A Fast Method of Event Mode Neutron Diffraction Data Reduction for Spallation Neutron Source

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ABSTRACT

Using cross correlation, a fast event mode data reduction algorithm is developed for time-of-flight neutron diffraction time focusing. The method is verified by event mode neutron powder diffraction simulation and has been implemented at the newly commissioned diffractometer VULCAN as well as the powder diffractometer POWGEN. It is proven to have time accuracy better than the time resolution of neutron events collected. The time focusing computational time is reduced by a factor of 1500 compared to conventional approaches. In general it is applicable for event mode time-of-flight neutron diffraction data reduction at spallation neutron source.

Introduction

The advent of extremely high neutron flux, unique time event data acquisition and novel instrumentation at the Spallation Neutron Source (SNS) open up new possibilities, especially for in-situ dynamic and kinetic studies. Conventional histogram mode neutron data collection at time-of-flight (TOF) neutron sources provides a fixed time bin for histogram in detector electronics(Jorgensen *et al.*, 1989). At SNS, advanced chopper-detector synchronization techniques and data acquisition system provide high efficiency and high temporal resolution of neutron data collection. Unlike conventional histogram data at other TOF neutron sources, the time event data acquisition scheme at SNS records neutrons with a high-resolution timestamp of 10^{-7}s. The spatial resolution of modern detector is smaller with more detector pixels in a bank and it takes longer time to have enough statics per detector pixel for reliable single pixel Rietveld refinement, although neutron flux is higher. Histogram mode diffraction data reduction including time focusing and calibration is not suitable for the event data mode because of the new data format as well as time consuming Rietveld refinement of single pixel spectrum. In this paper, we developed a new and fast time focusing algorithm for data reduction of event mode time-of-flight neutron diffraction.

Event mode time focusing by cross correlation

With SNS neutron data acquisition scheme, a neutron event is recorded when a recordable neutron collected by a detector pixel, the smallest detector unit in a detector module. A duo of time of flight of the neutron and identification number of the detector pixel is stored as one neutron event, which is real 32 bit type in binary. The new method of event mode time focusing algorithm is carried out directly on event data by applying the first order time-of-flight correction. The details of event mode time focusing are discussed below.

For one individual detector pixel in TOF powder neutron diffraction, the relationship between the d-spacing for a particular powder line and its TOF is (Larson and Von Dreele, 2000):

$$Tof_i = DIFC_i \cdot d + DIFA_i \cdot d^2 + ZERO_i$$

where, *Tof* is the time of flight of neutron, DIFC, DIFA and ZERO are characteristic of a given detector pixel on a TOF powder diffractometer. In time focusing the significant factor is the diffractometer constant DIFC, which is calculated from detector pixel geometry by de Broglie equation:

$$DIFC = 252.816 \cdot 2\sin\Theta\left(L_1 + \sqrt{L_2^2 + L_3^2/16}\right)$$

where, Θ is the Bragg angle, L_1 is the primary flight path, L_2 is sample to detector pixel center distance and L_3 is the height of the detector pixel. By omitting the constant ZERO and the second order items, the equation above can be simplified as:

$$Tof_i = DIFC_i \cdot d$$

by applying arithmetic logarithm on the equation, we have:

$$\log(Tof_i) = \log(DIFC_i) + \log(d)$$

and, the logarithmic time offset of pixel to a reference pixel is given by:

$$offset_i = \log(Tof_i) - \log(Tof_0) = \log(DIFC_i) - \log(DIFC_0)$$

With an offsets table for detector pixel arrays, the corrected time of flight referred to the reference pixel can be easily calculated as below:

$$\log(Tof_{ci}) = \log(Tof_i) - offset_i$$

and histogram of Tof_{ci} is time focused diffraction pattern in time of flight referred to the reference pixel, whose real $DIFC_0$, ZERO and DIFA can be calculated by standard crystalline powder refinement in a Rietveld program, such as GSAS (Larson and Von Dreele, 2000).

Using cross correlation, the time offset table can be refined by a standard calibration powder diffraction data with relative good statistics per pixel. Given the diffraction spectrum of one detector pixel, $Y_i[tof]$, the cross correlation of any two detector pixels can be written as:

$$s_{ij}(n) = \sum_{m=tof_{\min}}^{tof_{\max}} Y_i[m] \cdot Y_j[m+n]$$

Central n can be given by offsets estimated from the geometric value of the individual detector pixel. Within small change of n, $s_{ij}(n)$ yields a statistical distribution and a general peak position fit such as by Gaussian distribution calculates directly the time offset between the two detector pixels.

Event mode time-of-flight Mn powder neutron diffraction is simulated for a detector module (Crow *et al.*, 2004) of the powder diffraction instrument, POWGEN (Hodges and Crawford, 2000) at SNS. The module has 1234 (8 by 154) detector pixels and each detector is 57x5 mm in vertical and horizontal direction, respectively. The reference detector pixel taken is at the center of the module where the Bragg angle Θ = 20.55^o, L_1=60 m, L_2=3.8900 m, L_3=0.005 m, and the calculated $DIFC_0$= 11341.268 μs/Å.

The time offsets calculated from the detector pixels geometry and refined from cross correlation, and also the difference of them are shown in Figure 1. Two lines in the plot overlap each other indicating the new method is accurate, and furthermore the differences between them are trivial and less than the time resolution (0.1μs) of neutron events collected, which are normal errors of $s_{ij}(n)$ peak position fit.

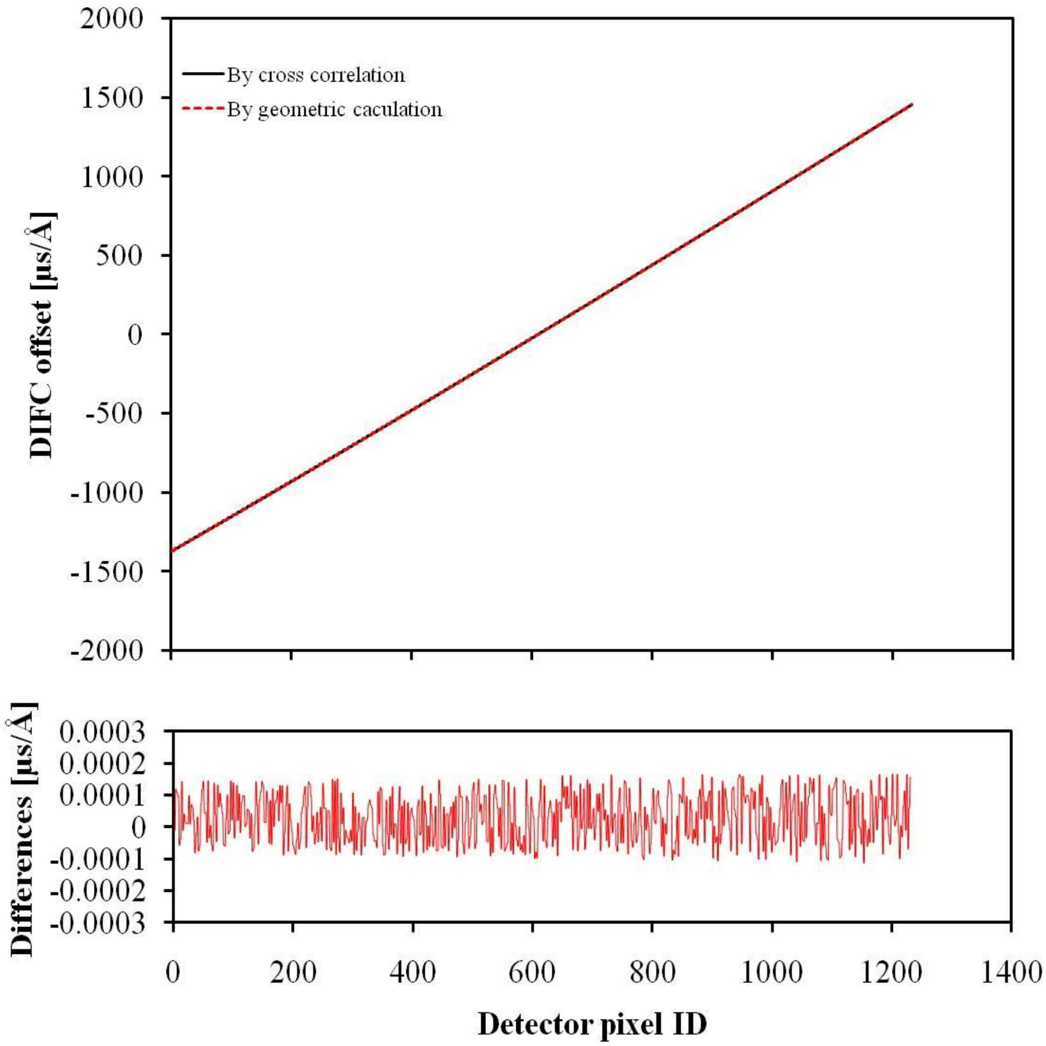

Figure 1, DIFC offset calculations from simulated neutron diffraction: comparison from cross correlation and geometric calculation.

Practical execution of this method has been successfully performed at the newly commissioned engineering diffraction instrument, VULCAN(Wang *et al.*, 2006), at SNS, which uses same type of detector modules. Figure 2 shows the measured and time-focused diamond powder from the 90 degree bank of 3 modules. Diamond powder diffraction was chosen for the practical execution of this method because of its strong neutron scattering and clearly separated sharp Bragg peaks. The diamond powder in a V can was collected for about 9 hours at 400kw accelerator power level. Raw time-of-flight neutron histogram of each pixel was generated. After subtracting the background of an empty V can, and being normalized to the highest peak intensity for each pixel, cross correlation was carried out for pixels in each module. Subsequently, after time-focusing neutrons in each module, Time offsets between the modules were refined by the same method. These different levels of offsets table allow flexible time-focusing over different groups of pixels and modules depending on the locations of the detector banks and experimental needs.

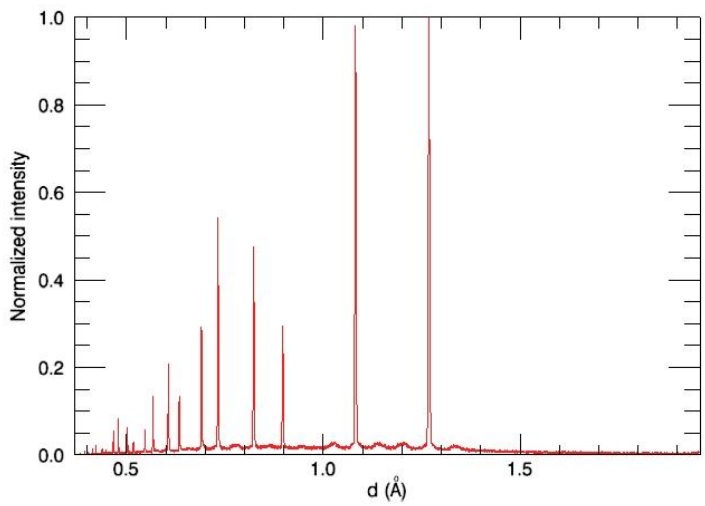

Figure 2, Time-focused diamond data collected at VULCAN instrument.

Advantages of the new method

Conventional time focusing of time-of-flight diffractions of detector tubes/pixels calculates the diffraction constant and other characteristics by full Rietveld refinement of individual tube/pixel spectrum, and corrections are applied on detector tubes/pixels histograms before time focusing. Furthermore, sometime interpolation of time bin is required during time-focusing due to fixed coarse time bin applied to the histogram. Both the refinement and time focusing is slow compared to the new method proposed here. Cross correlation is fast and only simple peak refinement is required to calculate the time offsets. The time focusing is directly applied to high time resolution event data and no interpolation is needed. In comparison of calibration refinements and time focusing, for a two-bank He tube detector system with 384 tubes at SMARTS (Bourke *et al.*, 2002), the engineering diffractometer at LANSCE, the refinement by SMARTSware (Bjørn Clausen) takes total 2 hour CPU time of a 2.4 GHz Intel processor (3.2 tubes/min). This cross correlation method yields 4928 pixels/min with an increased efficiency by factor of more than 1500. For an instrument like VULCAN which will have total 18 modules by 1232 pixels/module with relative good data statistics, the total cross correlation takes less than 5 mins; on the other hand with the conventional refinement, it may take up to 118 hours which is not feasible. For the extreme case of POWGEN with over 100 modules, the impact of this method is significant.

Once the offsets are generated, the offset table can be applied directly and quickly to event mode data either in real time or offline, and usually, the post time focusing takes less than one second for data set measured at VULCAN. This event mode time focusing also reserves the original high temporal resolution in event data. Alternative time bin from 1/60s to hours can be employed anytime for the nature of scientific needs, which is very valuable for dynamics and kinetic studies.

Conclusion

The new time-focusing method provides a fast event mode data reduction for TOF neutron diffraction time focusing. The method is verified to be accurate and efficient by neutron powder diffraction simulation. By the practical execution of this method at diffractometers at SNS. it was proven to be more feasible than conventional methods for modern TOF diffraction instruments with large scale detector coverage and fine spatial resolution detectors.

References

Bourke, M. A. M., Dunand, D. C. & Ustundag, E. (2002). pp. S1707-S1709.

Crow, M. L., Hodges, J. P. & Cooper, R. G. (2004). *Nuclear Instruments & Methods in Physics Research Section a-Accelerators Spectrometers Detectors and Associated Equipment* **529**, 287-292.

Jorgensen, J. D., Faber, J., Carpenter, J. M., Crawford, R. K., Haumann, J. R., Hitterman, R. L., Kleb, R., Ostrowski, G. E., Rotella, F. J. & Worlton, T. G. (1989). *Journal of Applied Crystallography* **22**, 321-333.

Wang, X. L., Holden, T. M., Rennich, G. Q., Stoica, A. D., Liaw, P. K., Choo, H. & Hubbard, C. R. (2006). *PHYSICA B-CONDENSED MATTER* **385**, 673-675.

Bjørn Clausen, SMARTSWare , http://public.lanl.gov/clausen/SMARTSware.html
Hodges, J.P. , Crawford, R.K., POWGEN3 conceptual design study, SNS Report No. IS-1.1.8.2-6034-RE-A-00, 2000
Larson, A.C. and Von Dreele, R.B. "General Structure Analysis System (GSAS)", Los Alamos National Laboratory Report LAUR 86-748 (2000).