Diffractometers GEM and Polaris: Instruments for Total Scattering from Crystalline Materials

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2nd UK-China Joint Workshop on Neutron Scattering
CSNS, Dongguan, China, 6-9th November 2016

Science & Technology Facilities Council ISIS

Total scattering from crystalline powders

$$g_{ij}(r) - 1 = \frac{1}{(2\pi)^3 \rho_0} \int_0^\infty 4\pi Q^2 [A_{ij}(Q) - 1] \frac{\sin Qr}{Qr} \, \mathrm{d}Q$$

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- As high a maximum Q as possible; correlations (bonds) can be sharp so high-Q data contains real information.

$$Q = 4\pi \sin\theta / \lambda$$

- As good a Q-resolution as possible; Bragg peaks are sharp!

$$\mathrm{d}Q / Q \simeq \mathrm{d}\theta / \tan\theta \; (+ \, \mathrm{d}\lambda / \lambda +)$$

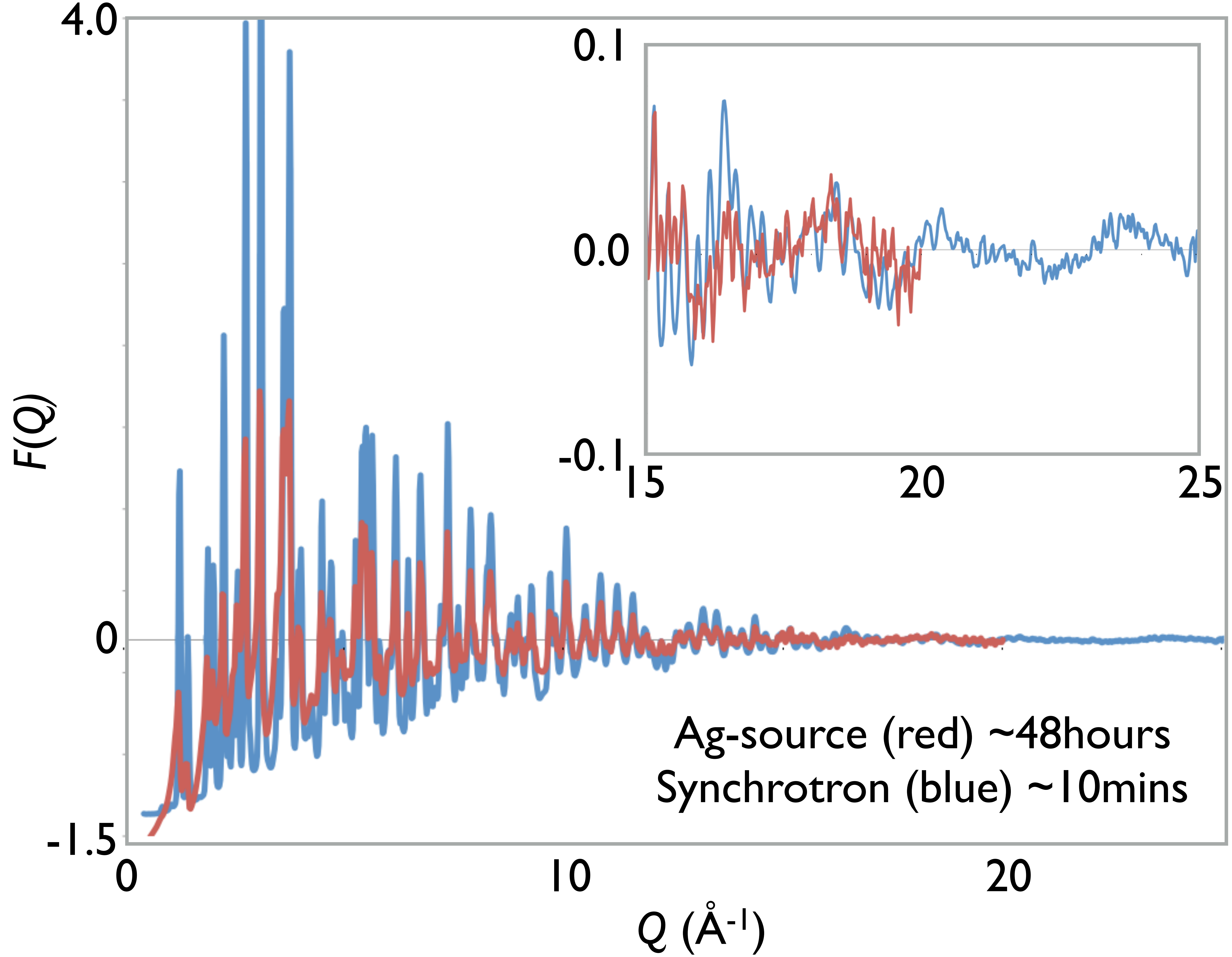
- Hence high scattering angles, 2θ, and short wavelengths, λ, are good.
- High count-rates; especially at high-Q

Total scattering from crystalline powders

Lab-source X-ray vs. Synchrotron X-ray

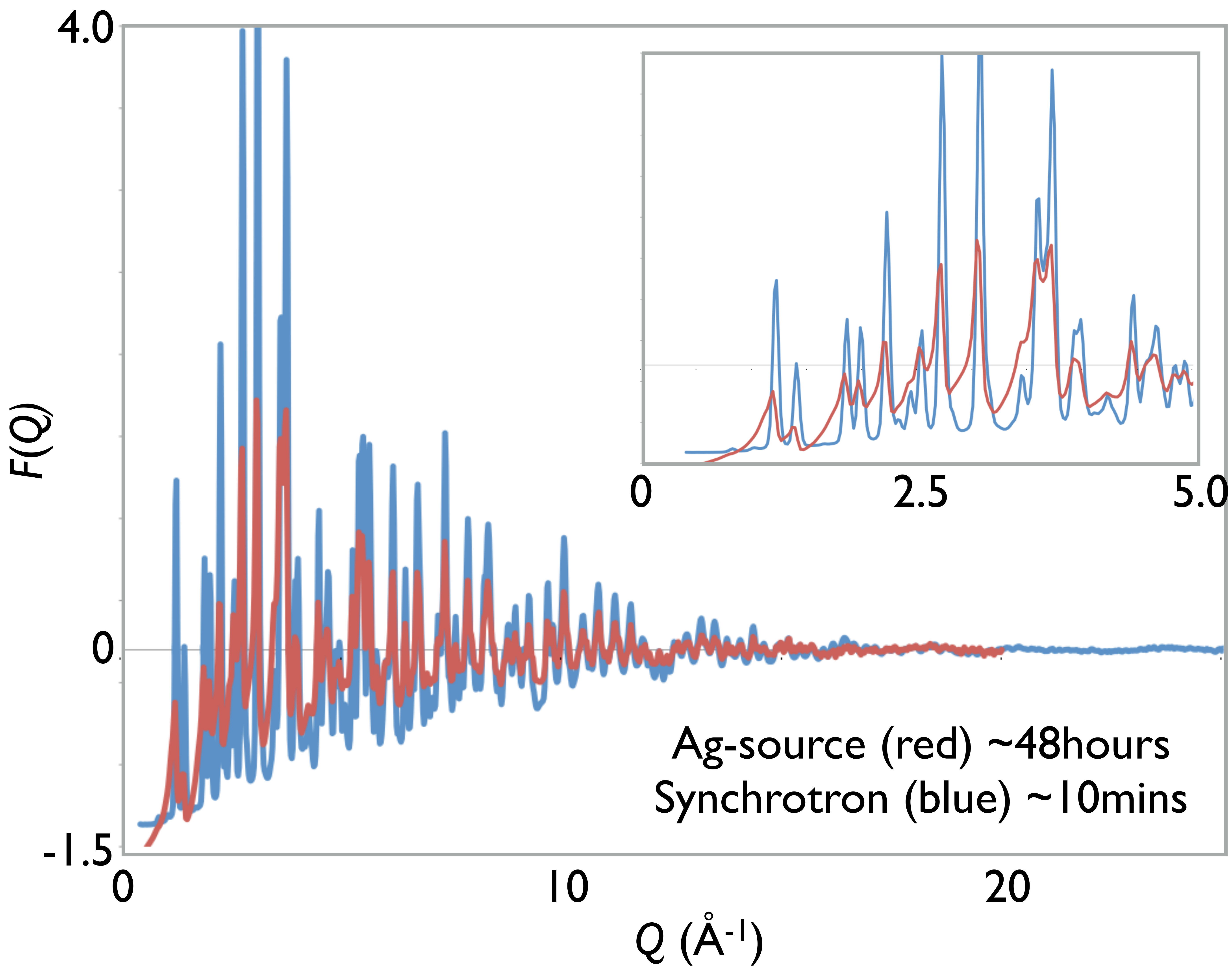
Total scattering from crystalline powders

Lab-source X-ray vs. Synchrotron X-ray



Total scattering from crystalline powders

Lab-source X-ray vs. Synchrotron X-ray



Total scattering from crystalline powders

Reactor neutron vs. Spallation neutron

Total scattering from crystalline powders

Reactor neutron vs. Spallation neutron

Reactor

- Single wavelength
- Low *Q*-resolution
- Limited Q_{max}~24Å^{-1}
- Higher neutron flux
- More restricted scattering geometry
- Simpler data analysis

Spallation source

- Polychromatic
- High *Q*-resolution
- High Q_{max}~50Å^{-1}
- Lower neutron flux
- Less restricted scattering geometry
- More complex data analysis

Total scattering from crystalline powders

Synchrotron X-ray vs. Spallation neutron

Total scattering from crystalline powders

Synchrotron X-ray vs. Spallation neutron

X-ray

- High Q-resolution
- Lower Q_{max}~35Å^{-1}
- High flux, rapid measurements
- Approximately normalised data

Neutron

- High Q-resolution
- High Q_{max}~50Å^{-1}
- Lower flux, longer measurements
- Absolutely normalised structure factors

…or combine X-ray and neutron measurements

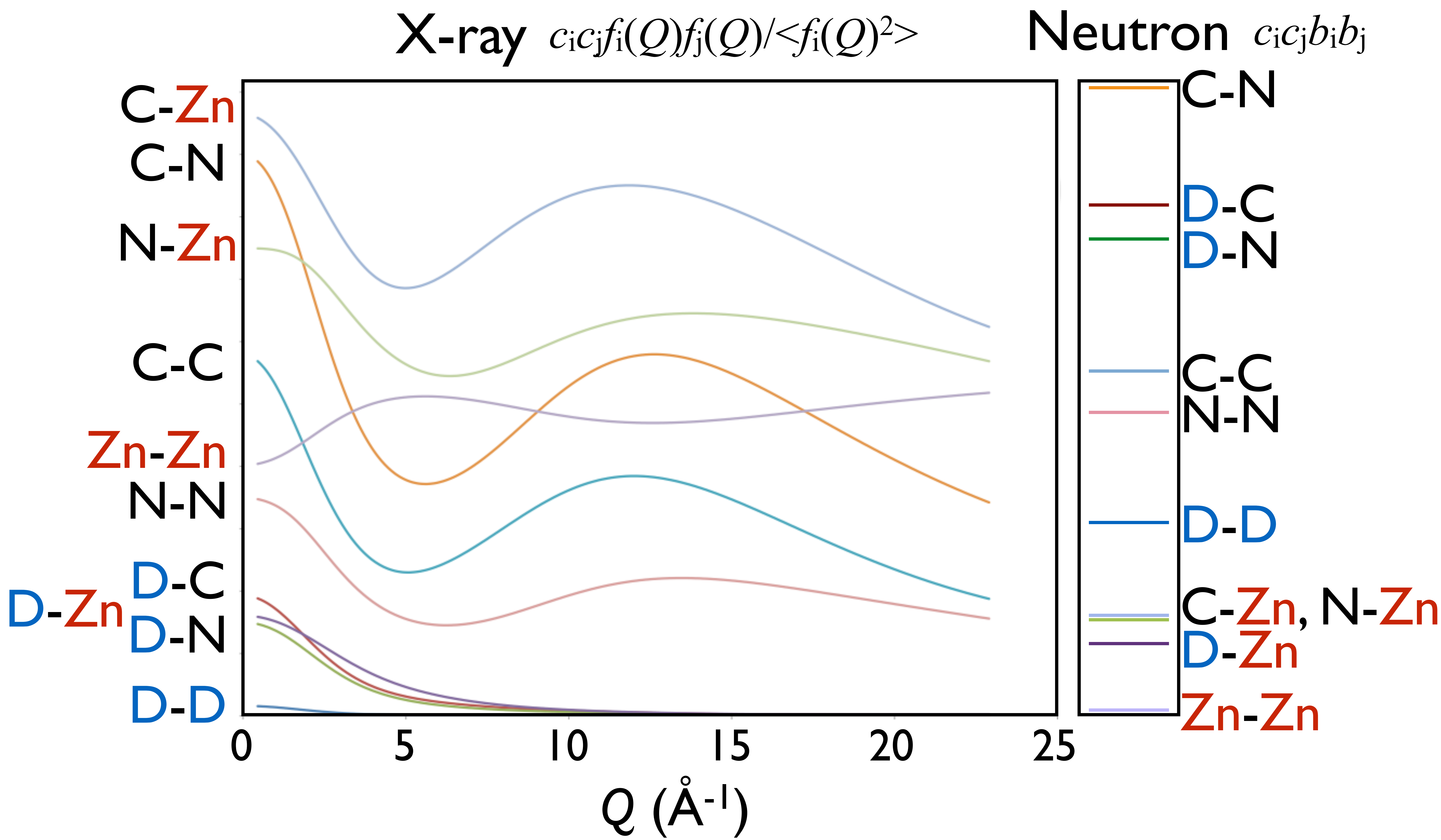
$$F(Q) = \sum_{i,j=1}^{n} c_i c_j \bar{b}_i \bar{b}_j [A_{ij}(Q) - 1]$$

e.g. MOF $Zn(C_3N_2H_3)_2$

…or combine X-ray and neutron measurements

$$F(Q) = \sum_{i,j=1}^{n} c_i c_j \bar{b}_i \bar{b}_j [A_{ij}(Q) - 1]$$

e.g. MOF $Zn(C_3N_2H_3)_2$



Summary: Use Central Facilities!

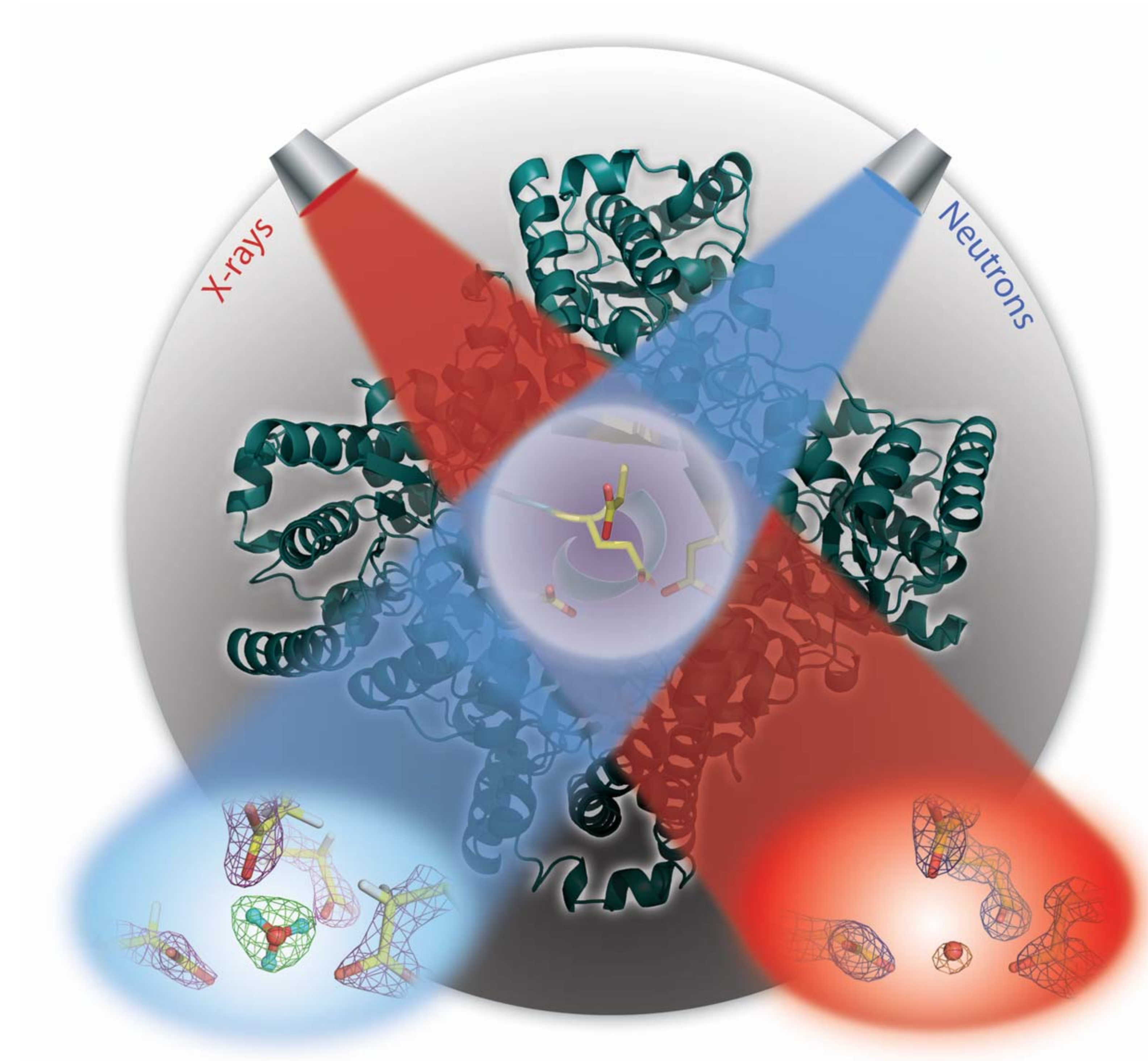
- Neutrons and x-rays 'see' things differently

$$F(Q) = \sum_{i,j=1}^{n} c_i c_j \bar{b}_i \bar{b}_j [A_{ij}(Q) - 1]$$

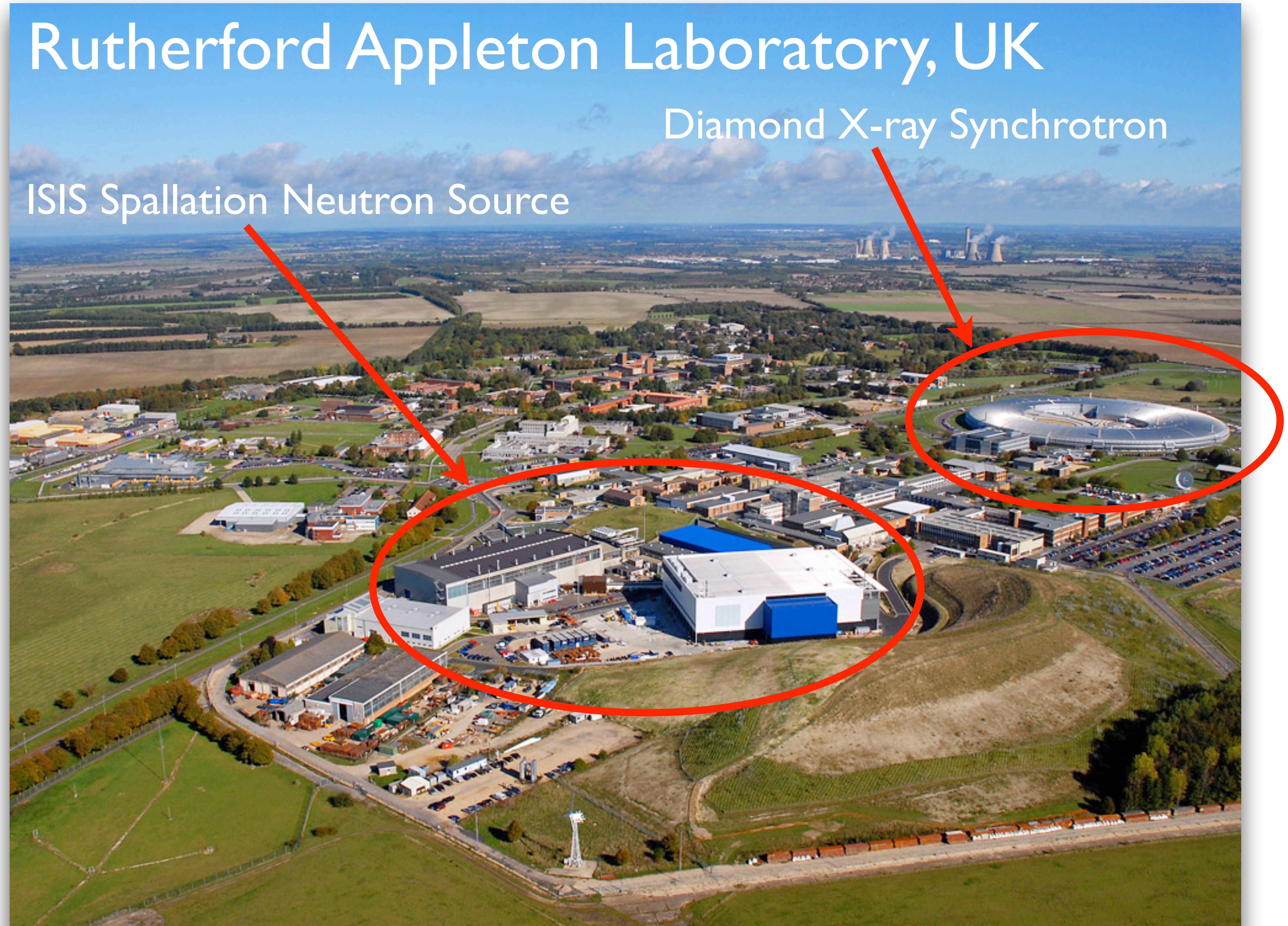
- Data may be measured faster, to higher resolution and with greater precision

$$g_{ij}(r) - 1 = \frac{1}{(2\pi)^3 \rho_0} \int_0^\infty 4\pi Q^2 [A_{ij}(Q) - 1] \frac{\sin Qr}{Qr} \, dQ$$

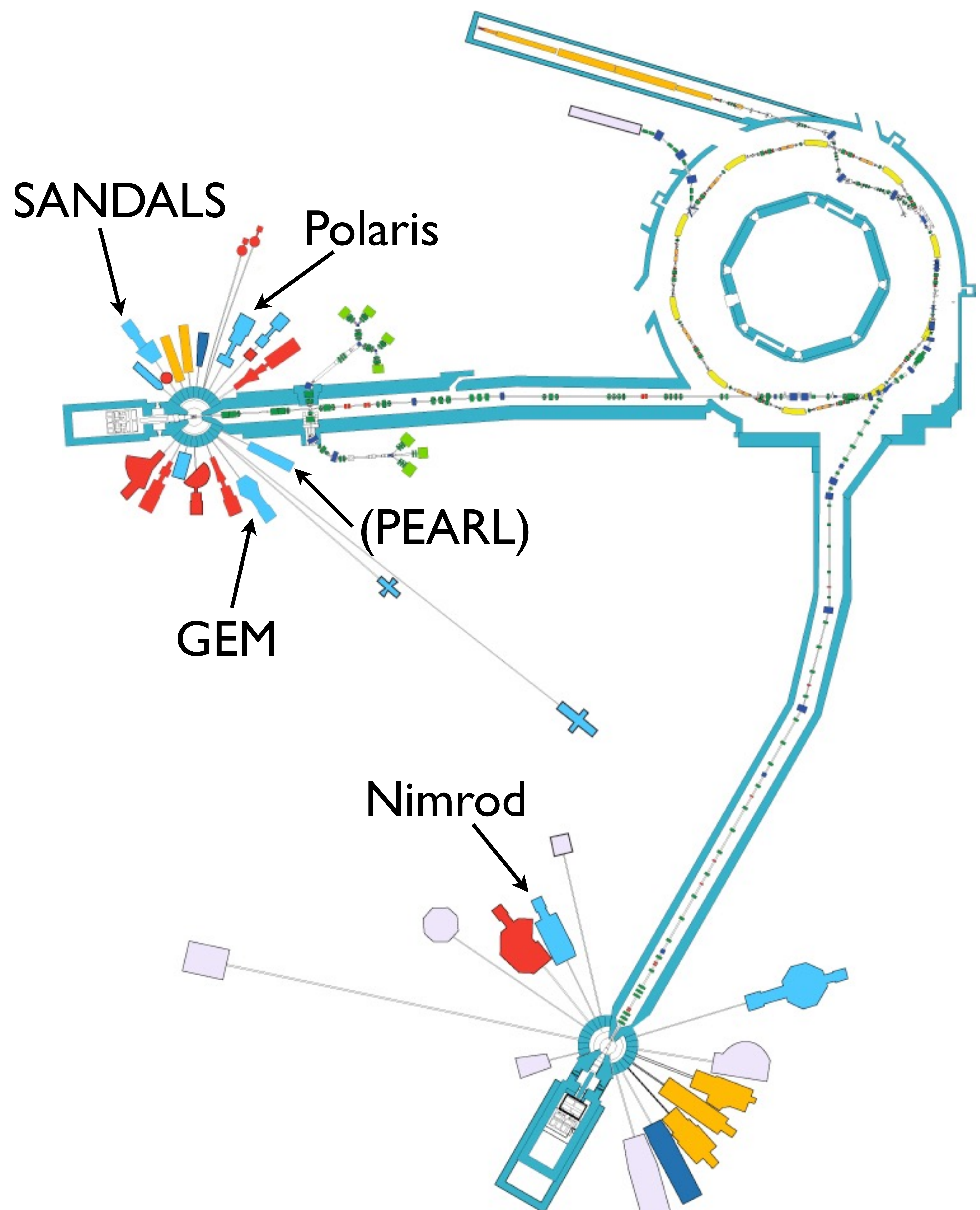
- (Spallation) neutron PDF is still the 'gold standard'!
- The resources are there...with more being built



Central facilities for total scattering

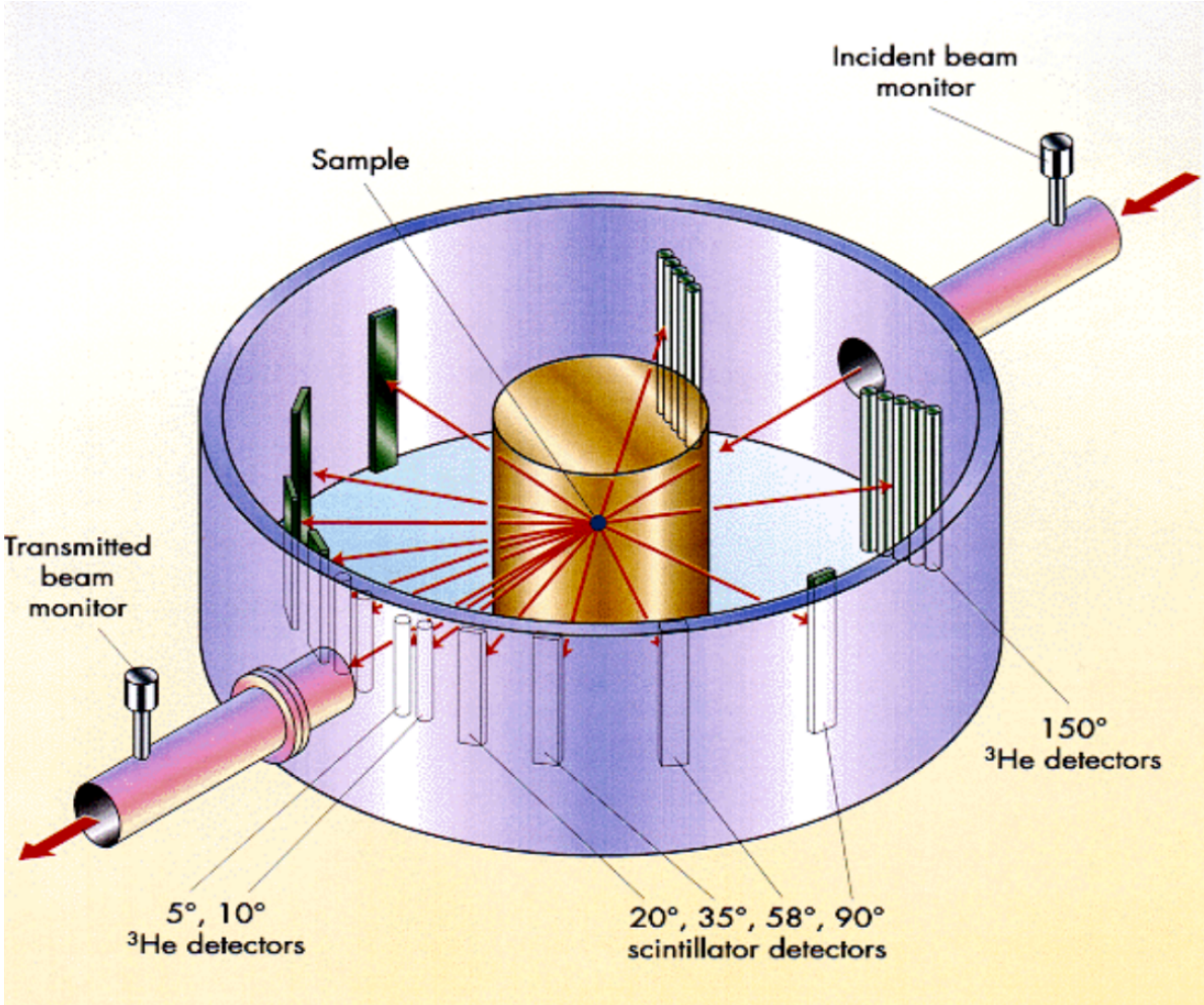


Total scattering instruments at ISIS

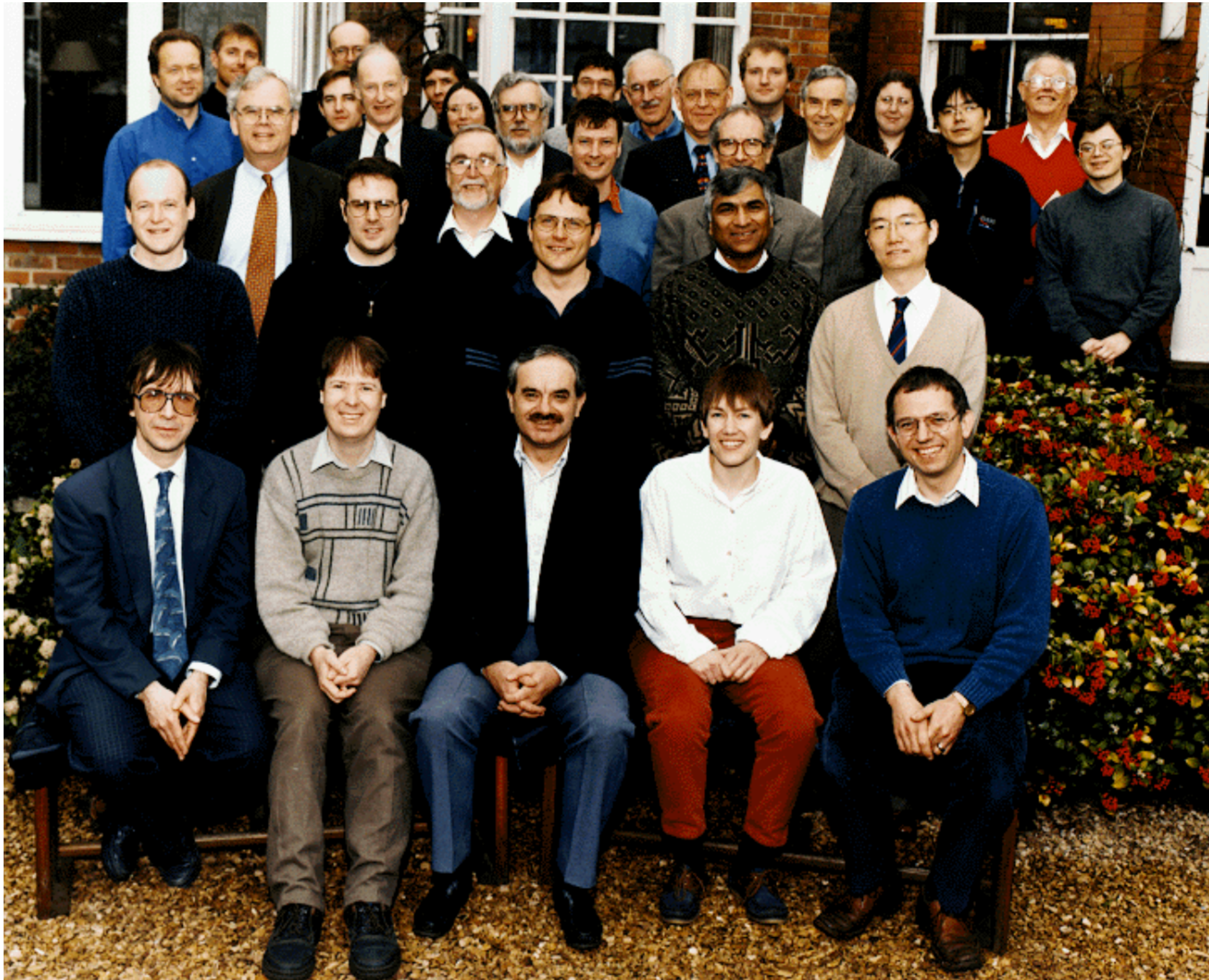


The Liquids and Amorphous Diffractometer (1982-1998)

The LAD diffractometer at ISIS (1984 - 1998)



LAD farewell meeting March 1999



LAD farewell meeting March 1999

LAD instrument parameters

	LAD (1984−1998)
• Moderator	Methane @110K
• Primary flight path	10 m
• Wavelength range	~0.1 − 10Å
• Detector banks	7
• Detector coverage	0.016 steradians
• Resolution DQ/Q	14 − 0.6%

Lessons learnt:

Stability

Low backgrounds

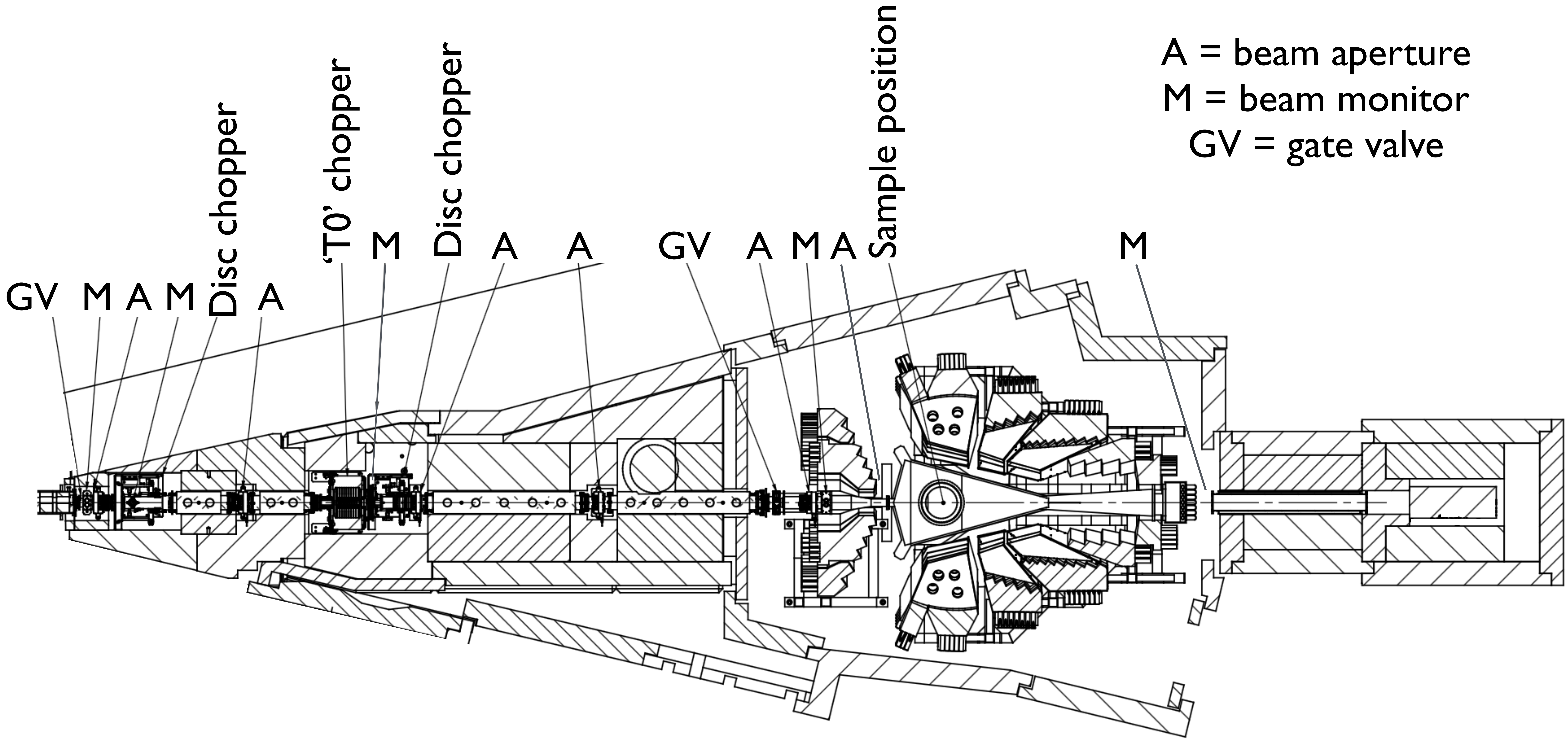
High count-rate

Good angle coverage

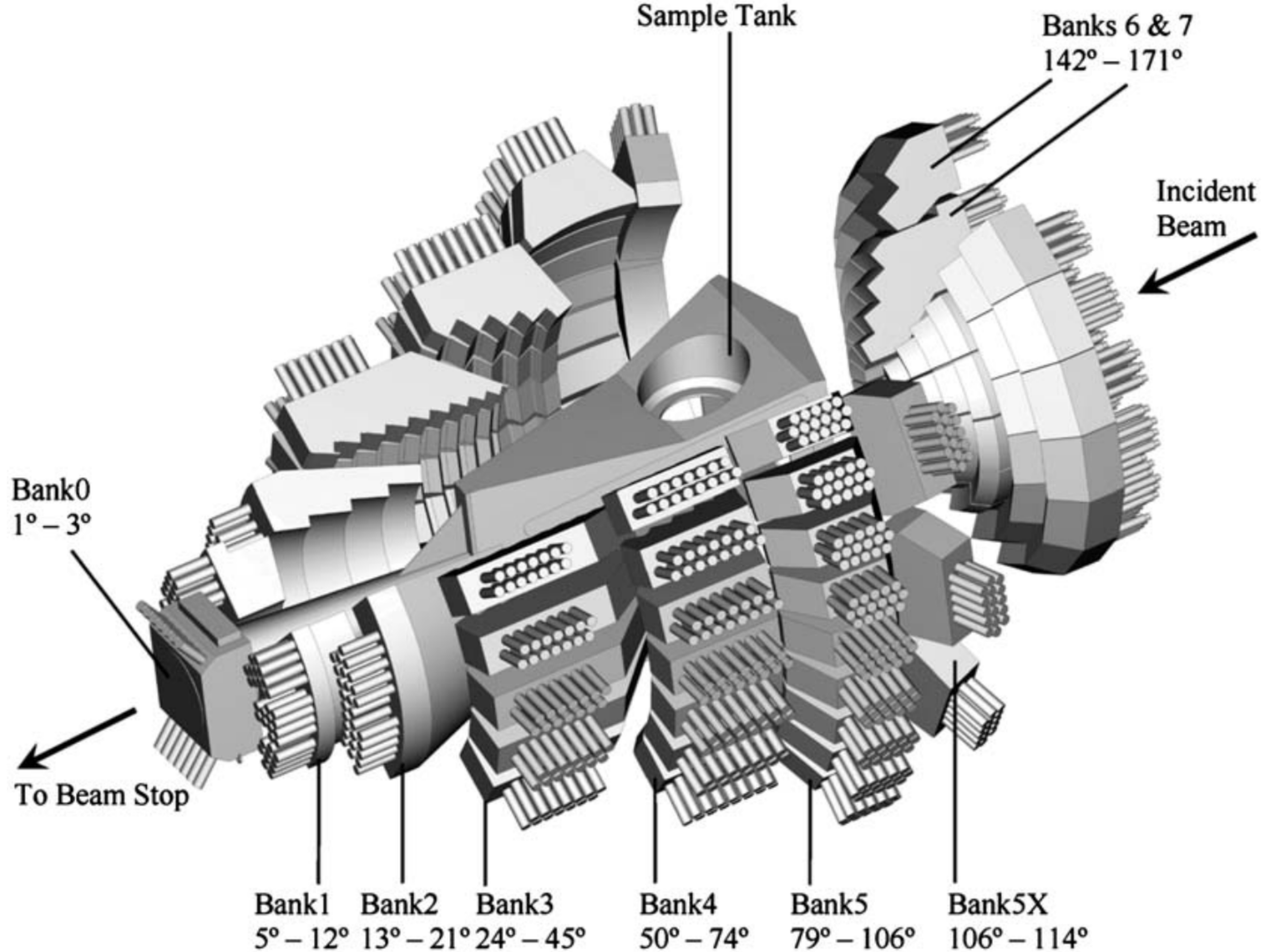
Short(er) wavelengths

W S Howells & A C Hannon *J Phys: Condensed Matter* **11** (1999) 9127

The GEM total scattering diffractometer at ISIS



The GEM total scattering diffractometer at ISIS



A C Hannon *Nucl Inst Meth A* **551** (2005) 88

GEM and LAD instrument parameters

	LAD (1984–1998)	*GEM (~1999–)*
• Moderator	Methane @110K	Methane @110K
• Primary flight path	10 m	17m
• Wavelength range	~0.1 – 10Å	~0.1 – 3.4Å
• Detector banks	7	7 (8)
• Detector coverage	0.016 steradians	3.86st
• Resolution DQ/Q	14 – 0.6%	~10 – 0.34%

A C Hannon *Nucl Inst Meth A* **551** (2005) 88

GEM detector coverage

Detector Bank	Scattering angle, 2θ (°)	Azimuthal angle, ø (°)	*L*2 (m)	Solid angle (st)	Δ*Q*/*Q* (%)	Q_{min} (Å^{-1})
0	1.2-3.2	±90	~2.76	0.008	5-10	0.04
1	5.3-12.7	±45	~2.37	0.056	4.7	0.17
2	13.4-21.6	±43.4	1.48-2.10	0.093	2.4	0.43
3	24.7-45.6	±42.5	1.08-1.89	0.478	1.7	0.79
4	50.1-74.7	±44.4	1.03-1.44	0.988	0.79	1.56
5	79.1-114.2	±44.5	~1.38	1.513	0.51	2.95
6	142.5-149.7	±69.3	1.54-1.74	0.280	0.34	3.50
7	150.0-171.4	±66.6	1.04-1.39	0.443	0.35	3.57

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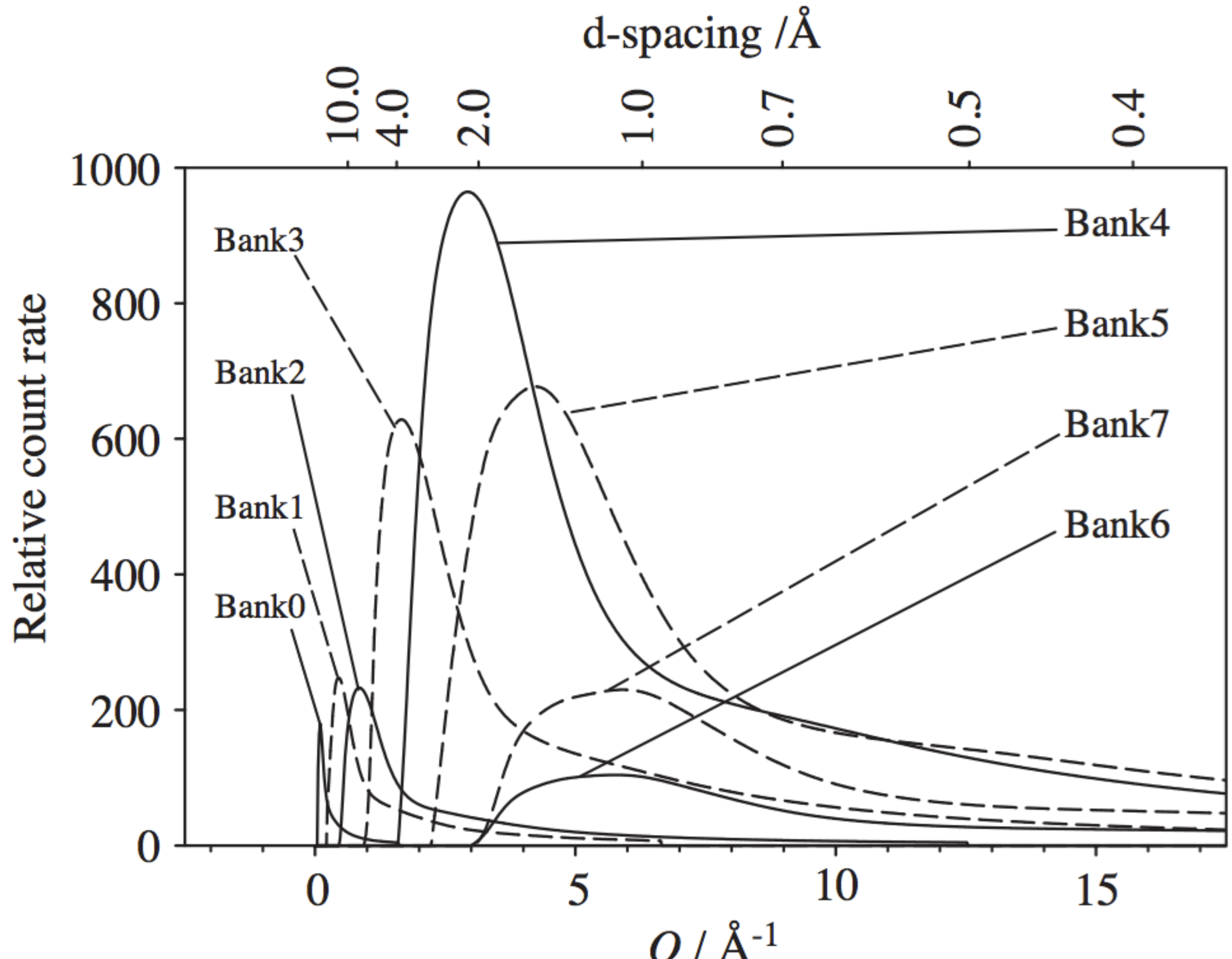
Relative count-rate of GEM detector banks



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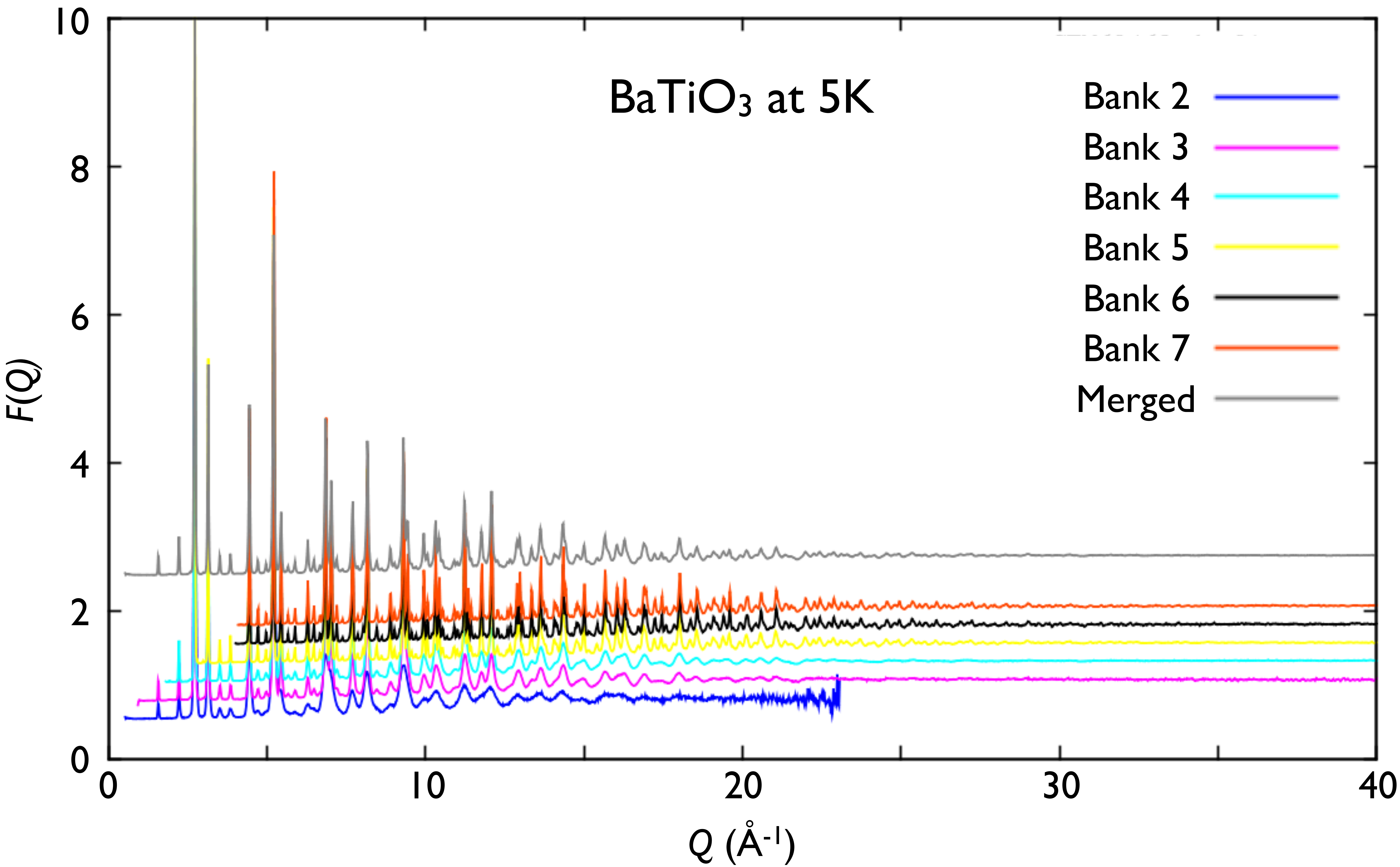
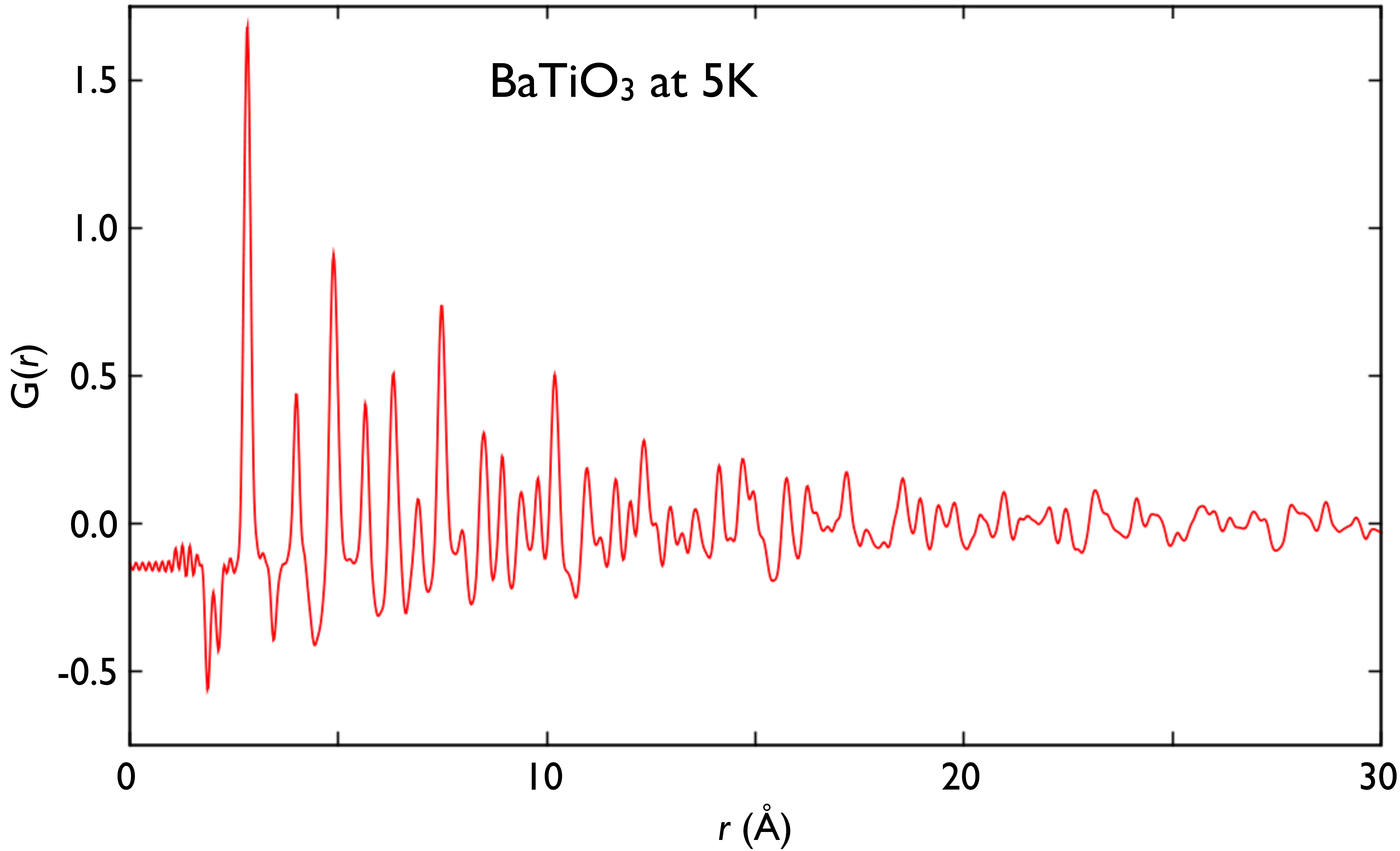
Corrected total scattering data from GEM



Corrected PDF data from GEM



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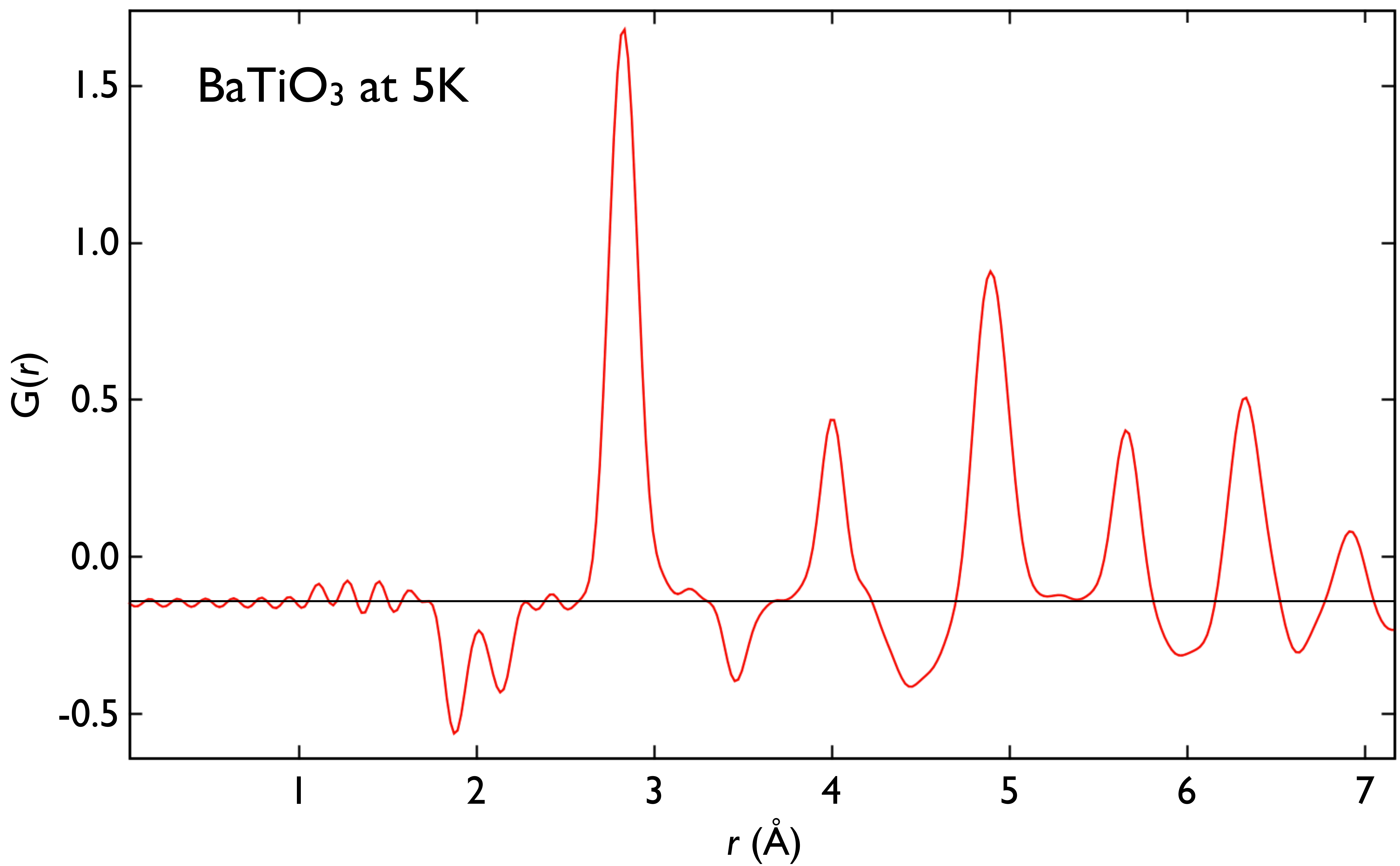

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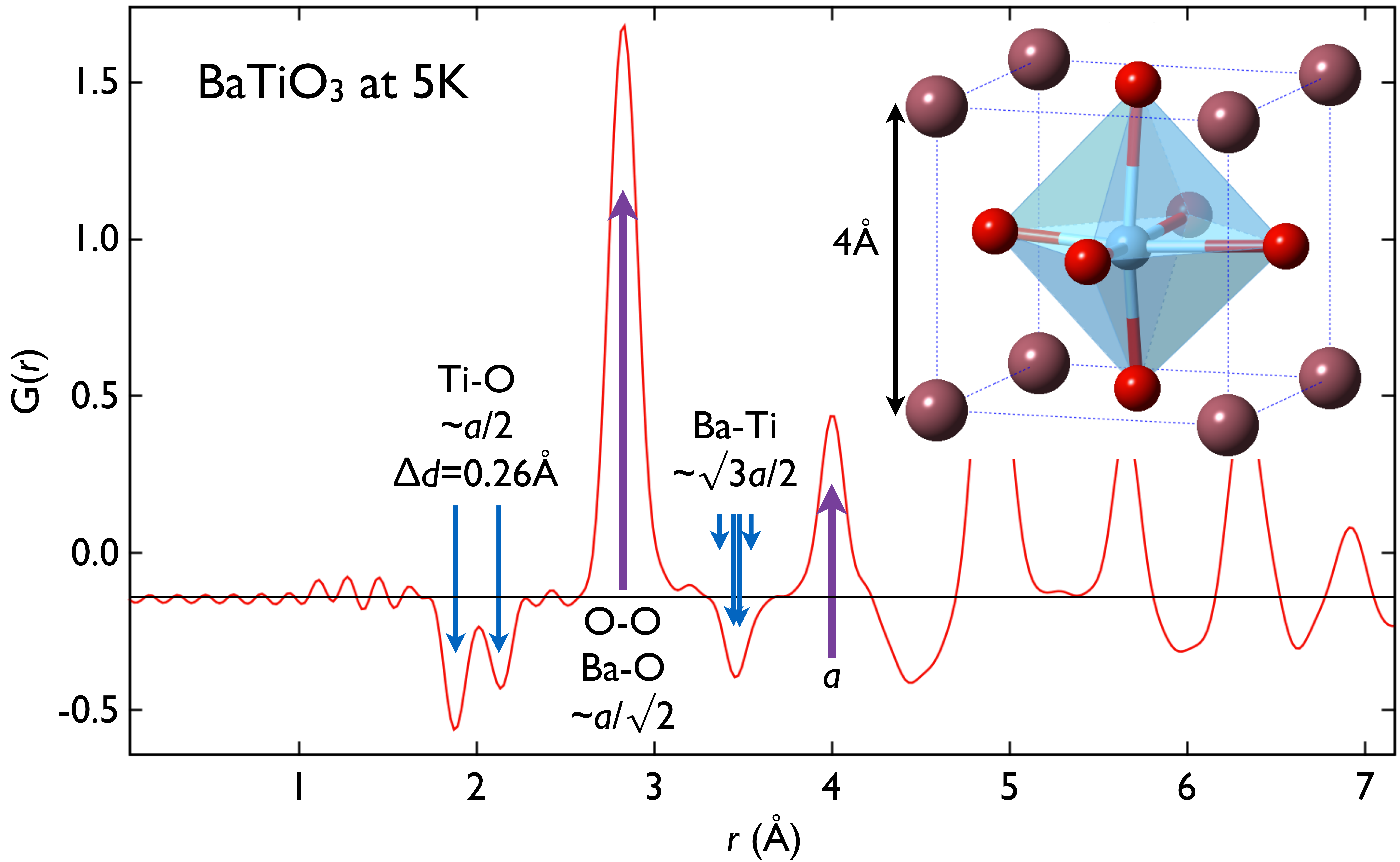
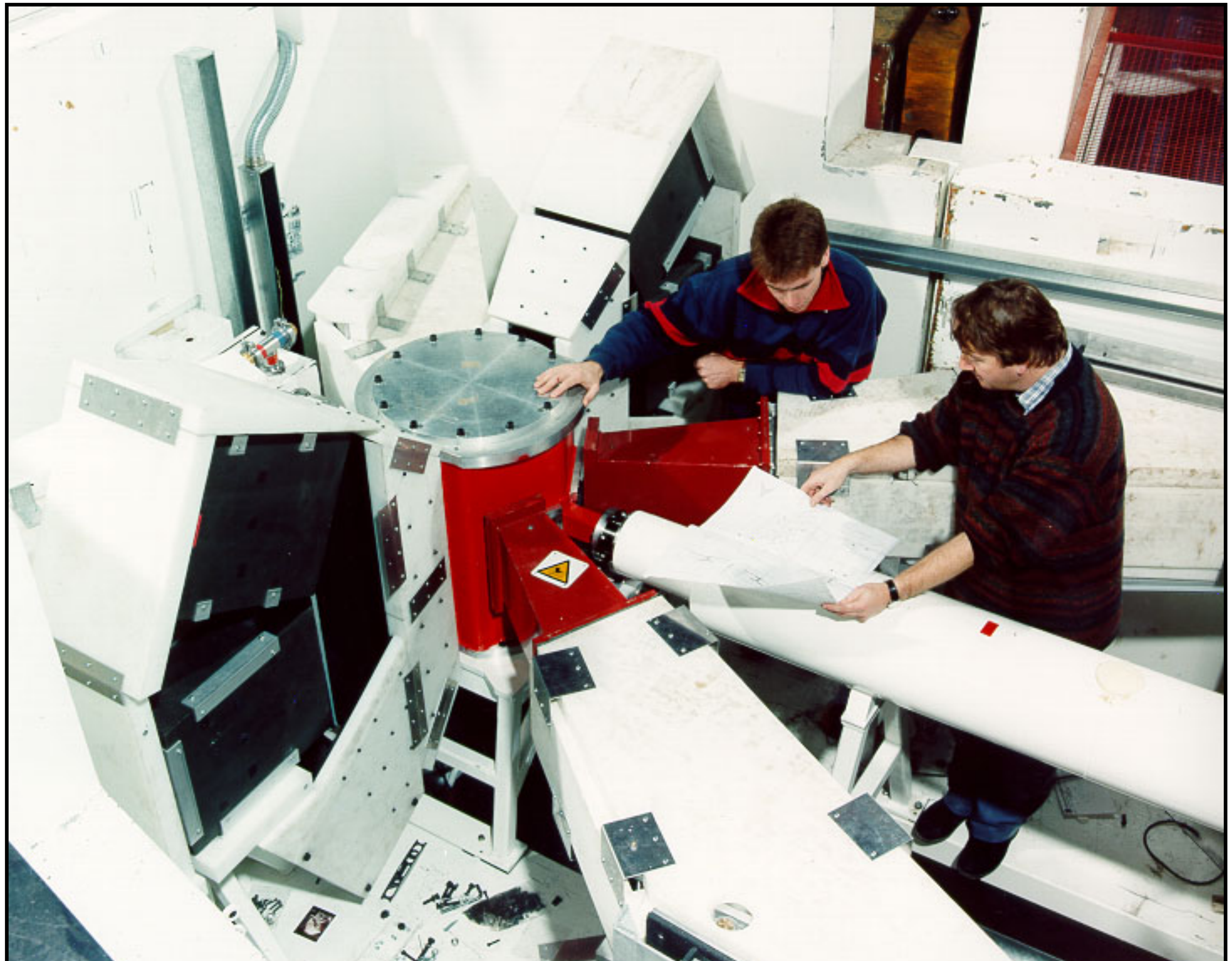
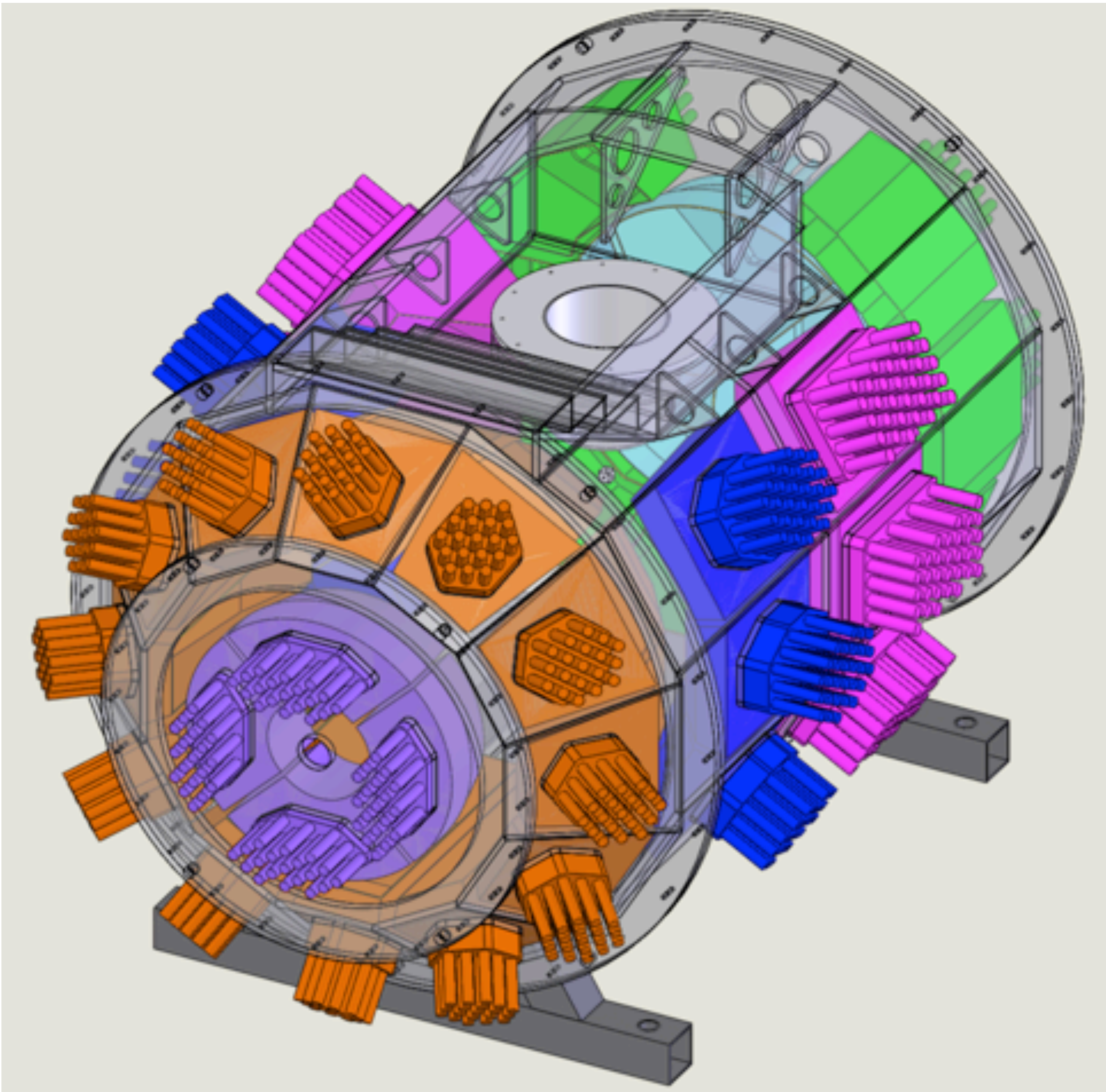


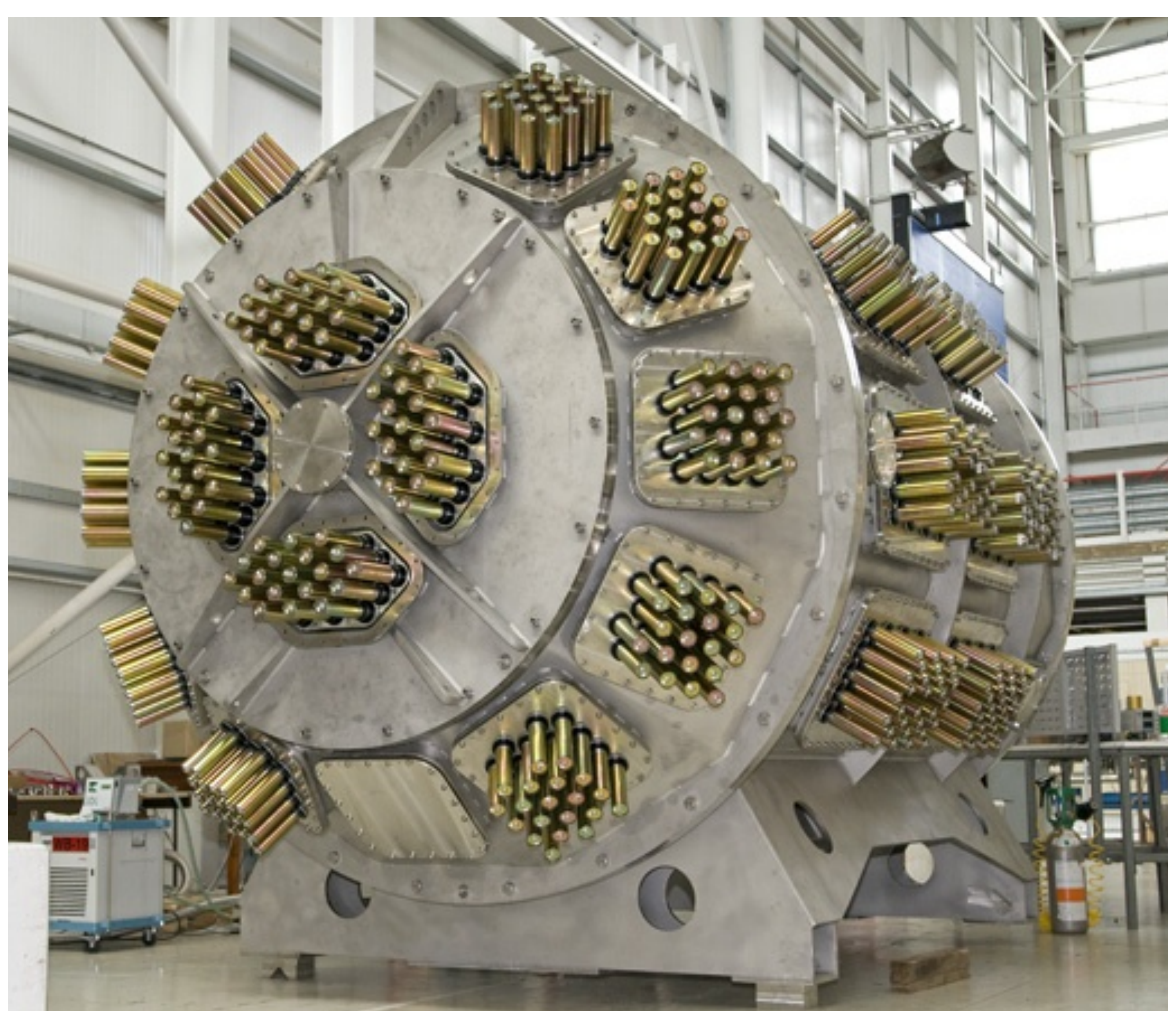
The “old” Polaris (1996-2006)

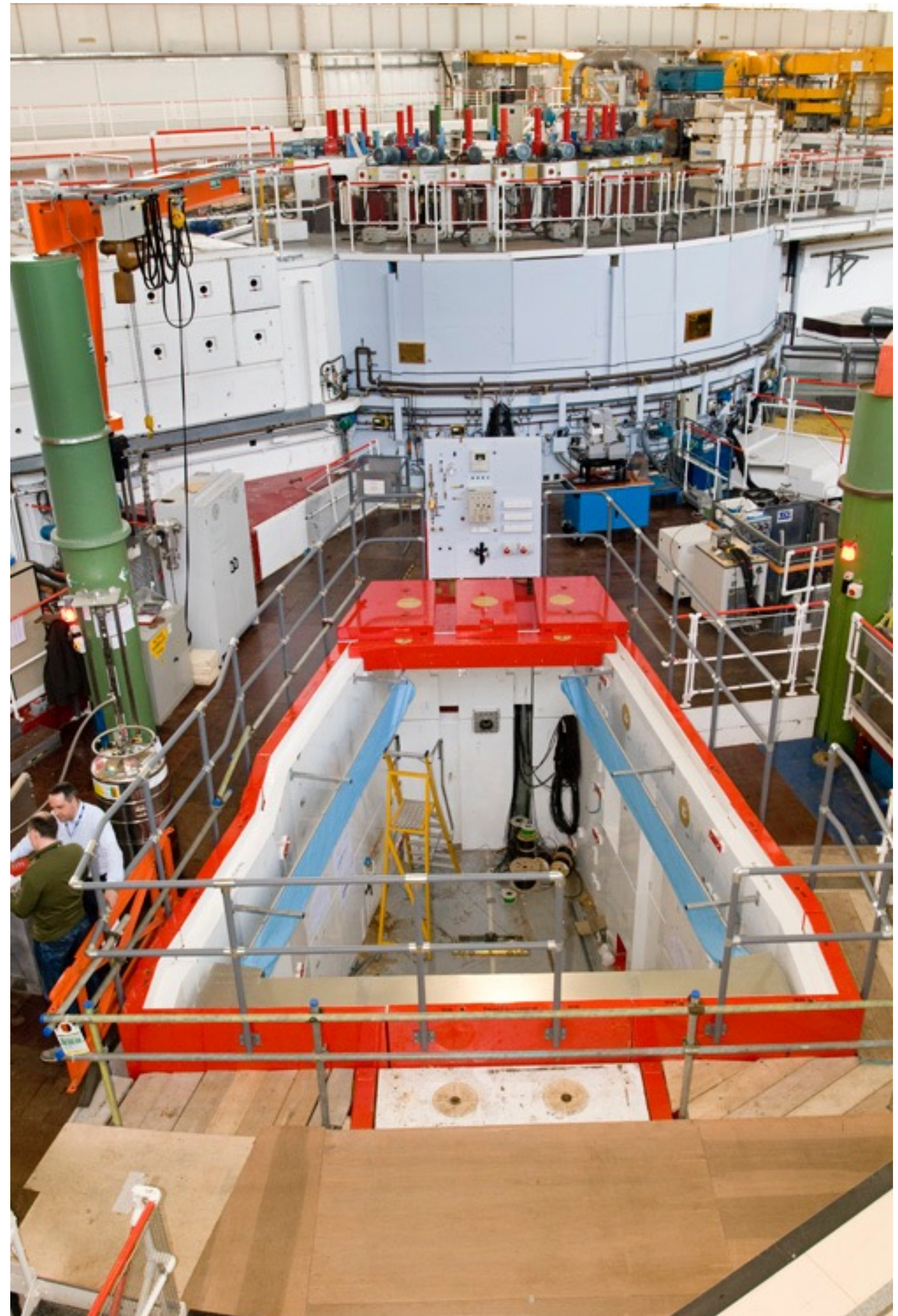


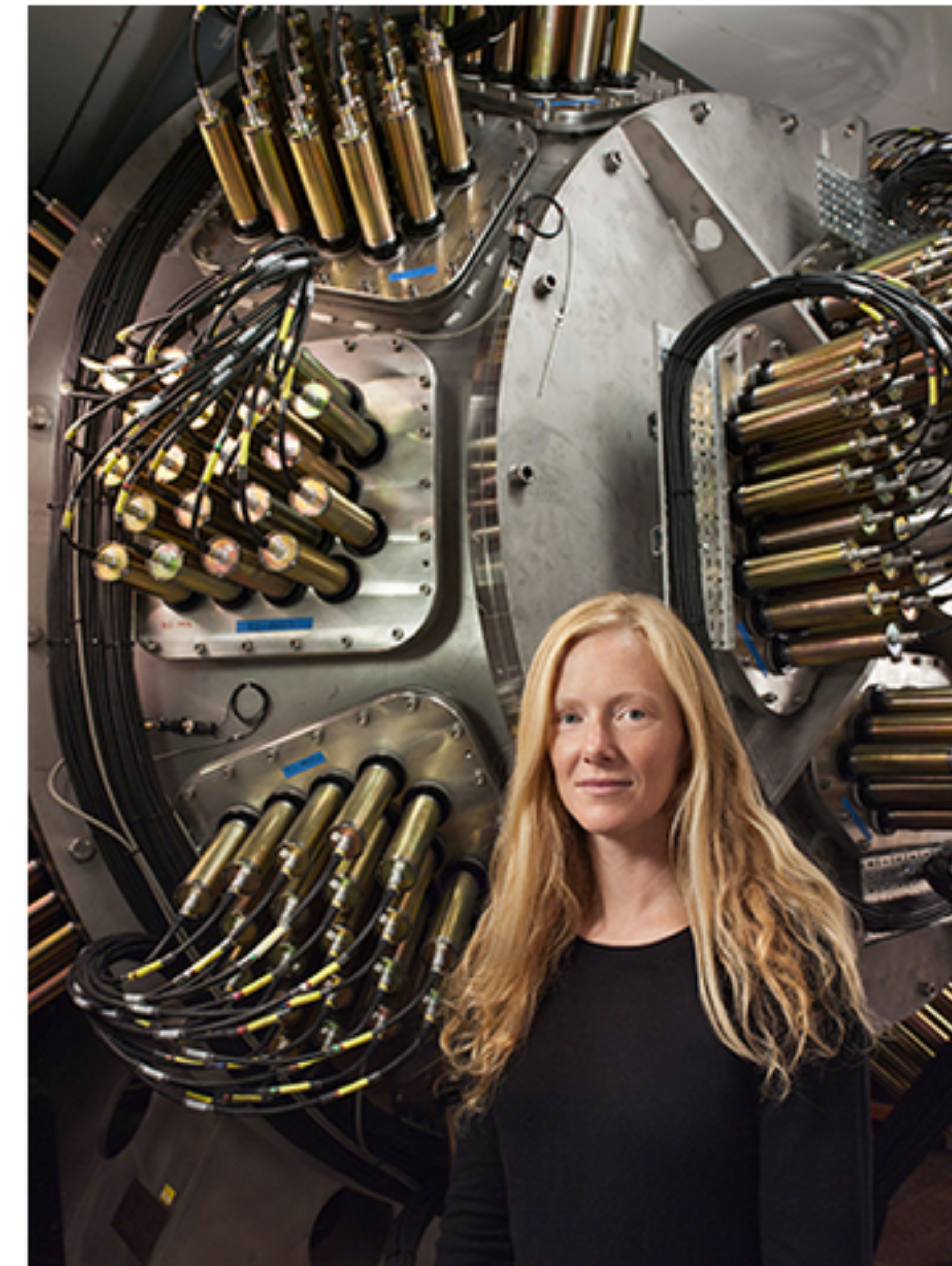
The Polaris total scattering diffractometer at ISIS

Available since late 2011









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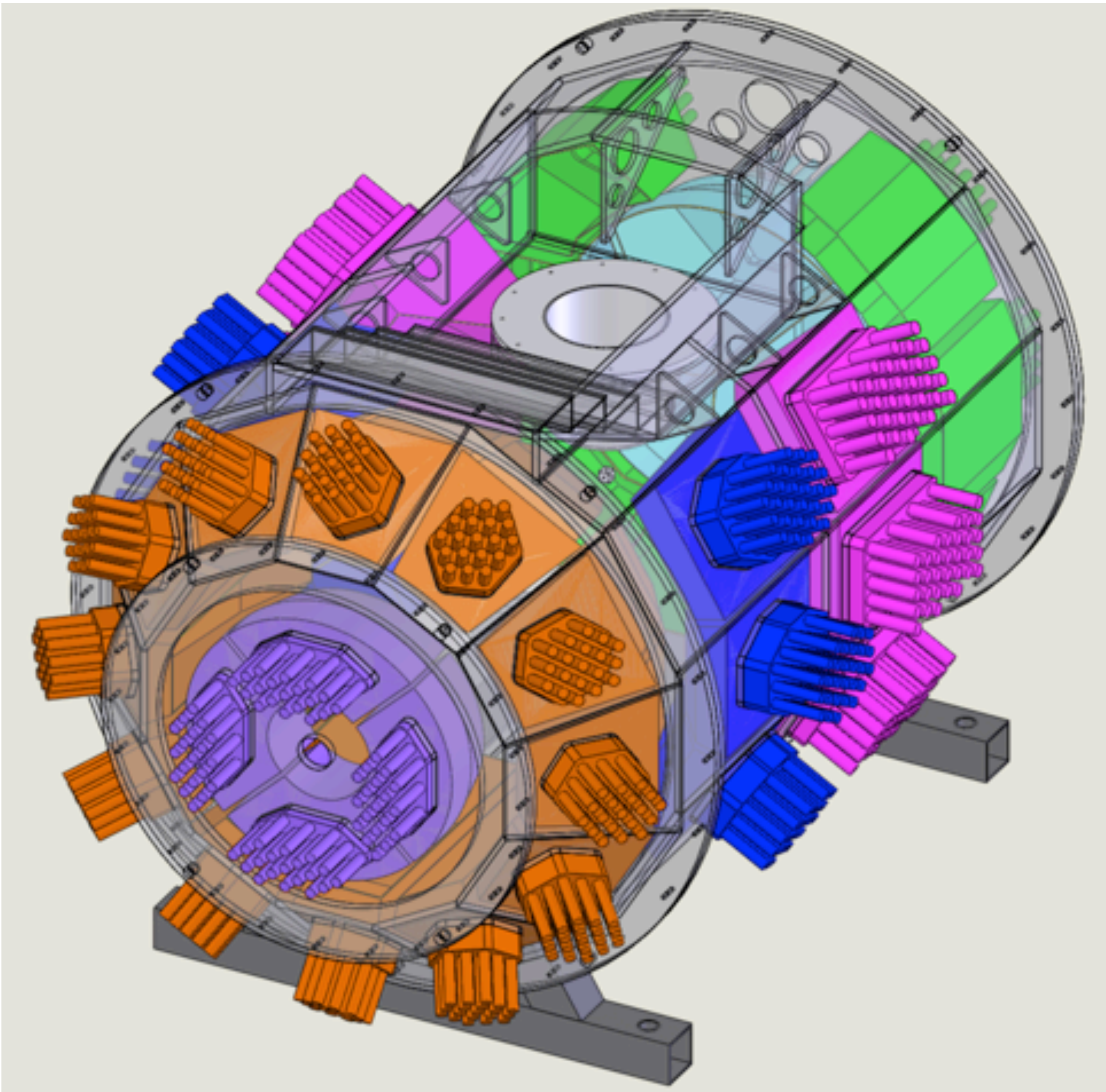
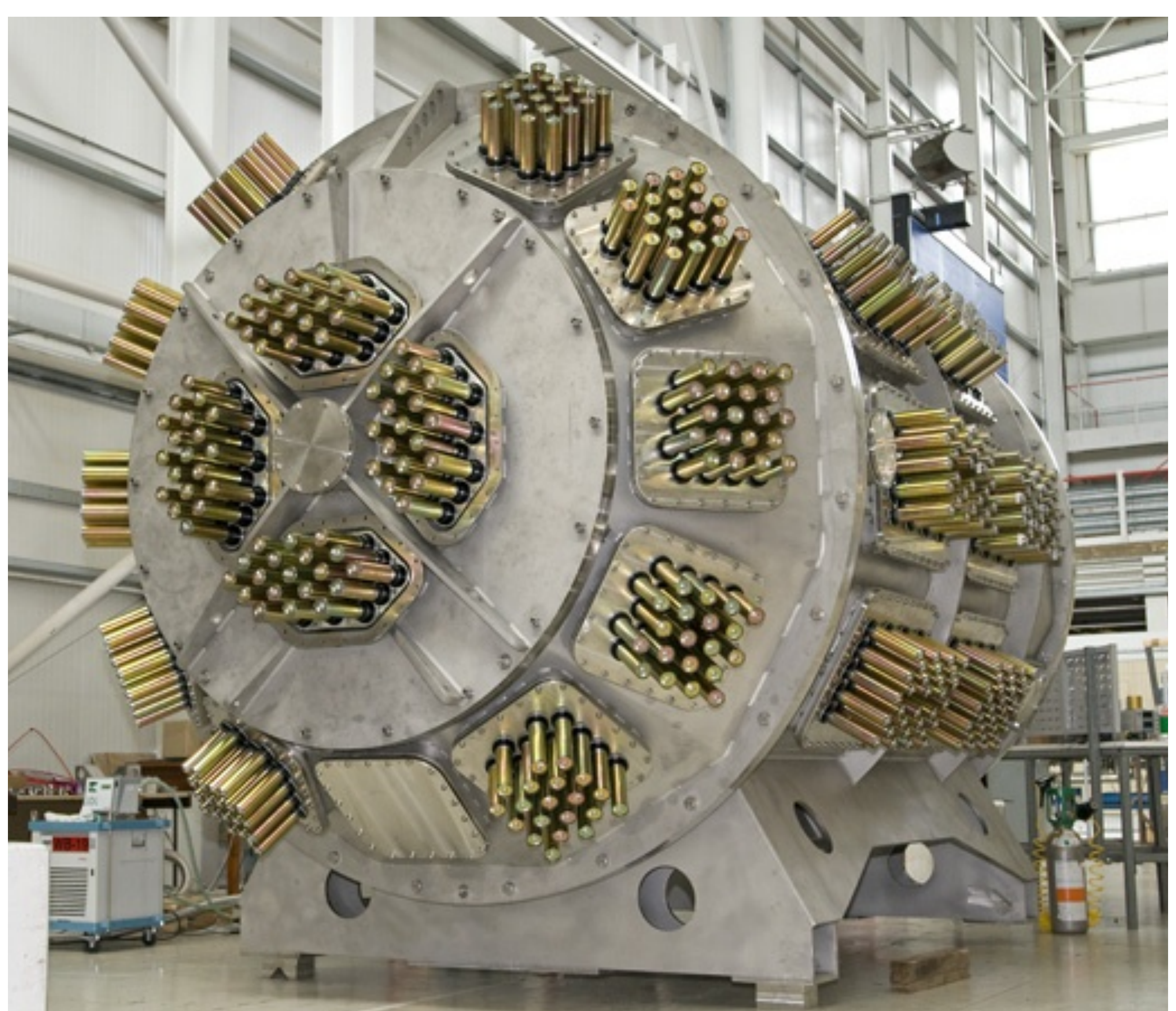
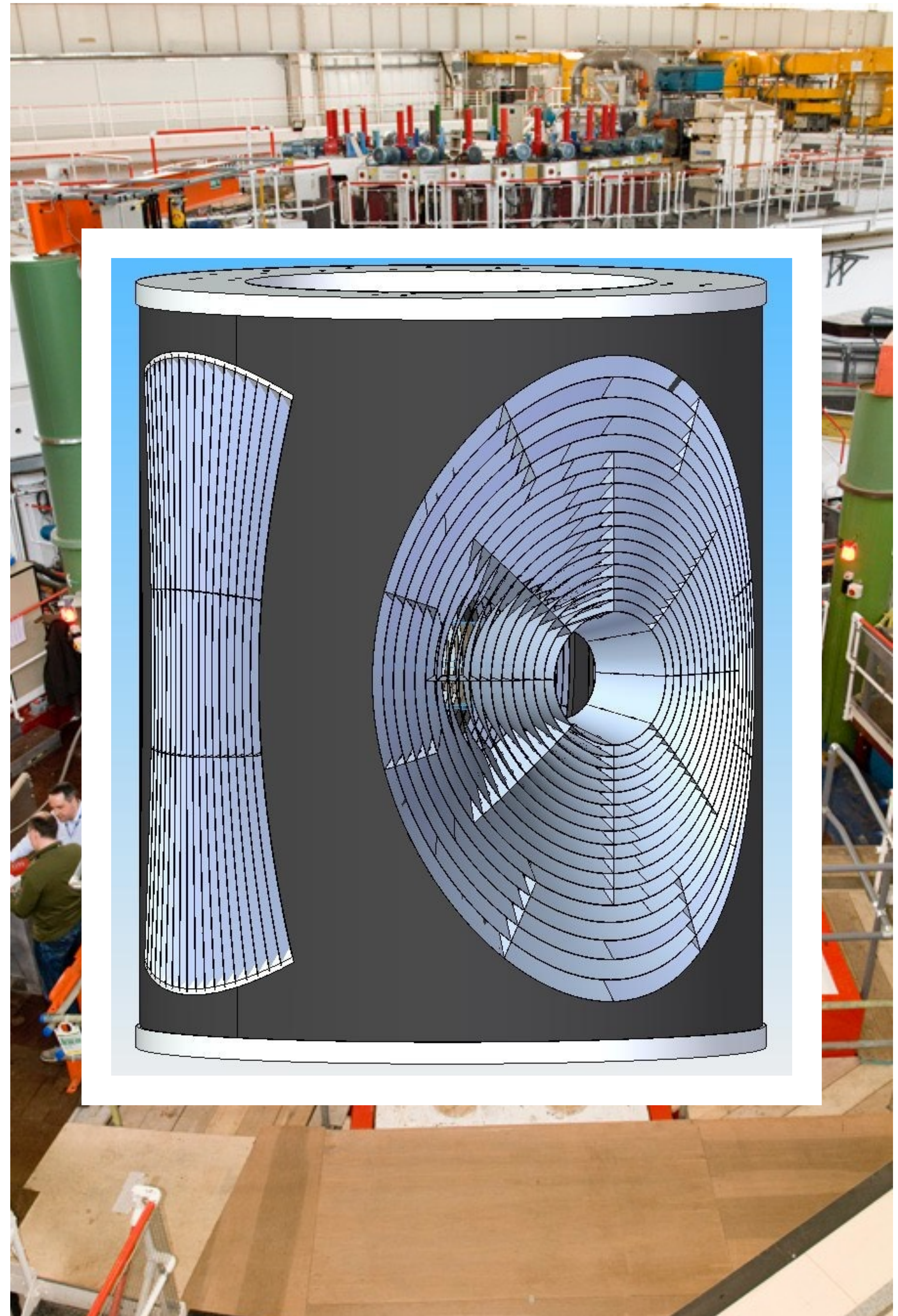
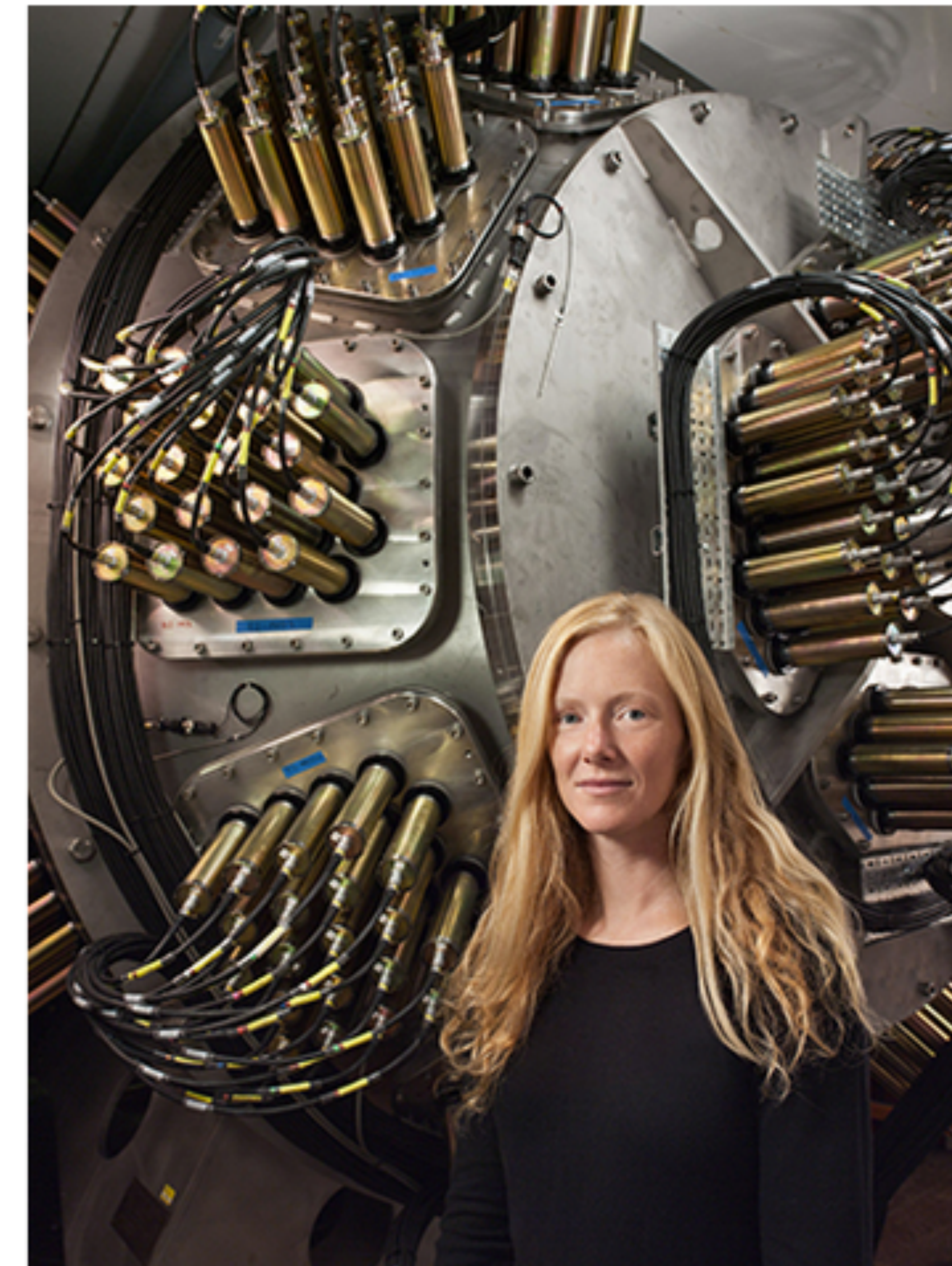








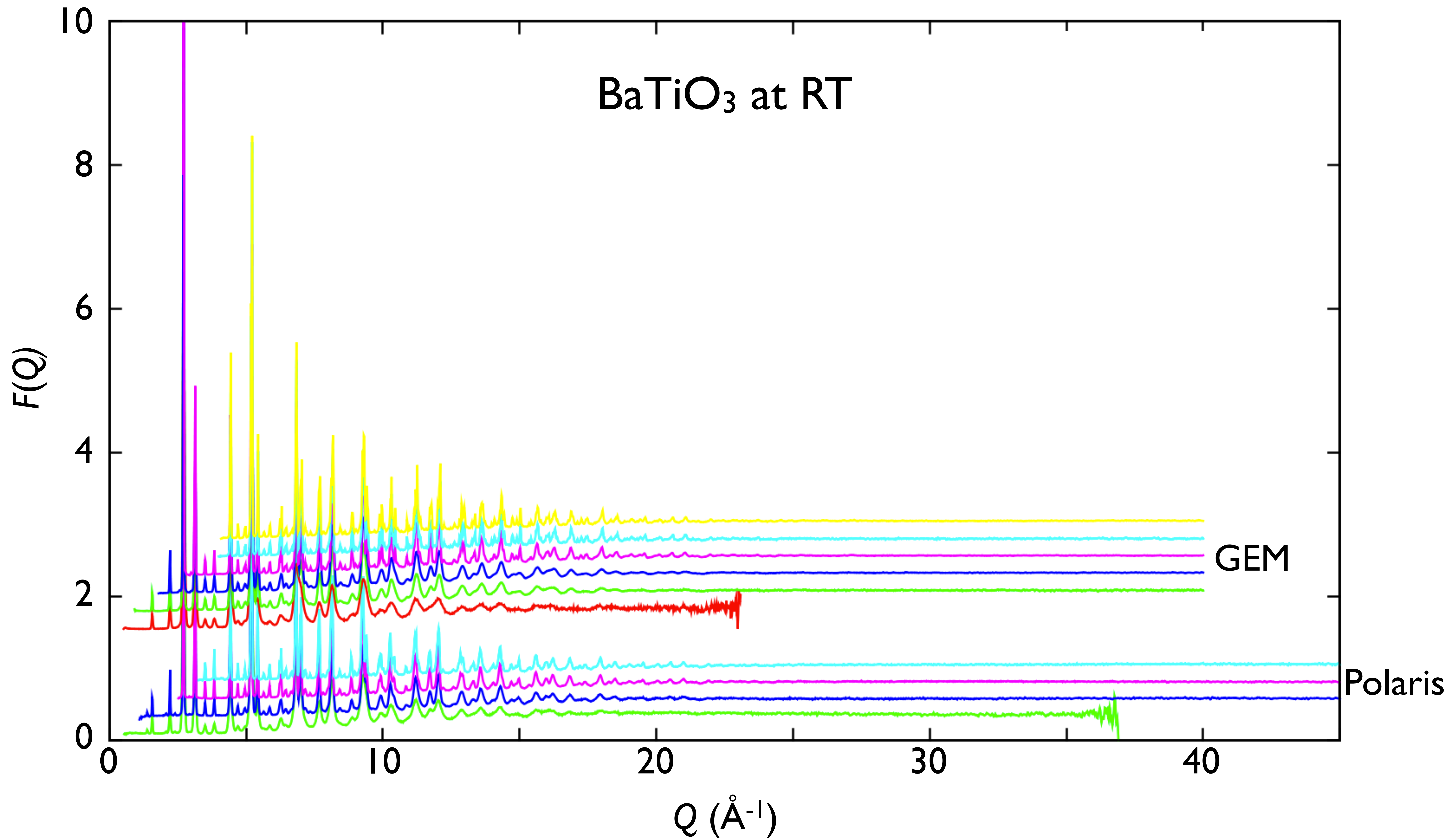
Polaris, GEM and LAD instrument parameters

	LAD (1984–1998)	*GEM (~1999–)*	*Polaris (2012–)*
• Moderator	Methane @110K	Methane @110K	Water @ 316K
• Primary flight path	10 m	17m	14m
• Wavelength range	~0.1 – 10Å	~0.1 – 3.4Å	~0.05 – 6.0Å
• Detector banks	7	7 (8)	5
• Detector coverage	0.016steradians	3.86st	5.67st
• Resolution DQ/Q	14 – 0.6%	~10 – 0.34%	~2.7 – 0.3%

Polaris detector coverage

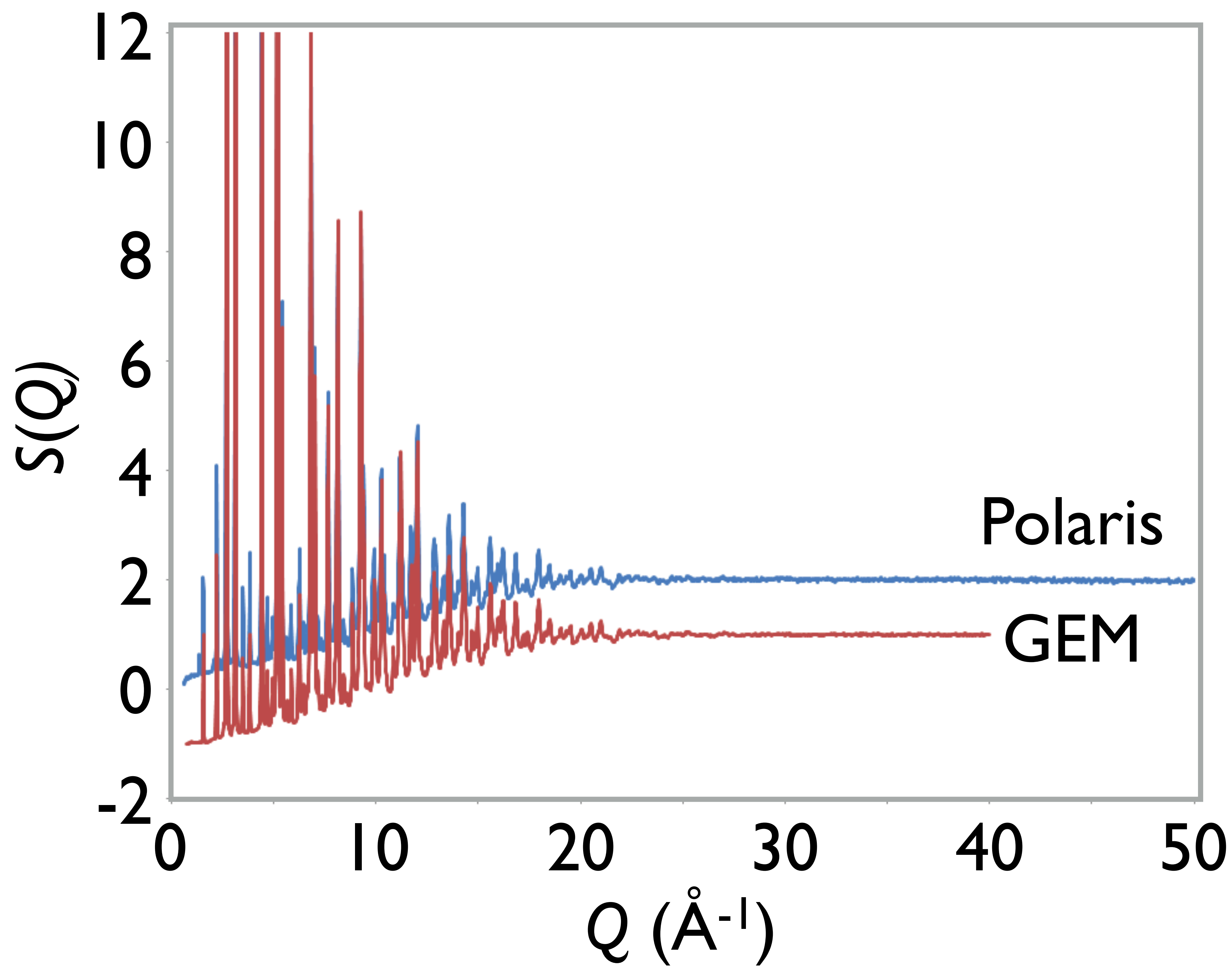
Detector Bank	Scattering angle, 2θ (°)	*L*2 (m)	Solid angle (st)	Δ*Q*/*Q* (%)	Q_{min} (Å^{-1})
1	6-14	2.25	0.26	2.7	~0.15
2	19-34	1.30-2.36	1.04	1.5	0.47
3	40-66	0.92-1.56	0.92	0.85	0.90
4 *(5 - GEM)*	75-113 *(79-114)*	0.71-1.08 *(~1.38)*	1.33 *(1.51)*	0.51 *(0.51)*	1.53 *(2.95)*
5	135-167	0.8-1.54	2.12	0.3	2.37

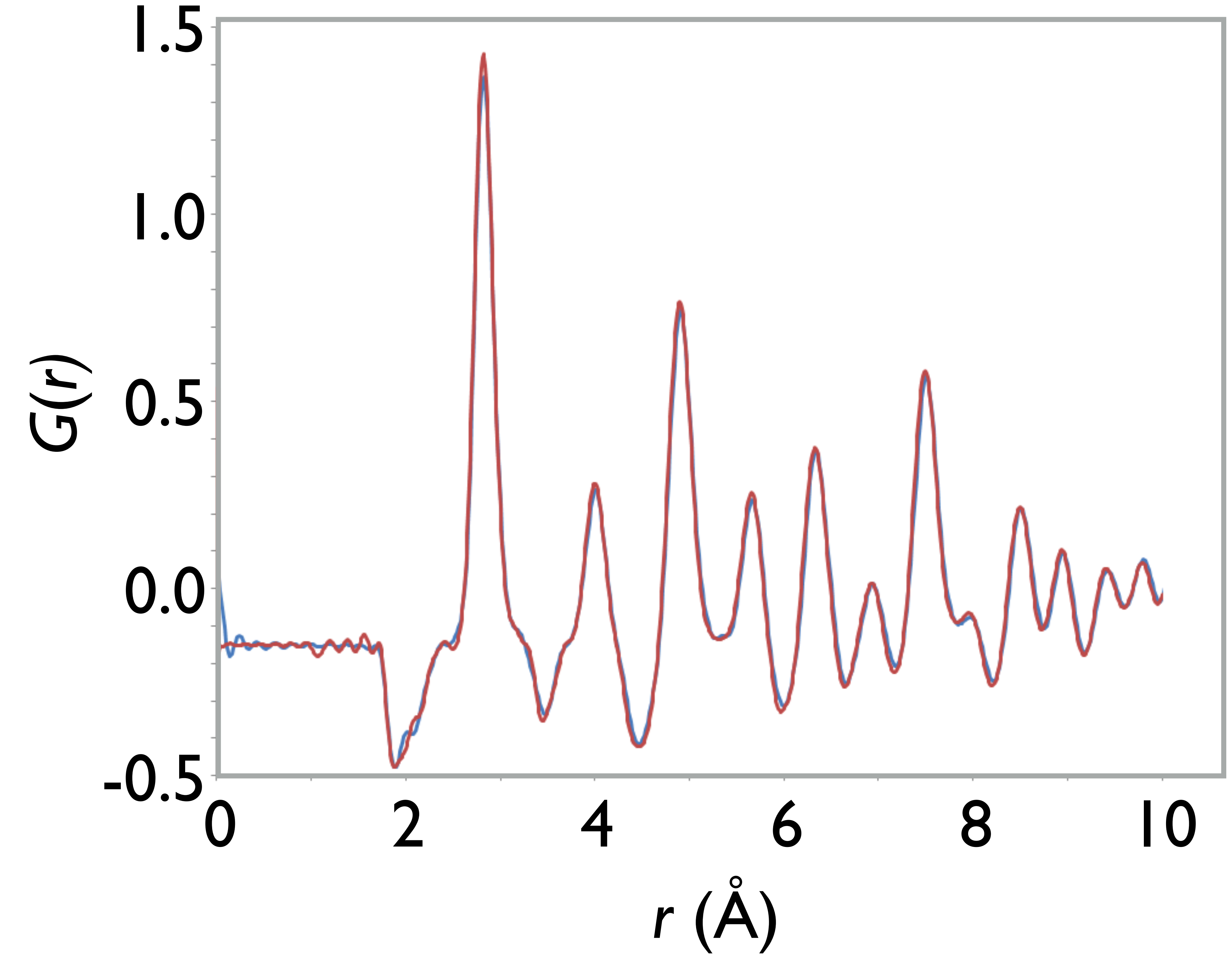
Corrected total scattering data from Polaris and GEM



Corrected *S*(*Q*) and PDF from Polaris and GEM

BaTiO$_3$ at RT





Possible future developments…

- Improved collimation and shielding and hence background
- Faster spinning Nimonic chopper to gain access to shorter wavelengths whilst keeping the beam completely blocked at T0
- Remodelling the GEM detector array following the experience gained from Polaris

Acknowledgements

Alex Hannon (GEM)

Ivan da Silva (GEM)

Helen Playford (Polaris)

Ron Smith (Polaris)

Steve Hull (Crystallography Group Leader)

Science & Technology Facilities Council

ISIS

Total Scattering Formalism – In more detail…!

Reciprocal Space

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

$$F(Q) = \sum_{i,j=1}^{n} c_i c_j \bar{b}_i \bar{b}_j [A_{ij}(Q) - 1]$$

$$A_{ij}(Q) - 1 = \rho_0 \int_0^\infty 4\pi r^2 [g_{ij}(r) - 1] \frac{\sin Qr}{Qr}\, \mathrm{d}r$$

F.T.

F.T. (n. only)

$$g_{ij}(r) - 1 = \frac{1}{(2\pi)^3 \rho_0} \int_0^\infty 4\pi Q^2 [A_{ij}(Q) - 1] \frac{\sin Qr}{Qr}\, \mathrm{d}Q$$

Real Space

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

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

$$g_{ij}(r) = \frac{n_{ij}(r)}{4\pi r^2\, \mathrm{d}r\, \rho_j}$$

D A Keen *J Appl Cryst* **34** 172 (2001)