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Modified cross-correlation for efficient white-beam inelastic neutron scattering spectroscopy

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ABSTRACT

We describe a method of white-beam inelastic neutron scattering for improved measurement efficiency. The method consists of matrix inversion and selective extraction. The former is to resolve each incident energy component from the white-beam data, and the latter eliminates contamination by elastic components, which produce strong backgrounds that otherwise obfuscate the inelastic scattering components. In this method, the optimal experimental condition to obtain high efficiency will strongly depend on the specific aim of the individual experiments.

1. Introduction

Inelastic neutron scattering has come to be recognized as indispensable in modern materials science, because a material's spin and lattice dynamics provides unique information about a system's Hamiltonian. A complete description of these excitations in momentum ($\boldsymbol{Q}$) and energy (E) space is needed to fully reconstruct the interactions that govern a material's behavior on the atomic scale. However, the technique normally requires a large volume of sample, often on the order of several cubic centimeters [1]. This is a very significant limitation in research to develop new materials with novel functions.

In neutron diffraction experiments, however, the development of time-of-flight (TOF) technique allowed the use of a white beam for increase in measurement efficiency compared with the conventional method using a monochromatic beam. Each wavelength (energy) component can be resolved by TOF, as shown in Fig. 1(a), and finally merged into a single diffraction $|\boldsymbol{Q}|$ pattern for a powder sample (time focusing) or a $\boldsymbol{Q}$ map for a single-crystal sample. Unfortunately, the same cannot be applied to inelastic scattering, because the different E_{i} (incident energy) and E_{f} (final energy) components are entangled at the same TOF for the same pixel on the detector, as shown in Fig. 1(b). Thus, it has been considered that either E_{i} or E_{f} must be monochromatized or must be analyzed, either of which incurs a large loss in neutron intensity.

Another remarkable method, called cross-correlation, was developed over four decades ago as an extension to the white-beam diffraction [2]. The method basically involves extracting the elastic components and removing the inelastic components [3]. As shown in Fig. 2, a special mechanical chopper modulates a white incident pulsed beam with a pseudorandom open/close sequence, and N-times cyclic phase shifts of the modulation generate a set of N data with different E_{i} contrast. Then, on the basis of the contrast, the data for each E_{i} can be mathematically resolved.

The mathematical formalization is given below. Here, for convenience, parameters and functions are renamed and redefined from those in the original papers [2,3]. The intensity detected at a specific TOF at a specific pixel of the detector, $I_{\mathrm{obs}}(p)$ ($p=1,\ldots,N$), is described by

$$I_{\mathrm{obs}}(p)=\sum_{j=1}^{N}F(j+p)I(j)+B \tag{1}$$

where p is the phase shift of the sequence (phase of sequence chopper), $F(k)$ is the kth element in the sequence F consisting of

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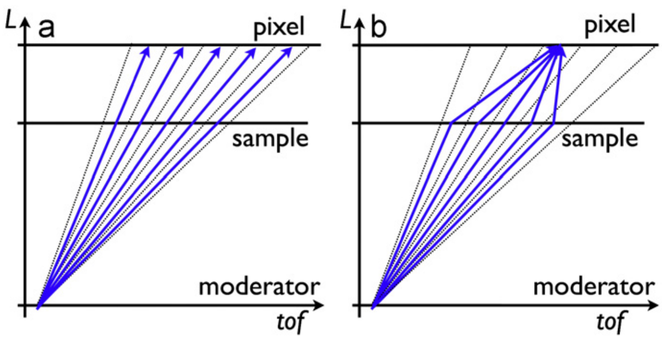

Fig. 1. (Color online) Plot of TOF against position without sequence chopper. Solid arrows indicate the most probable neutrons. Dotted lines sectionalize each E_i channel. (a) Diffraction. (b) Inelastic scattering.

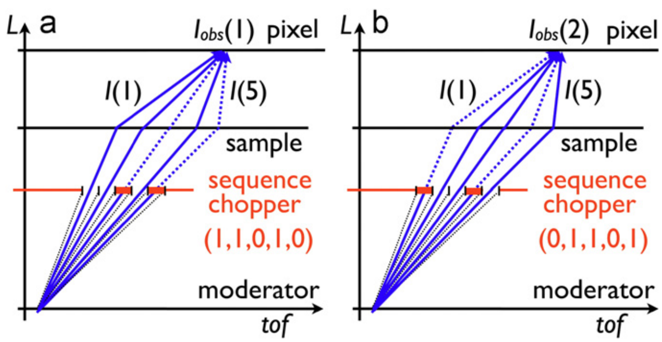

Fig. 2. (Color online) Example of TOF–position diagrams with sequence chopper ($N=5$) at sequence phases $p=1$ (a) and $p=2$ (b). By cyclically shifting the phase, a raw data set of $I_{obs}(1)$, $I_{obs}(2)$, $I_{obs}(3)$, $I_{obs}(4)$, and $I_{obs}(5)$ is obtained at a specific TOF and at a specific pixel on the detector. After the measurements, each E_i component of $I(1)$, $I(2)$, $I(3)$, $I(4)$, and $I(5)$ can be resolved mathematically.

only 0 (close) and 1 (open), $F(k+N)$ is defined to be equal to $F(k)$ for $k=1,\ldots,N$, j is an index for E_i, $I(j)$ is the intensity coming from the jth E_i component in a white incident pulsed beam for $j=1,\ldots,N$, and B is the background. The pseudorandom sequence F is restricted by

$$N=2^n-1 \quad (n:\text{integer}) \tag{2a}$$

$$F'(k)=2F(k)-1 \tag{2b}$$

$$\sum_{k=1}^{N} F'(k)=1 \tag{2c}$$

$$\sum_{k=1}^{N} F'(k)F'(k+k')=(N+1)\delta_{0,k'}-1. \tag{2d}$$

This type of sequence F is currently called a maximum length sequence, which is generated by a simple recurrence formula and is widely applied in the field of digital communications [4]. Combining the above equations, one can resolve each E_i component

$$I(j)=\frac{2}{N+1}\sum_{p=1}^{N}F'(j+p)I_{obs}(p)-\frac{2}{N+1}B. \tag{3}$$

It should be noted, however, that the method cannot be directly applied to inelastic scattering because the elastic components and their large statistical errors obfuscate the very weak inelastic components. This is probably why the method has not been realized thus far in an actual instrument dedicated to inelastic scattering. In fact, for the new CORELLI instrument under construction at the Spallation Neutron Source (SNS) in Oak Ridge, TN, the method will be used mainly to study diffusive elastic scattering such as in frustrated systems and ionic conductors [5].

This paper presents a modification to this method for a more practical white-beam inelastic neutron scattering setup. The proposed method has two novel aspects: the introduction of an inverse matrix representation and a proposed method for selective extraction. The former affords a different solution to Eq. (1) to resolve each E_i component in the white-beam data. The latter eliminates contamination by elastic components, which otherwise produce strong backgrounds. Finally, we present the estimates of some instrumental specifications for the TOF polarized neutron spectrometer, POLANO, being constructed at J-PARC.

2. Inverse matrix representation

We formalize an alternative solution to Eq. (1) without the conditions Eqs. (2a)–(2d). Here, the measurement of $I_{obs}(p)$ ($p=1,\ldots,N$) is the same as in the original method except for the kind of sequence. Ignoring B for simplicity, Eq. (1) can be represented by

$$\boldsymbol{I}_{obs}=\hat{F}\boldsymbol{I} \tag{4}$$

where $\boldsymbol{I}_{obs}$ is the vector $[I_{obs}(p)]$ ($p=1,\ldots,N$); $\hat{F}$ is the matrix $[\boldsymbol{F}_{p=1},\boldsymbol{F}_{p=2},\ldots,\boldsymbol{F}_{p=N}]$; $\boldsymbol{F}_p$ is the sequence vector $[F(j+p)]$ ($j=1,\ldots,N$); and $\boldsymbol{I}$ is the vector $[I(j)]$. Hence, one can resolve $\boldsymbol{I}$ by

$$\boldsymbol{I}=\hat{F}^{-1}\boldsymbol{I}_{obs}. \tag{5}$$

Consider, for example, the sequence (0, 1, 1, 0, 1) at $p=1$

$$\begin{bmatrix} I_{obs}(1)\\ I_{obs}(2)\\ I_{obs}(3)\\ I_{obs}(4)\\ I_{obs}(5)\end{bmatrix}=\begin{bmatrix} 0&1&1&0&1\\ 1&0&1&1&0\\ 0&1&0&1&1\\ 1&0&1&0&1\\ 1&1&0&1&0\end{bmatrix}\begin{bmatrix} I(1)\\ I(2)\\ I(3)\\ I(4)\\ I(5)\end{bmatrix}. \tag{6}$$

Hence, one can obtain

$$\begin{bmatrix} I(1)\\ I(2)\\ I(3)\\ I(4)\\ I(5)\end{bmatrix}=\frac{1}{3}\begin{bmatrix} -1&-1&-1&2&2\\ 2&-1&-1&-1&2\\ 2&2&-1&-1&-1\\ -1&2&2&-1&-1\\ -1&-1&2&2&-1\end{bmatrix}\begin{bmatrix} I_{obs}(1)\\ I_{obs}(2)\\ I_{obs}(3)\\ I_{obs}(4)\\ I_{obs}(5)\end{bmatrix}. \tag{7}$$

Thus, almost all types of sequences can be used as long as F^{-1} exists. Taking into account B again, one can also identify a sequence to minimize $|F^{-1}(B,B,B,B,B)|$, for example, by trial and error with many numerical trials.

It should be noted that this general matrix formalization is not considered superior to the maximum length sequence. However, the general matrix formalization does afford an advantage when the conditions of Eqs. (2a)–(2d) are not satisfied on actual instrumentation, for example, because of insufficient switching speed between 0 and 1 for high E_i range or high resolution. In this paper, we use the general matrix formalization only because selective extraction, proposed in the next section, also does not fulfill the conditions.

3. Selective extraction

We explain the proposed selective extraction method using the above example in Eq. (6). First, one needs to prepare another chopper with the inverted sequence—from open/close to close/open, that is, from 1/0 to 0/1. The inverted chopper gives another