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Investigation of the Correlation Technique at pulsed sources using Monte Carlo Simulations

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International Workshop on Applications of Advanced
Monte Carlo Simulations in Neutron Scattering

October 2-4, 2006, PSI, Switzerland

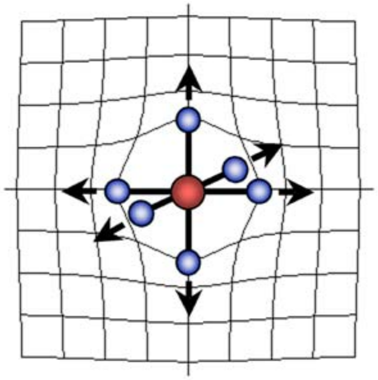
Scientific interest for developing new instrumentation

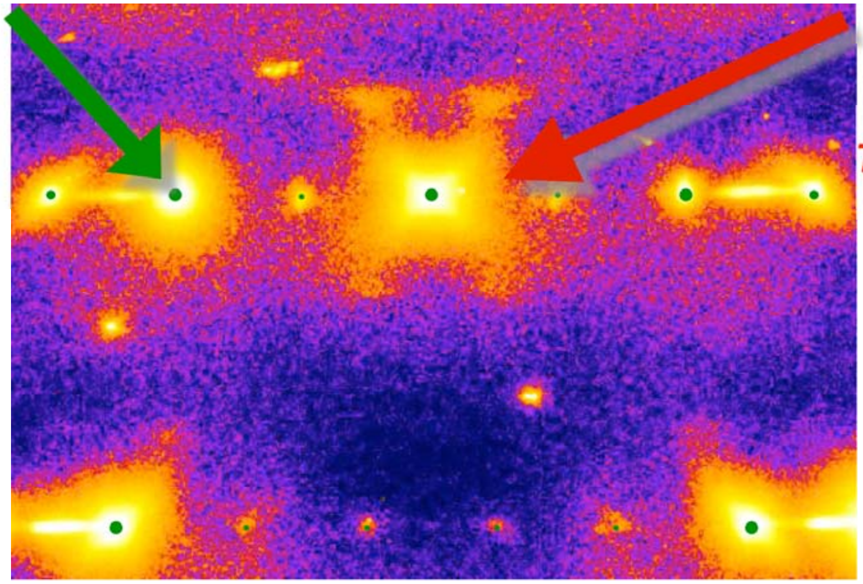
- **Many emerging phenomena are governed by complex disorder and short range correlations on the 1-10nm length scale**
    - Colossal Magnetoresistance
    - Geometric Frustration
    - Fast Ion Conduction
    - Nanoporous Host-Guest Frameworks
    - Relaxor Ferroelectricity
    - Molecular Crystals
    - Quasicrystals
    - Doped Semiconductors

Bragg Scattering

average structure

Jahn-Teller polarons in CMR manganites





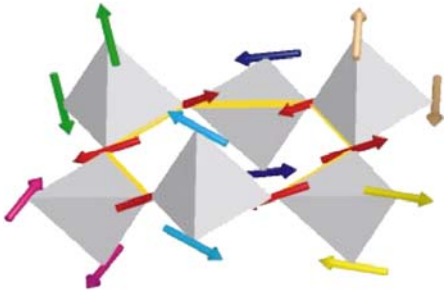
Diffuse Scattering

deviations from the average structure

short-range correlations

Topology of local structure

Hexagonal AF spin cluster in a geometrically frustrated magnet



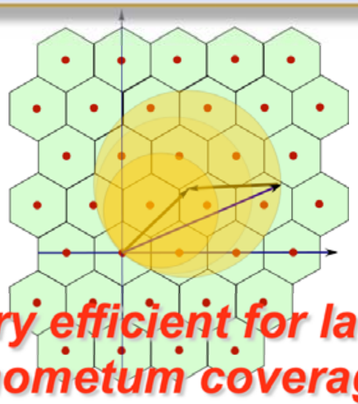
Challenges of Single Crystal Diffuse Scattering

- Diffuse scattering weak compared to Bragg signal.
- Requires both, high Q-resolution and large Q-coverage for accurate modeling.
- Energy discrimination required to separate quasi-static diffuse scattering from dynamic processes such as phonons.
- Quantitative analysis is usually confined to models of low complexity.

➠ Need to develop highly efficient tools for measuring and analyzing single crystal diffuse scattering from systems with *complex disorder*.

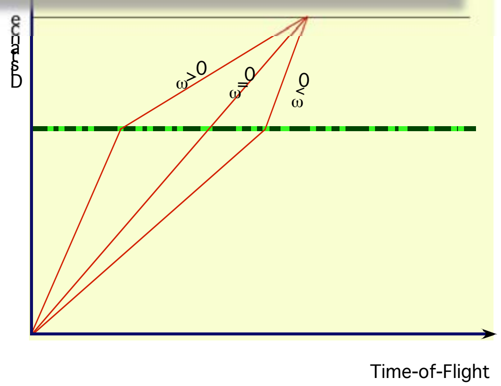
Proposal for highly efficient instrumentation

TOF Laue Diffractometer



Very efficient for large mometum coverage

Statistical Chopper



Intensity at detector when beam transmission is modulated in time by M(t)

$$I(t_0,t) = \int_0^{T_S} M(\tau - t_0) S(t,\tau) d\tau + B(t)$$

The scattering law can by reconstructed through the cross correlation

$$C(t,t_1) \equiv \frac{1}{T_S} \int_0^{T_S} M(t_1 - t_0) I(t_0,t) dt_0$$
$$= c_1 \cdot S(t,t_1) + c_2 \cdot B(t)$$

if M(t) obeys

$$A^{MM}(\tau) \equiv \frac{1}{T_S} \int_0^{T_S} M(t) M(t-\tau) dt = c_1 \cdot \delta(\tau) + c_2$$

L. Pal *et al*, *Neutron Inelastic Scattring*, p. 407 (Vienna, 1968)
R. Von Jan & R. Scherm, Nucl. Inst. Meth. **80**, 69 (1970)
P. Pellionicz *et al*, Nucl. Inst. Meth. **92**, 125 (1971)
W. Matthes, *Neutron Inelastic Scattering*, p. 773 (Vienna, 1972)
R. K. Crawford *et al,* ICANS-IX (1986)
L. D. Cussen *et al*, Nucl. Inst. Meth. A**314**, 155 (1992)

Cross Correlation Chopper

TOF Laue Diffractometer

- highly efficient data collection
- wide dynamic range in Q

Statistical Chopper

- elastic energy discrimination
- optimum use of white beam

Sample with :

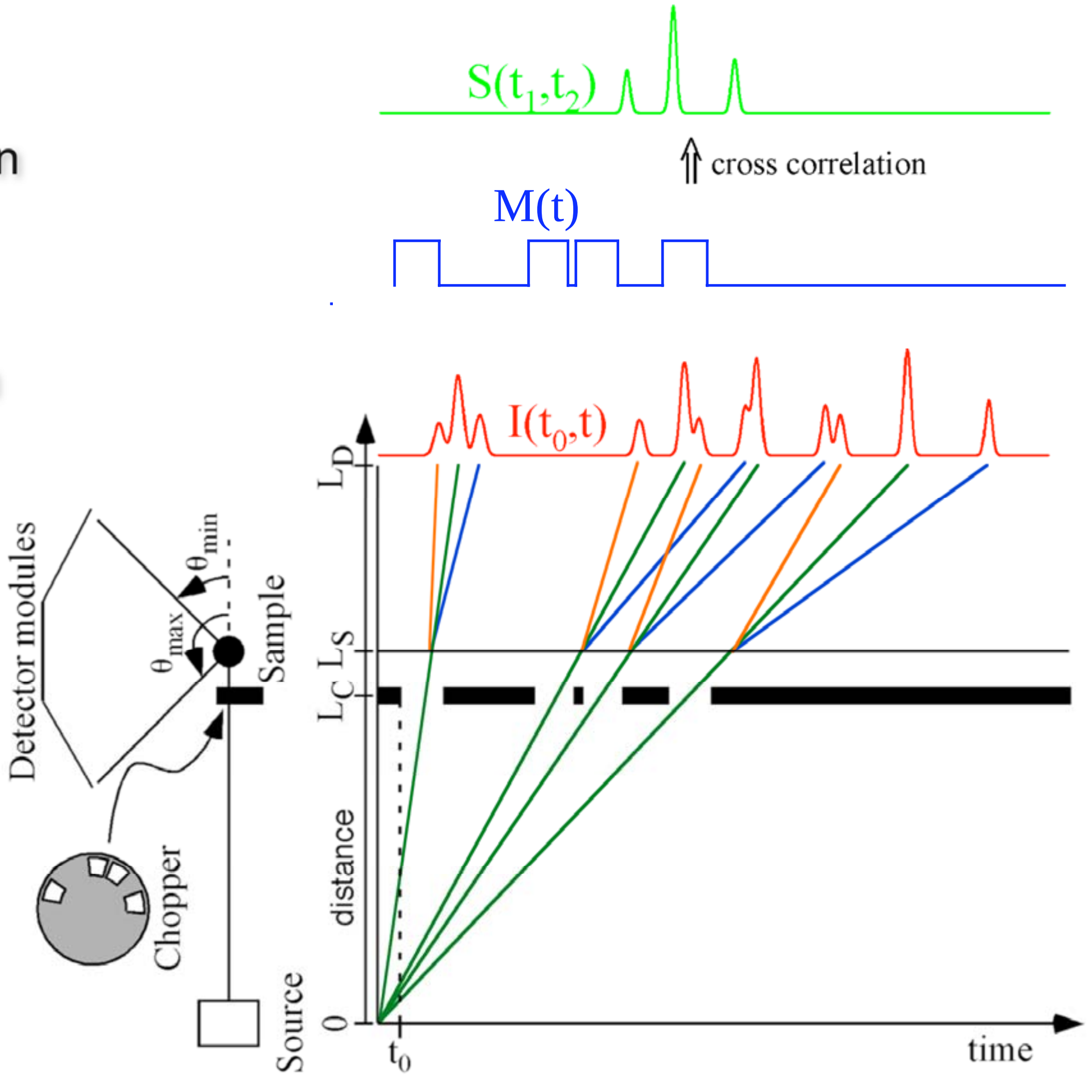
elastic scattering

$$\hbar\omega = 0$$

inelastic excitations

$$\hbar\omega = +E_0$$

$$\hbar\omega = - E_0$$



Statistics

Measured Intensity:

$$I(l,k) = \sum_{i} x(i-l)\,S(i,k) + b$$

Cross Correlation :

$$S(i,k) = \frac{1}{m(1-c)} \sum_{l} \left[x(i-l) - c\right] I(l,k) - b/m$$

$$\Delta S(i,k)^2 = \frac{1}{m(1-c)} \left\{ (1-2c)\,S(i,k) + c \sum_{l} S(l,k) + \left(1 - \frac{c}{m}\right) b \right\}$$

Gain Factor :

$$G = \frac{\left[\mathrm{rel.}\ \ var.\ \ S(i,k)\right]_{\mathrm{conv}}}{\left[\mathrm{rel.}\ \ var.\ \ S(i,k)\right]_{\mathrm{corr}}} \approx \frac{m(1-c)}{1 - 2c + \dfrac{2\beta c + cN}{\sigma_{ij} + \beta}}$$

Average Gain Factor:

$$G_{\mathrm{av}} \approx (1-c)\,\frac{1+\beta}{1 + \dfrac{\beta}{m}}$$

Potentially large gain for strong signals

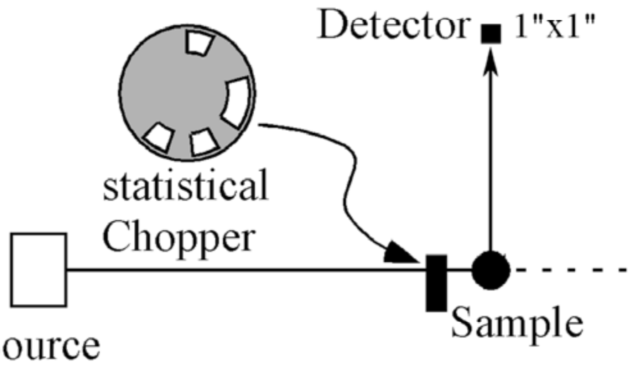
Full Experiment Simulations with McStas

Instrument : Christina (IPNS, Chex)

Chopper : N = 255; f = 198 Hz; R = 0.26 cm

Sample : harmonic oscillator

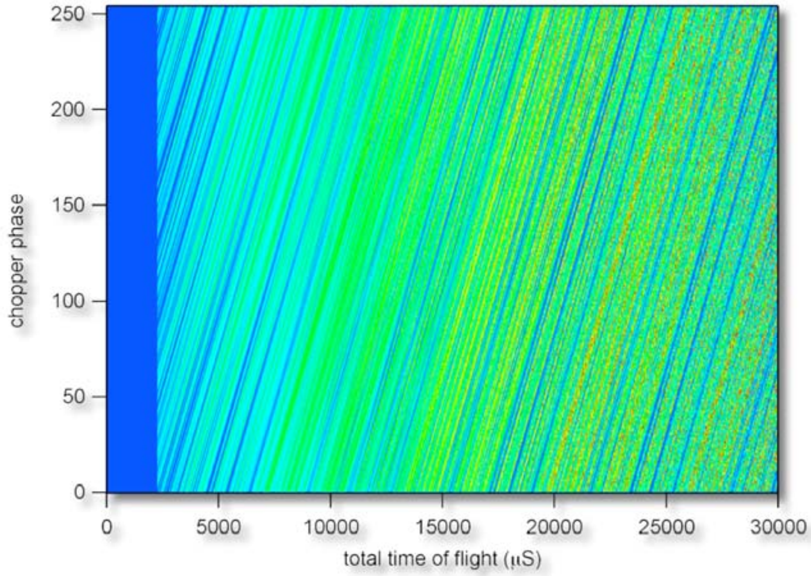
E0 = ±2 meV, M=60, T = 100K

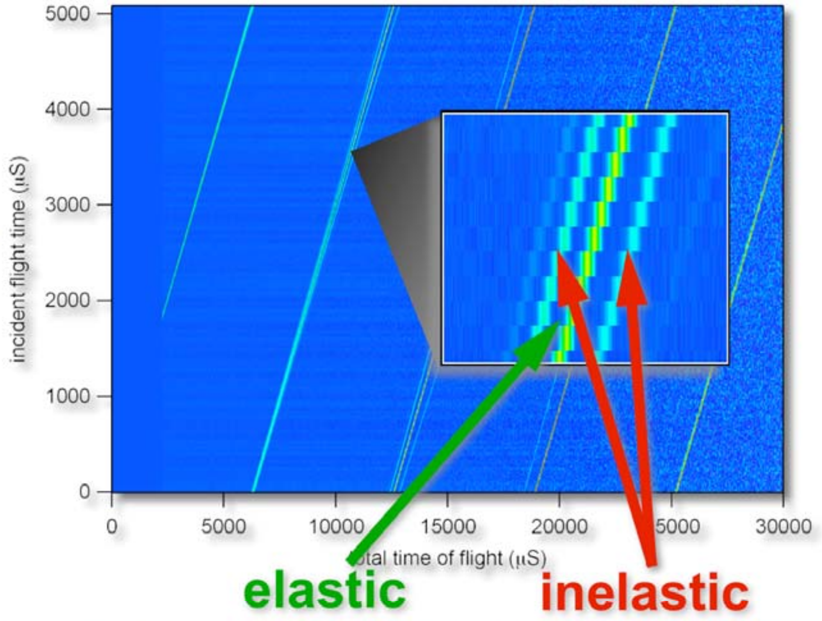


Raw Data

Cross Correlation

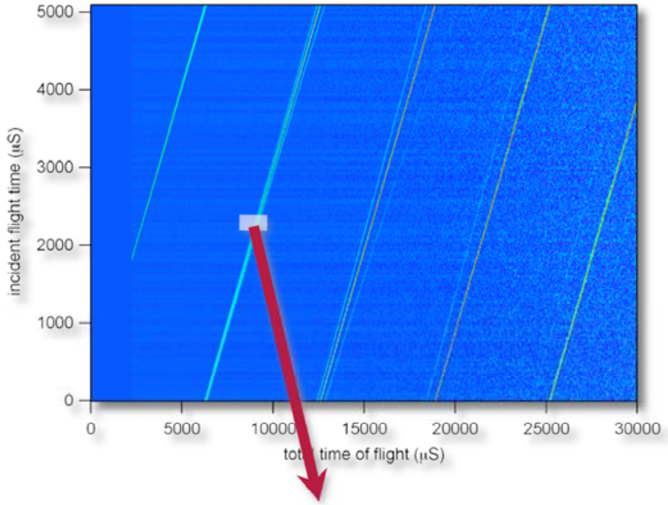
Reconstructed Scattering Function





Comparison with conventional chopper

Scattering Function



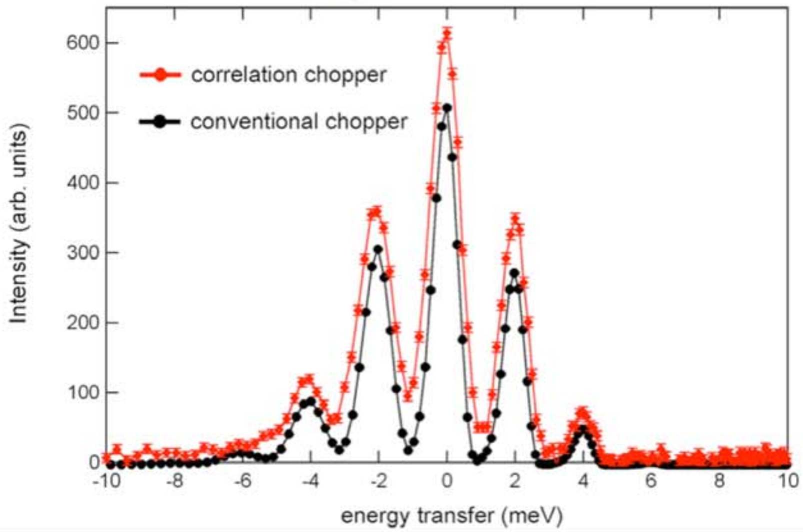
Sample: harmonic oscillator;

$E_0 = \pm 2$ meV; M=60; T = 100K

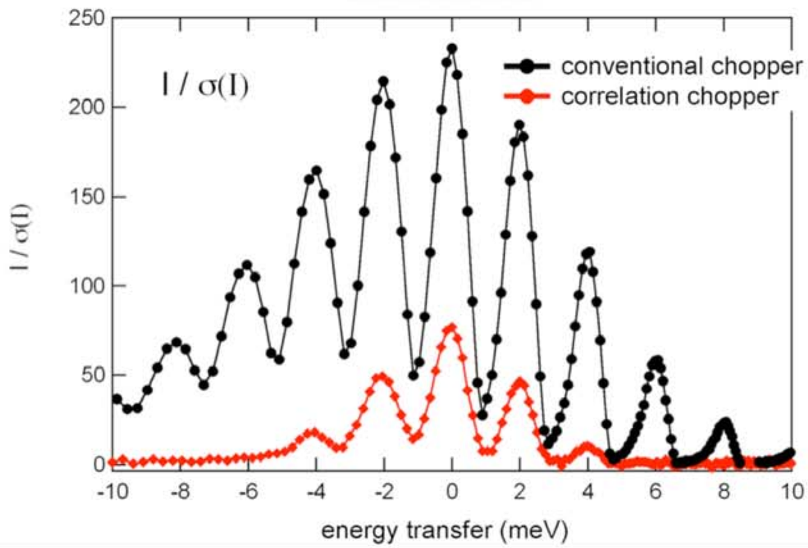
Instrument: CHEX (IPNS F-moderator; 30Hz)

Correlation Chopper : 198 Hz; N=255; c=50%

Conventional Chopper : 210 Hz; N=1; c~1%



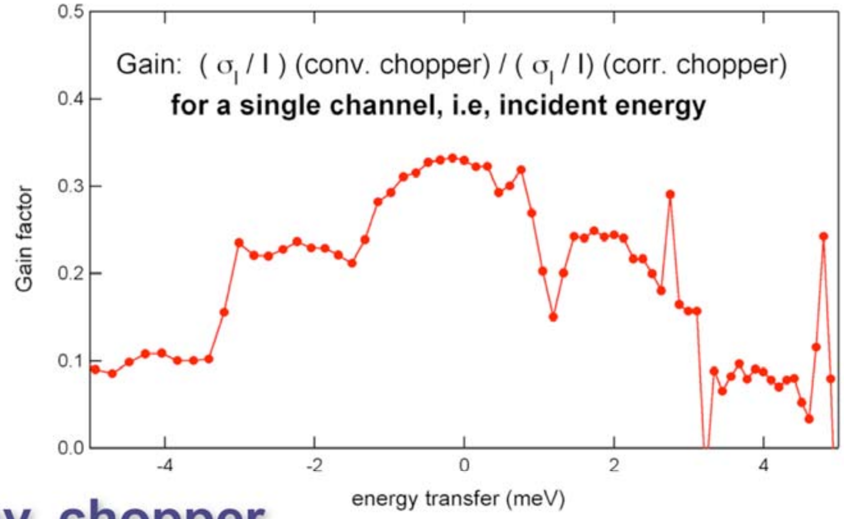
"Statistics"



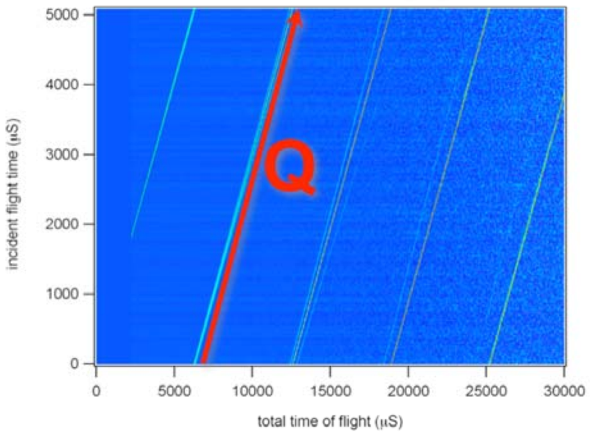
Gain in efficiency

$$\text{Gain} = \frac{\text{rel. var. S(conventional)}}{\text{rel. var. S(correlation)}}$$

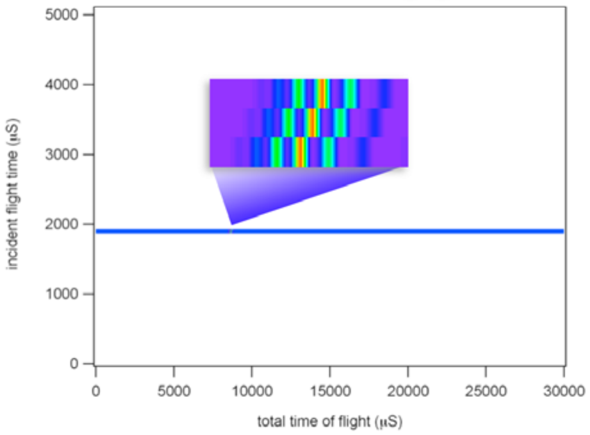
- Gain factor < 1 : less efficient
- However, Gain factor is for a single channel *but* the correlation chopper covers many channels simultaneously (N=255)

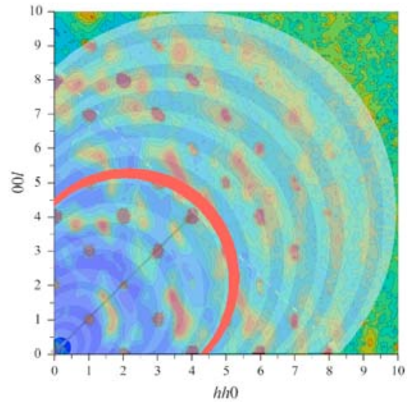


corr. chopper



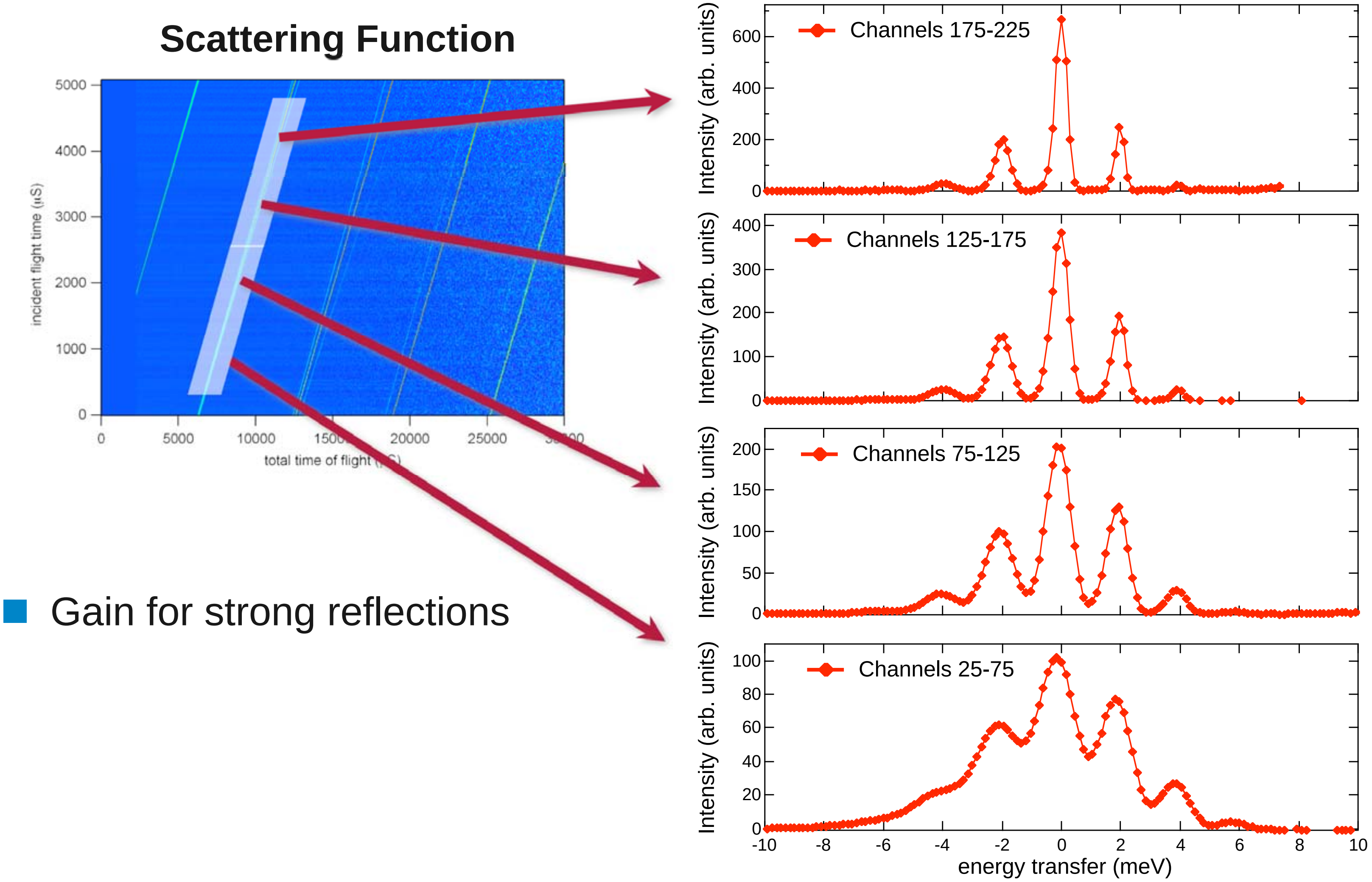
conv. chopper





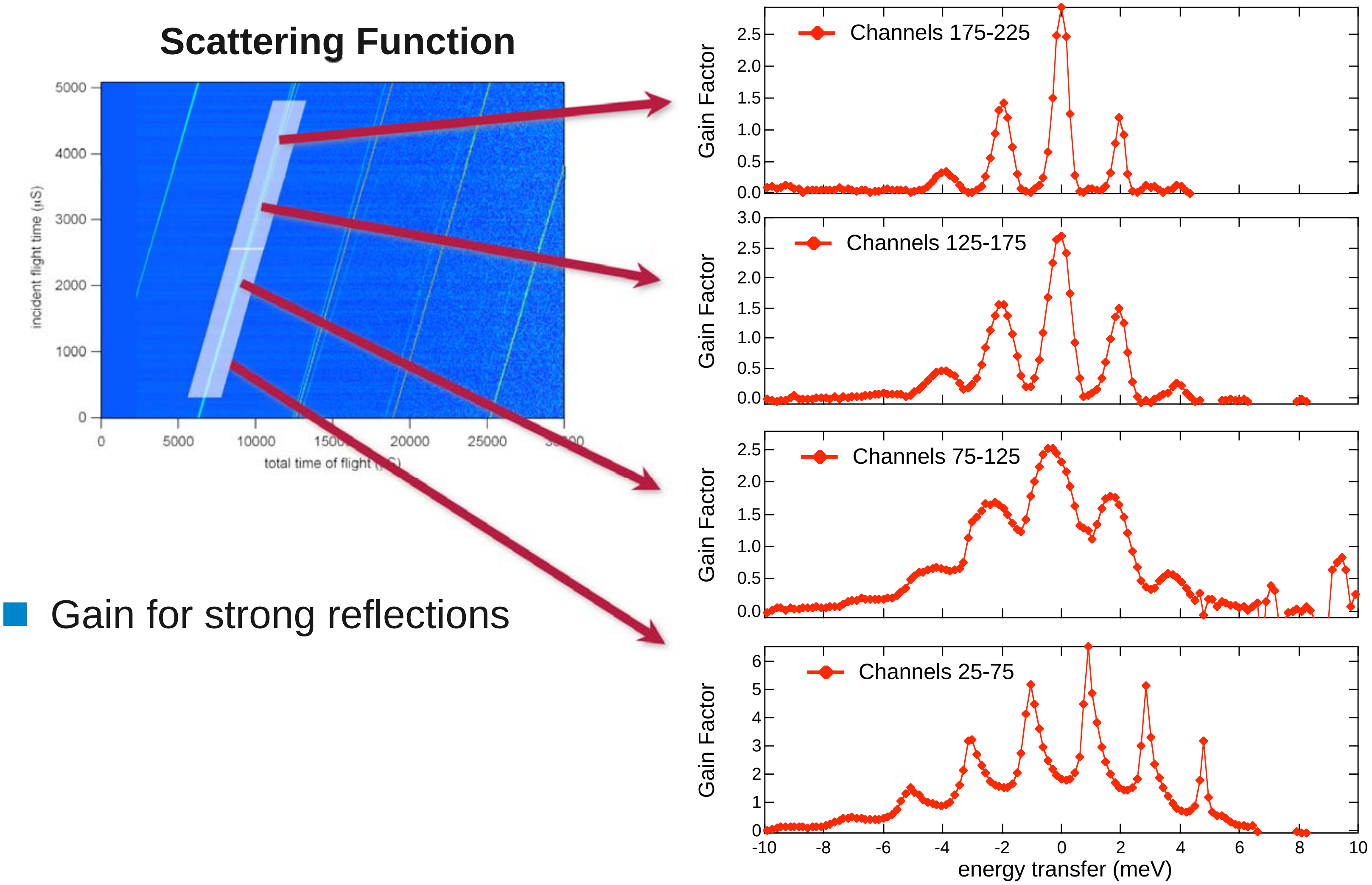
⇨ **Gain ~ N/10 ~ 30-50**

Gain Factor for Q-independent excitation



- Gain for strong reflections

Gain Factor for Q-independent excitation



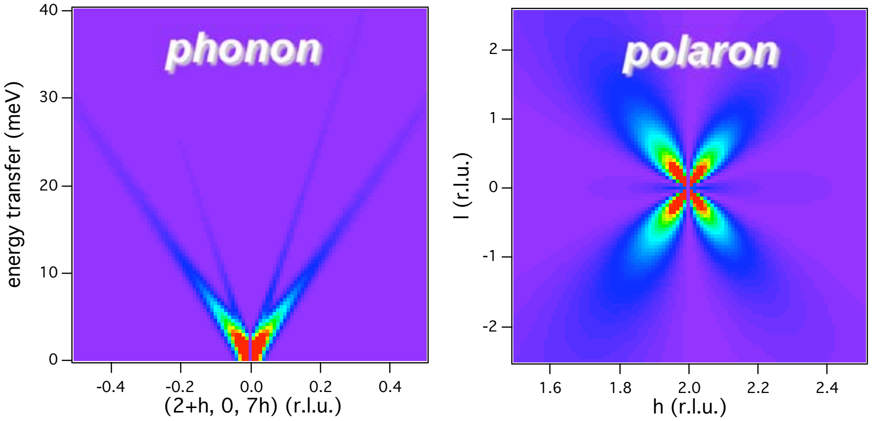
- Gain for strong reflections

Ongoing development

- Realistic single crystal diffuse scattering
  - ZrO_2
  - Manganites

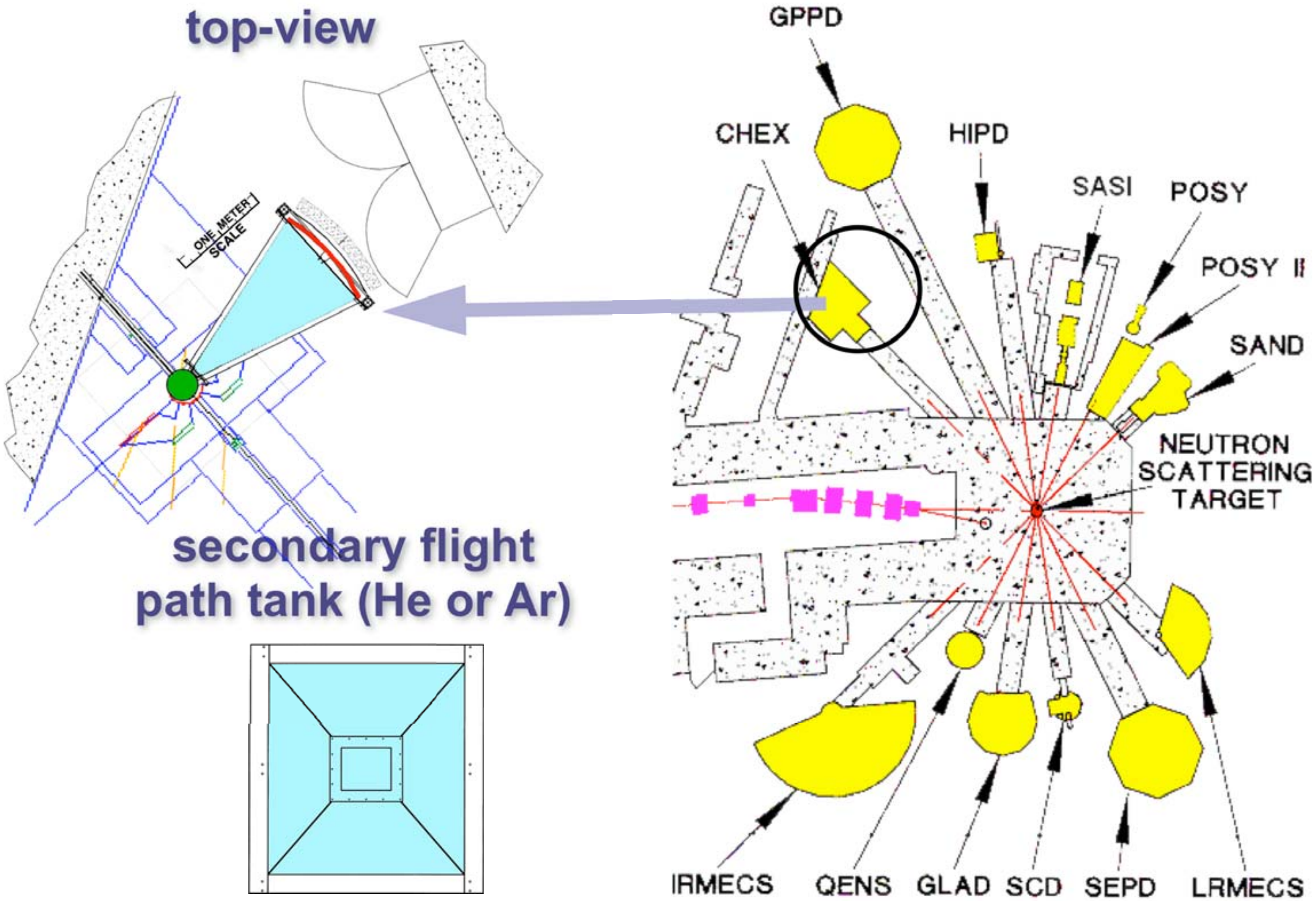
- Prototype
  - Comparison with simulation

- Proposal to SNS

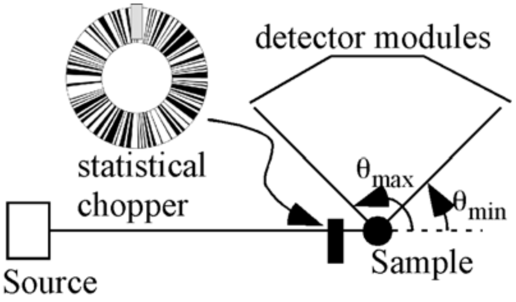


- Investigate other applications that could benefit using correlating technique
  - Energy and momentum dependence even for single scattering angle

Prototype Instrument at IPNS: Christina



Christina Design Parameters



- IPNS
  - 30 Hz
  - decoupled, poisoned moderator (CHEX - beamline)
- L_1 = 13.6 m
- Chopper: N=255, R~0.3m, 250Hz based on SNS bandwidth choppers (different disc design)
- L_2 = 2m, He (or Ar) filled secondary flight path
- ARCS-type detector
  - 40 Linear PSD's
  - ca 1"x1" spatial resolution
  - event-based histogram
- Resolution estimate (for E_i ~ 20meV)
  - dE ~ 2%
  - $dQ_{||} / Q_{||}$ (90°scattering angle) ~ 4.5×10^{-3}
- Energy resolution from simulation
  - conventional disc chopper: 2.1 %
  - correlation chopper : 2.5 %

SNS Instrument Proposal: CORELLI

Momentum range: $Q \sim 0.2 - 20$ Å^{-1} ➡ $E_i \sim 5 - 100$ meV

Resolution: $\Delta Q/Q \sim 3\times10^{-3}$ $\Delta\omega \sim 1$ meV

Sample size: $> 1\times1$ cm^2

Propose to build dedicated instrument at SNS:

- decoupled, poisoned moderator (short pulse)
- $L_1 \sim 20$m
- $L_2 \geq 2.5$m (energy resolution, sample size)
- ARCS type linear PSD's, large coverage
- correlation chopper with N ~ 256, c=0.5, R ≥ 0.3m, f ≥ 200 Hz

Estimated Resolution

E_i (meV)	$\Delta\varepsilon$ (meV)	$\Delta Q/Q$ (30°)	$\Delta Q/Q$ (60°)	$\Delta Q/Q$ (90°)	$\Delta Q/Q$ (150°)
10	0.11	6.4×10^{-3}	3.0×10^{-3}	1.8×10^{-3}	0.7×10^{-3}
20	0.31	6.4×10^{-3}	3.1×10^{-3}	1.9×10^{-3}	0.9×10^{-3}
50	1.18	6.5×10^{-3}	3.2×10^{-3}	2.0×10^{-3}	1.2×10^{-3}
75	2.17	6.5×10^{-3}	3.3×10^{-3}	2.2×10^{-3}	1.4×10^{-3}
100	3.33	6.6×10^{-3}	3.4×10^{-3}	2.3×10^{-3}	1.6×10^{-3}

Resolution estimates for *Corelli* based on: decoupled water moderator, $L_1 =$ 20m, $L_2 = 2.5$m, sample size 1x1cm^2, detector pixel size 2.5cm. The transverse resolution is generally much better except at very high scattering angle.

Conclusion

- Cross Correlation technique at pulsed source promises huge gains in efficiency for
  - Single crystal diffuse scattering with energy discrimination
  - Weakly Q-independent excitations
- Further investigations necessary to test efficiency for spectroscopy in constrained geometries
  - e.g. high pressure spectroscopy
- Prototype under construction
  - Limited flux, resolution, detector coverage
  - Sufficient for real experiments and further development of technique
- Proposal for dedicated instrument at SNS in preparation