New Trends in Thermal Neutron Spectrometry Getting High-Resolution Performances in Thermal Neutrons Spectrometry by Using Inverse Space Focusing

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Abstract. The characteristics of a Q-space focusing configuration are given. The procedure to get inverse space conditions is given also. Both the diffraction and inelastic scattering are considered. For the neutron diffractometry both instruments with one crystal monochromator and with two crystals monochromators are taken into consideration. The time of flight diffractometers both for a pulsed neutron source and a steady state one is considered also. For inelastic scattering the inverse space focusing conditions for triple axis spectrometers are given line-width.

Introduction
In neutron spectrometry the optimal use of the available neutrons is of the greatest importance. The general philosophy to do it is to maximize the neutron flux at sample by using neutron guides, supermirrors [1], or spatial focusing effects involving flexible configurations and curved crystals. A different approach [2], [3], is to obtain the required optimum experimental conditions not by getting focused beams at sample or anywhere else, but only by decreasing as much as possible the scan variable variances.

A Q-space focussing configuration is characterized by high-resolution even when no spatial focussing exist at sample position or anywhere else or is used quite open beam without any Soller collimators; only coarse collimators should be used to reduce the background level.

This is possible just by decreasing the scan variable variances.

The following steps should be followed:

- to define the scan variable
- to define the significant spatial variables as are the monochromator and sample width for example
- to express the scan variable, using specific geometry characteristics and the existing correlation between variables, through the spatial variables
- to cancel the significant contributions to the scans variable variances by cancelling the important variables coefficients, from the scan variable expression; when correlation between variable exists, as is the case for these kind of experimental configurations, such a coefficient has more then one term, not all of the same sign, appearing the possibility to be cancelled and therefore to cancel entirely the corresponding contribution of this spatial variable to the line-width.

Examples for neutron diffractometers and triple axis spectrometer are given bellow.
Neutron Diffractometers

2.1. The General Method

In the neutron diffractometry the scan variable is $X_1$, the $X$ vector component along the $Q_0$ direction, where $X=Q-Q_0$ and $Q = k_f - k_i$; $k_f, k_i$ are the wave vectors for the incoming respectively the diffracted neutrons and “0” refers to the corresponding most probable value. The scan variable is given by:

$$X_1 = 2 \cdot \Delta k \cdot \sin \theta_f + k \cdot (\gamma_f - \gamma_i)$$

(1)

where $\gamma_i, \gamma_f$ are the horizontal divergence angles from the most probable trajectories, $\Delta k$ is the wave vector spread, $2\theta_0$ is the scattering angle; the indices $i,f$ refer to the incoming respectively the scattered neutrons. The variables defining the neutron trajectories in phase space are all the spatial relevant coordinates to which has to be added, for the time-of-flight diffractometry, the temporal variables defining its emission respectively detection. All the spatial and temporal variables are measured from the corresponding most probable values. The relation defining the temporal variables is:

$$\frac{h}{2\pi m} (t_f - t_0) = \frac{\Delta L}{k} - \frac{L}{k} \frac{\Delta k}{k}$$

(2)

where $L$ is the neutron flight path from source (from chopper centre for a steady state source) to detector, $h$ is the Plank constant and $m$ is the neutron mass.

For the crystal diffractometry the Bragg constraints have to be considered:

$$\gamma_i - \gamma_f = 2 \cdot \frac{l_m}{R_m} \cdot \text{sign}(\theta_m + \chi_m)$$

$$\frac{\Delta k}{k} = \cot \theta_m \cdot \frac{\gamma_i - \gamma_f}{2}$$

(3)

The relation (2) and (3) defines the $\Delta k$ in (1) for the time-of-flight respectively the crystal diffractometry. To these relations must be added the relations giving the horizontal angular variables expressed through the spatial variables $l_0, l_s, l_d, l_m$ the source, sample, detector and monochromator lengths; $R_m$ is the crystal radius of curvature. The general procedure is to express the scan variable $X_1$ through spatial and, for time-of-flight diffractometry, temporal variables and to cancel the major contributions, i.e. the $l_0, l_s, l_d, l_m$ coefficients.

2.2. The Crystal Diffractometry

2.2.1. One Crystal Monochromator

For this geometry (see Figure 1), following the above-described procedure and cancelling the $l_m$ coefficient, one obtains:

$$R_m = \frac{2a}{2a-1} \cdot \frac{L \cdot \text{sign}(\theta_m + \chi_m)}{\sin(\theta_m - \chi_m)}$$

(4)

where

$$a = \frac{\text{sign} \theta}{\tan \theta_m}$$

Cancelling the “$l_0$” coefficient one obtains:
\[
\tan \chi_s = \cot \theta_s \cdot \left[ 1 - \frac{2}{1 + (2a - 1) \cdot \frac{L_c}{L_i}} \right] \\
\cos(2\theta_s) - (2a - 1) \cdot \frac{L_c}{L_i} \\
\cot \alpha_s = \frac{\sin(2\theta_s)}{2} \\
\alpha_s = \chi_s + \theta_s + \pi / 2
\] (5)

where \(\alpha_s\) as the inclination angle measured from the monochromatic beam direction.

The (5) condition is fulfilled if the sample is rotated during the experimental diffraction pattern raising; for each value of the scattering angle the sample has to be positioned accordingly to the (5) relation. In this case the contribution to the line-width of the sample width is cancelled. But, as in this situation, the sample effective volume where the neutrons are scattered, is different for each value of the scattering angle, the normal Rietveld experimental data processing procedure should be modified accordingly.

The (4) relation is the condition to cancel the monochromator width contribution to the line-width. But as the crystal reflectivity is strongly dependent on the radius of curvature, is not possible to be changed during the diffraction pattern raising; this would lead to a significant variation of the incident beam intensity and no data processing is possible in this situation. It is convenient to keep the monochromator radius of curvature constant, at a value corresponding to that given by (4) for a scattering angle \(2\theta_s\) in the range 90-110 degrees, where the overlapping problems are the most significant and therefore the line-width should be minimum. A value of around 10 m for the monochromator radius of curvature is quite convenient, giving a minimum line-width of 12-15 minutes and not more than 25 minutes for the rest of scattering angle values.

An important problem appears. The sample position given by (5) correspond to a certain value of the scattering angle \(2\theta_s\). The question is: if we keep the same sample position corresponding to a certain scattering angle value, how much is decreased the resolution if we measure the pattern for a certain
angular range, around this value? Can be used a position sensitive detector in the case of Q-space focussing configurations?

To find an answer to this important problem, computations were made, using the DAX program [4], for the configuration existing at INR Pitesti and a sample of 2.5 cm width, 3 cm height and .5 cm thickness, for 5 values of the scattering angle, 30°, 50°, 70°, 90° and 110°, for which the optimum sample orientation angles are 29°, 37°, 90°, 121°, 144° respectively; were computed the line widths for a range of 20° around the above mentioned scattering angle values. The results are given in Figure 2.

![Figure 2](image-url)

The conclusion is quite obvious: the resolution is still very good even for a range of 20° around the scattering angle for which the sample is in the optimum position; if still a better resolution is desired, the detector angular width can be lowered at 15°; that means that the diffraction pattern can be got from 5 -7 steps.

Considering the advantages offered by a Q-space focussing configuration in comparison with the high resolution conventional ones, we have to mention that the neutrons use efficiency is better with a factor of 10^3/5, were 10^3 is roughly the transmission of the Soller collimators system defining the conventional instrument considered; that means that with a lower flux of two order of magnitude can be obtained the same luminosity (the same measure time); for comparable neutrons flux the measure time can be reduced correspondingly.

2.2.2. Two Crystals Monochromator

For this geometry (see Figure 3), following the above-described procedure, tacking account that the Bragg constraints, for the two crystals, are:

\[
\gamma_0 + \gamma_1 = 2 \cdot \text{sign}(\theta_{m1} + \chi_{m1}) \cdot \frac{I_{m1}}{R_{m1}} \\
\gamma_2 + \gamma_1 = 2 \cdot \text{sign}(\theta_{m2} + \chi_{m2}) \cdot \frac{I_{m2}}{R_{m2}}
\]

(6)
\[
\frac{\Delta k_i}{k} = \left( \gamma_0 - \gamma_1 \right) \cdot \frac{\cot \theta_{m1}}{2} \quad \text{(7)}
\]
\[
\frac{\Delta k_2}{k} = \left( \gamma_1 - \gamma_2 \right) \cdot \frac{\cot \theta_{m2}}{2}
\]

to which it has to be added the condition that the neutrons reflected by the first crystal must have a proper wave-length to be reflected by the second crystal, i.e.
\[
\Delta k_i = \Delta k_f \\
\text{or} \\
\gamma_2 = \gamma_1 \cdot \left( 1 - a_m + a_m \cdot \gamma_0 \right)
\]

with
\[
a_m = \frac{\tan \theta_{m2}}{\tan \theta_{m1}}.
\]

Figure 3

One obtains the following focusing conditions, giving the crystals radii of curvature:
\[
\frac{1}{R_i} = \frac{a_m - 1}{a_m \cdot L_i} \cdot \text{sign}(\theta_{m1} + \chi_{m1}) \cdot \sin(\theta_{m1} - \chi_{m1})
\]
\[
\frac{1}{R_2} = -\frac{a_m - 1}{L_2} \cdot \text{sign}(\theta_{m2} + \chi_{m2}) \cdot \sin(\theta_{m2} + \chi_{m2})
\]

By cancelling the “\(l_0\)” and “\(l_s\)” coefficients, one obtains respectively
\[
\frac{L_2}{L_1} = \frac{1 + \frac{a_{m}}{2a}}{1 - a_{m}} \frac{\sin(\theta_m - \chi_m)}{\sin(\theta_m + \chi_m)}
\]
\[
\tan \chi_s = \cot \theta_s \cdot \frac{\xi - 1}{\xi + 1}
\]

with
\[
\xi = \frac{-L_2}{L_2} \cdot \frac{2 \cdot (a + a_{m}) - 1 - 2 \cdot \alpha_1 \cdot (1 - \frac{1}{a_{m}}) \cdot (a + a_{m} - 1)}{2a_{m} - 1 - 2 \cdot \alpha_1 \cdot \left(\frac{1 - a_{m}}{a_{m}}\right)^2 + \alpha_2 \cdot \left(1 + \alpha_1 \cdot \frac{2 - a_{m}}{a_{m}}\right)}
\]
\[
\alpha_1 = \frac{L_0}{L_1} \sin(\theta_m - \chi_m), \quad \alpha_2 = \frac{L_2}{L_1} \sin(\theta_m - \chi_m)
\]

2.3. The Time-of-Flight Diffractometry

2.3.1. Pulsed Source

For this geometry (see Figure 4) following the above-described procedure, tacking account that
\[
L = L_0 + L_1
\]
\[
\Delta L = -l_d \cdot \sin \chi_d - 2 \cdot I_s \cdot \sin \theta_s \cdot \cos \chi_s + l_0 \cdot \sin \chi_0,
\]
by cancelling the \(l_d\), \(l_s\), and \(l_0\) coefficients, one obtains, respectively:
\[
\tan \chi_d = \cot \theta_s \cdot \frac{L}{2 \cdot L_0}
\]
\[
\tan \chi_s = \frac{4 \cdot \tan^2 \theta_s + \frac{1}{L_1} + \frac{1}{L_0}}{\tan \theta_s \cdot \left(\frac{1}{L_0} - \frac{1}{L_1}\right)}
\]
\[
\tan \chi_0 = -\cot \theta_s \cdot \frac{L}{2 \cdot L_0}
\]

2.3.2. Steady State Source

For this geometry, (see Figure 5), following the above-described procedure, tacking account that in this case \(t_0\) is the moment when the neutron reaches the chopper centre, \(L = L_1 + L_2\),
\[ \Delta L = l_d \cdot \sin \chi_d - 2 \cdot l_s \cdot \cos \chi_d \cdot \sin \theta, \] that the constraint introduced by the chopper presence is given by \( t_0 = \gamma_2 / \omega \), cancelling the “l_0”, the “l_s” and “l_d” coefficients, one obtains respectively:

\[
\begin{align*}
\tan \theta_s &= -\frac{\pi m \omega}{h k} \cdot (L_1 + L_2) \\
\tan \chi_d &= -\frac{L \cdot \tan \theta_s}{2 \cdot L_s} \\
\tan \chi_d &= -\frac{L \cdot \tan \theta_s}{2 \cdot L_s}
\end{align*}
\]

(13)

**Three Axis Spectrometers**

The experimental setup is given in Figure 6. As the real focusing effects are possible only when no Soller collimators are present between the spectrometer components this situation is considered below. Let us consider the idealized case of a neutron spectrometer with ideal perfect crystals and sample, monochromator and analyzer of negligible thickness.

![Three Axis Spectrometers Diagram](image)

*Figure 6*

The spread of the scan variable can be written as a linear combinations between X vector components

\[ \Delta = X_d + C_1 \cdot X_1 + C_2 \cdot X_2 + C_3 \cdot X_3 \]

(14)

where X is the vector of components \((Q - Q_0, \omega - \omega_0)) \), \( Q = k_i - k_f \) and \( h \omega = E_i - E_f \); \( k_i, k_f, E_i, E_f \) are the wave vectors respectively the energy for the incoming and the diffracted neutrons, while the "0" index refers to the corresponding most probable values. The \( X_d \) and \( X_s \) components are along respectively normal to \( Q \), direction, all situated in the scattering plane. In the first approximation the \( X_s \) component, normal to the scattering plane, should not be taken into consideration as there is no correlation.
between horizontal and vertical variables. The variables defining the neutron trajectories in phase space are all the spatial relevant coordinates.

The expression giving the X components are:

\[ X_1 = \cos \phi \cdot \Delta k_i + k_{i0} \cdot \sin \phi \cdot \gamma_i - \cos \phi' \cdot \Delta k \cdot \sin \phi \cdot \gamma_f \]  
\[ X_2 = -\sin \phi \cdot \Delta k_i + k_{i0} \cdot \cos \phi \cdot \gamma_i + \sin \phi' \cdot \Delta k \cdot \cos \phi \cdot \gamma_f \]  
\[ X_3 = \frac{h}{m} \cdot (k_{i0} \cdot \Delta k_i - k_{f0} \cdot \Delta k_f) \]

where

\[ \tan \phi = \frac{k_{f0} \cdot \sin 2\theta_f}{k_{i0} - k_{f0} \cdot \cos 2\theta_i} \]  
\[ \phi' = \phi - 2 \cdot \theta_s \] (18)

and \( \gamma_i, \gamma_f \) are the horizontal divergence angles from the most probable trajectories, \( \Delta k_i, \Delta k_f \) are the corresponding wave vector spread, \( 2\theta_s \) is the scattering angle; the index i, f refers to the incoming respectively scattered neutrons; \( \phi \) is the angle between \( Q_0 \) and \( k_{i0} \).

To these relations must be added the Bragg constraints related to the monochromator and analyzer respectively:

\[ \gamma_0 + \gamma_1 = 2 \cdot l_m \cdot \rho_m \]  
\[ \frac{\Delta k_i}{k_{i0}} = \frac{\gamma_0 - \gamma_1}{2} \cdot \cot \theta_m \]  
\[ \gamma_2 + \gamma_3 = 2 \cdot l_a \cdot \rho_a \]  
\[ \frac{\Delta k_f}{k_{f0}} = \frac{\gamma_2 - \gamma_3}{2} \cdot \cot \theta_a \]

where \( \rho_m = 1/R_m, \rho_a = 1/R_a \) and \( R_m, R_a \) are the radius of curvature for the monochromator and analyzer respectively.

The geometry of the experimental configuration gives the following expressions for \( \gamma_0, \gamma_1, \gamma_2, \gamma_3 \):

\[ L_0 \cdot \gamma_0 = -l_0 + l_m \cdot \sin(\theta_m + \chi_m) \]  
\[ L_1 \cdot \gamma_1 = l_i \cdot \cos(\theta_i + \chi_i) + l_m \cdot \sin(\theta_m - \chi_m) \]  
\[ L_2 \cdot \gamma_2 = -l_i \cdot \cos(\theta_i - \chi_i) + l_a \cdot \sin(\theta_a + \chi_a) \]  
\[ L_3 \cdot \gamma_3 = l_a + l_a \cdot \sin(\theta_a - \chi_a) \]

The angles \( \chi_m, \chi_a \), all measured in trigonometric sense, represent the angle between to the lattice plane and the normal to the crystal surface, for monochromator and analyzer respectively.

Eliminating the angular variables \( \gamma \) and \( \gamma_i \) one obtains the following expressions for \( \Delta k_i, \Delta k_f \):

\[ \Delta k_i = \frac{k_{i0} \cdot \cot \theta_m \cdot (l_m \cdot \rho_m - \gamma_i)}{l_i} \]  
\[ \Delta k_f = \frac{k_{f0} \cdot \cot \theta_a \cdot (l_a \cdot \rho_a - \gamma_f)}{l_a} \]

Using (15), (16), (17) in (14) on obtains for the scan variable \( \Delta \):

\[ \Delta = \Delta k_i \cdot A_i - A_f \cdot \Delta k_f + k_{i0} \cdot B_i \cdot \gamma_1 - k_{f0} \cdot B_f \cdot \gamma_2 \]

where:
Using (19) and (20) the relation giving the scan variable $\Delta$ becomes:

$$
\Delta = k_{i0} \cdot \frac{h}{m} + C_1 \cdot \cos \phi - C_2 \cdot \sin \phi
$$

$$
A_i = k_{i0} \cdot \frac{h}{m} + C_1 \cdot \cos \phi - C_2 \cdot \sin \phi
$$

$$
A_f = k_{f0} \cdot \frac{h}{m} + C_1 \cdot \cos \phi' - C_2 \cdot \sin \phi'
$$

$$
B_i = C_1 \cdot \sin \phi + C_2 \cdot \cos \phi
$$

$$
B_f = C_1 \cdot \sin \phi' + C_2 \cdot \cos \phi'
$$

(23)

with:

$$
D_i = B_i - A_i \cdot \cot \theta_m \quad D_f = -(B_f + A_f \cdot \cot \theta_a)
$$

(24)

Cancelling the coefficients of $l_m$, $l_a$ and $l_i$, one obtains:

$$
\frac{L_4}{2 \cdot f_1} = 1 - \frac{B_i}{A_i} \cdot \tan \theta_m \quad \frac{L_2}{2 \cdot f_2} = 1 + \frac{B_f}{A_f} \cdot \tan \theta_a
$$

(25)

$$
\frac{\cos(\theta_a + \chi_a)}{\cos(\theta_a - \chi_a)} = \frac{L_4 \cdot k_{i0}}{L_2 \cdot k_{f0}} \cdot \frac{A_f \cdot \cot \theta_a + B_f}{A_i \cdot \cot \theta_m - B_i}
$$

(26)

The first two conditions can be fulfilled by properly choosing the radius of curvature $R_m$, $R_a$ for monochromator and analyzer respectively when varying $k_i$ and $k_f$, if $L_1$ and $L_2$ are fixed while the third one can be fulfilled by a proper sample orientation. The focusing conditions given by (26) assure the compensation of the contributions given by monochromator and analyzer respectively to the line width while the focusing condition (27) assures the sample contribution compensation. The focusing conditions deduced above for an idealized configuration can serve as a “0”-th order approximation in a numerical optimization process.

**Conclusions**

Very high resolution performances can be obtained through inverse-space focusing in thermal neutrons spectrometry if focusing conditions involving monochromator and analysis radius of curvature and the sample orientation are fulfilled.

For time-of-flight instruments the inverse space focusing conditions given in [5] should be considered.

Inverse space focusing represents a very promising way to get high-resolution instruments in neutron spectrometry. A new concept of high-resolution instrument based on inverse space focusing effects could be developed.

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