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The Inverse Scattering Problem for Spherical Polydispersion

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Abstract

It is shown how the mean form factor for Rayleigh–Gans scattering from a spherical polydispersion may be inverted to obtain the size distribution of the polydispersion.

The problem of the scattering of electromagnetic waves by spherical particles has seen a resurgence of activity in the last 20 years, although the basic solution was known 70 to 100 years ago (Clebsch 1863; Lorenz 1890; Mie 1908; Debye 1909). The present interest stems from the widespread application of the theory to the physical chemistry of macromolecules and colloids, to X-ray scattering, to atmospheric and space optics and to radar meteorology, which are reviewed, for example, in the books by Van der Hulst (1957) and Kerker (1969). One of the problems of current interest is that of determining the size distribution of a dilute polydisperse system from the properties of the scattered radiation. In this note it is shown that measurements of the angular distribution of intensity or polarization may be explicitly inverted to obtain the size distribution when the individual scatterers are uniform spheres, and the scattering from each is treated in the Rayleigh–Gans approximation.

In the Rayleigh–Gans approximation the scattering from a sphere of radius R, with a relative refractive index m, is described in terms of a form factor $P(k,\theta)$ given by (Rayleigh 1910, 1914, 1918; Debye 1915; Gans 1925)

$$P(k,\theta;R) = \tfrac{9}{2}\pi(xR)^{-3}J_{3/2}^2(xR) = P(xR), \tag{1}$$

where $x = 2k\sin\frac{1}{2}\theta$, θ is the scattering angle and $k = 2\pi/\lambda$ is the wave number. The form factor is related to the intensity I_1 for scattering of radiation polarized perpendicular to the scattering plane, and to the intensity I_2 for scattering of radiation polarized parallel to the scattering plane, by

$$I_1 = \frac{k^4}{r^2}R^6\left|\frac{m^2-1}{m^2+2}\right|^2 P(k,\theta;R) \qquad \text{and} \qquad I_2 = \cos^2\theta\, I_1, \tag{2a, b}$$

where m is the relative refractive index of the scatterer and r is the distance to the observer. From these expressions the Stokes parameters may be derived in the usual way (Newton 1966; Kerker 1969).

Observations on scattering from a polydisperse system will measure an average form factor, defined by

$$\bar{P}(k,\theta) = \int_0^\infty R^6\, P(k,\theta;R)\, n(R)\, \mathrm{d}R\,, \tag{3}$$

where $n(R)$ is the size distribution function of the polydispersion. The fundamental inverse problem is the solution of the expression (3) regarded as an integral equation for $n(R)$. This problem occurs, for example, in the determination of the size distribution from small angle X-ray scattering (Letcher and Schmidt 1966) and in the determination of the size distribution of the atmosphere aerosol (Kuriyan and Sekera 1974; Kuriyan *et al.* 1974). For the case in which the form factor $P(k,\theta;R)$ is given by the Rayleigh–Gans expression (1), we can rewrite equation (3) as

$$\bar{P}(x) = \int_0^\infty P(xR)\, N(R)\, \mathrm{d}R\,, \tag{4}$$

where $N(R) = R^6\, n(R)$. This equation has a solution in terms of Mellin transforms,

$$N(R) = \int_0^\infty W(xR)\, \bar{P}(x)\, \mathrm{d}x\,, \tag{5}$$

where

$$P^{\mathrm{M}}(s)\, W^{\mathrm{M}}(1-s) = 1 \tag{6}$$

and $f^{\mathrm{M}}(s)$ is the Mellin transform of $f(z)$:

$$f^{\mathrm{M}}(s) = \int_{-\infty}^{\infty} f(z)\, z^{s-1}\, \mathrm{d}z\,, \tag{7a}$$

$$f(z) = (2\pi\mathrm{i})^{-1} \int_{C-\mathrm{i}\infty}^{C+\mathrm{i}\infty} f^{\mathrm{M}}(s)\, z^{-s}\, \mathrm{d}s\,. \tag{7b}$$

This solution of equations of the form (4) is derived, for example, by Titchmarsh (1937) and Bracewell (1965). For the particular $P(z)$ defined by equation (1) it is straightforward to construct

$$P^{\mathrm{M}}(s) = \tfrac{9}{4}\pi^{\frac{1}{2}}\,\Gamma(\tfrac{1}{2}s)/\,(3-\tfrac{1}{2}s)(2-\tfrac{1}{2}s)\Gamma(\tfrac{5}{2}-\tfrac{1}{2}s)\,, \tag{8}$$

using standard Mellin transform tables (Erdélyi *et al.* 1954). The construction of $W(z)$ is a little more tedious, but one finds eventually

$$W(z) = \tfrac{16}{9}\pi^{-1} z^{7}\{(\tfrac{9}{4}z^{-\frac{1}{2}}-1)z^{-\frac{1}{4}} j_1(2z^{\frac{1}{4}}) - \tfrac{3}{2}z^{-\frac{3}{4}} j_0(2z^{\frac{1}{4}})\}\,, \tag{9}$$

where $j_l(z)$ is the spherical Bessel function of order l.

The solution of equation (4) can also be given in an alternative form by writing this equation as

$$-\tfrac{1}{9}\pi^{-\frac{1}{2}} x^3\, \bar{P}(x) = -\int_0^\infty k_{3/2}(xR)\, m(R)\, \mathrm{d}R\,, \tag{10}$$

where

$$k_{3/2}(z) = \tfrac{1}{2}\pi^{\frac{1}{2}}\, \mathrm{d}\big(zJ_{3/2}^2(z)\big)/\mathrm{d}z \tag{11}$$

is Bateman's kernel (Bateman 1906; Titchmarsh 1937) and

$$\mathrm{d}m/\mathrm{d}R = R^2 n(R). \tag{12}$$

Equations of the type (10) were solved by Bateman. In the present case we obtain

$$m(R) = \tfrac{1}{9}\int_0^\infty J_{3/2}(\tfrac{1}{2}xR)\, Y_{3/2}(\tfrac{1}{2}xR)\, x^3\, \bar{P}(x)\, \mathrm{d}x. \tag{13}$$

As a check on the above construction, the expression for $\bar{P}(x)$ given by Kuriyan and Sekera (1974) for the Deirmendjian (1969) haze H distribution function

$$n(R) = aR^2 \exp(-bR) \tag{14}$$

has been explicitly inverted to recover the original distribution function.

It should be emphasized that the inversions given by equations (5) and (13) formally require a knowledge of $\bar{P}(x)$ for all x in the range $0 < x < \infty$, while in any practical situation $\bar{P}(x)$ can be measured for only a finite range of values of x. An important question is the extent to which this lack of knowledge of $\bar{P}(x)$ over the whole range of x affects the uniqueness of the inversions (5) and (13). As an extreme example that this restriction of the range can limit the power of the inversion technique, we note that (McKellar 1974)

$$\bar{P}(x) = \bar{P}(0)\{1 - \tfrac{1}{5}\rho^2 x^2 \dots\},$$

where

$$\rho^2 = \int_0^\infty R^8\, n(R)\, \mathrm{d}R \bigg/ \int_0^\infty R^6\, n(R)\, \mathrm{d}R.$$

This implies that a knowledge of $\bar{P}(x)/\bar{P}(0)$ only for very small x (which is sometimes the experimental situation) determines just the parameter ρ^2 of the distribution.

At this stage the inversion formulae (5) and (13) should be regarded as formal solutions of the inversion problem for scattering by polydispersions. It is proposed in a later paper to discuss the problem of converting these formal solutions into practical tools, and the related problems of deciding which of the inversion formulae is of more use in practical situations.

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