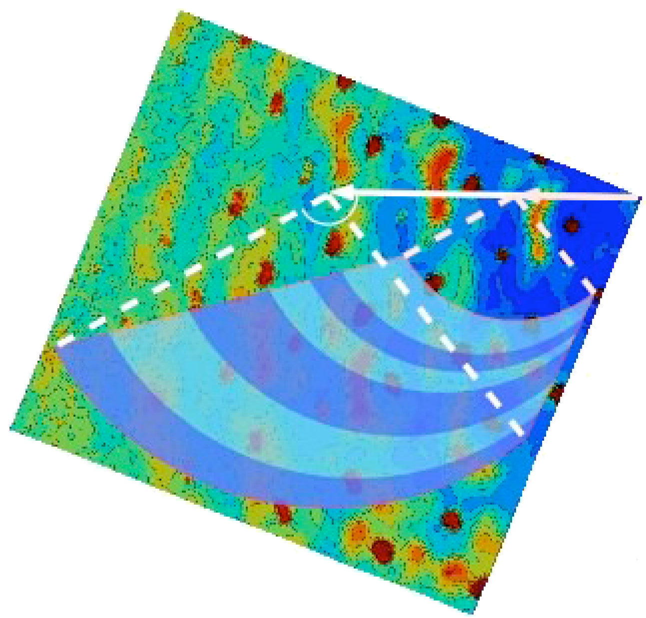
Cross Correlation Spectroscopy at Pulsed Neutron Sources

Stephan Rosenkranz and Ray Osborn



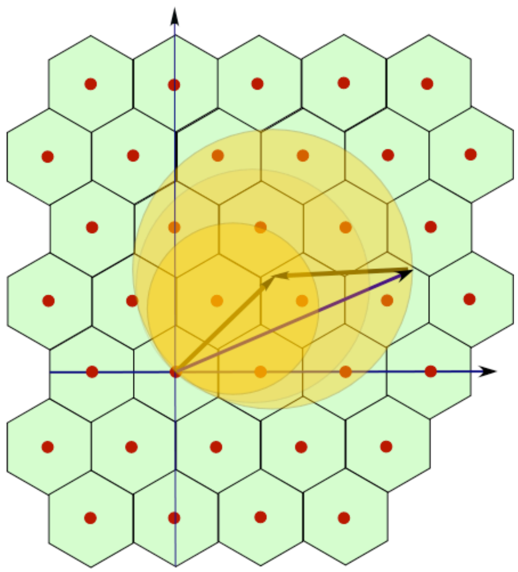
Goal

- Measure single crystal diffuse scattering S(Q,□=0) at a pulsed neutron scource
  - Over a large volume of reciprocal space
  - With energy discrimination
  - Good Q-resolution (at low Q's)
  - "large" samples (~cm's)
  - Low background

⇨ Cross-correlation Spectrometer

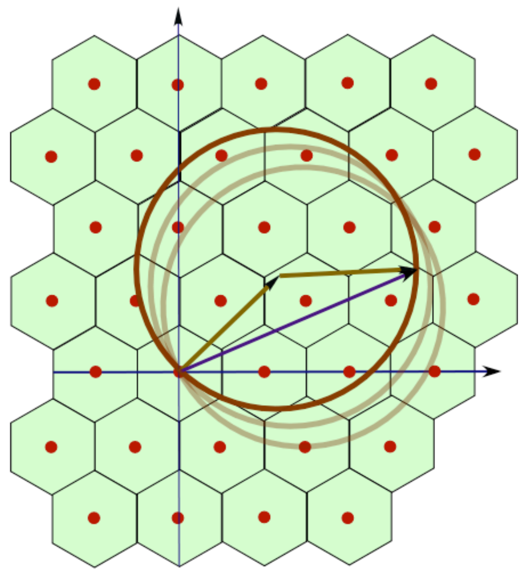
Conventional Techniques

White Beam



NO energy discrimination

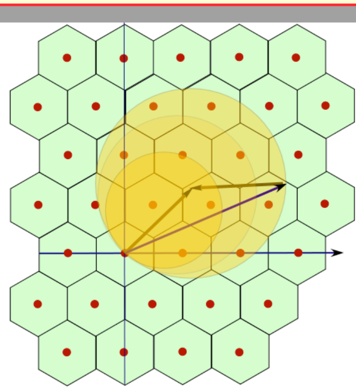
Fixed k_i



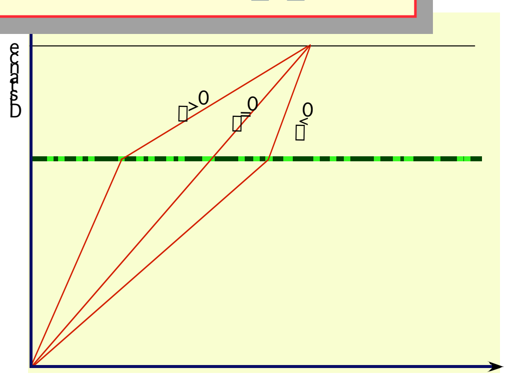
NOT efficient

Proposal

TOF Laue Diffractometer



Statistical Chopper



"combine white beam and fixed k_i"

1. TOF Laue Diffractometer
   - highly efficient data collection
   - wide dynamic range in Q
2. Statistical Chopper
   - elastic energy discrimination
   - optimum use of white beam

$$C(j,k) = \sum_{i=i_{\min}}^{i_{\min}+N} A(i-k)S(i,j-i) + B(j)$$

$$S(i,j) = \frac{2}{N+1}\sum_{k=1}^{N} A(i-k)C(i+j,k) - \frac{2}{N+1}B(i+j)$$

Principal of Correlation Technique

- First proposals and tests for high-efficiency time-of-flight methods ~ 1968
- Beam transmission modulated in time by M(t) with 0 ≤ M(t) ≤ 1
- Autocorrelation for pseudorandom sequence

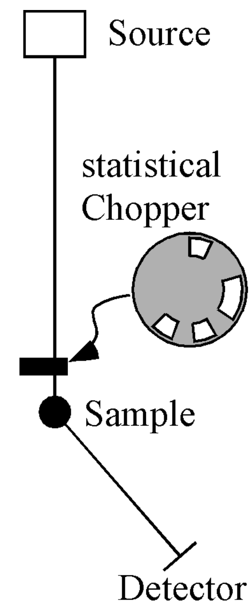
$$A^{MM}(\tau) \equiv \frac{1}{T_S}\int_0^{T_S} M(t)M(t-\tau)dt = c_1 \cdot \delta(\tau)$$

- Signal $I(t_0,t)$ due to sample scattering $S(t,t_1)$ and background B(t)

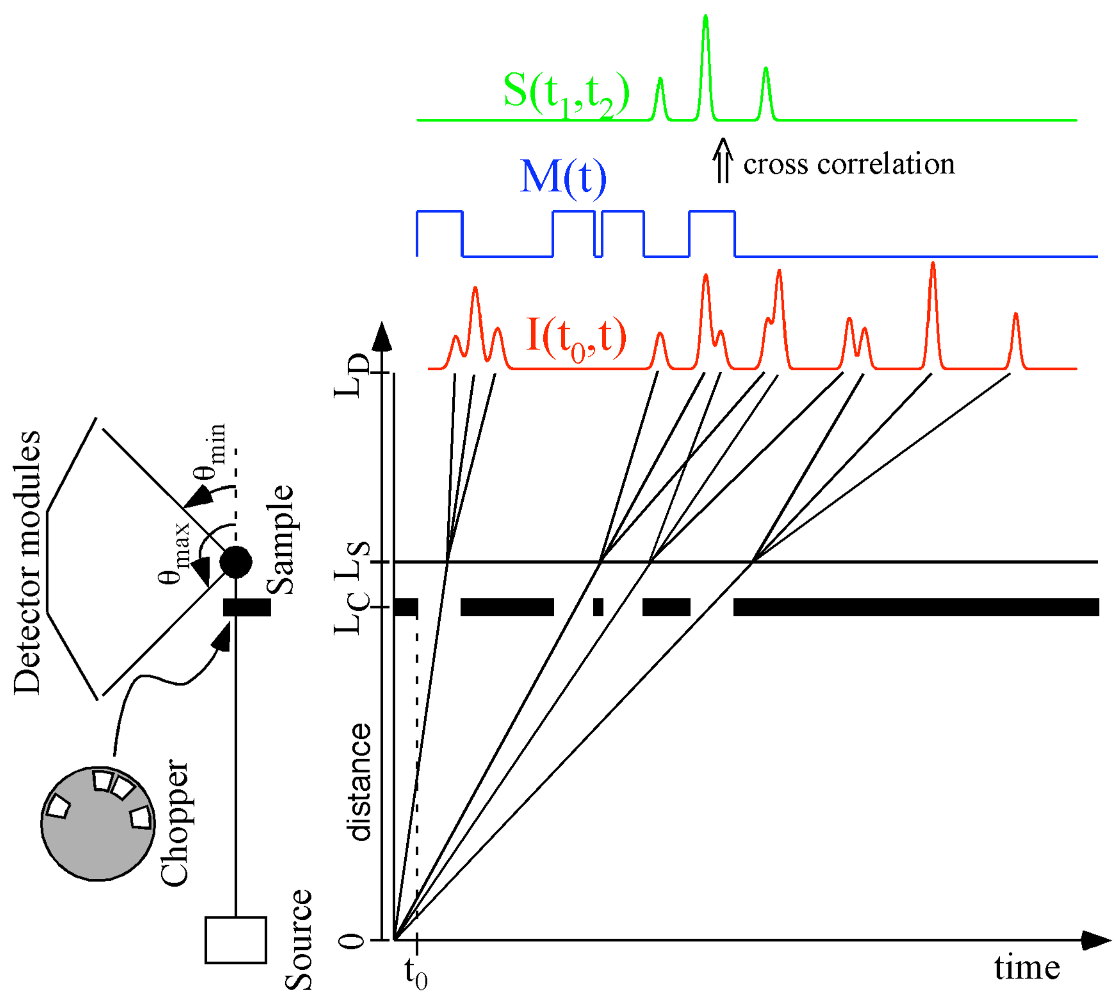
$$I(t_0,t) = \int_0^{T_S} M(\tau - t_0)S(t,\tau)d\tau + B(t)$$

- Cross correlation

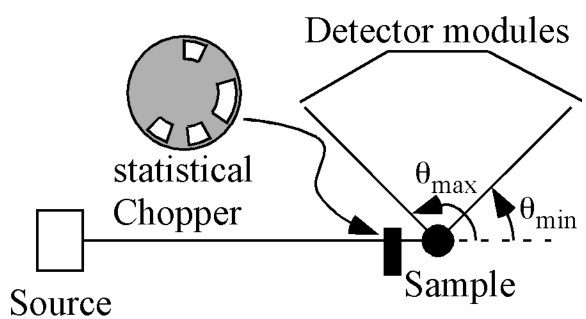
$$C(t,t_1) \equiv \frac{1}{T_S}\int_0^{T_S} M(t_1 - t_0)I(t_0,t)dt_0 = c_1 \cdot S(t,t_1) + c_2 \cdot B(t)$$

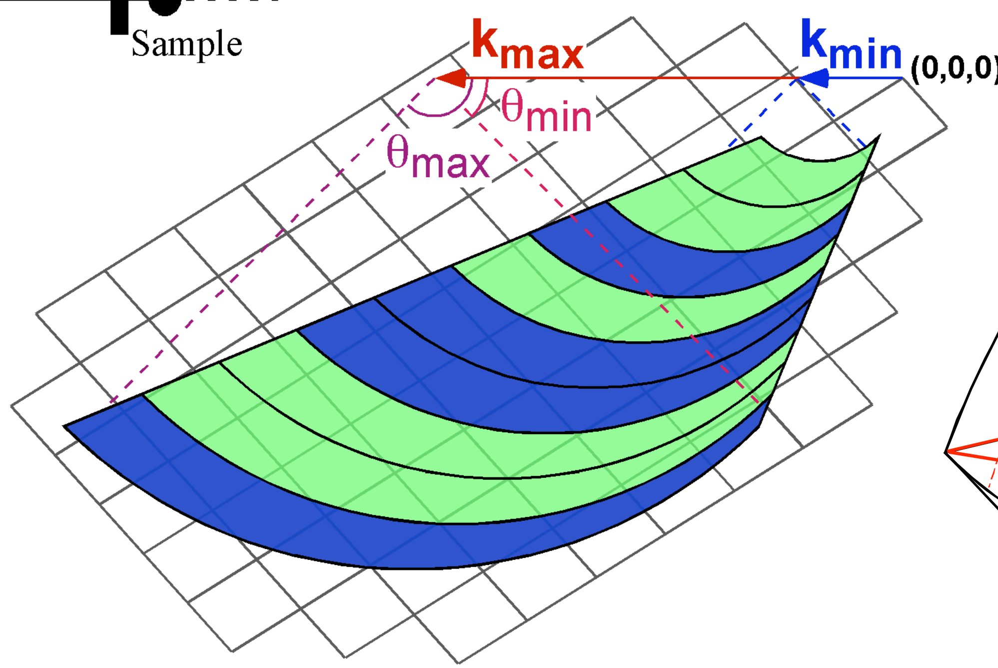


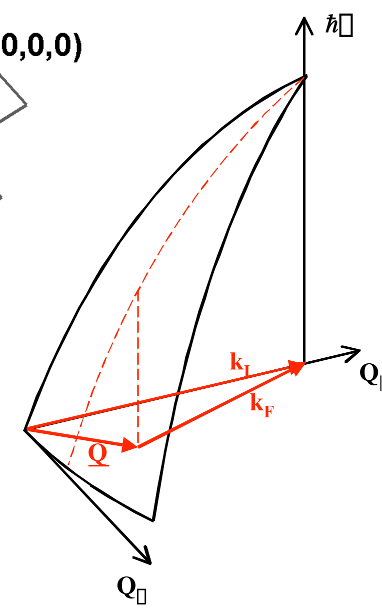
Cross Correlation t-D diagram



Elastic coverage







Statistics

- time-channel notation

$$C(j,k) = \frac{1}{N}\sum_{l=1}^{N} M(j-l)\,I(l, j-k)$$

$$S(i,j) = \frac{2}{N+1}\sum_{k=1}^{N} A(i-k)\,C(i+j,k) - \frac{2}{N+1} B(i+j)$$

- Relative signals

$$\square_{ij} = \frac{S(i,j)}{\bar{S}}, \quad \square_{j} = \frac{B(j)}{\bar{S}}$$

- Gain Factor

$$G = \frac{[\text{rel. var. } S(i,j)]_{conv}}{[\text{rel. var. } S(i,j)]_{corr}} \square \frac{m(1-c)}{1-2c+\dfrac{2\square c+cN}{\square_{ij}+\square}}$$

- Average gain Factor

$$G_{av} \square (1-c)\frac{1+\square}{1+\dfrac{\square}{m}}$$

Does it work?

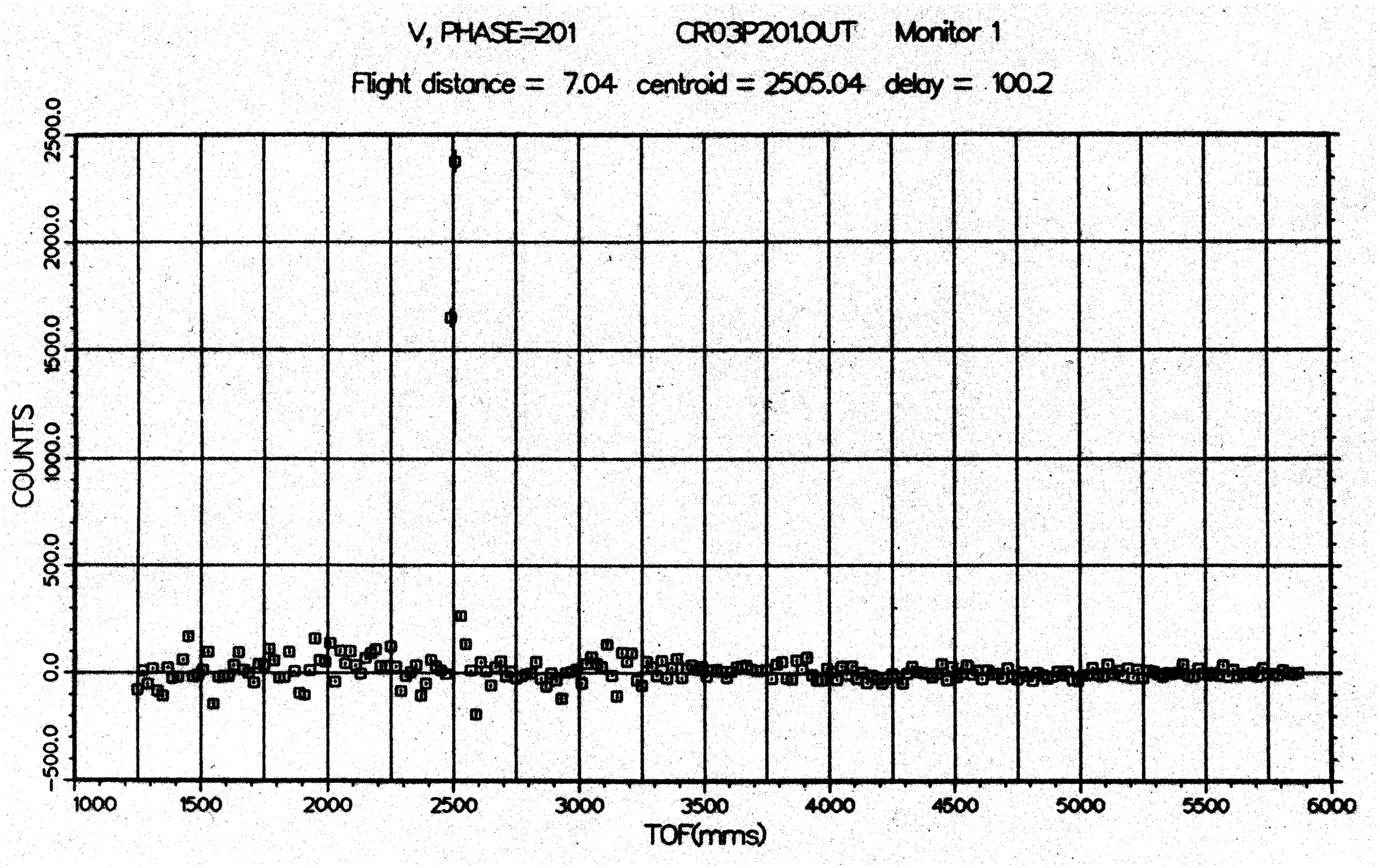
- Test performed on LRMECS at IPNS
- Chopper with 251 elements running at ~ 200Hz, asynchronously with the source (30Hz)
- Modified data acquisition module
- Measured standard vanadium sample for 4.5h
  (Test's terminated because of accelerator breakdown)

TEST OF A CORRELATION CHOPPER AT A PULSED

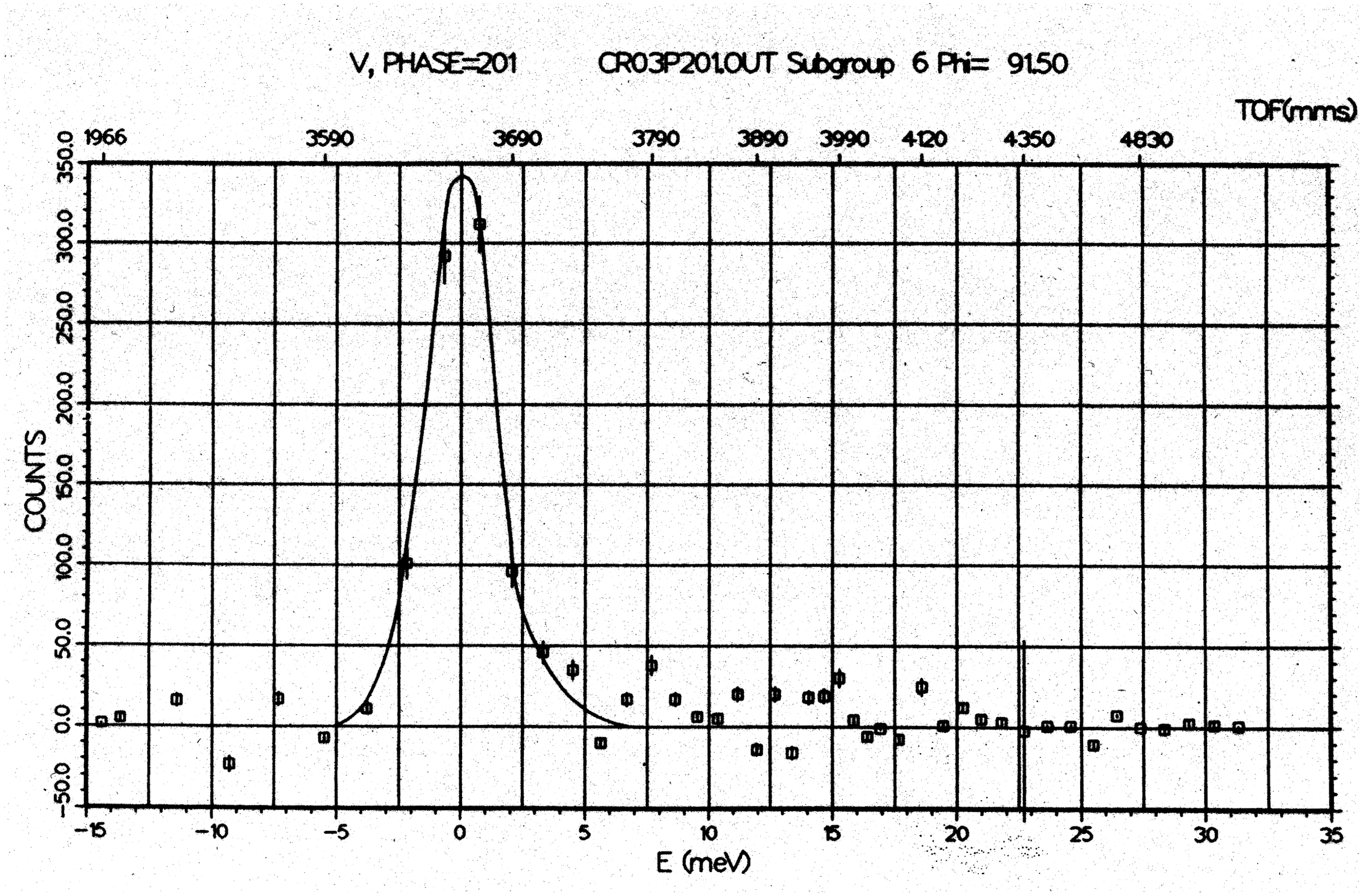
SPALLATION NEUTRON SOURCE*

R. K. Crawford,** J. R. Haumann,+ G. E. Ostrowski,**
D. L. Price,++ and K. Sköldt†
Argonne National Laboratory
Argonne, Illinois 60439

Monitor

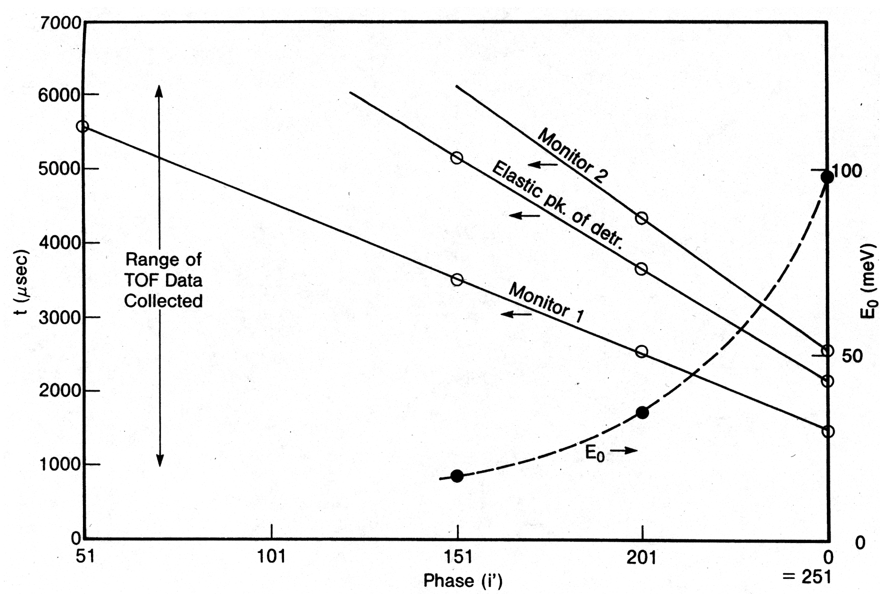

V, PHASE=201
CR03P201.OUT
Monitor 1
Flight distance = 7.04 centroid = 2505.04 delay = 100.2
COUNTS
-500.0
0.0
500.0
1000.0
1500.0
2000.0
2500.0
1000
1500
2000
2500
3000
3500
4000
4500
5000
5500
6000
TOF(mms)

Vanadium

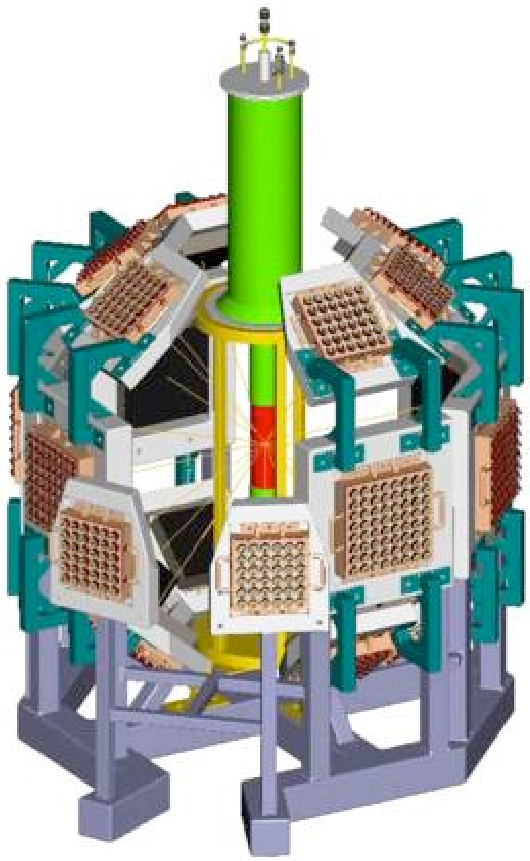

V, PHASE=201 CR03P201.OUT Subgroup 6 Phi= 91.50
TOF(mms)
1966
3590
3690
3790
3890
3990
4120
4350
4830
COUNTS
−50.0
0.0
50.0
100.0
150.0
200.0
250.0
300.0
350.0
−15
−10
−5
0
5
10
15
20
25
30
35
E (meV)

Elastic peaks

- Within accuracy of experiment, source pulse at t=0 for all neutrons
- Energy resolutions = 1.5 (E_0=17.5), 3.2 (25), 16.2 meV FWHM, slightly worse than typical value for Fermi choppers

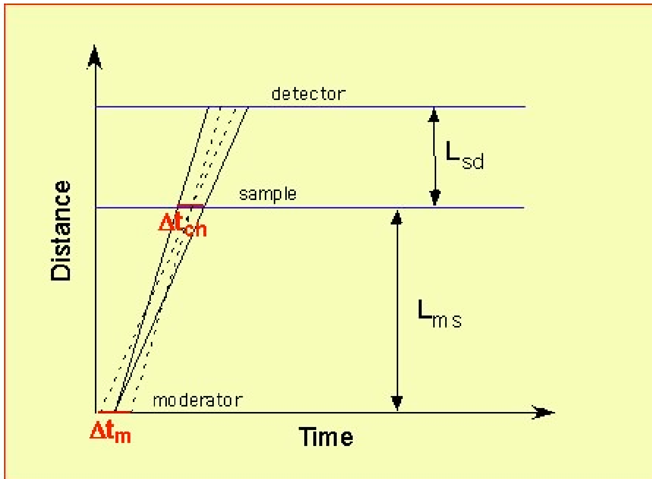


Option for existing Instruments?



- Diffuse scattering makes particularly stringent demands on instrumental backgrounds
  - The scattering is very weak
  - It is broadly distributed in Q-space
  - It is not easily identified *e.g.,* by Brillouin zone symmetries
- The current design of SCD envisages an open instrument
  - Bragg peaks are relatively easy to identify and integrate
  - However, there will be backgrounds from:
    - Air scattering
    - Extra sample environment windows
- We believe that it will be essential to have an evacuated secondary flight path

Resolution



$$\frac{\Delta\omega}{E_i} = \left[\left(2\frac{\Delta t_{ch}}{t_{ch}}\left(1+\frac{L_{ms}}{L_{sd}}\right)\right)^2 + \left(2\frac{\Delta t_m}{t_{ch}}\right)^2\right]^{\frac{1}{2}}$$

Q-resolution

SCD plans to have reasonable resolution

$\Delta d/d \sim 0.01$

with L_{ms} ~ 15m and L_{sd} ~ 1m

There are two problems with this value:

1. It is calculated at 90°.
   - We need good resolution at low Q
2. It neglects sample size.
   - We must accommodate large samples.

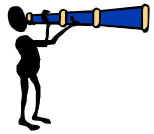
ω-resolution

- The secondary flight path of 1m is too small to give adequate energy resolution.

Summary

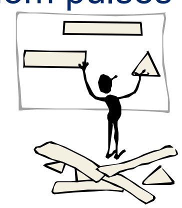
- It is unlikely that the demands of the crystallography and diffuse scattering communities can be satisfied by a single diffractometer.

  Diffuse scattering requires:
  - **low backgrounds**
  - **good resolution at low-Q**
  - **a longer secondary flight path for adequate energy resolution**
- Furthermore, the potential scientific demand for diffuse scattering is sufficient to justify a dedicated diffractometer, since:
  - **it overcomes previous technical limitations**
  - **there is increased interest in the science of complex disorder**
- We plan to hold a workshop NOW to strengthen the scientific case for such an instrument and establish the relevant scientific community.

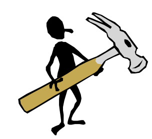


Future developments

- Thorough analysis of cross-correlation technique
  - Optimized statistical sequences required for different kinds of diffuse scattering
  - Best method fo generating and phasing the pseudo-random pulses to the source
  - Most practical wavelength range
  - Design parameters for an optimized chopper



- Build prototype Chopper and test performance for measuring single crystal diffuse scattering



We just received funding from DOE and IPNS