Determination of Correlation Functions of Scattering Potentials of Stochastic Media from Scattering Experiments

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The classic ''Ewald-sphere construction'' for determining the structure of crystalline objects from x-ray and neutron diffraction experiments is generalized to determine the correlation functions of scattering potentials of stationary random media from scattering experiments.

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One of the oldest, very powerful, and useful methods for determining the structure of crystalline objects is the well-known technique of x-ray diffraction. The x-ray diffraction method was introduced in classic works of von Laue, Friedrich, and Knipping [1] and of Ewald [2,3] in the 1920's, and since then it has became one of the chief tools for the study of the structure of materials. Later on, this technique has also come to be used with slow neutrons (see for example [4,5]). Basic papers on this subject are reprinted in Ref. [6].

In the present Letter, we show that this technique may be generalized to determine, approximately, the correlation functions of scattering potentials of random (stochastic) static media from scattering experiments. The essence of the technique consists of measurements of a correlation function (the so-called cross-spectral density function) of the far field, generated by scattering of a coherent plane wave on the medium and to calculate, from the measured data, an approximation to the correlation function of the scattering potential of the medium by means of multiple Fourier transforms. In the classic theory of x-ray diffraction by crystals, the range of accessible Fourier components of the crystal structure is determined elegantly with the help of the well-known ''Ewald-sphere construction'' [2,7]. We show that some of the low spatial-frequency components of the correlation function of the scattering potential of a static stochastic medium may be determined with the help of two spheres, each of which is analogous to the Ewald limiting sphere of the classic theory of the x-ray diffraction by crystals.

Several attempts to obtain information about scattering potentials of random media involve the measurement of the spectrum of the scattered light in the far zone (see, for example, [8–11]). Because the spectrum is a function of position (i.e., it is a ''single point'' quantity), whereas the correlation function of the scattering potential is a function of two points, these methods are only applicable when the medium has some special properties, such as homogeneity. In a paper published about two decades ago [12], general expressions were obtained for the cross-spectral density function, for the spectral degree of coherence, and for the spectral density of the light in the far zone generated by scattering on a random medium; i.e., it deals with the direct scattering problem. In this Letter, we present solution to an inverse problem. We show how to obtain information about the scattering potential of the medium from measurements of the cross-spectral density function of the scattered light in the far zone, using a generalization of the Ewald-sphere construction. The proposed method allows the determination of some, if not all, of the low spatial-frequency components of the correlation function of the scattering potential of weak scatterers of a wide class [13].

In the absence of symmetry properties of the scattering medium, our method is computationally rather intensive. But the theory reveals an elegant analogy with the classic theory of structure determination of crystalline objects and provides a method for determining an approximation to the correlation functions of scattering potentials of static random media.

Suppose that a coherent polychromatic plane light wave, propagating in a direction specified by a real unit vector $\mathbf{s}_0$, is incident on a statistically stationary random medium, occupying a finite domain D (see Fig. 1). The incident light at a point $\mathbf{r}$ may be characterized by a statistical ensemble $\{U^{(i)}(\mathbf{r}, \mathbf{s}_0; \omega) \exp[-i\omega t]\}$ of monochromatic realizations $U^{(i)}(\mathbf{r}, \mathbf{s}_0; \omega)$, all of frequency ω, in the sense of coherence theory in the space-frequency domain (see Ref. [15], Chapter 4). Here,

$$U^{(i)}(\mathbf{r}, \mathbf{s}_0; \omega) = a(\omega) \exp(ik\mathbf{s}_0 \cdot \mathbf{r}), \quad (1)$$

where $a(\omega)$ is, in general, complex and $k = \omega/c$, c being the speed of light in vacuum. The cross-spectral density function of the incident light at a pair of points, specified by position vectors $\mathbf{r}_1$ and $\mathbf{r}_2$, may be expressed in the form ([15], Sec. 4.1)

$$W^{(i)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{s}_0; \omega) = \langle U^{(i)*}(\mathbf{r}_1, \mathbf{s}_0; \omega) U^{(i)}(\mathbf{r}_2, \mathbf{s}_0; \omega)\rangle, \quad (2)$$

where the asterisk denotes the complex conjugate and the angular brackets denote ensemble average. From Eqs. (1) and (2), it follows that the cross-spectral density function

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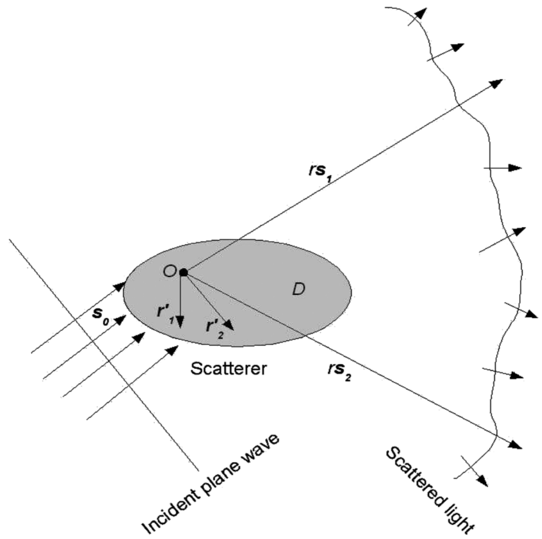

FIG. 1. Illustrating the notations. $\mathbf{s}_0$ is a unit vector in the direction of propagation of the incident plane wave, $\mathbf{s}_1$ and $\mathbf{s}_2$ are unit vectors in directions of scattering. r is a distance from an origin in the domain occupied by the scatterer to a point in the far zone.

of the incident light may be expressed as

$$W^{(i)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{s}_0; \omega) = S^{(i)}(\omega) \exp[ik\mathbf{s}_0 \cdot (\mathbf{r}_2 - \mathbf{r}_1)], \quad (3)$$

where $S^{(i)}(\omega) = \langle a^*(\omega)a(\omega)\rangle$ is the spectral density of the incident light.

The scattering potential of a deterministic medium, at a point specified by a position vector $\mathbf{r}'$ within the scatterer, is defined by the formula {[16], Sec. 13.1.1, Eq. (6)}

$$F(\mathbf{r}'; \omega) = \frac{k^2}{4\pi}[n^2(\mathbf{r}'; \omega) - 1], \quad (4)$$

where $n(\mathbf{r}', \omega)$ is the refractive index of the medium, at frequency ω of the incident light. For a random medium $F(\mathbf{r}'; \omega)$ is, of course, a random function $\mathbf{r}'$. The correlation function of the scattering potential at a pair of points, specified by position vectors $\mathbf{r}'_1$ and $\mathbf{r}'_2$ in the scattering medium, is defined by the formula

$$C_F(\mathbf{r}'_1, \mathbf{r}'_2; \omega) = \langle F^*(\mathbf{r}'_1; \omega)F(\mathbf{r}'_2; \omega)\rangle_m, \quad (5)$$

where the angular brackets with subscript m denote the average, taken over the ensemble of the random medium. We assume that the medium is a weak scatterer so that the scattering may be analyzed within the accuracy of first Born approximation ([16], Sec. 13.1.2). The cross-spectral density function of the scattered light in the far zone, at two points specified by position vectors $r\mathbf{s}_1$ and $r\mathbf{s}_2$ ($\mathbf{s}_1^2 = 1$, $\mathbf{s}_2^2 = 1$) (see Fig. 1), is given by the formula {[15], Sec. 6.3.2, Eq. (9)}

$$W_s^{(\infty)}(r\mathbf{s}_1, r\mathbf{s}_2, \mathbf{s}_0; \omega) = \frac{S^{(i)}(\omega)}{r^2} \times \tilde{C}_F[-k(\mathbf{s}_1 - \mathbf{s}_0), k(\mathbf{s}_2 - \mathbf{s}_0); \omega]. \quad (6)$$

Here,

$$\tilde{C}_F[\mathbf{K}_1, \mathbf{K}_2; \omega] = \int_D \int_D C_F(\mathbf{r}'_1, \mathbf{r}'_2; \omega) \times \exp[-i(\mathbf{K}_1 \cdot \mathbf{r}'_1 + \mathbf{K}_2 \cdot \mathbf{r}'_2)]d^3r'_1 d^3r'_2 \quad (7)$$

is the six-dimensional Fourier transform of the correlation function (5) of the scattering potential, and

$$\mathbf{K}_1 = -k(\mathbf{s}_1 - \mathbf{s}_0), \qquad \mathbf{K}_2 = k(\mathbf{s}_2 - \mathbf{s}_0). \quad (8)$$

The vectors $\mathbf{K}_1$, and $\mathbf{K}_2$ are analogous to the momentum transfer vector of quantum mechanical theory of potential scattering. From Eqs. (8), it follows that as the direction $\mathbf{s}_1$ varies over all possible directions, with the direction $\mathbf{s}_0$ of incidence being kept fixed, the end points of the vectors $\mathbf{K}_1$ move on the surface of a sphere of radius k centered at the point $k\mathbf{s}_0$. Similarly, with $\mathbf{s}_0$ kept fixed, and the direction $\mathbf{s}_2$ changing, the end points of the vector $\mathbf{K}_2$ move on the surface of another sphere of radius k centered at the point with position vector $-k\mathbf{s}_0$ (see Fig. 2). Each of these spheres are analogous to the classic "Ewald sphere of reflection" used in the theory of x-ray diffraction of crystals [2,7]. It is clear from the definitions (8) that

$$0 \le |\mathbf{K}_1| \le 2k = \frac{4\pi}{\lambda}, \qquad 0 \le |\mathbf{K}_2| \le 2k = \frac{4\pi}{\lambda}, \quad (9)$$

where λ is the wavelength corresponding to the frequency ω. Figure 2 illustrates the domain of the accessible Fourier components, labeled by vectors $\mathbf{K}_1$ and $\mathbf{K}_2$, given by Eqs. (8), of the two point spatial correlation function of the scattering potential, for a particular direction of incidence $\mathbf{s}_0$. Consequently, for a fixed direction of incidence $\mathbf{s}_0$, measurements of the cross-spectral density of the scattered field at all pairs of points ($r\mathbf{s}_1$, $r\mathbf{s}_2$) in the far zone allows the determination of all of those six-dimensional Fourier components ($\mathbf{K}_1$, $\mathbf{K}_2$) for which $\mathbf{K}_1$ and $\mathbf{K}_2$ lie on their respective Ewald spheres of reflection. However, it can be seen from Eqs. (8) and from Fig. 2 that $\mathbf{K}_1$ and $\mathbf{K}_2$ are coupled because both depend on the vector $\mathbf{s}_0$ and cannot be determined independently, because their Ewald spheres of reflection rotate in unison as the direction of incidence $\mathbf{s}_0$ is varied. The vectors $\mathbf{K}_1$ and $\mathbf{K}_2$ satisfy another inequality in addition to the inequalities (9), viz.,

$$|\mathbf{K}_1 + \mathbf{K}_2| = k|\mathbf{s}_2 - \mathbf{s}_1| \le \frac{4\pi}{\lambda}. \quad (10)$$

Nonetheless, as $\mathbf{s}_0$ is varied, the vectors ($\mathbf{K}_1$, $\mathbf{K}_2$) sweep out a continuous, albeit complicated six-dimensional volume in Fourier space.

Equation (6) makes it possible to express the six-dimensional Fourier transform of the correlation function of the scattering potential in terms of the cross-spectral density function of the far field, which may be determined experimentally (see Ref. [15], Sec. 4.2). Using Eqs. (6) and (7), one can obtain an estimate of the correlation function of the scattering potential, denoted by $\hat{C}_F(\mathbf{r}'_1, \mathbf{r}'_2; \omega)$ in terms of the inverse Fourier transform of the cross-spectral density function as

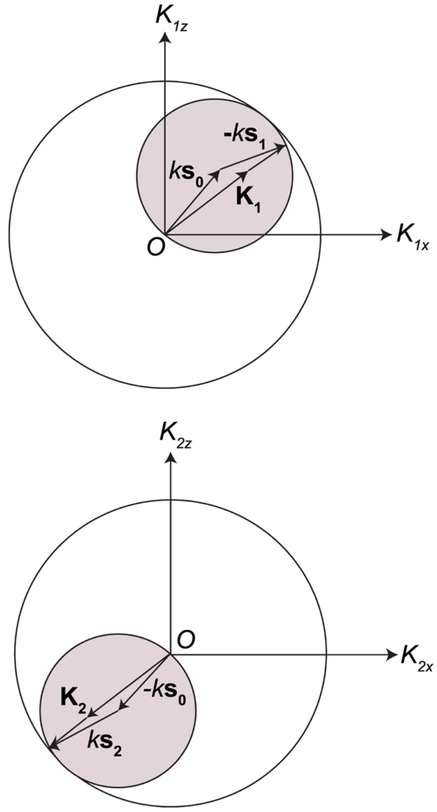

FIG. 2 (color online). The small shaded spheres are analogous to the ''Ewald's spheres of reflection'' for a particular direction $\mathbf{s}_0$ of incidence of a plane wave, with wave number k. These spheres are the region of accessible Fourier components for a particular direction of incidence $\mathbf{s}_0$. The large spheres are analogous to the ''Ewald's limiting sphere."

$$\hat{C}_F(\mathbf{r}_1', \mathbf{r}_2'; \omega) = \frac{r^2}{(2\pi)^6 S^{(i)}(\omega)} \times \int_{V(\mathbf{K}_1)} \int_{V(\mathbf{K}_2)} W_s^{(\infty)}(r\mathbf{s}_1, r\mathbf{s}_2, \mathbf{s}_0; \omega) \times \exp[i(\mathbf{K}_1 \cdot \mathbf{r}_1' + \mathbf{K}_2 \cdot \mathbf{r}_2')] d^3K_1 d^3K_2. \quad (11)$$

Here, $V(\mathbf{K}_1)$ and $V(\mathbf{K}_2)$ denote the regions of integration of $\mathbf{K}_1$ and $\mathbf{K}_2$, respectively.

We will illustrate this technique by determining the correlation function of the scattering potentials of a quasi-homogeneous medium [17]. The correlation function of the scattering potentials of such a medium has the form

$$C_F(\mathbf{r}_1', \mathbf{r}_2'; \omega) = C_R(\mathbf{R}', \omega) C_r(\mathbf{r}'; \omega), \quad (12)$$

where function $C_R(\mathbf{R}', \omega)$ varies much more slowly with $\mathbf{R}' = (\mathbf{r}_1' + \mathbf{r}_2')/2$ than the function $C_r(\mathbf{r}', \omega)$ varies with $\mathbf{r}' = \mathbf{r}_2' - \mathbf{r}_1'$. Substituting Eq. (12) into Eq. (7), we find that the six-dimensional Fourier transform of the correlation function has the factorized form

$$\tilde{C}_F(\mathbf{K}_1, \mathbf{K}_2; \omega) = \tilde{C}_R(\mathbf{K}_1 + \mathbf{K}_2; \omega) \tilde{C}_r[(\mathbf{K}_2 - \mathbf{K}_1)/2; \omega]. \quad (13)$$

In this case, the six-dimensional spatial-frequency vector is most naturally represented (using sum and difference vectors) as $\mathbf{K} = (\mathbf{K}_S, \mathbf{K}_D)$, where

$$\mathbf{K}_S = \mathbf{K}_1 + \mathbf{K}_2 = k(\mathbf{s}_2 - \mathbf{s}_1), \quad (14)$$

$$\mathbf{K}_D = (\mathbf{K}_2 - \mathbf{K}_1)/2 = k[(\mathbf{s}_1 + \mathbf{s}_2)/2 - \mathbf{s}_0]. \quad (15)$$

We see that the two three-dimensional spatial-frequency vectors, $\mathbf{K}_S$ and $\mathbf{K}_D$, are uncoupled and can independently sweep out two Ewald limiting spheres of radius $2k$, i.e., $V(\mathbf{K}_S) = \{|\mathbf{K}_S| \leq 2k\}$ and $V(\mathbf{K}_D) = \{|\mathbf{K}_D| \leq 2k\}$. Hence, using Eq. (11), one can obtain an estimate of the low spectral frequency part of the correlation function of the scattering potential of the random medium.

To illustrate the inversion, we will assume that $C_R(\mathbf{R}')$ and $C_r(\mathbf{r}')$ have Gaussian forms, viz.

$$C_R(\mathbf{R}') = C_0 \exp\left[-\frac{|\mathbf{R}'|^2}{2\sigma_R^2}\right], \quad (16a)$$

$$C_r(\mathbf{r}') = \exp\left[-\frac{|\mathbf{r}'|^2}{2\sigma_r^2}\right], \quad (16b)$$

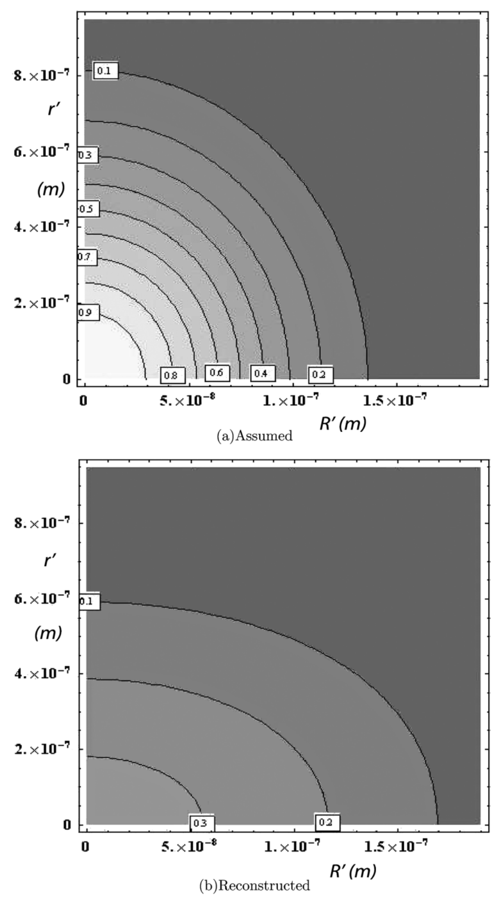

FIG. 3. The assumed (a) and the reconstructed (b) normalized low spatial-frequency part $C_F^{(\mathrm{LP})}/C_0$ of the correlation function of the scattering potential plotted as functions of $|\mathbf{R}'| = R'$ and $|r'| = r'$, for a medium with correlation function of scattering potential given by Eqs. (12) and (16). The parameters were chosen as $\sigma_r = 0.1\lambda$ and $\sigma_R = 0.6\lambda$.

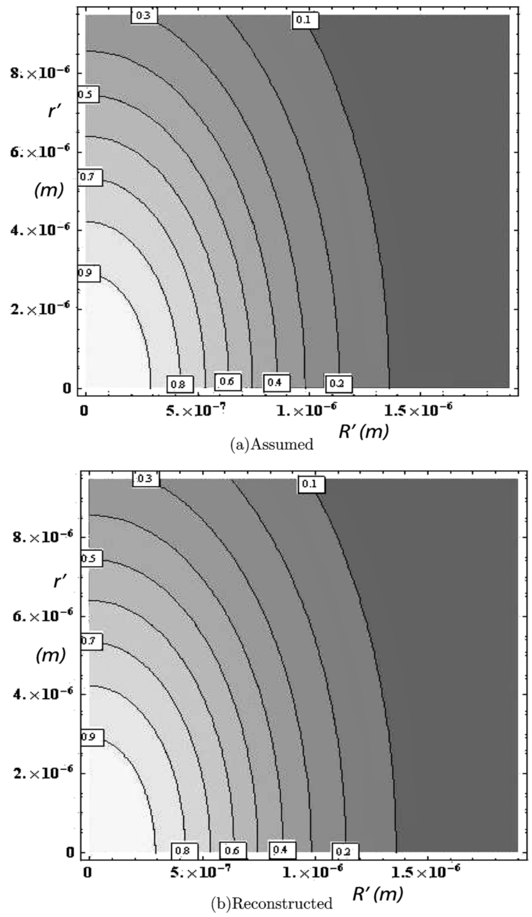

FIG. 4. Same as Fig. 3, but with parameters $\sigma_r = \lambda$ and $\sigma_R = 10\lambda$.

where $\sigma_R \gg \sigma_r$. From Eqs. (6) and (7), one readily finds that, in this case,

$$W_s^{(\infty)}(r\mathbf{s}_1, r\mathbf{s}_2, \mathbf{s}_0; \omega) = \frac{(2\pi\sigma_R\sigma_r)^3 S^{(i)}(\omega)}{r^2} C_0 \times \exp\left[-\frac{1}{2}|\mathbf{K}_S|^2\sigma_R^2\right] \times \exp\left[-\frac{1}{2}|\mathbf{K}_D|^2\sigma_r^2\right]. \quad (17)$$

The cross-spectral density function $W_s^{(\infty)}$ $(r\mathbf{s}_1, r\mathbf{s}_2, r\mathbf{s}_0; \omega)$ is proportional to the degree of coherence, and hence can be determined from interference experiments (see [15], Sec. 4.2). Using Eqs. (11) and (17), one can then determine the low spatial-frequency part $C_F^{(\mathrm{LP})}$ $(\mathbf{r}_1', \mathbf{r}_2'; \omega)$ of the correlation function of the scattering potential. The reconstruction may be expected to be reasonably accurate, when the parameters σ_R and σ_r are appreciably greater than the wavelength, as is the case illustrated by the Figs. 3 and 4.

We conclude by saying that we have described a method for determining an approximate form of the correlation function of scattering potential of stochastic random media from measurements on the correlation function of the scattered field in the far zone. The method may be regarded as a generalized analogue of the classic technique due to von Laue and Ewald of determining the structure of crystalline objects from x-ray diffraction experiments. We illustrated the theory by an example.

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