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Strategies for removing multiple scattering effects revisited

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ABSTRACT

The multiple scattering problem of small angle neutron scattering (SANS) is revisited using differential equations from which different contributions are derived. The coherent scattering is connected to multiple scattering events and the ideal single scattering cross sections can be related to the apparent scattering cross section. The multiple scattering problem of the incoherent scattering is more demanding because the sample geometry – I assumed a slab – matters. I tried to solve this problem analytically, and used Monte Carlo simulations. From all concepts I derived a strategy for a computer program that is capable to remove multiple scattering effects. As a side aspect, I could also remove resolution effects.

In this article the multiple scattering problem for small angle scattering is revisited. In early times the methods were developed by Schelten and Schmatz [1], and a program for simulating or removing multiple scattering was proposed by Monkenbusch [2]. The proposed formalism is capable to work on anisotropic scattering patterns. The important prerequisite is that the scattering appears at small angles where the Ewald sphere is flat, and the sample thickness does not vary as a function of the scattering angle. These methods basically apply for the coherent scattering portion while the incoherent scattering needs to be considered separately. For neutrons the incoherent scattering emerges from point scatterers, which facilitates the handling. Whatever radiation is used in the experiment, for my purpose, I assume that the incoherent scattering is rather flat in the experimental q-range, which means that the underlying scattering centers are small compared to $q_{\max}^{-1}$, i.e. the reciprocal largest scattering vector $q_{\max}$. Apart from that, the sample thickness usually varies as a function of the scattering angle. For the slab geometry, basically no signal is detectable in the lateral direction. Early studies from Chandrasekhar [3] focused on the multiple scattering problem quite generally, but the proposed mathematical algorithms do not converge very well. So, even for simple incoherent scattering, the math stays quite difficult.

Our approach uses differential equations for describing the multiple scattering problem. I assume that the different scattering events take place at well-separated points inside the sample, i.e. that the neutron leaves the coherence volume before it is scattered a second time. Coherent multiple scattering would lead to more complicated effects that are known as Mie scattering for light and are also discussed for ultra small angle X-ray scattering [4]. Differential equations make the transfer of neutrons between different channels very clear and the incoherent scattering problem can be implemented easily (see Fig. 1). I distinguish within the channels the primary intensity I_0, the coherently scattered intensity i_1 (as a function of the scattering vector), the incoherent scattered intensity in the forward direction j_+ (as a function of scattering angle), and the incoherent scattered intensity in the backward direction j_-. From all the channels there are outgoing probabilities (proportional to the corresponding scattering cross section), and possibly incoming probabilities. I assumed that a once incoherently scattered neutron does not experience considerably large changes by coherent scattering. The primary intensity, and the coherent scattering can be derived analytically. For the incoherent scattering I tried an analytic approach that makes it difficult to implement boundary conditions. Therefore, I applied Monte Carlo computer simulations to describe the overall multiple scattering of a sample. I chose a microemulsion that scatters strongly enough to generate multiple scattering. From the example I learned which properties of the scattering curves are essential. Then, a general strategy for removing multiple scattering is proposed.

1. Analytical approach for the slab geometry

When describing multiple scattering phenomena analytically, the best way is using differential equations for characterizing the transfer of intensity between different channels. I assumed the slab geometry for the sample with a thickness d. All channels are a function of the normal position x and of the scattering angle ϑ, which also connects to the scattering vector $\mathbf{q}$ with the modulus $q = |\mathbf{q}| = 4\pi \sin(\vartheta/2)/\lambda$. The wavelength of the neutron is λ. Probabilities for the transfer between different channels are described by a cross section and the intensity, from which the contribution emerges. I know the following cross sections for coherent scattering Σ_c, for incoherent scattering Σ_i and for absorption Σ_a (and abbreviate $\Sigma_{ia} = \Sigma_i + \Sigma_a$ and $\Sigma_t = \Sigma_{ia} + \Sigma_c$).

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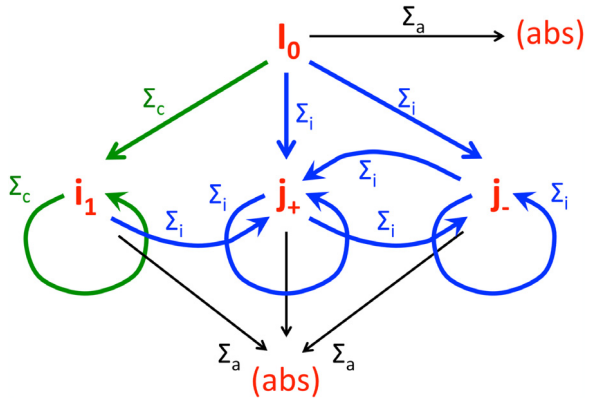

Fig. 1. Explanation for the different channels that neutrons can take from different scattering events. All intensity emerges from the primary intensity I_0, and can end as coherent (i_1) or incoherent scattering in the forward (j_+) or backward (j_-) direction. The scattering probabilities are proportional to the scattering cross sections Σ_x, with x being 'c' for coherent, 'i' for incoherent and 'a' for absorption.

All contributions are summed up, partially from different emerging channels and/or directions, and describe the change of the channel intensity with the position x. In this way, I obtain the following integro differential equations:

$$\partial_x I_0(x) = -\Sigma_t I_0(x) \tag{1}$$

$$\partial_x i_1(x,\mathbf{q}) = \frac{d\Sigma_c}{d\Omega}(\mathbf{q}) I_0(x) + \int \frac{d\Sigma_c}{d\Omega}(\mathbf{q}'-\mathbf{q}) i_1(x,\mathbf{q}') d^2\Omega' - \Sigma_t i_1(x,\mathbf{q}) \tag{2}$$

$$\partial_x j_+(x,\vartheta) = \frac{\Sigma_i}{4\pi}\left(I_0(x) + I_1(x) + \int \frac{j_+ + j_-}{\cos\vartheta} d^2\Omega\right) - \Sigma_{ia}\frac{j_+(x,\vartheta)}{\cos\vartheta} \tag{3}$$

$$-\partial_x j_-(x,\vartheta) = \frac{\Sigma_i}{4\pi}\left(I_0(x) + I_1(x) + \int \frac{j_+ + j_-}{\cos\vartheta} d^2\Omega\right) - \Sigma_{ia}\frac{j_-(x,\vartheta)}{\cos\vartheta} \tag{4}$$

Mathematically, I can distinguish between more channels than experimentally possible (see Fig. 1). The primary intensity I_0 is interpreted as the source beam that only gets weaker inside the sample from scattering processes. The decay of the primary intensity along the normal direction of the slab is described by the differential operator ∂_x, while the probability of the decay is proportional to the total scattering cross section Σ_t and the primary intensity itself. The primary beam takes the full primary intensity $\hat{I}$ at the entrance as one boundary condition. The coherent scattering channels i_1 collect intensity from the primary beam and the other coherent channels due to multiple scattering, and lose intensity for the same reason. The cross-talk (i.e. coherent multiple scattering) between the different scattering channels $i_1(x,\mathbf{q})$ emerges from all coherent scattering channels $i_1(x,\mathbf{q}')$ with the cross talk probability $d\Sigma_c/d\Omega(\mathbf{q}'-\mathbf{q})$. And the range of $\mathbf{q}'$ is connected to the angular range Ω'. On this level, I idealize the 2-dimensional $\mathbf{q}$-plane as flat in contrast to the real Ewald sphere. This means, that I stay in the SANS regime for the coherent single scattering and the multiple scattering. The two incoherent functions $j_\pm$ cover the full angular space in the forward and back direction. So the angle ϑ only covers the half-space (0 to $\pi/2$). The sign of Eq. (4) takes care of the correct orientation of the solid angle. I separated the two half spaces from each other for reasons of the boundary conditions: The back-scattering intensity is zero at the exit of the sample $x = d$, while the forward-scattering takes zero at the entrance $x = 0$ (so does i_1). The effectively longer paths differing from the x-directions are taken into account by the cosine terms (for the incoherent scattering only). One has to keep in mind that, later in the experiment, the coherent scattering and the incoherent scattering in the forward direction superimpose and cannot be distinguished as a bare intensity anymore.

I can define the overall coherent scattering intensity by a similar integration $I_1(x) = \int i_1(x,\mathbf{q}) d^2\Omega = \frac{\lambda^2}{(2\pi)^2}\int i_1(x,\mathbf{q}) d^2\mathbf{q}$. The according differential equation (see Eq. (2)) would read then $\partial_x I_1 = \Sigma_c I_0 - \Sigma_t I_1 + \Sigma_c I_1$. This indicates that the multiple scattering processes do not affect the behavior of the integral coherent intensity. The integral coherent scattering intensity collects intensity from the primary beam and loses intensity to the incoherent channels. Eqs. (3)–(4) treat redistribution effects due to the incoherent scattering only. This means that after a first incoherent scattering process a second coherent scattering process will not show a huge effect. If the main coherent scattering is concentrated at small angles this approximation is quite good and so these possible corrections can be neglected. A wide-angle scattering law introduces atomic and/or molecular structures that are usually observed at large angles and beyond the Ewald sphere. I assume that these contributions are small compared to the incoherent signal, and do especially not show significant multiple scattering. Usually, the content of hydrogenous materials needs to be reasonably high to cause multiple scattering and then the atomistic structures scatter weakly [5].

The different scattering contributions can be solved sequentially. For the primary intensity I obtain $I_0 = \hat{I}\exp(-\Sigma_t x)$. The total coherent scattering is described by $I_1 = \hat{I}(-\exp(-\Sigma_t x) + \exp(-\Sigma_{ia} x))$. It has a maximum at $x = \ln(\Sigma_t/\Sigma_{ia})/\Sigma_c$ and the ideal sample thickness d is chosen accordingly for maximum information of the scattering experiment.

The multiple coherent scattering solution is obtained by using the Fourier transformation. Any function in $\mathbf{q}$-space will be transformed to reciprocal $\mathbf{r}$-space via $\tilde{a}(\mathbf{r}) = \frac{1}{2\pi}\int a(\mathbf{q})\exp(\mathrm{i}\mathbf{q}\mathbf{r}) d^2\mathbf{q}$. The back-transformation is done by the expression $a(\mathbf{q}) = \frac{1}{2\pi}\int \tilde{a}(\mathbf{r})\exp(-\mathrm{i}\mathbf{q}\mathbf{r}) d^2\mathbf{r}$. At this point, the formalism describes anisotropic scattering completely right. For isotropic scattering, the Fourier transformation can be carried out in one dimension according to $\tilde{a}(r) = \int_0^\infty a(q) q J_0(qr) dq$ and for the back-transformation according to $a(q) = \int_0^\infty \tilde{a}(r) r J_0(qr) dr$, both known as Hankel transformation of zeroth order. At any point, the reader may go back to anisotropic scattering by dealing with the full vectorial dependence on $\mathbf{r}$. The already well-known [1,2] analytical solution reads then:

$$\tilde{i}_1(d,r) = \hat{I}\frac{2\pi}{\lambda^2}\left(\exp\left(\frac{\lambda^2}{2\pi}\frac{\widetilde{d\Sigma_c}}{d\Omega}(r) d\right) - 1\right)\exp(-\Sigma_t d) \tag{5}$$

This solution also includes the integral intensity solution I_1 according to $\Sigma_c = \frac{\lambda^2}{2\pi}\frac{\widetilde{d\Sigma_c}}{d\Omega}(0)$ and $I_1 = \frac{\lambda^2}{2\pi}\tilde{i}_1(d,0)$. Note, that for small scattering signals the single scattering solution is obtained asymptotically ($\exp(\epsilon) - 1 \approx \epsilon$). The difficulty of this equation is the separation of the incoherent and coherent scattering (say in terms of Σ_c and Σ_i or i_1 and j_+) that will be observed as a sum on the detector. The manuscript will deal with some considerations how to separate these contributíons best. The transmission $T = \exp(-\Sigma_t d)$ only measures the total scattering probability Σ_t. The whole Eq. (5) can be solved for the desired macroscopic cross section Σ_c according to:

$$\frac{\widetilde{d\Sigma_c}}{d\Omega}(r) = d^{-1}\cdot\frac{2\pi}{\lambda^2}\cdot\ln\left(\frac{\lambda^2}{2\pi}\frac{\tilde{i}_1(d,r)}{\hat{I}Td} d + 1\right) \tag{6}$$

Again for single scattering processes, the simplification $\ln(\epsilon+1) \approx \epsilon$ leads to the simple absolute calibration formula $d\Sigma_c/d\Omega(q) = i_1(d,q)/(\hat{I}Td)$ that is usually applied to any small angle scattering data. Following this idea, I can define the apparent macroscopic cross section with multiple scattering included.

In the following I discuss how the analytically separated channels add up on the detector and what finite resolution will do to them. The total intensity on the detector is a simple sum of the primary intensity, the coherently and incoherently scattered intensity, according to:

$$i_{tot}(d,\vartheta) = I_0(d)\delta(\vartheta) + i_1(d,q) + j_+(d,\vartheta) \tag{7}$$