On the Inverse Problem for the Multiple Small-Angle Neutron Scattering

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We consider the small-angle multiple neutron scattering and a possibility of its model-free analysis by the inverse problem method. We show that the ill-defined inverse problem is essentially regularized by the use of a planar detector of neutrons without a “beam-stop” facility. We analyze the obtained expressions with the examples of different models of scatterers.

Keywords : multiple small-angle neutron scattering, inverse problem

The small angle neutron scattering (SANS) technique is an important tool for studying large-scale inhomogeneities in condensed matter. The profile of the intensity of passing neutrons around the (0,0,0) reflection provides an information about the size and shape of the inhomogeneities. As discussed, e.g., in [1], the procedure of extracting this useful information may be complicated by different factors, the depletion of the incident beam and multiple scattering among them.

Different ways of analysis of the multiple scattering were proposed up to now [2,3], however all of them depend to some extent on the particular assumptions. In fact, the assumptions are almost inevitable, since the analysis of multiple SANS data deals essentially with an inverse problem, which is mathematically ill-defined.

In this paper we show that the degree of uncertainty in the analysis of multiple scattering can be substantially reduced, when one uses the neutron detector without a so-called “beam-stop” facility. This latter item is implemented in many experimental setups in order to decrease the presumably useless contribution of neutrons passed without a collision. We argue that when a notable part of neutrons scatter in the specimen a few times, this usually discarded intensity can provide a crucial ingredient for the model-free data analysis. Mathematically, taking into account the intensity at the zero angle corresponds to the substantial regularization of the inverse problem found in the multiple SANS.

The rest of this paper is organized as follows. First we rederive the basic equation known in the Bethe-Molière theory of multiple scattering. Our treatment here is similar to one in [1]. We show an intermediate formula which is more suitable for the desired inversion. Formally inverting this formula, we discuss the obtained expression to some detail. The form of functions expected in some models of scattering is shown and discussed in the last part of the paper.

The scattering probability is determined by the single scattering cross section \( \sigma = d\sigma/d\Omega \) with the momentum transfer \( \mathbf{Q} \). Given the momentum of the incident neutron \( k_i \), it is convenient to define the transverse “momentum” \( \mathbf{q} = \mathbf{Q}/k_i \) and the normalized quantity \( \sigma(\mathbf{q}) = \sigma(\mathbf{q})/(\int d^2q \tilde{\sigma}(\mathbf{q})) \). The value \( \sigma(\mathbf{q}) \) is a conditional probability of the scattering at the angle \( \mathbf{q} \). The total scattering cross section \( \sigma_0 = \int d^2q \tilde{\sigma}(\mathbf{q}) \) enters the definition of a mean-free path, \( l \), in a usual way, \( l = (\sigma_0 n_i)^{-1} \), where \( n_i \) is the scatterers’ density.

In the diffractional limit, when \( l \) is much larger than the typical size of inhomogeneities, the probability of multiple scattering is given by a Poisson distribution so that \( n \) scattering events on the path \( L \) are expected for the fraction \( p_n \) of incoming neutrons, \( p_n = (L/l)^n \exp(-L/l) \). The part \( p_0 = \exp(-L/l) \) corresponds to the non-scattered neutrons.

The total multiple scattering intensity is given by a sum

\[
I(q, L) = \sum_{n=0}^{\infty} p_n I_n(q)
\]  

with the recursively defined \( I_n(q) \):

\[
I_0(q) = \delta(q)
\]

\[
I_n(q) = \int d^2k I_{n-1}(q-k)\sigma(k), \quad n \geq 1
\]

In terms of 2D Fourier transform \( A(\mathbf{r}) = \int d\mathbf{q} e^{i\mathbf{q}\cdot\mathbf{r}} A(q) \) we rewrite

\[
I(\mathbf{r}, L) = \sum_{n=0}^{\infty} p_n I_n(\mathbf{r})
\]

\[
I_n(\mathbf{r}) = [\sigma(\mathbf{r})]^n, \quad n \geq 0
\]

The sum (1) is explicitly represented in the form

\[
I(\mathbf{r}, L) = \exp(-L/l) \sum_{n=0}^{\infty} \left( \frac{L}{l} \sigma(\mathbf{r}) \right)^n
\]

\[
= \exp \left[ -\frac{L}{l} (1 - \sigma(\mathbf{r})) \right]
\]

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Note that, according to our definition, $\sigma(\mathbf{r} = 0) = 1$. Let us assume now that the scattering is isotropic, i.e., $\sigma(\mathbf{q})$ does not depend on the direction of $\mathbf{q}$. It follows then that the quantity $\sigma(\mathbf{r})$ depends only on the absolute value of $\mathbf{r}$, and we can transform (5) by going to the “momentum” representation and integrating over the angle in $\mathbf{r}$--plane. As a result, we obtain

$$I(q, L) = \int_0^\infty \frac{r \, dr}{2\pi} J_0(qr) \exp \left[ -\frac{L}{r} \frac{L}{r} (1 - \sigma(r)) \right]$$

$$= \delta(q) e^{-L/|r|} + \int_0^\infty \frac{r \, dr}{2\pi} J_0(qr) \left[ e^{\sigma(r)L/|r|} - 1 \right] e^{-L/|r|}$$

with the Bessel function $J_0(x)$. In the second line of (5) the integral is regularized by explicitly subtracting the singular contribution of the direct beam. One easily verifies the equivalence of the expression (5) to the Bethe’s formula for the small-angle scattering.

The Eq. (5) has two disadvantages. First, it demands the isotropy of the scattering in the $xy$--plane, which could be violated, e.g., for the textured compounds. Second, Eq. (5) is rather awkward, which makes hard its subsequent analysis with taking into the account the experimental resolution function. This latter shortcoming of (5) cannot be cured if one works experimentally with the stripe (one-dimensional) neutron dectector which was considered in (8). Indeed, given our assumptions above, we see that if the quantity $I_{exp}(\mathbf{r}, L)$ is determined for two samples with thicknesses $L_1$, $L_2$, then the ratio $I_{exp}(\mathbf{r}, L_1)/I_{exp}(\mathbf{r}, L_2)$ does not depend on $F(\mathbf{r})$. It allows us to invert the equations (5), (11) and to write

$$\frac{1}{L_2 - L_1} \log \frac{I_{exp}(\mathbf{r}, L_1)}{I_{exp}(\mathbf{r}, L_2)} = \left[ \frac{1 - \sigma(\mathbf{r})}{l} + \frac{1}{l_a} \right] = f(\mathbf{r})$$

This equation is the main result of our paper. Let us discuss it in more detail.

1) The rhs of Eq.(12) does not depend on the sample thickness $L_{1,2}$, therefore one can combine the scattering data, obtained for different $L_i$ and to improve the determination of the shape of $f(\mathbf{r})$.

2) For the case of the isotropic scattering, $\sigma(\mathbf{r}) = \sigma(\mathbf{r})$, one averages over the directions in $xy$--plane in (12) and not in (5).

3) Generally, $I_{exp}(\mathbf{r}, L)$ is a complex-valued function. However, the ratio $I_{exp}(\mathbf{r}, L)/I_{exp}(\mathbf{r}, 0)$ entering (12) should be real, provided $\sigma(\mathbf{k}) = \sigma(-\mathbf{k})$. This condition means simply the inversion symmetry which is generally present on the SANS scale (violating probably in some special cases, e.g. for some substances in the magnetic field). Adopting that $\sigma(\mathbf{k}) = \sigma(-\mathbf{k})$, we note that the imaginary part of logarithm in (12) can stem from two reasons. The first reason is the statistical noise and the second one is the displacement of the direct beam ($\mathbf{q} = 0$) during the measurements with different $L_i$. If $\mathbf{q}_{disp}$ is the displacement of the direct beam, then $I_{exp}(\mathbf{r}, L)/I_{exp}(\mathbf{r}, 0)$ acquires the factor $\exp[i\mathbf{q}_{disp}\mathbf{r}]$. This factor can be easily determined by the appropriate analysis of the data. The remaining complexity in the value of the discussed ratio is due to the statistical noise. Both discussed sources of the complex quantity in the lhs of (12) are irrelevant to the physical quantity $\sigma(\mathbf{r})$ and can be stripped off by writing

$$f_{reg}(\mathbf{r}) = \frac{1}{L_2 - L_1} \log \left| \frac{I_{exp}(\mathbf{r}, L_1)}{I_{exp}(\mathbf{r}, L_2)} \right|$$

This definition of $f_{reg}(\mathbf{r})$ is the partial regularization of our inverse problem, since it projects the experimental data into a definite (symmetrical) class of the functions $\sigma(\mathbf{q})$. Hereafter we assume that $f(\mathbf{r})$ is regularized according to (13) and drop the subscript in the function $f_{reg}(\mathbf{r})$. In fact, the regularization (13) is not very restrictive. The crucial regularization of the problem appeared in (14), corresponding to the assumption of the planar detector without ”beam-stop”.
4) Two basic features in the determined shape of \( f(r) \) are of interest. First, since \( \sigma(0) = 1 \) one has \( f(0) = 1/l_a \), i.e. the absorption length is found in the same experiment as the other quantities. Second, one expects that \( \sigma(\infty) = 0 \) (see below) so that \( f(\infty) = 1/l + 1/l_a \) and hence the value of \( l_a \) is determined.

5) Knowing \( l \) and \( l_a \), we find \( \sigma(r) \) and can compare the form of this function with the different models of scatterers, partly discussed below. The characteristic scale \( r_0 \) of the variation of \( \sigma(r) \) is connected with the size of the inhomogeneities \( R \) by a simple relation \( R = r_0/k \).

6) The above limit \( \sigma(\infty) = 0 \) corresponds to the vanishing angle between the neighboring neutron detectors, which is evidently never realized. In practice, one deals with the finite Fourier transform, and the finite-size effects appear at largest \( r \), determined by the resolution \( \Delta q \). One may judge confidently about the limiting value \( f(\infty) \) when the curve \( f(r) \) saturates at \( r < 1/\Delta q \). This saturation takes place when the average size of the scatterers is smaller than \( 1/\Delta q \). In this case one uniquely determines two main parameters \( l \) and \( r_0 \), along with the whole \( r \)-dependence of \( \sigma(r) \).

7) It was proposed in [3] to analyze the multiple scattering by inverting Eq. (9) with the subtracted \( \delta \)-function. We note here that such procedure requires the knowledge of the ratio \( L/l \) and should be sensitive to its choice. The inversion of (9) is also vulnerable due to the ignorance of the absorption, \( l_a \), and the experimental resolution.

It is worth to provide here some examples of the possible shapes of \( \sigma(r) \). We do it first for the scattering on the spheres and for the phenomenological “Ornstein-Zernike-squared” cross-section.

In the first case, one has the spherical scatterers of one radius \( r_0 \), and the differential cross-section in the diffraction limit reads as

\[
\sigma_{sph}(q) \propto q^{-3}I_{3/2}(qr_0)
\]  

For this function one finds the image \( \sigma(r) \) in the form

\[
\sigma_{sph}(r) = g_{sph}(r^2/4r_0^2)
\]

with \( g_{sph}(x) = \sqrt{1-x}(1 + x/2) \)

\[
-\frac{x}{2}\left(1 - \frac{x}{2}\right) \ln \left[ \frac{1 + \sqrt{1-x}}{\sqrt{x}} \right]
\]

at \( x \leq 1 \) and \( g_{sph}(x) = 0 \) otherwise.

In the second case, one writes

\[
\sigma_{OS2}(q) \propto (q^2r_0^2 + 9/2)^{-2}
\]

with the choice of the factor 9/2 explained below. The image \( \sigma(r) \) is found as

\[
\sigma_{OS2}(r) = g_{OS2}(r/2r_0)
\]

\[
g_{OS2}(x) = (3x/\sqrt{2})K_1(3x/\sqrt{2})
\]

with the modified Bessel function \( K_1(x) \). We note that the expression \( \left(\frac{1}{2}\right) \) corresponds to the Guinier radius \( R_G^2 = 4r_0^2/3 \), which is approximately two times larger than in the case of spheres, \( R_S^2 = 3r_0^2/5 \).

The behavior of the corresponding functions \((1-\sigma(r))\), entering eq. (14), is shown in Fig. 1. An obvious saturation at large \( r \) takes place in the case of spherical scatterers, while this feature is not so prominent for the function \((1-\sigma_{OS2}(r))\). On the other hand, the point of intersection of both curves at \((1-\sigma(r)) \simeq 0.88 \) at \( r \simeq 1.4r_0 \) turns out to be semi-invariant.

To explain the last statement, we remind that in practice one can rarely expect that all the scatterers are of the same size. To model the situation, we suggest that the sizes of the spherical scatterers are distributed within some range, with the probability \( P(R) \) to find a sphere of radius \( R \). The function \( P(R) \) is normalized and the mean square of \( R \) is \( r_0^2 \):

\[
\int_0^\infty P(R)dR = 1, \quad \int_0^\infty R^2P(R)dR = r_0^2
\]

Then one can write

\[
\sigma(r) = \int_0^\infty dR g_{sph}(r^2/4R^2)P(R)
\]

The simplest one-parameter family of functions \( P(R) \), obeying (20), is given by

\[
P_\alpha(R) = \frac{2}{R}F_\alpha \left( \frac{R^2}{r_0^2} \right), \quad F_\alpha(x) = \frac{(x\alpha)^\alpha}{\Gamma[\alpha]} e^{-x\alpha}
\]

The parameter \( \alpha \) characterizes the width of distribution, according to a relation \( \langle R^4 \rangle - \langle R^2 \rangle^2 = r_0^4/\alpha \), with \( \langle \ldots \rangle \) standing for the average. Thus the case \( \alpha \to \infty \) corresponds to the previous case of the monodispersed situation, \( P_\infty(x) = \delta(R-r_0) \), while \( P_\alpha(x) \) becomes uniform distribution in the limit \( \alpha \to 0 \).

Substituting (22) into (21), we find the family of curves \( \sigma_\alpha(r) \) in the form

\[
\sigma_\alpha(r) = \sqrt{\pi}G_{\frac{30}{23}} \left( \frac{r^2\alpha}{4r_0^2} \right)^{3/2, \alpha} 0, 1, \alpha
\]

with the Meijer function, implemented, e.g., in Mathematica. We numerically found that the above point \((1-\sigma(r)) \simeq 0.88 \) and \( r \simeq 1.4r_0 \) is semi-invariant for the family (23) for \( 0.4 \leq \alpha \leq 15 \). For completeness, we also present here the form of \((\sigma(q)) \). Up to the overall coefficient one has:

\[
\sigma_\alpha(q) = q^{-2}G_{\frac{31}{23}} \left( \frac{\alpha}{q^2r_0^2} \right)^{0, 3/2, \alpha} 1, \alpha
\]

Let us now discuss our choice of the factor 9/2 in Eq. (14). This factor appears quite naturally when one poses oneself a following question. Which distribution \( P(s) \) in
leads to the intensity profile of the form $(q^2 r^2 + 1)^{-2}$ and what is the relation between $r_0$ and $r$ here?

The answer to this question is based on the observation, that eq. (21) is the Mellin convolution of the functions $g_{ph}(x)$ and $P(x)$ and, in principle, may be inverted for the known $\sigma(r)$. The corresponding derivation involves the theory of Mellin-Barnes integrals, discussed, e.g. in [5]. Without going into details, we quote here the final result of the form

$$P(R) = \left(2/R\right) F \left(R^2/r_0^2\right)$$

$$F(x) = \left(\frac{9x}{2}\right)^2 e^{-3\sqrt{x}} \left(1 + \frac{1}{3\sqrt{2x}}\right). \tag{25}$$

The behavior of this function is shown in Fig. 2. Therefore the distribution (25) of spherical scatterers with average $\langle R^2 \rangle = r_0^2$ leads to the intensity profile (17), (18).

Consider also a situation when the inhomogeneities in the specimen form clusters, whose size is described by fractal statistics. [6] In the case of simple volume fractals, the commonly adopted density-density correlation function is given by

$$\langle \rho(r)\rho(0) \rangle \propto r^{D-3} \exp(-r/\xi), \tag{26}$$

with a fractal dimension $D$ and maximum cluster size $\xi$. Given the function (26), one finds the form of the single scattering cross section $d\sigma/d\Omega$, exhibited elsewhere [6]. Its counterpart in $r$–space, entering our eq. (12) reads as

$$\sigma(r) = G_{13}^2 \left(\frac{r^2}{4\xi^2}\right)^{1/2} \left(\frac{1}{2}, \frac{D-2}{2}, \frac{D-1}{2}\right). \tag{27}$$

The saturation of this function at large $r$ is realized when $r > \xi$. In particular, it means that one cannot uniquely determine the parameters $\xi$, $l$, $l_a$ in the case when $\Delta q > 1$.

In conclusion, we discuss the inverse problem for the multiple small-angle neutron scattering. We show that the consideration of the forward-going neutrons corresponds to the essential regularization of this problem. The proposed formalism may be applied in the region where the sample thickness is of order of a few mean-free paths $l$.

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