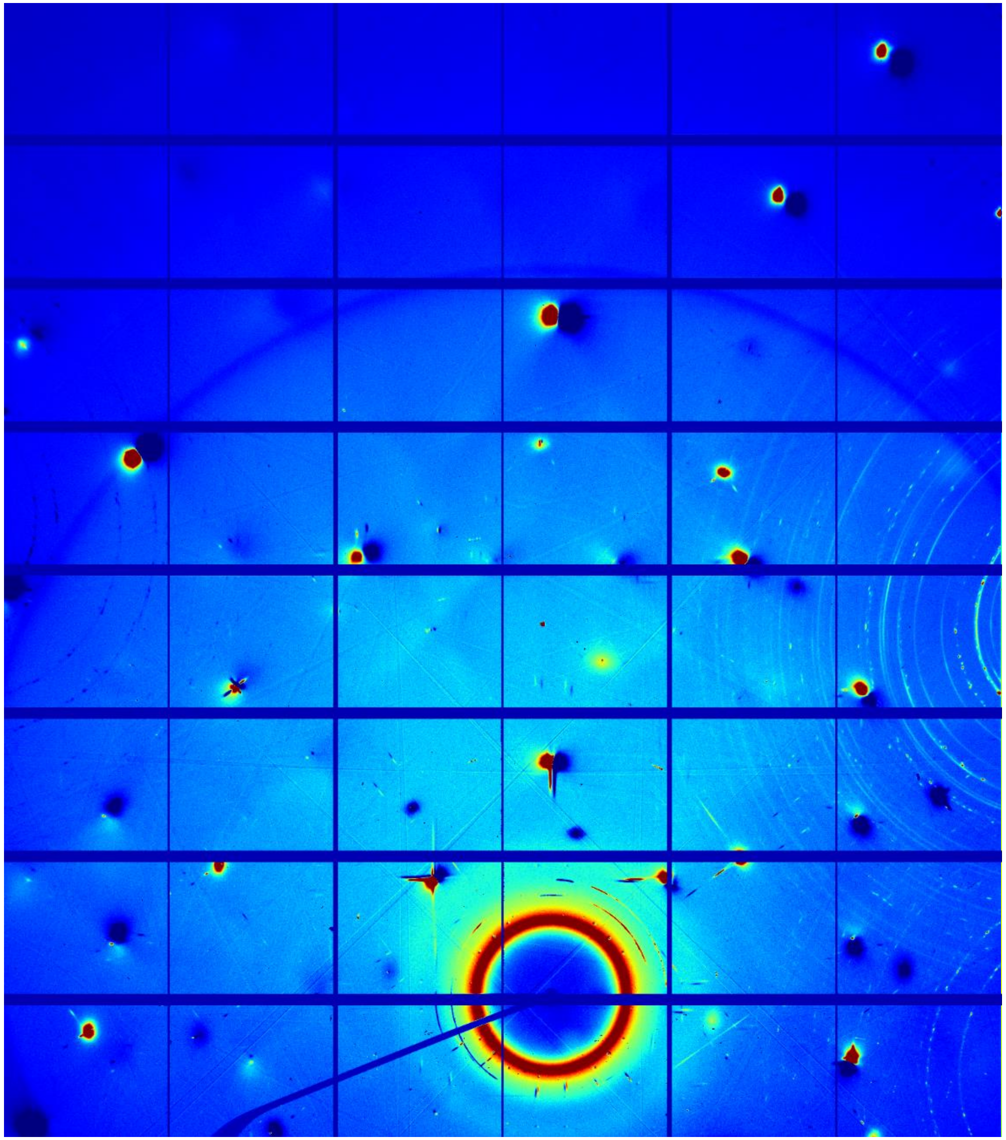
ANALYTICAL VS. SEMI-EMPIRICAL BACKGROUND SUBTRACTION

Inelastic Backgrounds can be subtracted by either

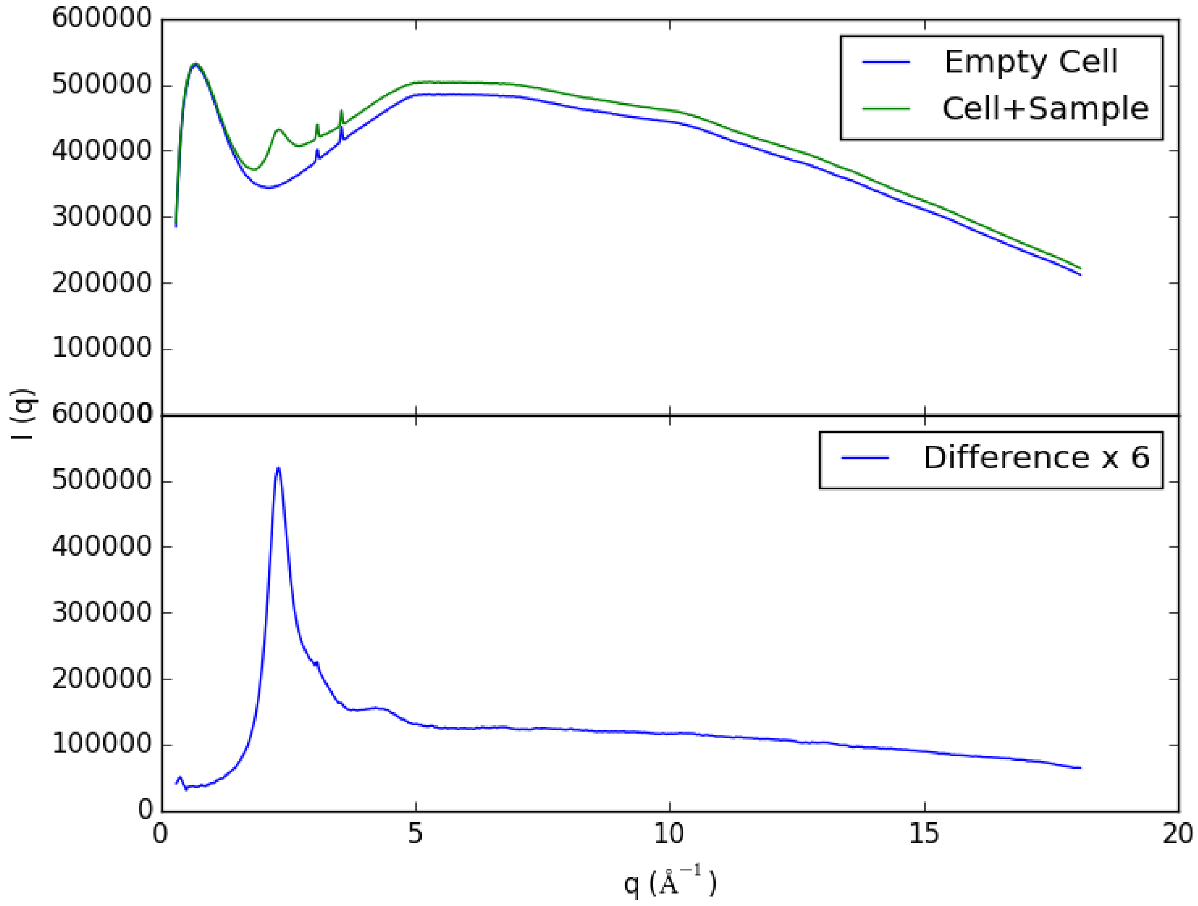
- **Analytical**
  - Correct form calculated and removed
- **Semi-Empirical**
  - Polynomial or spline representation for the effects
  - Form of the function respects analytical form



PDF IN DIFFICULT CIRCUMSTANCES

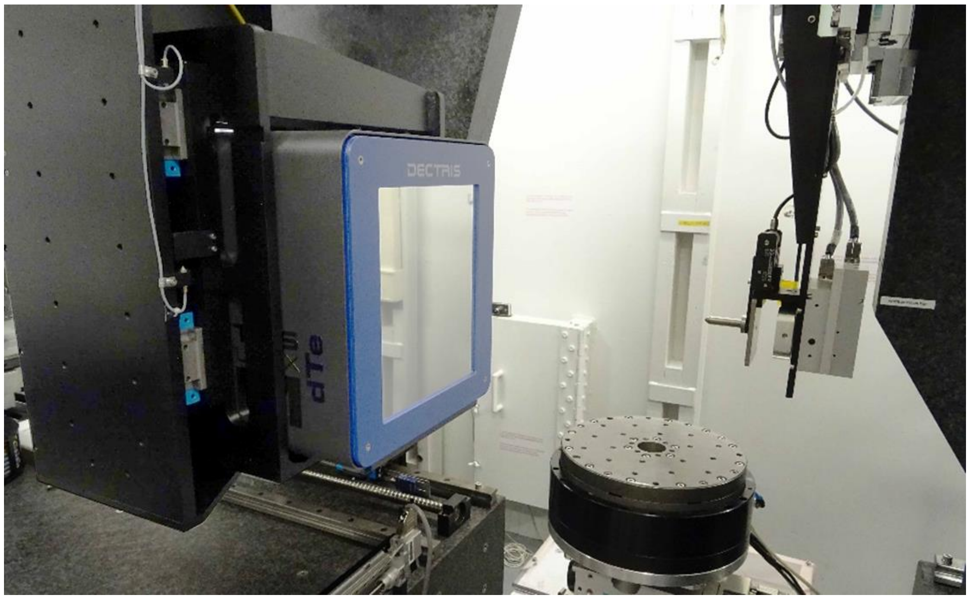


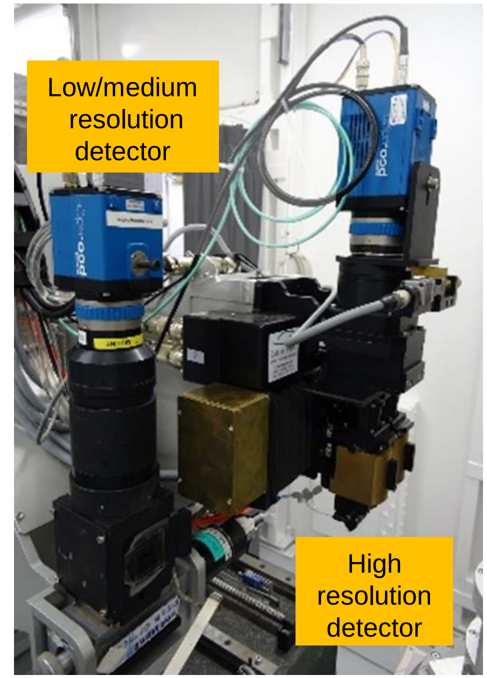
PDF IN DACS



Signal from the material of interest is a small fraction of the total signal

PARALLEL OPTICS AND DETECTORS



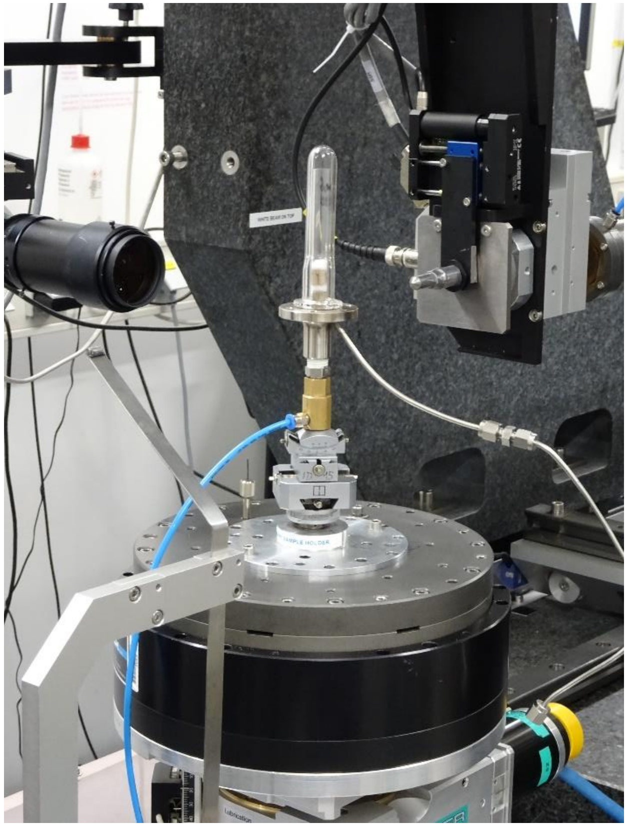


Parallel mounting of diffraction detector and 3 imaging detectors for rapid change of configuration. Permanently pre-aligned.

Time to change between different configurations (energy/beam size/detector)
On the order of 1 minute.

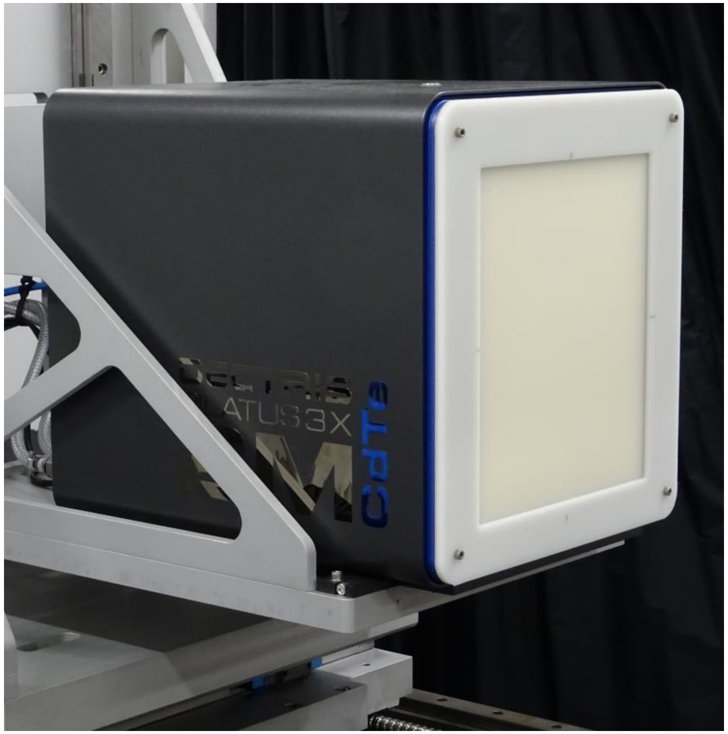
EXPERIMENTAL SETUP

Experimental setup



- High speed high precision rotation and translation stages
- Gas heaters or furnace
- Mass Spectrometer
- Gas delivery system

Pilatus3X 2M CdTe



- Pixel size 172µm x 172µm
- Single photon counting
- 20-bit dynamic range
- Linear up to more than 1Mcps
- Maximum frame rate 250Hz (500 with ROI)

CONCLUSION

- **Every new advance in data quality reveals new problems to resolve**
- **Data quality from 2d detectors is now approaching that of point detectors/analyser crystal**
  - ms resolution is now possible
  - Sub-micron resolution already achieved