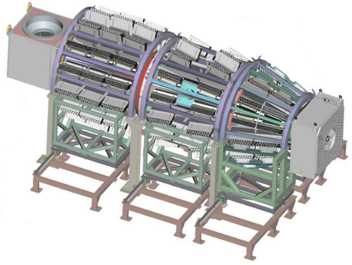
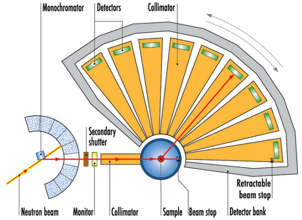
Experimental procedure and data reduction: Neutron Diffraction

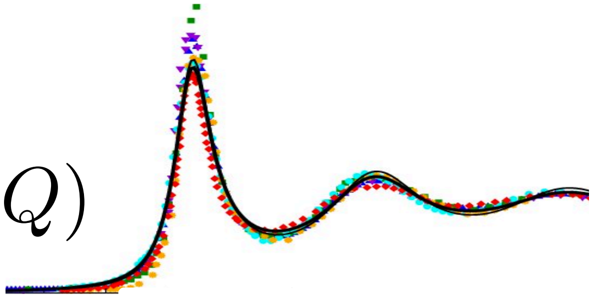


Gabriel Cuello

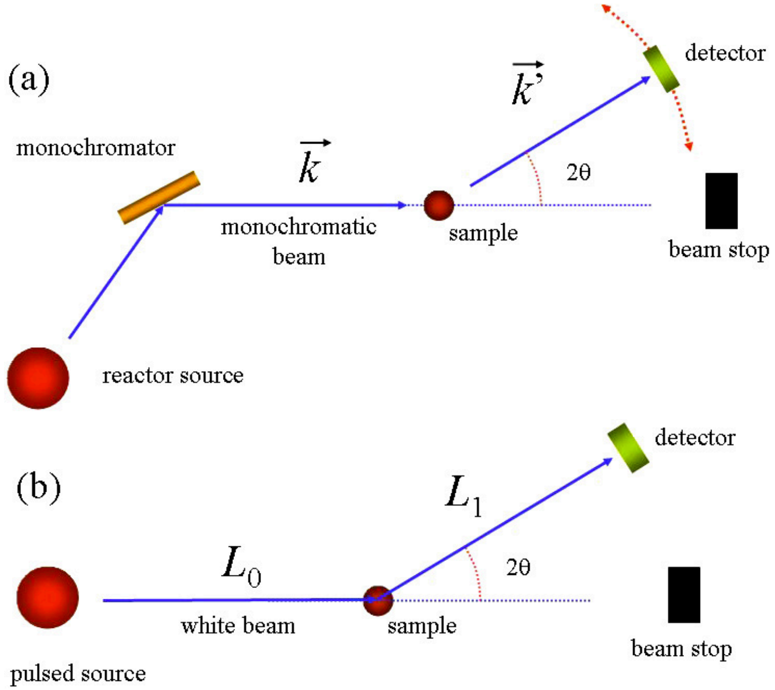
ILL

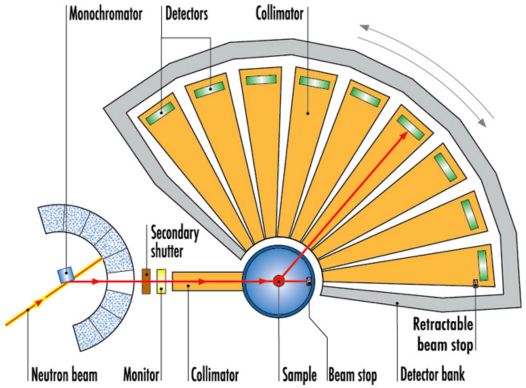


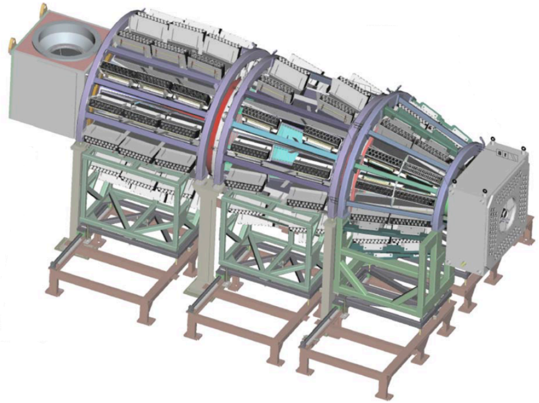
$$I(Q) \longrightarrow S(Q)$$



CW and TOF experiments





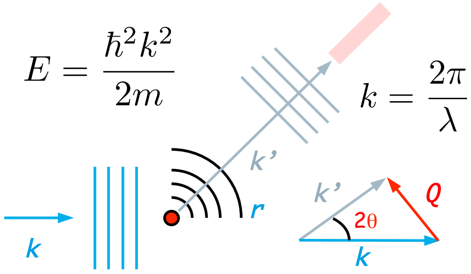


Structure factor

$$I_{2\theta,\lambda}(Q,\omega) = C\ \Phi_0(\lambda)\ N\ \frac{\lambda}{\lambda'}\ \frac{\sigma}{4\pi}\ S(Q,\omega)\ \epsilon(\lambda')$$

$$I_{2\theta,\lambda}(Q) = C\ \Phi_0(\lambda)\ N\ \frac{\sigma}{4\pi} \int_{-\infty}^{E_{\max}} \mathrm{d}\omega\ \frac{\lambda}{\lambda'}\ S(Q,\omega)\ \epsilon(\lambda')$$

$$E = \frac{\hbar^2 k^2}{2m} \qquad k = \frac{2\pi}{\lambda}$$



$$\frac{\hbar^2 Q^2}{2m} = 2E + \hbar\omega - 2E\sqrt{1 + \frac{\hbar\omega}{E}}\cos 2\theta$$

Elastic scattering

$$\lambda' = \lambda$$

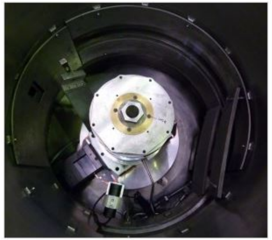
$$Q = \frac{4\pi}{\lambda}\ \sin\theta$$

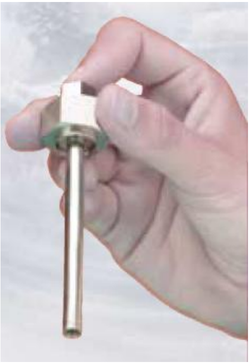
$$Q = \frac{2mL}{ht}\ \sin\theta$$

$$I_{2\theta,\lambda}(Q) = C\ \Phi_0(\lambda)\ \frac{N}{4\pi}\ (\sigma_{\mathrm{coh}}\ S(Q) + \sigma_{\mathrm{inc}})\ \epsilon(\lambda)$$

$$\sigma_{\mathrm{coh}}\ S(Q) = \frac{I_{2\theta,\lambda}(Q)}{C\ \Phi_0(\lambda)\ \epsilon(\lambda)}\ \frac{4\pi}{N} - \sigma_{\mathrm{inc}}$$

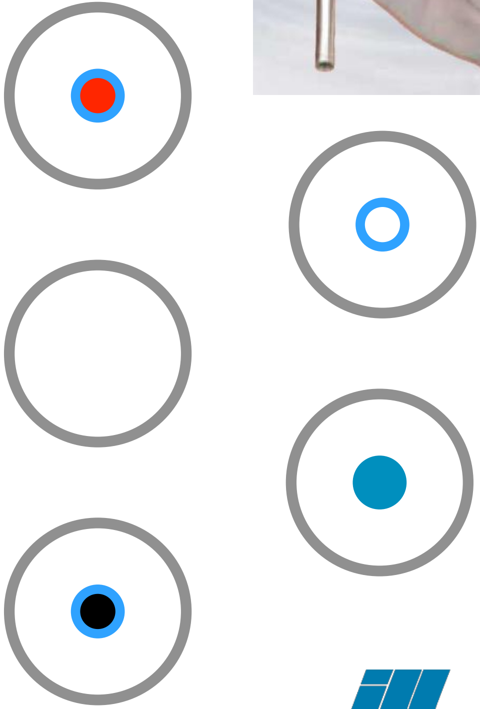
Typical experiment





Required measurements

- Sample + container + environment $I_{\mathrm{SCE}}(Q)$
- Empty container + environment $I_{\mathrm{CE}}(Q)$
- Empty environment $I_{\mathrm{E}}(Q)$
- Vanadium + environment $I_{\mathrm{VE}}(Q)$
- Absorbent + container + environment $I_{\mathrm{ACE}}(Q)$



Background subtraction

P. S. Salmon et al., J. Phys. F: Met. Phys. 18, 2345 (1988)

$$F(Q) = b_{\mathrm{coh}}^2 \left(S(Q) + P(Q)\right) + b_{\mathrm{inc}}^2 \left(1 + P(Q)\right)$$

$$F(Q) = \frac{1}{R_1(2\theta)} \left[\left(\frac{I_{\mathrm{SCE}}(Q)}{a(2\theta)} - M_{\mathrm{SCE}}(Q) \right) - R_2(2\theta) \left(\frac{I_{\mathrm{CE}}(Q)}{a(2\theta)} - M_{\mathrm{CE}}(Q) \right) - R_3(2\theta) \left(\frac{I_{\mathrm{E}}(Q)}{a(2\theta)} - M_{\mathrm{E}}(Q) \right) \right]$$

$$R_1(2\theta) = N\ A_{\mathrm{S,SCE}}(2\theta) \qquad R_2(2\theta) = \frac{A_{\mathrm{C,SCE}}(2\theta)}{A_{\mathrm{C,CE}}(2\theta)} \qquad R_3(2\theta) = \frac{1}{A_{\mathrm{E,E}}(2\theta)} \left(A_{\mathrm{E,SCE}}(2\theta) - \frac{A_{\mathrm{C,SCE}}(2\theta)\ A_{\mathrm{E,CE}}(2\theta)}{A_{\mathrm{C,CE}}(2\theta)} \right)$$

Attenuation factors

$$I_{\rm m}(Q) = A_{\rm m,\mathbb{M}}(2\theta)\ I_{\rm m}^{\rm theo}(Q)$$

$$\mathbb{M} = [{\rm S}, {\rm C}, {\rm E}_1, {\rm E}_2, \ldots]$$

H. H. Paalman and C. J. Pings
J. Appl. Phys. **33**, 2635 (1962)

$$\begin{bmatrix} A_{\rm S,SCE}(2\theta) & A_{\rm C,SCE}(2\theta) & A_{\rm E,SCE}(2\theta) \\ & A_{\rm C,CE}(2\theta) & A_{\rm E,CE}(2\theta) \\ A_{\rm V,VE}(2\theta) & & A_{\rm E,VE}(2\theta) \\ & & A_{\rm E,E}(2\theta) \end{bmatrix}$$

$$\mu(\lambda) = \rho\sigma(\lambda) = \rho(\sigma_{\rm sca} + \sigma_{\rm abs}(\lambda))$$

In CW experiments, these coefficients are wavelength dependent because of inelasticity

In TOF experiments, these coefficients are naturally wavelength dependent

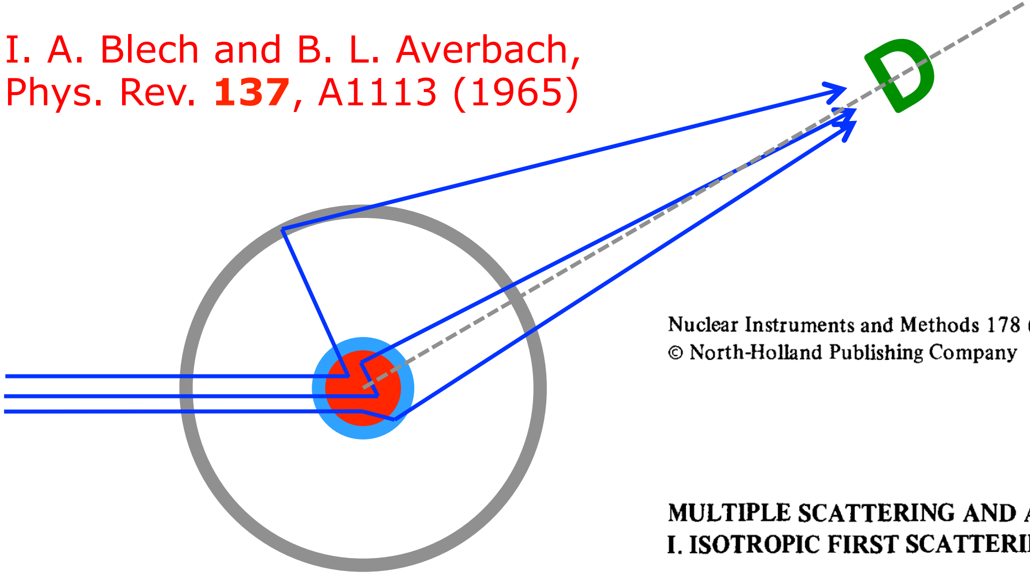
Generalised by P. F. J. Poncet
PhD Thesis, University of Reading (1976)

$$A_{\rm m,\mathbb{M}}(2\theta) = \frac{1}{V_{\rm m}} \int_{V_{\rm m}} {\rm d}V_{\rm m}\ \exp\left(-\sum_{k\in\mathbb{M}} \mu_k \ell_k(\vec{r}, 2\theta)\right)$$

- Elastic scattering approximation
- 2D calculation
- Not fully illuminated cylinders
- Uniform beam

m.s. and attenuation

I. A. Blech and B. L. Averbach,
Phys. Rev. **137**, A1113 (1965)



$$\begin{bmatrix} M_{\mathrm{SCE}}(Q) & M_{\mathrm{CE}}(Q) \\ M_{\mathrm{VE}}(Q) & M_{\mathrm{E}}(Q) \end{bmatrix}$$

$$M(Q) = \Delta\, I_1(Q)$$

Nuclear Instruments and Methods 178 (1980) 415–425

MULTIPLE SCATTERING AND ATTENUATION OF NEUTRONS IN CONCENTRIC CYLINDERS: I. ISOTROPIC FIRST SCATTERING

Alan K. SOPER * and P.A. EGELSTAFF
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CORRECT, GUDRUN, GO, …

Background subtraction

P. S. Salmon et al., J. Phys. F: Met. Phys. 18, 2345 (1988)

$$F(Q) = b^2_{\mathrm{coh}} \left(S(Q) + P(Q)\right) + b^2_{\mathrm{inc}} \left(1 + P(Q)\right)$$

$$F(Q) = \frac{1}{R_1(2\theta)} \left[\left(\frac{I_{\mathrm{SCE}}(Q)}{a(2\theta)} - M_{\mathrm{SCE}}(Q) \right) - R_2(2\theta) \left(\frac{I_{\mathrm{CE}}(Q)}{a(2\theta)} - M_{\mathrm{CE}}(Q) \right) - R_3(2\theta) \left(\frac{I_{\mathrm{E}}(Q)}{a(2\theta)} - M_{\mathrm{E}}(Q) \right) \right]$$

$$R_1(2\theta) = N\ A_{\mathrm{S,SCE}}(2\theta) \qquad R_2(2\theta) = \frac{A_{\mathrm{C,SCE}}(2\theta)}{A_{\mathrm{C,CE}}(2\theta)} \qquad R_3(2\theta) = \frac{1}{A_{\mathrm{E,E}}(2\theta)} \left(A_{\mathrm{E,SCE}}(2\theta) - \frac{A_{\mathrm{C,SCE}}(2\theta)\ A_{\mathrm{E,CE}}(2\theta)}{A_{\mathrm{C,CE}}(2\theta)} \right)$$

$$a(2\theta) = \frac{I_{\mathrm{VE}}(2\theta) - \frac{A_{\mathrm{E,VE}}(2\theta)}{A_{\mathrm{E,E}}(2\theta)} I_{\mathrm{E}}(2\theta)}{A_{\mathrm{V,VE}}(2\theta)\ N_{\mathrm{V}}\ b^2_{\mathrm{inc,V}} \left[1 + P_{\mathrm{V}}(Q)\right] + M_{\mathrm{VE}}(Q) - \frac{A_{\mathrm{E,VE}}(2\theta)}{A_{\mathrm{E,E}}(2\theta)} M_{\mathrm{E}}(Q)}$$

Strongly attenuating samples or complex sample environment

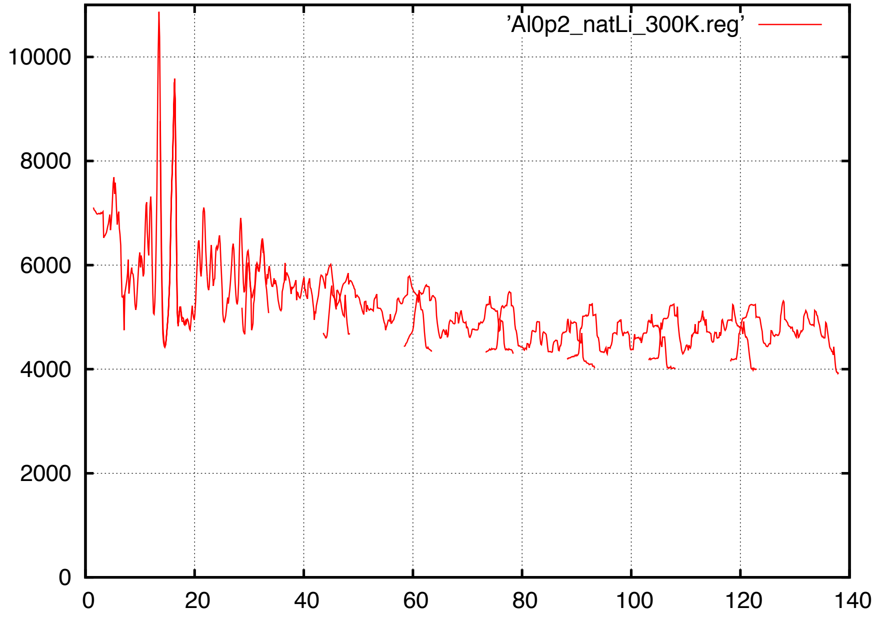
$$I'_{\mathrm{S}}(Q) = I_{\mathrm{SCE}} - \left[\overline{A_{\mathrm{S,SC}}(2\theta)}\ I_{\mathrm{ACE}}(Q) + \left(1 - \overline{A_{\mathrm{S,SC}}(2\theta)}\right)\ I_{\mathrm{CE}}(Q) \right]$$

Still needs to be corrected by auto-attenuation and m.s.

Example D4: $Li_{6.4}Al_{0.2}La_3Zr_2$

Solid electrolytes

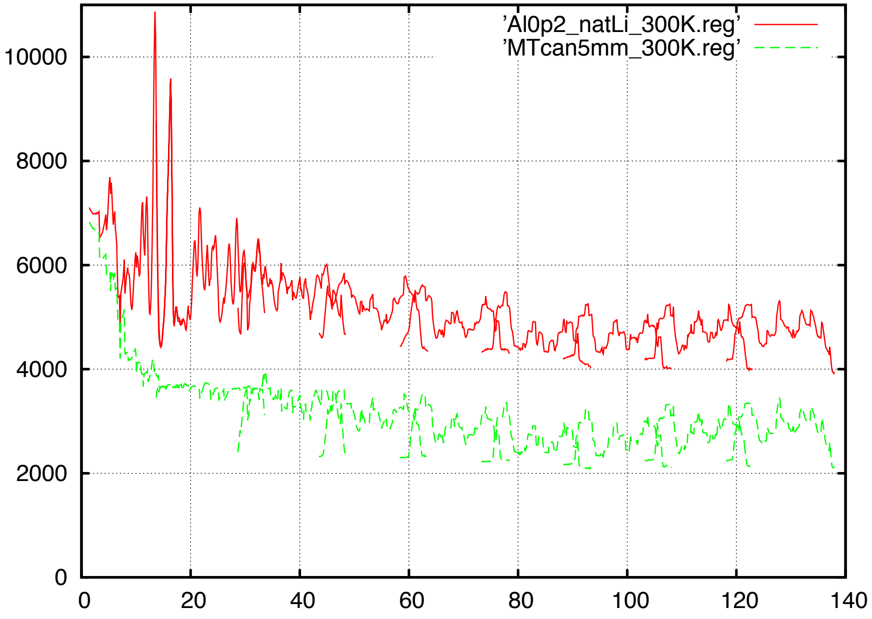
Raw data



Scattering angle

Example D4: $Li_{6.4}Al_{0.2}La_3Zr_2$

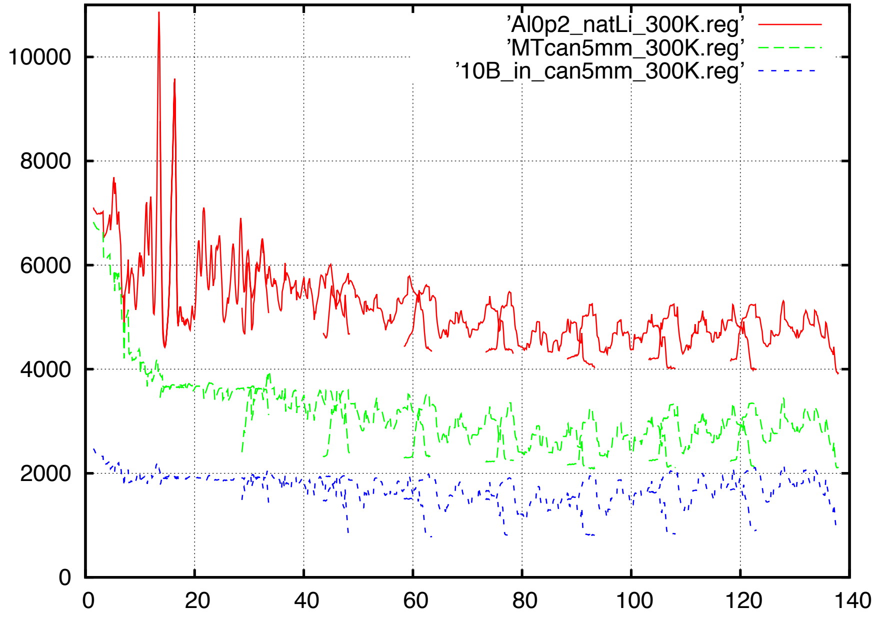
Raw data



Scattering angle

Example D4: $Li_{6.4}Al_{0.2}La_3Zr_2$

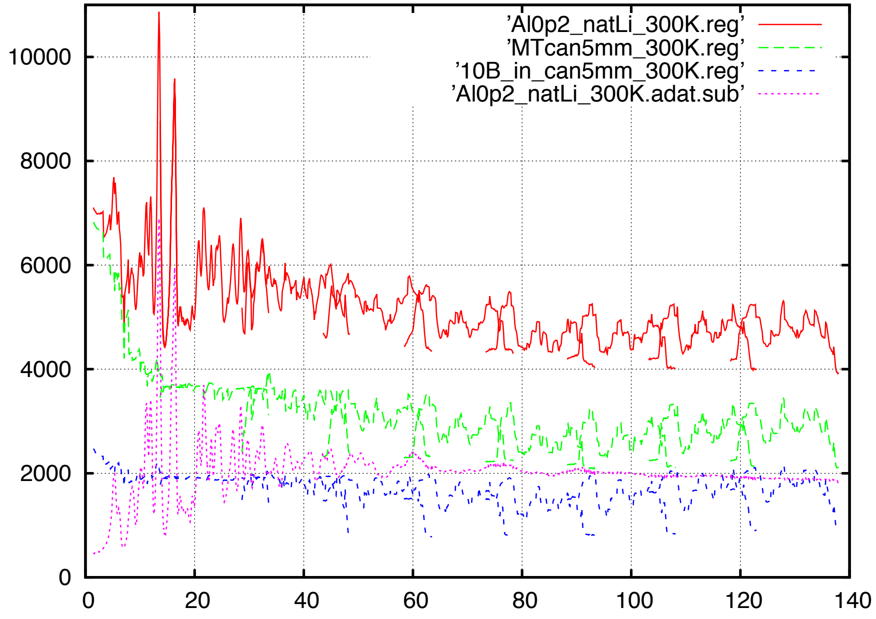
Raw data



Scattering angle

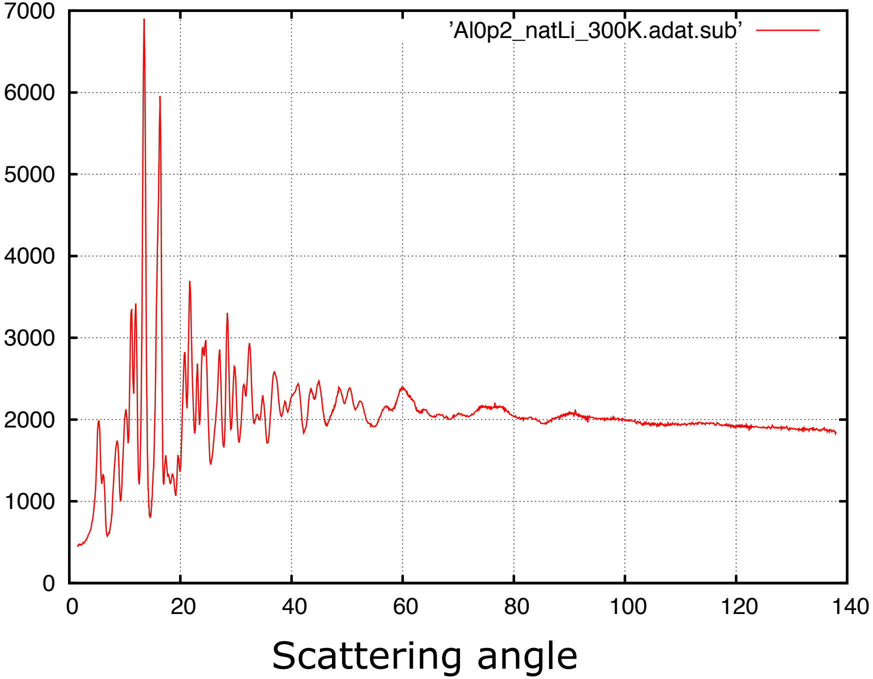
Example D4: $Li_{6.4}Al_{0.2}La_3Zr_2$

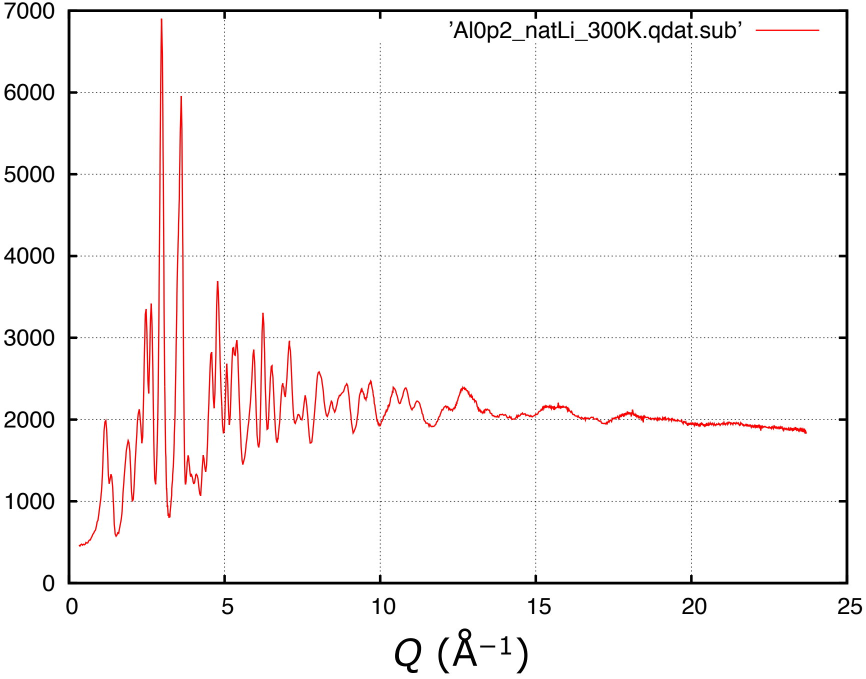
Raw data



Scattering angle

Example D4: $Li_{6.4}Al_{0.2}La_3Zr_2$



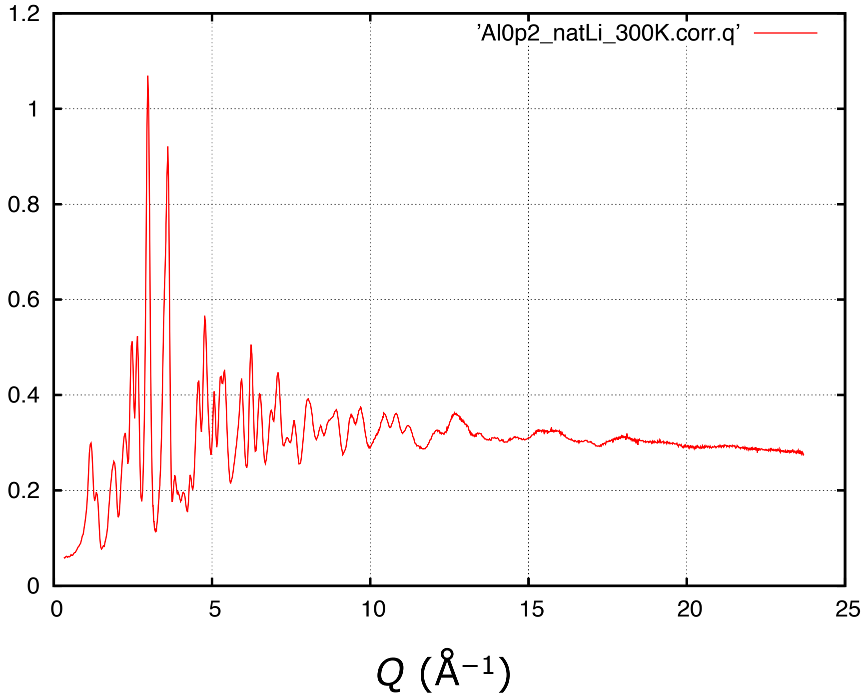


$$I'_{\mathrm{S}}(Q) = I_{\mathrm{SCE}} - \left[\overline{A_{\mathrm{S,SC}}(2\theta)}\, I_{\mathrm{ACE}}(Q) + \left(1 - \overline{A_{\mathrm{S,SC}}(2\theta)}\right) I_{\mathrm{CE}}(Q)\right]$$

Example D4: $Li_{6.4}Al_{0.2}La_3Zr_2$

After CORRECT

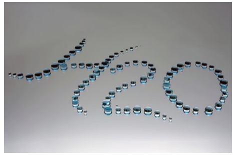
Auto-attenuation
Multiple scattering

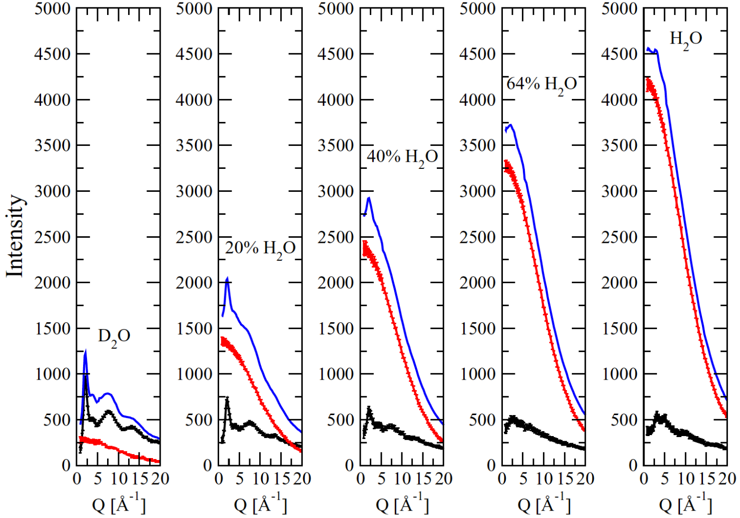


Inelasticity

$$S(Q) = 1 + \frac{F(Q)}{b^2_{\text{coh}}} - \left(1 + \frac{b^2_{\text{inc}}}{b^2_{\text{coh}}}\right)[1 + P(Q)]$$

P. S. Salmon et al., J. Phys. F: Met. Phys. 18, 2345 (1988)





Two ways to correct for inelasticity

- Empirical
  - Polynomial in *Q*
  - Linear combination Gaussian/Lorentzian for light isotopes
    η *G*(*Q*) + (1 − η) *L*(*Q*)
- Analytical
  - Placzek's expansion

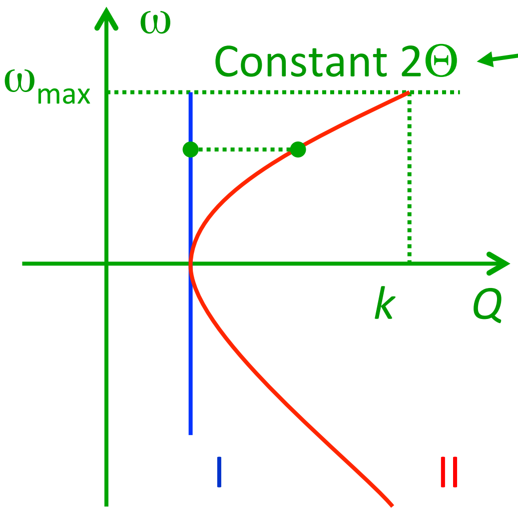
Inelasticity

$$I(2\theta) = C\ \Phi_0\ N\ \frac{\sigma}{4\pi} \int_{-\infty}^{E_{\max}} \frac{k'}{k}\ S(Q,\omega)\ \epsilon(k')\ \mathrm{d}\omega$$

$E_{\max} \rightarrow \infty$

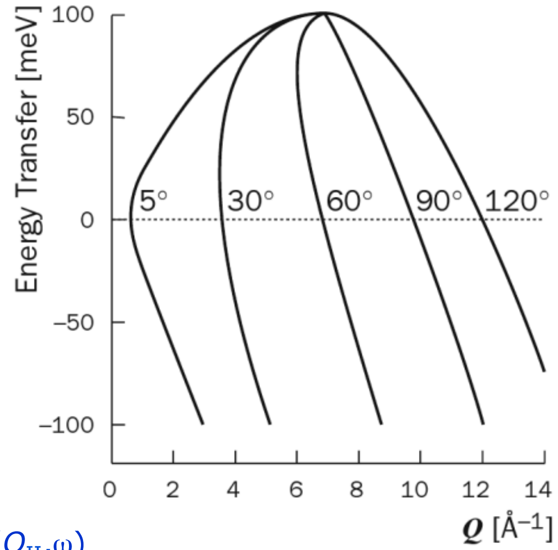
$\epsilon(k') \rightarrow$ ε(*k*)

Constant 2θ



$$\frac{\hbar^2 Q^2}{2m} = 2E + \hbar\omega - 2\sqrt{E^2 + \hbar\omega E}\ \cos 2\theta$$

These effects are closely associated to the detector efficiency



- Taylor expansion of $S(Q_I,\omega)$ around $(Q_I,\omega) \longrightarrow S(Q_{II},\omega)$
- Expansion of $Q_{II}^2 - Q_I^2$, $\varepsilon(k')$ and k'/k in powers of ω/ω_{max}
- Energy integration

Placzek's expansion

$$S(Q) = 1 + \frac{F(Q)}{\epsilon_0 b_{\mathrm{coh}}^2} - \left(1 + \frac{b_{\mathrm{inc}}^2}{b_{\mathrm{coh}}^2}\right)[1 + P(Q)]$$

P. S. Salmon et al., J. Phys. F: Met. Phys. 18, 2345 (1988)

$$P(Q) = -C_1\delta + C_2\delta^2 - C_3\delta\gamma + \frac{m}{2M}(\delta + \gamma)$$

$$\delta = \frac{E_{\mathrm{rec}}}{E} = \frac{\frac{\hbar^2 Q^2}{2M}}{E} \qquad \gamma = \frac{k_{\mathrm{B}}T}{E}$$

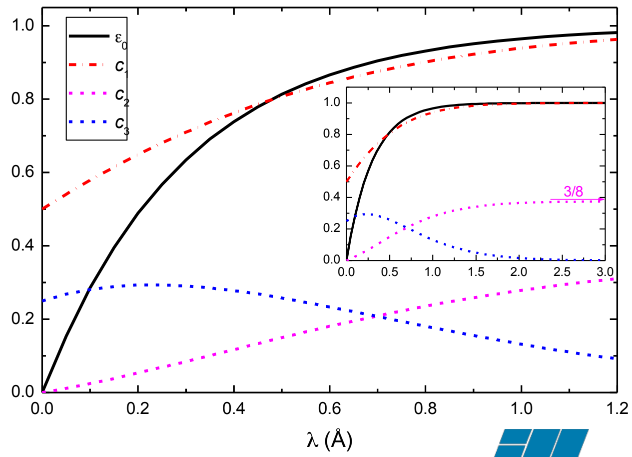
Efficiency

- Black detector, $\varepsilon(E) = 1$
- $1/v$ detector, $\varepsilon(E) \propto E^{-1/2}$
- Exponential detector, $\varepsilon(E) = 1 - \exp\{-\alpha(E/E')^{1/2}\}$

For an exponential detector

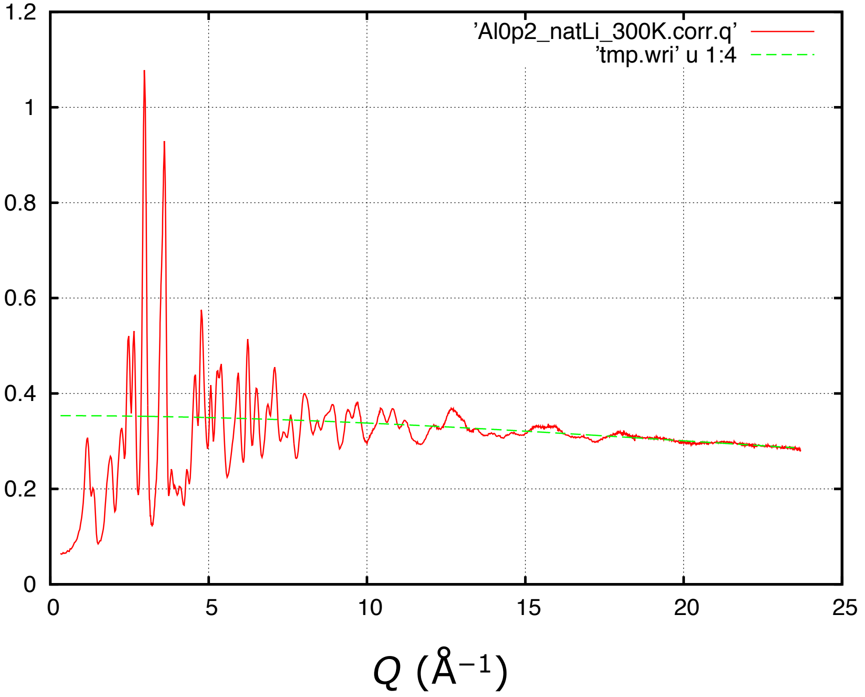
$$C_1 = 1 - \frac{\alpha/2}{e^{\alpha} - 1} \qquad C_2 = \frac{3}{8} - \frac{\alpha(\alpha+3)}{8(e^{\alpha}-1)} \qquad C_3 = \frac{\alpha(\alpha+1)}{4(e^{\alpha}-1)}$$

Coefficients for Placzek correction
(Yarnell *et al.*, PRA **7**, 2130 (1973))



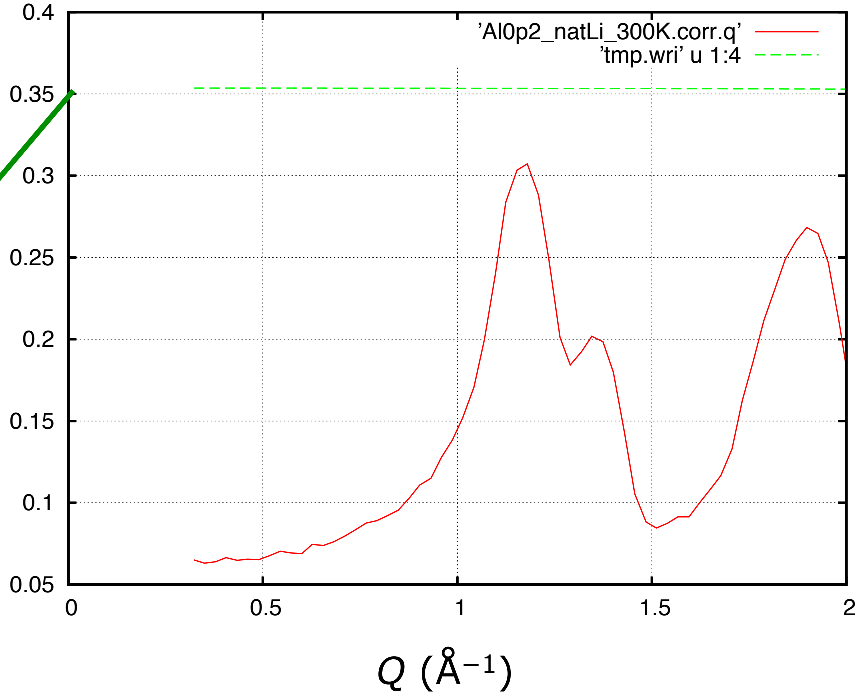
Example D4: $Li_{6.4}Al_{0.2}La_3Zr_2$

Inelasticity



Example D4: $Li_{6.4}Al_{0.2}La_3Zr_2$

Normalisation in *Q*-space



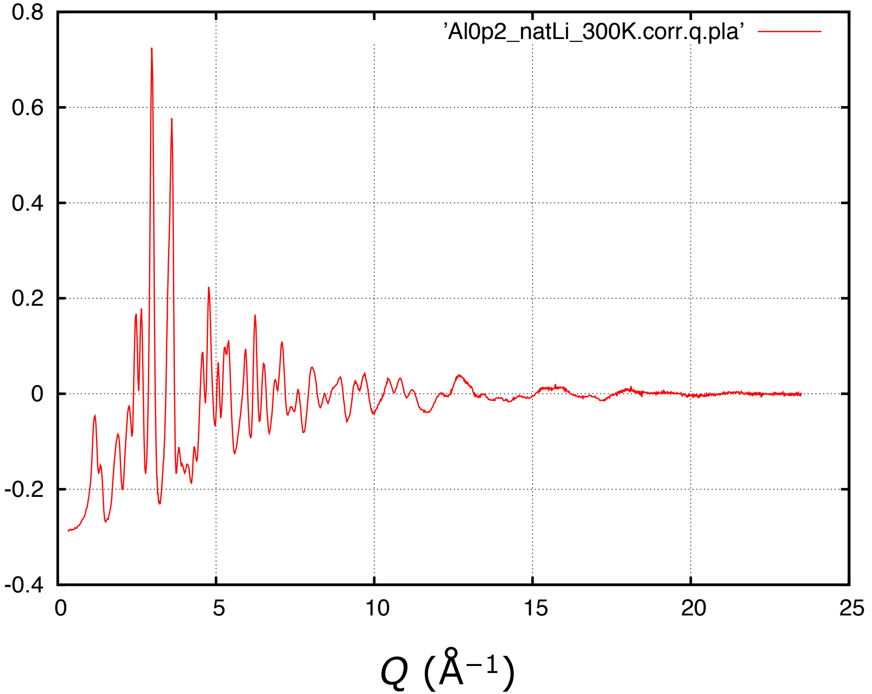
Limiting value:
bound cross section

Number density
Packing fraction
Container fullness
Sample composition

Example D4: $Li_{6.4}Al_{0.2}La_3Zr_2$

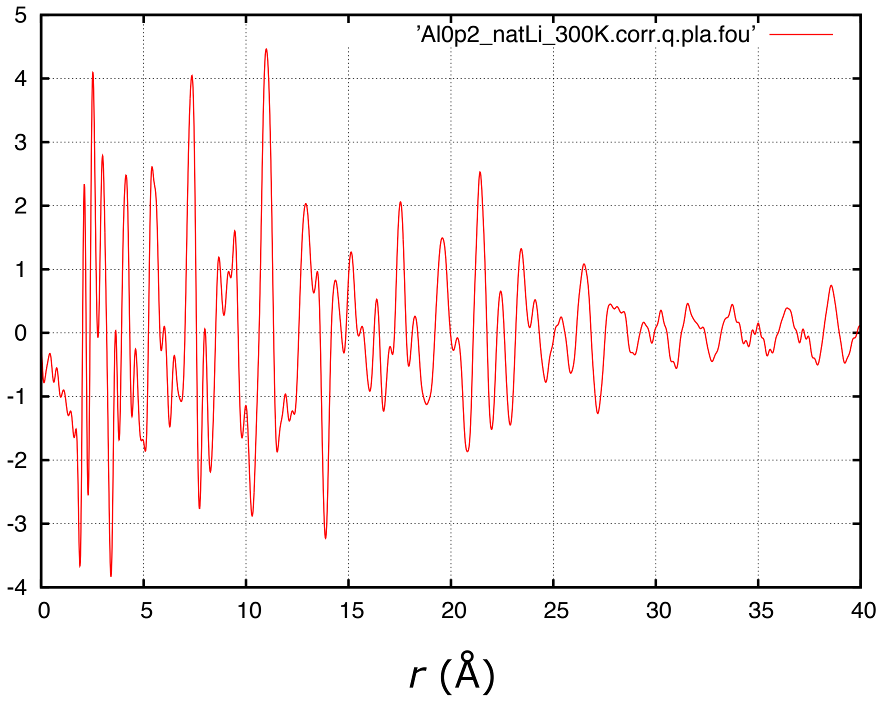
Final structure factor:

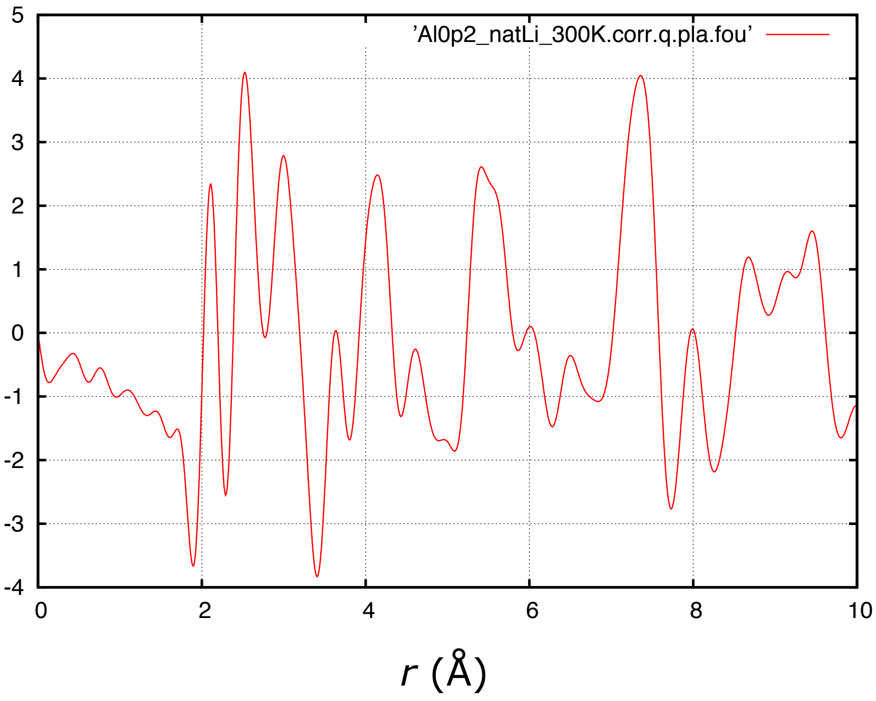
$S(Q)-1$



Example D4: $Li_{6.4}Al_{0.2}La_3Zr_2$

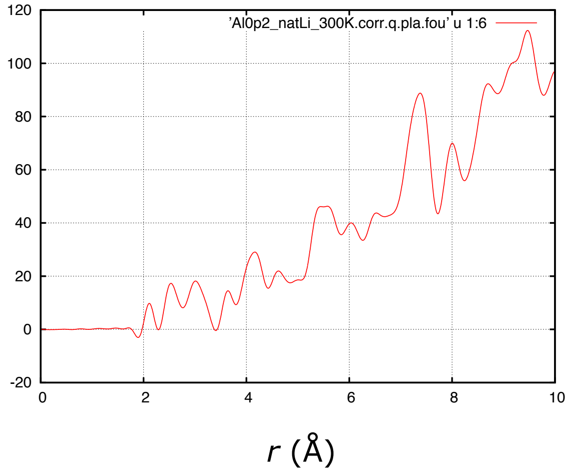
PDF: *G*(*r*), *D*(*r*)

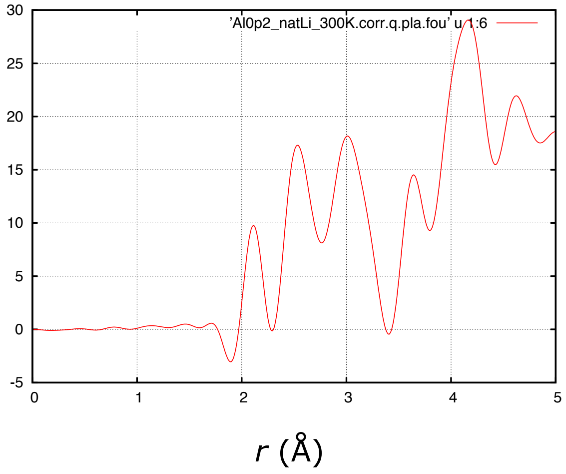


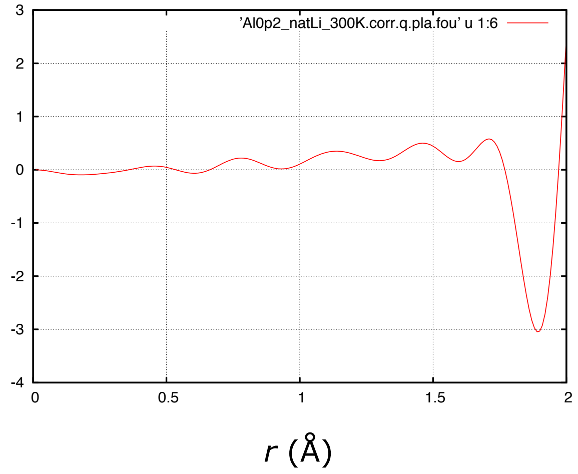


Example D4: $Li_{6.4}Al_{0.2}La_3Zr_2$

RDF







Normalisation in real space

Summary

$I_{\mathrm{SCE}}(Q)$ $I_{\mathrm{CE}}(Q)$ $I_{\mathrm{E}}(Q)$

$I_{\mathrm{VE}}(Q)$ $I_{\mathrm{ACE}}(Q)$

- At least 4 or 5 measurements
- Correction programs: CORRECT, GUDRUN, GO, …

  (not to be used as black boxes)
- Consistent normalisation, in *Q* and *r* spaces
- Patience, perseverance and imagination

Thanks for your attention!