Multiple small-angle scattering. A probe for large inhomogeneities
S. Mazumder

To cite this version:
S. Mazumder. Multiple small-angle scattering. A probe for large inhomogeneities. Journal de Physique IV Colloque, 1993, 03 (C8), pp.C8-519-C8-522. <10.1051/jp4:19938108>. <jpa-00252239>

HAL Id: jpa-00252239
https://hal.archives-ouvertes.fr/jpa-00252239
Submitted on 1 Jan 1993

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
Multiple small-angle scattering. A probe for large inhomogeneities

S. MAZUMDER

Solid State Physics Division, Bhabha Atomic Research Centre, Bombay 85, India

Abstract

In conventional small-angle scattering technique, multiple scattering is considered to be of nuisance value and one looks for a correction factor to eliminate its effect. But it is possible to expand the scope of small-angle scattering beyond its conventional limit if multiple scattering is suitably exploited. This paper intends to illustrate this point.

Introduction

In traditional small-angle scattering (SAS) experiments, one can study the structural features of inhomogeneities with sizes ranging between 1μm and 1nm depending upon the resolution of the instrument. The upper limit of the extractable size range bears a reciprocal relation to the minimum value of the accessible wave-vector transfer q which is generally limited by the finite width of the direct beam. A primary feature of multiple scattering is the broadening of the scattering profile. It should therefore be possible to exploit this beam broadening feature of multiple scattering, using suitably thick samples, to probe very large size inhomogeneities which are otherwise inaccessible to a measurement because of the fact their single scattering profile falls well within the incident beam profile. Further, due to the high thickness of the sample in multiple scattering experiment, the incident beam can get scattered almost completely, yielding much improved signal to background ratios down to very high wave-vector transfer.

The traditional SAS technique has been largely exploited to study thin samples for which the single scattering approximation, for the radiation-matrix interaction, holds good. The common practice is to obtain correction factors to eliminate the effect of multiple scattering in analysing the scattering data. The single scattering approximation is invalid when the thickness of the sample far exceeds the mean free path between SAS events. So the validity of scattering laws based on single scattering approximation remains a question mark.

We have developed a formalism, summarised in a review article [1], on multiple small-angle scattering (MSAS) which delineates various aspects of multiple small-angle scattering. The results of an experimental investigation [2] on two bidisperse alumina samples agree very well with those predicted by this formalism. In this paper, we will highlight some of the results of the formalism for an easy exploitation and interpretation of multiple small-angle scattering. We will describe the behaviour of multiple scattering laws taking the special case of spherical particles. We will also describe how the multiple scattering laws, in association with variational technique like the maximum entropy, can be exploited to extract particle size distribution.

Scattering laws for multiple small-angle scattering

1. Monodisperse system

In multiple small-angle scattering (MSAS), scattering laws are described by normalised density
function \( F(q) \) \([\int F(q)\,d\Omega = 1]\).

The angular distribution of the radiation, for \( \delta(q) \) incident beam, passing through the sample is given by

\[
\delta(q) \cdot \exp(-\mu Z N) + (1-e^{-\mu})\exp(-\mu Z N)F(q)
\]

where

\( \mu \) is macroscopic absorption co-efficient of the sample, \( N = g \zeta Z \lambda^2 D R^3 \) is the average number of SAS interaction radiation has undergone while passing through the sample of thickness \( Z \), \( \zeta \) is the number density of the inhomogeneity of radius \( R \) in the sample, \( \lambda \) is the wave length of the radiation and \( D \) is the scattering length density contrast of the inhomogeneity vis-a-vis that of the otherwise homogeneous matrix.

The fraction of the incident radiation absorbed in the sample is given by \( (1-e^{-\mu Z}) \).

Differential scattering cross section \( S(q) \) of the sample is given by

\[
S(q) = \text{Area of the sample surface exposed to the incident beam.}(1-e^{-\mu})\ F(q).
\]

Differential scattering cross section \( S_{\text{p}}(q) \) per unit volume of the sample is \( (1-e^{-\mu})\ F(q)/Z \)

For conventional SAS, \( 1-e^{-\mu Z} \approx N \), whereas for NSAS \( 1-e^{-\mu Z} \approx 1 \).

a) Guinier regime

\( k \) denoting the wave number of the radiation, for monodisperse population of spheres of radius \( R \),

\[
\lim_{q\to 0}F_{\text{MAS}}(q) = 2\pi k^2/(5\mu N) \cdot \exp(-\pi^2 q^2/5N).
\]

The above relation depicts the beam broadening feature of multiple scattering and indicates the following facts:

i) Guinier law is valid, under a suitable scaling down of wave-vector transfer from \( q \to q/N \)

ii) Guinier plot provides information about rescaled radius \( R/N \).

b) Porod regime

The scattering cross section in the Porod regime is given by

\[
\lim_{q\to 0}S_{\text{MAS}}(q) = (4\pi/3)(R^2)q^{-4}. \text{ Area of the sample surface exposed to the incident beam.}
\]

In contrast, the corresponding single scattering cross section is given by

\[
\lim_{q\to 0}S_{\text{MAS}}(q) = 2\pi d^2(4\pi R^2)q^{-4} \text{ where, } m \text{ is the number of particles bathed in the beam.}
\]

The expression for \( S_{\text{MAS}}(q) \) indicates that Porod Law remains invariant as far as the functional dependence of the scattering cross section on wave vector transfer is concerned.

2. Polydisperse system

Now let us consider the nature of the scattering profile for a polydisperse system. For the present discussion, we assume that the sample under study consists of a large variety of spherical scattering particles. The volume fraction of the \( i \)-th type of particles in the sample is denoted by \( p_i \) while \( R_i, \zeta_i \) and \( \mu_i \) are respectively its radius, number density and macroscopic absorption co-efficient. The subscript \( i \) runs from 1 to \( n \) where \( n \) indicates the degree of polydispersity of the medium.

When the linear dimensions of the particles are negligible in comparison with the mean free path of
the radiation inside the medium, it is possible to define an effective effective single scattering cross section and absorption coefficient of the medium. Such polydisperse medium is termed as "effective medium".

We will restrict our present discussion on scattering profile from a polydisperse effective medium only. We assume that different particles have same scattering length density contrast $D$.

a) Guinier regime

The scattering profile in the Guinier regime is represented by

$$\lim_{q \to 0} F_{\text{MSAS}}(q) = 2q^2 R^2 / (15R^2)(q^2)^2 \exp\left(-2q^2 R^2 / (15R^2)^2 \right)$$

where, $\tau$ denotes the packing fraction of the sample. The above relation indicates that the extractable information from the Guinier regime of a MSAS profile from a polydisperse system is given by

$$\langle R^3 / R^2 \rangle = \frac{\tau}{\langle R \rangle} \frac{\langle R \rangle}{\langle R^2 \rangle} = \frac{\langle R \rangle}{\langle R \rangle} \frac{\langle R^2 \rangle}{\langle R \rangle}$$

for a continuous distribution of particles.

b) Porod regime

The scattering profile in the Porod regime of the profile is given by

$$\lim_{q \to \infty} F_{\text{SAS}}(q) = (2\pi \rho_0 \lambda_0)^2 \langle R^2 / R^3 \rangle q^{-4}$$

indicating that the nature of the extractable information from the Porod regime of the multiple scattering profile is given by $\langle R^4 / R^2 \rangle$.

In contrast, the corresponding single scattering profile is given by

$$\lim_{q \to \infty} F_{\text{SAS}}(q) = (2\pi \rho_0 \lambda_0)^2 \langle R^2 / R^3 \rangle q^{-4}$$

indicating $\langle R^2 / R^3 \rangle$ is the extractable information from the Porod regime of SAS profile of a polydisperse system.

c) Particle size distribution

Now let us investigate how to obtain the particle size distribution using the extractable moment ratios from the MSAS profile.

If we assume that the particle size distribution is log-normal in nature, then $\rho(R)$ is given by

$$\rho_{\text{LN}}(R) = \frac{1}{\langle R \rangle (2\pi)^{1/2}} \exp\left(-1/2(\ln R - \ln \tau)^2\right)$$

where $\tau$ and $\gamma$ are solutions of the following simultaneous equations

$$\langle R^3 / R^2 \rangle = \exp(5/2\gamma^2 - \gamma/\tau) = a \text{ (say)}$$

and

$$\langle R^4 / R^2 \rangle = \exp(6/\gamma^2 - 2\gamma/\tau) = b \text{ (say)}.$$

To obtain the maximum entropy distribution, one has to search that $\rho(R)$ for which the entropy functional

$$S = \int \rho(R) \ln(\rho(R) / q(R)) dR$$
is maximum and which also satisfies the constraints
\[ \left( \frac{\rho(R)}{R^3} \right) dR / \left( \frac{\rho(R)}{R^2} \right) dR = a \quad \text{and} \quad \left( \frac{\rho(R)}{R^4} \right) dR / \left( \frac{\rho(R)}{R^2} \right) dR = b. \]

In the expression of entropy functional, \( g(R) \) is the prior distribution. In the present case \( g(R) \) is such that \( \rho(R) \to 0 \) for \( R \to 0 \). To be noted that the condition \( \rho(R) \to 0 \) for \( R \to \infty \) is already satisfied even for \( g(R) = 1 \).

\( \rho(R) \) is given by the following expression
\[ \rho(R) \propto g(R) \exp \left( -\chi \left( R^2 - aR^2 \right) - \theta \left( R^2 - bR^2 \right) \right) \]
where \( \chi \) and \( \theta \) are solutions of the following simultaneous equations
\[ \int (R^2 - aR^2) \rho(R) dR = 0 \quad \text{and} \quad \int (R^4 - bR^2) \rho(R) dR = 0. \]

To appreciate the difference between the two distributions namely the log normal (LN) one and maximum entropy (ME) one, let us consider the result of the numerical calculation with \( g(R) = R^2 \) as depicted in figure 1. As expected the ME distribution is relatively flatter.

![Fig.1. Estimation of particle size distribution \( \rho(R) \) from the assumed values of the moment ratios \( \langle R^3 \rangle / \langle R^2 \rangle = 1.62 \times 10^6 \text{ A} \) and \( \langle R^4 \rangle / \langle R^2 \rangle = 2.7 \times 10^8 \text{ A}^2 \). Curve 1 and 2 respectively depict the log-normal and maximum entropy distributions. For curve 1 \( \rho(R) \) maximizes at 1466.7 \text{ A} whereas for curve 2 the corresponding value is 1614 \text{ A}. The first and the second moments of the two distributions are following: \( \langle R \rangle_{\text{ME}} = 1481 \text{ A}, \langle R^2 \rangle_{\text{ME}} = 2313000 \text{ A}^2, \langle R \rangle_{\text{LN}} = 1531 \text{ A}, \text{ and } \langle R^2 \rangle_{\text{LN}} = 2410000 \text{ A}^2 \).](image)

**Conclusion**

We have discussed few key results of a formalism on multiple small-angle scattering. Some of these results have been tested experimentally. By and large, the results are open for exploitation and further investigation both experimentally and theoretically.

**References:**
