Multiple neutron scattering corrections. Some general equations to do fast evaluations.

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Abstract. An efficient Monte Carlo algorithm has been developed aimed at simulating neutron tracing in a liquid or amorphous sample, during a diffraction experiment. At odds with approximated analytical estimates, such a method allows to easily take into account some physical and geometrical features, which play an important role in determining the measured scattered neutron intensity.

Some general laws giving the multiple scattering intensity, in terms of few adimensional parameters, are provided. In particular, it results that the most relevant quantity involved in the neutron transport is the mean free path (defined as the inverse of the attenuation factor), which can be helpfully assumed as natural length unit. This yields to some important consequences: for instance, in the case of absorbing or structured samples, the mean free path can strongly depend on the neutron energy and this should be considered in the choice of the experimental apparatus.

1. Introduction
The diffraction of thermal neutrons is widely used to study the interatomic structure of isotropic systems [1]. As a matter of fact, the general theory predicts that, in case of neutrons scattered at most once, the intensity of detected radiation is directly proportional to the differential cross section as a function of momentum transferred, giving a measure of the atomic ordering within the system. This implies that, in reality, the measured data should be corrected for neutrons scattered more than once, in order to get the static structure factor.

Analytical estimates of multiple (more than one) scattering have been provided since 1954 [2, 3, 4]: all of these are based on the assumption that the angular distribution of primary scattered neutrons is nearly isotropic so that it makes sense to assume that the ratio of next scattering probabilities is constant. More recently, an analytical algorithm to extract single scattering from measured profiles in small angle experiment has been proposed [5]: this is based on the formalism developed for investigating large scale structures [6] and the main issue is the estimation of the scattering mean free path, which depends on the size of scatterers and the density contrast. Here we consider a quite different case: the scatterers are point, and no inhomogeneities are taken into account so that the small angle scattering (SAS) is null and the mean free path depends only on the strenght of interaction between the nuclei of the sample and the probing radiation. While the last feature is a simplification vis-a-vis the SAS, to our best knowledge the need of considering an interatomic structure factor does not lead any inversion method, to get the single scattering from the measured detected beam.
In particular, in the following, we’ll point out that when considering a real experiment, consisting of a finite-size neutron beam, incident on a finite-size specimen, which scatters differently at different angles, the analytical approach carried out for diffraction data analysis may break down. Instead of it, numerical integrations of Boltzmann neutron transport equation, have traditionally been used to tackle real cases. The more general numerical approach is given by Monte Carlo simulations, which have been widely used to include all the features of the experiment (from [7] to [8]). But even nowadays, running simulations to get a rough but reliable real-time correction could be too slow. Here we propose to recover an analytical approach, based on equations fitting a plenty of simulated data. As shown in the following for elastic scattering, some universal scaling laws hold. This is due to the fact that a natural length unit, depending on the energy, can be introduced, so that many scattering signals coming from different samples collapse into few general behaviours.

Even if this research has been assigned to a specific task, i.e. to improve and speed up the analysis on data collected at diffractometers at ISIS [9], its results could be easily generalized, or give suggestions for further developments. Moreover, it’s noteworthy that modern diffractometers cover wider and wider Q-ranges, detecting both large-scale and microscopic structure, so in next future an integrated analytic method shall hopefully be developed.

2. From neutron transport theory to numerical evaluation of scattered neutron intensity

A brief review on the neutron transport theory could be helpful to introduce the implemented algorithm, which is similar, but not identical, to some previous code [10].

Consider a neutron, which enters at \( \vec{r}_0 \) and goes out of the sample at \( \vec{r}_{\text{out}} \). It may have \( n \) collisions in the sample, at positions \( \vec{r}_1, \vec{r}_2, \vec{r}_3, \ldots \vec{r}_n \).

After each collision, the neutron changes flight direction (see Fig.1) so that

\[
\vec{r}_{j} = \vec{r}_{j-1} + \vec{l}_j \quad \text{being} \quad \vec{l}_j = l_j \begin{pmatrix}
\sin \theta_{j-1} \cos \phi_{j-1} \\
\sin \theta_{j-1} \sin \phi_{j-1} \\
\cos \theta_{j-1}
\end{pmatrix}
\]
While \((\theta_j, \phi_j)\) gives the direction after the \(j\)-collision in the lab reference frame, the direction after the \(j\)-collision in the neutron reference frame are \((\theta_j^*, \phi_j^*)\). In particular, the angle formed by the neutron flight directions after and before the \(j\)-collision, \(\theta_j^* = \arccos(\hat{r}_j \cdot \hat{r}_{j-1})\), is called scattering angle.

In the following, we’ll tackle only elastic scattering events, i.e. we’ll suppose that neutron energy doesn’t change after each collision in the sample.

Under this hypothesis, the neutron beam detected at \(\theta_D\), due to \(n\) collisions, can be written as \([4]\):

\[
\begin{align*}
T_n(\theta_D|\lambda) &= \left(\frac{\mu_s}{4\pi}\right)^n \prod_{i=1}^{n-1} \int_0^{l_{\text{max}}} dl_i e^{-\mu_T l_i} \int_0^{4\pi} d\Omega_i \left[ \frac{\mu_{\text{coh}}}{\mu_s} S(Q_i) + \frac{\mu_{\text{inc}}}{\mu_s} \right] I_0(\vec{r}_i, \lambda) \\
&= \frac{\mu_s}{4\pi} \int_0^{l_{\text{max}}} dl_1 e^{-\mu_T (l_1 + \text{out})} \left[ \frac{\mu_{\text{coh}}}{\mu_s} S(Q_1) + \frac{\mu_{\text{inc}}}{\mu_s} \right] I_0(\vec{r}_0, \lambda)
\end{align*}
\]

where \(Q_i = \frac{4\pi}{\lambda^2} \sin \theta_i^*\) is the momentum transferred in the \(i\)-th collision, \(\mu_T, \mu_{\text{coh}}\) and \(\mu_{\text{inc}}\) are the attenuation, coherent and incoherent coefficients (\(\mu_{\text{inc}} + \mu_{\text{coh}} = \mu_s\)), \(l_{\text{max}}\) is the longest distance a neutron can travel along the \(\Omega_{i-1} = (\theta_{i-1}, \phi_{i-1})\) direction, depending on its own on \(\vec{r}_{i-1}\) and \(\Omega_{i-1}\). This integral means that all the possible histories \((l_1, \Omega_1, l_2, \Omega_2, \ldots, l_{n-1}, \Omega_{n-1}, l_n)\), corresponding to \(n\)-th elastic collisions of neutrons having a wavelength \(\lambda\) and going out at a detected angle \(\theta_D\) should be considered. For \(n = 1\), Eq. (1) becomes:

\[
T_1(\theta_D|\lambda) = \frac{\mu_s}{4\pi} \int_0^{l_{\text{max}}} dl_1 e^{-\mu_T (l_1 + \text{out})} \left[ \frac{\mu_{\text{coh}}}{\mu_s} S(Q_1) + \frac{\mu_{\text{inc}}}{\mu_s} \right] I_0(\vec{r}_0, \lambda)
\]

which, in the case of extremely thin sample, reduces to \(T_1(\theta_D|\lambda) = \frac{\mu_{\text{coh}}}{4\pi} S(Q_1) + \frac{\mu_{\text{inc}}}{4\pi} I_0(\vec{r}_0, \lambda)\). Dividing \(T_1(\theta_D|\lambda)\) by the incident flux, \(I_0(\vec{r}_0, \lambda)\), and the sample density, \(\rho\), one gets the differential cross section: \(\frac{d\sigma}{d\Omega}(\theta_D|\lambda) = \frac{\sigma_{\text{coh}}}{4\pi} S(Q_1) + \frac{\sigma_{\text{inc}}}{4\pi}\).

In (1) \(\mu_T\) indicates the total attenuation coefficient; this is given by the product of the sample density, \(\rho\), and the total cross section \(\sigma_T\): \(\mu_T = \rho \cdot \sigma_T\).

The total cross section can be calculated by tables \([11]\) as \(\sigma_T(\lambda) = \sigma_s + A_{\text{abs}} \lambda\) being \(\sigma_s\) the scattering cross section, \(A_{\text{abs}}\) the ratio between the thermal absorption cross section at \(\lambda^* = 1.7979 A\) and \(\lambda^*\), and \(\lambda\) the neutron wavelength. Otherwise, the total cross section can be evaluated from the intensity measured by the transmission monitor, \(T_0\), as: \(\sigma_T(\lambda) = \frac{1}{\text{th}} \ln T_0(\lambda)\), being \(\text{th}\) is the thickness of the sample.

Similarly, \(\mu_s\) can be calculated from tables, being \(\mu_s = \rho \sigma_s\), or from the experimental value of \(\mu_T\): \(\mu_s = \mu_T - \rho \cdot A_{\text{abs}} \lambda\).

When the sample is contained in a container, an “average effective” sample should be simulated, i.e. the total attenuation coefficient of the sample in the container is calculated as a weighted average of the attenuation coefficients of the sample (*) and the container (**):

\[
\mu_T = \frac{\sigma_T^* \rho^* \text{th}^* + \sigma_T^c \rho^c \text{th}^c}{\text{th}^* + \text{th}^c}
\]

and, analogously, the absorption coefficient is given by

\[
\rho A_{\text{abs}} = \frac{A_{\text{abs}}^* \rho^* \text{th}^* + A_{\text{abs}}^c \rho^c \text{th}^c}{\text{th}^* + \text{th}^c}.
\]

The integral (1) can be evaluated by running a Monte Carlo (MC) simulation of the neutron tracing: this consists in following the history of a plenty of neutrons in the sample. For each neutron, for each collision, but the last,

- the free path before collision \(l\)
- the new direction of flight \((\theta, \phi)\)

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3
Given the position of the detector $D$, for the last collision only the free path before it should be chosen.

Figure 2. Given the position of the detector $D$, for the last collision only the free path before it should be chosen. For the last collision, the final flight direction, that pointing the detector, is given, as sketched in Fig. 2; so only the free path, $l_n$, before the last collision should be chosen, according to (1), where the integral runs over the last free path, given the final direction.

The evaluation of the generic term of the product of the sequence in (1)

$$
\int_0^{l_{\text{max}}} dl_j e^{-\mu_T l_j} \int_0^{2\pi} d\phi_j^s \int_0^\pi d\theta_j^s \sin \theta_j^s \left[ \frac{\mu_{\text{coh}}}{\mu_s} S(Q_j) + \frac{\mu_{\text{inc}}}{\mu_s} \right]
$$

(2)

can be done through an importance-sampling [12] of the neutron free path before the $j$-th collision, $l_j$, and the scattering angles $\theta_j^s$, $\phi_j^s$. This consists in forcing successive scattering events to occur within the sample and by calculating the score at each scattering point. The optimization of the numerical evaluation of (2) can be achieved by changing integration variables. Be:

$$
t_1 = \frac{1 - e^{-\mu_T l_j}}{1 - e^{-\mu_T l_{\text{max}}}},
$$

(3)

$$
t_2 = \frac{\phi_j^s}{2\pi},
$$

(4)

$$
t_3 = \frac{1 - \cos \theta_j^s}{2},
$$

(5)

$$
t_4 = P(Q_j) \quad \text{where} \quad P(Q_j) = \frac{I(Q_j)}{I(2k)} \quad \text{and} \quad I(x) = \int_0^x dQ_j S(Q_j)
$$

(6)

In terms of $t_1, t_2, t_3, t_4$, Eq.(2) can be written as

$$
\left[ \frac{1 - e^{-\mu_T l_{\text{max}}}}{\mu_T} \int_0^1 dt_1 \right] \left[ 2\pi \int_0^1 dt_2 \right] \left[ 2\frac{\sigma_{\text{inc}}}{\sigma_s} \int_0^1 dt_3 + 2\frac{\sigma_{\text{coh}}}{\sigma_s} \frac{I(2k)}{k^2} \int_0^1 dt_4 \right]
$$

(7)

This suggests how the corresponding Monte Carlo algorithm works.

(i) Given a random number $t_1$ uniformly distributed in $[0, 1]$, from (3) the free path before the $j$-th collision, $l_j$, is found:

$$
l_j = -\frac{1}{\mu_T} \ln (1 - (1 - e^{-\mu_T l_{\text{max}}})t_1).
$$
Eq. (7) requires to scale the neutron weight:

\[ \text{weight} = \text{weight} \cdot \frac{1 - e^{-\mu_T l_{\text{max}}}}{\mu_T}. \]

(ii) Given a random number \( t_2 \) uniformly distributed in [0, 1], \( \phi_j^s \) can be found from (4):

\[ \phi_j^s = 2\pi t_2. \]

According to Eq. (7) the neutron weight should be scaled:

\[ \text{weight} = \text{weight} \cdot 2\pi. \]

(iii) Two possible scattering events may occur: according to Eq. (5), given a random number \( t_5 \) uniformly distributed in [0, 1],

- if \( t_5 < \frac{\mu_{\text{inc}}}{\mu_s} \), an incoherent elastic scattering is scored, by rescaling the neutron weight as

\[ \text{weight} = \text{weight} \cdot 2 \]

and selecting the scattering angle as

\[ \theta_j^s = \arccos (1 - 2t_3), \]

being \( t_3 \) a random number uniformly distributed in [0, 1].

- Otherwise, a random number \( t_4 \) uniformly distributed in [0, 1] is chosen, a \textit{coherent} elastic scattering is scored, the scattering angle is calculated as

\[ \theta_j^s = P^{-1}(t_4) \]

and the weight is updated as

\[ \text{weight} = \text{weight} \cdot 2 \frac{I(2k)}{k^2}. \]

The maximum scattering order is not decided \textit{a priori} but according to its weight in the total multiple scattering (MS) intensity, given a certain level of accuracy.

To avoid correlations between events, each neutron is used once (i.e. for one scattering order, for one wavelength). A faster algorithm could be implemented to exploit a neutron history for all the scattering orders.

3. \textbf{Natural units and relevant parameters}

The neutron scattering depends on both the physical properties of the sample (the density, \( \rho \), the total and the absorption cross section, \( \sigma_t, \sigma_{\text{abs}} \)) and the geometrical features of the source (the beam size, \( b \), and the incident neutron wavelength, \( \lambda \)), the sample itself (shape and size) and the detectors (size and positions).

The general expression of the neutron beam, having wavelength \( \lambda \), detected at \( \theta_D \), emerging after \( n \) collisions, is given by Eq. (1). It’s worth noting that the integrations on the free paths \( l_i \) can be substituted with the corresponding integration on the adimensional variable \( \xi_i = \mu_l l_i \) so that

\[ T_{n}(\theta_D|\lambda) = \left( \frac{\mu_s}{\mu_{\text{tot}}} \right)^n \prod_{i=1}^{n-1} \int_0^{\xi_{\text{max}}} d\xi_i e^{-\xi_i} \int_0^{4\pi} d\Omega_i^s \int_{0}^{\xi_{\text{max}}} d\xi_n e^{-\xi_n} \int_{0}^{4\pi} d\Omega_n^s \left[ \frac{\mu_{\text{coh}}}{\mu_s} S(Q_n) + \frac{\mu_{\text{inc}}}{\mu_s} \right] I_0(\overrightarrow{r_0}, \lambda). \]
This means that, at given \( \lambda \), the absolute linear dimensions are not relevant but their products with the total attenuation \( \mu_t = \mu_t(\lambda) \). In other words, at given wavelength \( \lambda \), the mean free path \( mfp \) defined as
\[
mfp(\lambda) = \frac{1}{\rho \sigma_t(\lambda)},
\]
can be assumed as “natural” length unit. As a matter of fact, the distinction between thin and thick samples contained in a slab can be given \([4, 13]\) using the adimensional parameter
\[
\tau = \frac{\text{thickness}}{mfp},
\]
so that a sample is said thin if \( \tau \ll 1 \), thick is \( \tau \geq 1 \). Note that, in general, the mean free path is a function of \( \lambda \), as the fastest neutrons have the longest typical path. So, in principle, a sample could be thin for high-energy neutrons and thick for low-energy ones.

Generalizing the normalization done for the thickness, all the linear geometrical features can be usefully expressed in \( mfp \) units. Moreover, to take into account the finite size of the beam we introduce the adimensional beam size
\[
\beta = \frac{\text{beam size}}{mfp}.
\]
Regarding the geometry of the detecting apparatus, the detector size is usually negligible in comparison with the sample-detector distance and, if the sample size is small with respect to the detecting apparatus, the detector position can be taken into account considering only the detecting angle, \( \theta_D \), defined with respect to the incident beam direction.

Another adimensional parameter to take into account is the percentage of absorption
\[
\alpha = \frac{\sigma_{abs}}{\sigma_t}.
\]
To evaluate the single and multiple scattering, it’s important to recall that these intensities can be easily calculated if we consider a sample for which
- the structure factor is flat: \( S(Q) = 1 \);
- the transversal size is infinite;
- incident neutron beam is extremely collimated \( (\beta = 0) \);
- absorption is negligible \( (\alpha = 0) \).

From now on, this case will be assumed as reference, for both the slab and the cylinder geometry. Some formulas will be provided to get the multiple scattering intensity from the single scattering intensity. In the following, we’ll see how to correct the multiple scattering calculated for the reference case, to get a prediction for the general case: by processing a plenty of data coming from Monte Carlo simulations, some general laws, reported in the next pages, have been found.

### 4. Slab geometry

In case of sample contained in a slab, having a square as transversal face, the linear dimensions are the thickness and the width, which are, respectively, the size along the direction of the incident beam and the size along the transversal directions. In \( mfp \) units, these correspond to the adimensional parameters:
\[
\tau = \frac{\text{thickness}}{mfp}, \quad \omega = \frac{\text{width}}{mfp}.
\]

The meaning of adimensional parameters \( \alpha, \beta, \omega, \tau, \theta_D \) is sketched in Fig.3.

For the real cases I’ve taken into account (vanadium, empty slab container of silica or titanium-zirconium, water in a slab, measured on ISIS diffractomet \([9]\) ), the variability ranges of the relevant parameters are reported in Tab.1.
4.1. Angular and thickness dependence for the reference case

In the reference case we can analytically calculate the intensity of neutrons scattered once, i.e.

\[ T_1(\theta_D|\lambda) = (1 - \alpha) \frac{e^{-\tau \sec \theta_D} - e^{-\tau}}{4\pi (1 - \sec \theta_D)} I_0(\lambda) \]

where \( \theta_D \) is the scattering angle, \( \mu_t = \rho \sigma_t \) the total attenuation coefficient, \( \mu_s = \mu_t - A_{abs} \lambda \), \( th \) is the thickness, \( l_{out} = (th - z) \sec(\theta_D) \) the distance to exit from \( z \) at \( \theta_D \) angle, \( I_0 \) the incident flux (put \( I_0 = 1 \)). This can be used to test to goodness of the Monte Carlo simulations (Fig.4).

For the smallest detecting angles, the single intensity is the highest as the attenuation is the lowest. Increasing \( \theta_D \), the single intensity decreases. Moreover, by increasing \( \tau \), the angular dependence gets stronger, as the range between the shortest and the longest path through the sample is enhanced. Note that while \( T_1 \) increases for \( \tau \leq 1 \) and decreases for \( \tau > 1 \) (Fig.5), the beam of multiple scattered neutrons, \( T_m \), defined as:

\[ T_m = \sum_{i=2}^{n} T_i \] (for \( n \) as large as required by the accuracy level)

increases for all \( \tau \) and has a weaker angular dependence for high \( \tau \) values, as the incident direction is not relevant for high order scattering events (Fig.6).

Instead of considering \( T_1 \) and \( T_m \) separately, look at the ratio \( \frac{T_m}{T_1} \) (Fig.7). As a function of \( \theta \), this value is almost constant for low \( \tau \) values, while it’s slightly increasing with \( \theta_D \) for high \( \tau \) values. Incidentally, in SAS experiment, the functional dependence of the scattered intensity in the Porod region of momentum transfer has been found to remain invariant under multiple
scattering [13], that is to say that the scattering profiles of two samples of the same substance but different thickness differ from one another only for a scale factor.

We propose to fit $T_m/T_1$ by a linear function of $\theta_D$: $T_m/T_1 \simeq C_\tau + B_\tau \theta$.

By fitting a plenty of data in the examined range (see Fig.7), we get

$$C_\tau = 1.54 \tau + 0.137 \tau^3 \quad B_\tau = 0.0 \cdot 10^{-3} \tau + 1.1 \cdot 10^{-3} \tau^3,$$

as showed in Fig.8. In conclusion, the $\theta$-dependence is very weak, with respect to the $\tau$-dependence, so the multiple scattering can be estimated by:

$$T_m^{ref}(\lambda) \simeq (1.54 \tau(\lambda) + 0.137 \tau(\lambda)^3) T_1^{ref}(\lambda) \quad (8)$$

This result is consistent with previous analytic estimates by Sears and Vineyard (see Fig.9).

Furthermore, the expansion of $T_m$ in odd powers of $\tau$ can be justified by considering that the most probable scattering angle is $\pi/2$; this means that at small $\theta_D$ are detected neutrons which have had two or four collisions at about $\pi/2$ in the sample. As $T_m/T_1 \sim \tau^{n-1}$, the formula (8) is a reasonable approximation.
4.2. Dependence on the finite size of the sample

4.2.1. Squat samples  Let us consider a finite slab containing a not absorbing sample, wet by an extremely collimated neutron beam ($\alpha = 0$, $\beta = 0$). In such a case, the ratio $\frac{T_m}{T_1}$ is a function of $\tau$, $\theta_D$, $\omega$. In particular, the single is not sensitive on the finite size if $\theta_D$ is so small that $tg\theta_D < \frac{\omega}{2\tau}$. On the contrary, the angular dependence is relevant for thick samples with a small width. In Figg.10,11,12, the single, the multiple scattering intensities and their ratio are reported for $\tau = 2.4$. Note that in the cases under study (Tab.1), $\frac{\omega}{2\tau} = \frac{\text{width}}{\text{thickness}} > 5$ so the angular dependence of the single scattering intensity for a finite slab does not depend on the width. In any case, it results that the angular dependence of the ratio $\frac{T_m}{T_1}$ is less important than the dependence on the width, so it can be neglected, also in this case, at least in a first approximation:

$$\frac{T_m}{T_1} \simeq C(\tau, \omega).$$

As usual, we’ll refer to the ratio between multiple and single scattering $C = \frac{T_m}{T_1}$, as it’s more flat vs $\theta_D$ than both the single intensity and the multiple intensity.
Note that if $\theta_{D,max} < tg \frac{\omega}{\tau}$ (as usually happens), the single scattering is not $\omega$-dependent, i.e. $T_1(\omega, \tau) = T_1(\infty, \tau)$ so $\frac{T_m(\tau, \omega)}{T_m(\tau, \infty)} = \frac{C(\tau, \omega)}{C(\tau, \infty)}$. A description of the $\tau$ and $\omega$ behaviour of $T_m$ can be achieved by considering the quantity:

$$1 - \frac{C(\tau, \omega)}{C(\tau, \infty)} = f(\tau; \omega),$$

where $f$ is a decreasing function going from 1 (for $\omega = 0$) to 0 (for $\omega = \infty$). A good fit of a plenty of numerical data produced by the Monte Carlo simulation can be achieved using a stretched exponential function in $\frac{\omega}{\tau}$ (Fig.13). In particular,

$$1 - \frac{C(\tau, \omega)}{C(\tau, \infty)} \simeq \exp \left\{ -A_\tau \left( \frac{\omega}{\tau} \right)^{d_\tau} \right\}$$

(9)

where $A_\tau$ and $d_\tau$ are power laws in $\tau$. Fitting $d_\tau$ and $A_\tau$ in the current range, it results:

$$d_\tau = 1.0\tau^{0.19} \quad A_\tau = 0.52\tau^{0.15}.$$  

It’s not surprising that the main parameter to be considered in the case of a finite slab should be the ratio between the lateral and the longitudinal sizes $\frac{\omega}{\tau} = \frac{\text{width}}{\text{thickness}}$; so that, in first approximation, a slab can be considered infinite if $\text{width} \gg \text{thickness}$. Summarizing the helpful formula is

$$T_m(\tau, \omega) = \left[ 1 - \exp \left( -0.52\tau^{0.15} \left( \frac{\text{width}}{\text{thickness}} \right)^{0.19} \right) \right] T^\text{ref}_m(\tau)$$

(10)

4.3. Absorption dependence

Come back to an infinite slab ($\omega = \infty$). So far we have considered a not absorbing sample. Be $\alpha = \sigma_{\text{abs}} / \sigma_{\text{t}}$ (between 0 and 1) the absorption parameter.

In the case of $\beta = 0$, the ratio $\frac{T_m}{T_1}$ can be compared with the corresponding not-absorbing case, and approximated with a polynomial in $\alpha$ with degree 4 (Fig.14). It results:

$$1 - \frac{T_m(\alpha)}{T_1(\alpha = 0)} = A_\tau \alpha + B_\tau \alpha^2 + C_\tau \alpha^3 + D_\tau \alpha^4$$

(11)
The coefficients of the polynomial series as function of $\tau$ are shown in Fig.27.

In conclusion, the absorption can be taken into account by using:

$$T_m(\alpha) = T_{m}^{\text{ref}} \cdot \left[1 - \alpha (1 + 1.5 \cdot \tau) + 2.6  \alpha^2 + O(\tau^2, \alpha^3)\right]$$  \hspace{1cm} (12)

4.4. Dependence on the beam size

Further calculations involve a real beam, i.e. having a finite size. Introducing

$$\gamma = \frac{\beta}{\omega} = \frac{\text{beam size}}{\text{width}}$$

we can estimate how important is the effect of the finite beam size. Put:

$$D(\gamma, \omega, \tau, \theta) = \frac{C(\gamma, \omega, \tau, \theta)}{C(0, \omega, \tau, \theta)}$$

being $C(\gamma, \omega, \tau, \theta) = \frac{T_m(\gamma, \omega, \tau, \theta)}{T_{m}^{\text{ref}}}$). $D(\gamma, \omega, \tau, \theta)$ is almost independent on $\theta$, so, from now on, we assume $D(\gamma, \omega, \tau, \theta) = D(\gamma, \omega, \tau)$.

The quantity $1 - D(\gamma, \omega, \tau)$ may be fitted with an even polynomial in $\gamma$:

$$1 - D(\gamma, \omega, \tau) = A_{\omega,\tau} \gamma^2 + 0.05 \gamma^4.$$  \hspace{1cm} (13)

In Fig.16, the results for $\tau = 2$ are shown. From these, it results that

- $D(\gamma, \omega, \tau)$ is always decreasing with $\gamma$, meaning that a less collimated and wider beam makes the ratio between multiple and single scattering lower;
- at $\gamma$ fixed, $D(\gamma, \omega, \tau)$ is not monotonic with $\omega$: it decreases for $\omega < \tau$ while it increases for $\omega > \tau$.
- at $\gamma$ and $\omega$ fixed, $D(\gamma, \omega, \tau)$ variation is enhanced for $\tau$ increasing.

The coefficient $A_{\omega,\tau}$ can be fitted as a function of $\omega$, which is 0 for $\omega = 0$ and $\omega \rightarrow \infty$:

$$A_{\omega,\tau}(\omega) = A_{\tau}\omega e^{-C_{\tau}\omega\tau}.$$  \hspace{1cm} (14)

The fits are reported in Fig.17.
5. Cylinder geometry

Consider now a sample contained in a cylinder, the axis of which is transversal to the incident beam. In this case we introduce the adimensional diameter given by

\[
2\rho = \frac{\text{diameter}}{mfp},
\]

and the adimensional height, defined by:

\[
\eta = \frac{\text{height}}{mfp}.
\]

In this geometry, the reference case is obtained for \( \eta = \infty, \beta = 0, \alpha = 0 \).
5.1. Some results on the reference case

Many remarks done for the slab are still valid for cylinder: while $T_1$ and $T_m$ may depend on the detected angle $\theta_D$, the ratio $\frac{T_m}{T_1}$ is much less dependent on $\theta_D$ (Fig.20). So $\frac{T_m}{T_1}$ may be well fitted with a function of $\rho$, as shown in Fig.21.

5.1.1. Comparison with the reference slab

Note that the intensities of scattered neutrons should be quite different in the two geometries as, by definition, the transversal sizes are equal in case of a square slab while are different in the case of a cylinder: one is the diameter, the other one is the height (see Fig.19). In other words, by comparing the two reference cases, an infinite slab is a $\infty^2$ object while an infinite cylinder is $\infty^1$ object.
It follows that the ratio $\frac{T_m}{T_1}$ is lower for an infinite cylinder of radius $R$ than for a slab having thickness equal to $2R$ (Fig. 21).

5.2. Dependence on the finite size of the sample
Let us consider cylinders which have finite height, say $\eta = 0.1, 1, 10$. While the single and multiple scattering intensities are dependent on the detected angle (Fig. 22), it results that the ratios $\delta = \frac{T_2}{T_1}$ and $\Delta = \frac{T_m}{T_1}$ are almost independent on the position of the detectors (Fig. 23).

![Figure 22. Angular dependence of single and multiple intensity for finite cylindrical samples.](image)

![Figure 23. Angular dependence of the ratios $\delta = \frac{T_2}{T_1}$ and $\Delta = \frac{T_m}{T_1}$ for finite cylindrical samples. The symbols are the same of Fig. 22.](image)

Moreover, $\Delta$ can be fitted as an odd polynomial function of $\delta$, being the coefficients almost, but not exactly, independent on $\eta$, as shown in Fig. 24. Here we propose an alternative approach which follows just what we’ve done for specimens contained in slabs.

So, introducing

$$C(\rho, \eta) = \frac{T_m(\rho, \eta)}{T_1(\rho, \eta)},$$
and processing many numerical data, a fit can be performed looking at

\[ 1 - \frac{C(\rho, \eta)}{C(\rho, \infty)} = f(\rho, \eta), \]

where \( f \) is a decreasing function going from 1 (for \( \eta = 0 \)) to 0 (for \( \eta = \infty \)). A good fit can be achieved using a stretched exponential function of \( \frac{\eta}{\rho} \) (Fig.25). In particular,

\[ 1 - \frac{C(\rho, \eta)}{C(\rho, \infty)} \simeq \exp \left\{ -A_\rho \left( \frac{\eta}{\rho} \right)^{d_\rho} \right\} \tag{15} \]

where \( A_\rho \) and \( d_\rho \) are power laws in \( \rho \). Fitting \( d_\rho \) and \( A_\rho \) in the current range, it results:

\[ d_\rho = 0.99\rho^{0.008} \quad A_\rho = -0.079\rho^2 + 0.85. \]

### 5.3. Absorption dependence

Come back to an infinite cylinder (\( \eta = \infty \)). So far we have considered a not absorbing sample.

Be \( \alpha = \frac{\sigma_{\text{abs}}}{\sigma_t} \) (between 0 and 1) the absorption parameter.

In the case of \( \beta = 0 \), the ratio \( \frac{T_m}{T_1} \) can be compared with the corresponding not-absorbing case, and approximated with a polynomial in \( \alpha \) with degree 4 (Fig.26).

It results:

\[ 1 - \frac{T_m(\alpha)}{T_1(\alpha = 0)} = A_\rho \alpha + B_\rho \alpha^2 + C_\rho \alpha^3 + D_\rho \alpha^4 \tag{16} \]

The coefficients of the polynomial series, as functions of \( \rho \), are shown in Fig.27.

In conclusion, the absorption can be taken into account by using:

\[ T_m(\alpha) = T_{m\text{ref}} \cdot \left[ 1 - \alpha(1 + 1.5 \cdot \rho) - 1.5\rho\alpha^2 + O(\rho^2, \alpha^3) \right] \tag{17} \]

Note that Eq.(17) is very similar to Eq.(12); this implies that if the sample is not very absorbent (\( \alpha \ll 1 \)) and quite thin (\( \tau \ll 1 \) or \( 2\rho \ll 1 \)) the absorption is almost independent on the shape of the sample.
5.4. Comparison with previous analytical results
In early studies [3, 4] approximated results were found for the cylinder geometry. In particular, \( \delta = \frac{T_2}{T_1} \) was tabulated as a function of \( \rho \) and \( \eta \). The present estimates give quite similar results, showing some limits of the previous.

First of all when a squat cylinder is considered (\( \rho \sim \eta \)) the angular dependence is not negligible (see Fig.28).

Secondly, approximating with a constant mean value, I get the values shown in Fig.29, which are slightly different from those obtained from Sears. Here, the analytical estimates for rods and discs are shown and the result tabulated by Sears[4] for the same case is shown for comparison.

I’ve also done a comparison with the results given by Blech-Averbach [3] for rod having \( \rho = 0.1, 0.2, 0.3, 0.4 \) and \( \eta = 10 \cdot \rho \) (Fig.30).

6. How to use the fitting curves to predict the multiple scattering
In the following I’ll show an example illustrating how to calculate quickly the multiple scattering corrections: this is the case of a typical silica slab, as measured on NIMROD diffractometer at ISIS-TS2 [9].

The input data needed for evaluation, as reported in the previous pages, are:

(i) the sample density, \( \rho = 0.0663 \frac{\text{atoms}}{\text{Å}^3} \).
(ii) the thickness, 0.4 cm;
(iii) the width, 4 cm;
(iv) the data collected by the transmission monitor, giving $\sigma_t(\lambda)$;
(v) the absorption $\sigma_{abs}(1.7979\text{Å}) = 0.057$ barn;
(vi) the beam size, 3 cm.

The relevant parameters, $\tau$, $\omega$, $\alpha$ and $\beta$, as functions of $\lambda$, are shown in Fig.31. The not-trivial $\lambda$-behaviour is due to the fact that $\sigma_t(\lambda)$, and so the mean free path, are not flat. The sample can be considered thin for almost all wavelengths and the absorption is at most 10% of the scattering power. The adimensional width $\omega$ is not greater than 1 (which should be for being the infinite slab approximation valid) so a large correction is due to the finite size of the slab. Also the beam size is not negligible in comparison with the sample width ($\beta \simeq 0.75$).

The evaluation of the multiple scattering can be done according to the following steps, performed for each $\lambda$ value, at $\theta_D = 0$.

(i) Calculation of the single and multiple scattering intensities for the reference case (according to Eq.(8)):

$$\tau(\lambda) = \rho \cdot \text{thickness} \cdot \sigma_t(\lambda)$$

$$T^\text{ref}_1(\lambda) = \frac{\tau e^{-\tau}}{4\pi}$$

$$T^\text{ref}_m(\lambda) = 1.52\tau T^\text{ref}_1(\lambda).$$
(ii) Correction for the absorption (according to Eq.(12)): 

$$\sigma_{\text{abs}}(\lambda) = \frac{\sigma_{\text{abs}}(1.7979\text{Å})}{1.7979} \lambda \quad \alpha(\lambda) = \frac{\sigma_{\text{abs}}(\lambda)}{\sigma_{t}(\lambda)}$$

$$f(\alpha) = \alpha(1 + 1.5 \cdot \tau) - 2.6\tau\alpha^2 + \ldots \quad T_{m}^{\alpha}(\lambda) = (1 - f(\alpha))T_{m}^{e\gamma}(\lambda)$$

(iii) Correction for the finite width (according to Eq.10):

$$T_{m}^{\omega,\alpha}(\lambda) = \left[ 1 - \exp\left( -0.52\tau^{0.15} (10)^{1.0^{0.19}} \right) \right] T_{m}^{\alpha}(\lambda).$$

(iv) Correction for the finite size of the beam (according to Eq.13 e 14):

$$T_{m}^{\omega,\alpha}(\lambda) = T_{m}^{\omega,\alpha}(\lambda) \left[ 1 - \left( 10e^{-0.8\tau\omega} \cdot e^{-5e^{-0.5\omega} - e^{-0.4} - 0.3} \right) \gamma^2 + 0.05\gamma^4 \right]$$

The results of these next steps are reported in Fig.33.

Figure 32. Single and multiple scattering intensities calculated by the equations here proposed at $\theta_D = 0.$

Figure 33. The effect of the next corrections on the multiple scattering intensity at $\theta_D = 0.$

7. Conclusions

A method to get an approximated analytical estimate of multiple scattering intensity of thermal neutron beam in a diffraction experiment has been presented. Even if the formulas provided are valid only for the specimens and instruments here studied, namely in the variability range given in Tab.1, the general approach could be followed aimed at implementing a fast real-time data analysis. It’s worth noting that each geometrical feature can be expressed in $mfp$ (standing for mean free path) units, so that a sample is said thin or thick, short or long, according to its physical features, in particular depending on its total cross section. Given an experimental setup and a scientific case (source, detectors, sample containers and specimens of interest), Monte Carlo simulations can be run in order to get fitting curves dependent on the adimensional parameters. As a matter of fact, these scaling laws can provide a quick and reliable evaluation of the multiple scattering correction, at least when the sample is quite disordered.

Interestingly, an analytical inversion algorithm for SAS experiment has been found elsewhere, by Mazumder et al [5]: this shall be considered in next future when further developments will be required to get an unique method for analysing data, collected on wide $Q$ range diffractometers.
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