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Acoustic Wave Correlation Tomography of Time-Varying Disordered Structures

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An original model based on first principles is constructed for the temporal correlation of acoustic waves propagating in random scattering media. The model describes the dynamics of wave fields in a previously unexplored, moderately strong (mesoscopic) scattering regime, intermediate between those of weak scattering, on the one hand, and diffusing waves, on the other. It is shown that by considering the wave vector as a free parameter that can vary at will, one can provide an additional dimension to the data, resulting in a tomographic-type reconstruction of the full space-time dynamics of a complex structure, instead of a plain spectroscopic technique. In Fourier space, the problem is reduced to a spherical mean transform defined for a family of spheres containing the origin, and therefore is easily invertible. The results may be useful in probing the statistical structure of various random media with both spatial and temporal resolution.

Keywords: wave scattering; random media; time correlation; tomographic reconstruction.

1. Introduction

The temporal correlation of waves scattered off, or transmitted through, a random medium has long been a very efficient tool in the study of the space-time dynamics of a great variety of complex structures (Rytov *et al.*, 1989; Berne, Pecora, 1976; Ishimaru, 1978). At the microscale, typical examples include the solutions of macromolecules and colloidal suspensions, gels, foams, granular materials, biological tissues, and laboratory plasma. Macroscopic structures range from atmospheric turbulence, internal waves in the ocean and heterogeneous Earth to ionospheric and interstellar plasmas. The basic quantity measured in such experiments is usually the temporal autocorrelation function (ACF), defined as

$$\Gamma(\tau) = \langle u(t)u^*(t+\tau)\rangle, \tag{1}$$

where $u(t)$ is the complex amplitude of the field (either scattered or total), and the angular brackets mean the ensemble average. In some situations, the analysis is based on the power spectrum of field fluctuations, which is given by the Fourier transformation of the temporal ACF.

The random medium, in its turn, is typically characterized by some constitutive parameter $\widetilde{\varepsilon}(\mathbf{r},t)$. The physical content of $\widetilde{\varepsilon}(\mathbf{r},t)$ depends on the specific problem under study: for example, (fluctuations of the) permittivity in electromagnetic applications, or slowness for acoustic waves, but will generally be referred to as the *scattering potential* in the sequel. The quantity we should recover is usually the two-point correlation function $B_\varepsilon(\boldsymbol{\rho},\tau)$ of the scattering potential, or a corresponding spectral density $\Psi_\varepsilon(\mathbf{q},\Omega)$, obtained by Fourier transforming $B_\varepsilon(\boldsymbol{\rho},\tau)$ in both space and time (see Appendix).

Two extreme regimes of wave scattering are typically considered. When the scattering is rather weak, the wave field can be described by using a single-scattering (Born) approximation. The resulting theory relates the power spectrum measured for a wave propagating initially in the direction of vector $\mathbf{k}_i$ and then scattered in the direction of vector $\mathbf{k}_s$, to the value of spectral density $\Psi_\varepsilon(\mathbf{q},\Omega)$, taken at the Bragg (momentum transfer) vector $\mathbf{q} = \mathbf{k}_s - \mathbf{k}_i$ (Rytov *et al.*, 1989). Quasi-elastic scattering implies the condition $|\mathbf{k}_i| = |\mathbf{k}_s| = k$, where k is the wave number of the radiation in a background medium. This concept is illustrated by mapping the appropriately constrained equation $\mathbf{q} = \mathbf{k}_s - \mathbf{k}_i$ onto the Ewald diagram (Devaney, 2012), see Fig. 1. In this regime, the selectivity of the wave-matter interaction in the **q**-space (Lahiri *et al.*, 2009) permits, in principle, reconstruction of the full space-time dynamics of the structure, by varying both the radiation frequency and the propagation directions of the incident and scattered waves (Berne, Pecora, 1976; Wolf, 1996). Apart from the classical Dynamic Light Scattering (DLS) (Berne, Pecora, 1976) and Dynamic Sound Scattering (DSS) (Cowan *et al.*, 2016; Igarashi *et al.*, 2014; Kohyama *et al.*, 2009; Konno *et al.*, 2016) techniques, new modalities exploring the same weak scattering regime have been proposed recently. For example, Differential Dynamic Microscopy (DDM) may provide valuable information on the temporal dynamics of random media at different values (and directions) of vector **q** (Cerbino, Trappe, 2008; Giavazzi *et al.*, 2009; Reufer *et al.*, 2012).

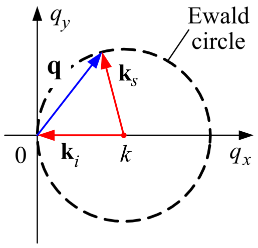

Fig. 1. Two-dimensional version of the Ewald construction. The points of the Ewald sphere for a given wave vector $\mathbf{k}_i$ determine all possible spectral components that could resonantly transform the incident wave into a scattered wave, with wave vector $\mathbf{k}_s$.

The condition of weak scattering imposes severe limitations on the disordered system and the wavelength of the radiation. Indeed, fluctuations in the scattering potential (for instance, the contrast of the constituent phases in composite media) must be very small, or the typical size ℓ_ε of the scatterers must be much smaller than the wavelength λ of the radiation applied to probe the structure. This precludes use of the corresponding methods in the study of densely packed systems beyond the limits of the Born approximation, i.e., in the regime of multiple scattering, when ℓ_ε is of the same order of magnitude as λ. On the other hand, probing the structures with multiply-scattered waves may be much more effective, since, in principle, they are sensitive to very small displacements in the medium. However, in order to realize this potential, we should have a rather accurate model relating the measured quantity, $\Gamma(\tau)$, to the statistics of the scattering medium.

Taking the multiple scattering into account is usually performed by completely changing the paradigm, namely by replacing the wave equation, as a basic tool, with a radiation transfer equation or its asymptotics, e.g., a diffusion model. One such technique, termed Diffusing Wave Spectroscopy (DWS), was proposed more than three decades ago, initially in optics (Maret, Wolf, 1987; Pine *et al.*, 1988; Weitz, Pine, 1993) and was extended later to acoustics (Cowan *et al.*, 2000) and seismology (Snieder, 2006). At present, applications of the DWS technique and its derivatives are not limited to the study of the dynamics of relatively simple structures, such as colloidal suspensions, but may be efficiently implemented, for example, in the functional imaging of brain activity (Li *et al.*, 2005).

Actually, DWS is a phenomenological theory, which models the transport of light as a random walk between scatterers. A partial ACF evaluated for a specific path length s is averaged over all possible values of s, with the photon (phonon) time-of-flight distribution $P(s)$ as a weighting factor. In the original version of the DWS theory, $P(s)$ is given by the isotropic diffusion model (Weitz, Pine, 1993). As is known, this model is unsuitable for describing light propagation along relatively short paths which, in turn, results in underestimating the field correlation for long times. Some attempts to correct the time-of-flight distribution $P(s)$ have been performed on the basis of solving the original version of the radiative transfer equation (Carminati *et al.*, 2004).

Nevertheless, the main problem of DWS is in the construction of the partial ACF itself, which is performed in a purely heuristic way, assigning to each specific path of length s a phase accumulated by the photon (phonon) on its way from one *point scatterer* to another. Hence, DWS provides us with a somewhat limiting amount of information. In contrast to the single scattering, inverting diffuse wave data permits estimation of only the mean square displacement of the scattering particle in time τ, instead of the full space-time dynamics described by the correlation function $B_\varepsilon(\boldsymbol{\rho}, \tau)$. Although the importance of *structural details* coupled with the wave dynamics was realized shortly after the DWS technique had been proposed (Nilsen, Gast, 1994; Ladd *et al.*, 1995; Stephen, 1988; MacKintosh, John, 1989), no essential progress in this direction has been observed so far.

In the present paper, we construct an original model of temporal correlations, which is based on first principles and therefore is free of the intrinsic limitations of the DWS phenomenology. At the same time, our model is *complementary* to both single scattering approximation and DWS, since it is designed to describe an intermediate, moderately strong (mesoscopic) scattering regime, in which the coherence of the wave process is still preserved.

Two main questions are explored here. First of all, the measured temporal ACF of the wave field is presented as a subtle integral transform of the spectral density $\Psi_\varepsilon(\mathbf{q}, \Omega)$, taking into account the multiple scattering effects. Second and more importantly, we show that by measuring the normalized ACF $\Gamma(\mathbf{k},\tau)$ as a function of both time delay τ and the wave vector $\mathbf{k}$, and applying a tomographic technique, it is possible to reconstruct (of course, only in a statistical sense) the full space-time dynamics of the scattering structure. Unlike all known wave tomography methods, the relevant $\mathbf{q}$-space spectral components of the scattering potential contributing to the measured field constitute a manifold of the same dimensionality as the ambient space. It will be shown, however, that the classical methods of integral geometry are still relevant here. This technique, tentatively called Correlation Wave Tomography (CWT), can be applied to the noninvasive study of density fluctuations in a variety of disordered systems.

The outline of the paper is as follows. The solution of the direct problem, i.e., finding the integral transform relating the temporal ACF $\Gamma(\mathbf{k},\tau)$ to the spectral density $\Psi_\varepsilon(\mathbf{q},\Omega)$, is presented in Sec. 2. The general solution obtained is analyzed in Sec. 3, especially for a statistically isotropic three-dimensional random medium. The concept of CWT based on the inversion of the integral transform is discussed in Sec. 4. Finally, the main results are summarized in Sec. 5.

2. Temporal correlation

Our starting point is the reduced Helmholtz equation,

$$\nabla^2 G(\mathbf{r},t) + k^2\left[1+\widetilde{\varepsilon}(\mathbf{r},t)\right]G(\mathbf{r},t) = -\delta(\mathbf{r}), \qquad (2)$$

written for a point source located in an unbounded, statistically homogeneous medium. In this expression, it is assumed implicitly that the medium is practically frozen during the typical time interval characterizing the wave-matter interaction (the so-called quasi-stationary approximation).

The formal solution of Eq. (2) is given in terms of the Feynman-Garrod path integral (Samelsohn, Mazar, 1996):

$$G(\mathbf{r},t) = \frac{i}{2k}\int_0^\infty \mathrm{d}\sigma \exp(ik\sigma/2)\int_{\mathbf{r}(0)=0}^{\mathbf{r}(\sigma)=\mathbf{r}} D\mathbf{r}(\xi) \cdot\exp\left[i\frac{k}{2}\int_0^\sigma \mathrm{d}\xi\left\{\dot{\mathbf{r}}^2(\xi)+\widetilde{\varepsilon}[\mathbf{r}(\xi),t]\right\}\right]. \qquad (3)$$

Here, wave number k contains an (infinitesimally small) positive imaginary part that enforces the radiation condition at infinity, and provides the convergence of the integral over pseudotime σ. The integration $\int D\mathbf{r}(\xi)$ in the continuum of all admissible paths is interpreted as the sum of contributions of arbitrary trajectories, over which a wave propagates from point $\mathbf{r}_0$ at $\sigma = 0$ to point $\mathbf{r}$ at the "moment" σ, and the expression in the exponent may be considered as an "action functional", which is related to the phase accumulated along the corresponding path.

Equation (3) is especially suitable for constructing statistical moments of the Green's function $G(\mathbf{r},t)$. Indeed, irrespective of the statistics of $\widetilde{\varepsilon}$ itself, the integral of $\widetilde{\varepsilon}$ along the path traversing many uncorrelated inhomogeneities, may be considered as a Gaussian random variable. As a result, the wave field correlator is presented in terms of the correlation function $B_\varepsilon(\boldsymbol{\rho},\tau)$ of the scattering potential. At the next step, as proposed by Samelsohn and Mazar (1996), the σ integral may be evaluated by the method of stationary phase, and the path integral replaced with a first cumulant approximation.

In the past, this procedure was verified for the statistical moments of first and second order. It was shown, in particular, that the mean field constructed in such a way coincides exactly with the Bourret approximation of the Dyson equation (Samelsohn, Mazar, 1996). The solution for the mean intensity, combined with the idea of spectral filtering, was used to describe the wave localization phenomenon in random media, with both isotropic and anisotropic disorder (Samelsohn *et al.*, 1999; Samelsohn, Freilikher, 2004). Also, the same path integral technique was employed to construct the two-frequency mutual coherence function (frequency field-field correlator) (Samelsohn, Freilikher, 2003; Samelsohn *et al.*, 2008). As is known, the Fourier transformation of this correlator leads to an impulse response function (time-of-flight distribution), measured for a short narrowband pulse (Ishimaru, 1978). To evaluate this impulse response, a cumulant expansion of a double path integral was applied (Samelsohn *et al.*, 2008). It was shown that even the first cumulant is capable of reproducing the two-scale structure of the coherence function, which corresponds to weakly and strongly scattered components of the wavefield, coexisting in the impulse response, even in the regime of multiple scattering.

It would be natural to extend this approach to the evaluation of the coherence function accounting for the temporal dynamics of the medium. In particular, here we evaluate the temporal ACF defined as

$$\Gamma(\mathbf{k},\tau) = \langle G(\mathbf{r},t)\; G^*(\mathbf{r},t+\tau)\rangle. \qquad (4)$$

Note that unlike the single-scattering approximation, we define the ACF for the total, not scattered, wavefield. In Eq. (4), the obvious fact is emphasized that this quantity should depend on the wave vector $\mathbf{k} = k\mathbf{r}/r$, directed along the line connecting the source with the observation point.

The generalization of the approach mentioned above to time-varying media is straightforward, and the calculations may be performed following the steps described in (Samelsohn *et al.*, 2008). Stationary phase approximation applied to the integrals over pseudotime and subsequent averaging lead to a double path integral of $\exp(-X)$, where the functional X has the form

$$X[\mathbf{r}_1(\xi),\mathbf{r}_2(\xi);\tau] = \frac{k^2}{8}\int_0^L \mathrm{d}\xi_1 \int_0^L \mathrm{d}\xi_2 \cdot [B_\varepsilon(\mathbf{r}_1(\xi_1)-\mathbf{r}_1(\xi_2),0) - 2B_\varepsilon(\mathbf{r}_1(\xi_1)-\mathbf{r}_2(\xi_2),\tau) + B_\varepsilon(\mathbf{r}_2(\xi_1)-\mathbf{r}_2(\xi_2),0)], \quad (5)$$

and $L = |\mathbf{r}|$ is the path length. Then, the cumulant expansion of the path integral is applied, where only the first term is kept. Omitting the details of the derivation, we concentrate on the final expression for the normalized ACF:

$$\gamma(\mathbf{k},\tau) \equiv \frac{\Gamma(\mathbf{k},\tau)}{\Gamma(\mathbf{k},0)} = \exp[-\chi(\mathbf{k},\tau)]. \quad (6)$$

The decrement $\chi(\mathbf{k},\tau)$ in the first cumulant approximation is calculated by replacing the correlation functions in Eq. (5) with their spectral expansions; see Eq. (36). This results in

$$\chi(\mathbf{k},\tau) = 2\int \mathrm{d}\Omega\,[1-\exp(-i\Omega\tau)]\,\widehat{\chi}(\mathbf{k},\Omega). \quad (7)$$

In its turn, $\hat{\chi}(\mathbf{k},\Omega)$ entering the latter equation is an integral transform of the spectral density,

$$\widehat{\chi}(\mathbf{k},\Omega) = k^3 L \int \mathrm{d}\mathbf{q}\, f(\mathbf{q},\mathbf{k})\,\Psi_\varepsilon(\mathbf{q},\Omega), \quad (8)$$

with kernel $f(\mathbf{q},\mathbf{k})$ of the form

$$f(\mathbf{q},\mathbf{k}) = (4kL)^{-1}\int_0^L \mathrm{d}\xi_1 \int_0^L \mathrm{d}\xi_2 \exp\left[\frac{i(\xi_1-\xi_2)\,\mathbf{q}\cdot\mathbf{k}}{k}\right] \cdot \exp\left\{-i\frac{[\xi_1(1-\xi_1/L)-\xi_2(1-\xi_2/L)]\,q^2}{2k}\right\}. \quad (9)$$

Note that a finite value of the ACF for $\tau\to\infty$ is due to a contribution of the mean field (the latter is, of course, exponentially small in the mesoscopic scattering regime we are interested in here).

To simplify the expressions for the filtering function, we introduce a new pair of integration variables,

$$\Xi = (\xi_1+\xi_2)/2, \qquad \xi = \xi_1 - \xi_2. \quad (10)$$

Then, the integration over Ξ is performed exactly,

$$f(\mathbf{q},\mathbf{k}) = K^{-2}\int_0^L \mathrm{d}\xi\,\xi^{-1}\cos\left(\frac{\xi\,\mathbf{q}\cdot\mathbf{k}}{k}\right) \cdot \sin\left[\frac{\xi(1-\xi/L)\,q^2}{2k}\right]. \quad (11)$$

The solution obtained is valid far from the source, under the conditions of weak to moderate scattering (Samelsohn *et al.*, 2008). Its detailed analysis in the mesoscopic regime is performed in Sec. 3.

3. Analysis

Further simplification of the filtering function (11) could be achieved if we increase the path length by setting $L\to\infty$. Mathematically, this is possible due to the factor $\cos(\xi\mathbf{q}\cdot\mathbf{k}/k)$, which becomes highly oscillatory for large ξ. Thus, we neglect the contributions of the ξ/L term with respect to unity, and extend the upper limit of the integral in Eq. (11) to infinity. By changing also the integration variable as $\xi = 2kt/q$, and using the integral representation of the Heaviside unit step function,

$$\vartheta(x-a) = \frac{2}{\pi}\int_0^\infty \mathrm{d}t\,t^{-1}\sin(xt)\cos(at), \qquad a\geq 0, \quad (12)$$

we finally obtain

$$\widehat{\chi}(\mathbf{k},\Omega) = \frac{\pi}{8}k^3 L\int \mathrm{d}\mathbf{q}\,q^{-2}\vartheta\left(q - \left|\frac{2\mathbf{k}\cdot\mathbf{q}}{q}\right|\right)\Psi_\varepsilon(\mathbf{q},\Omega). \quad (13)$$

In fact, the procedure of deriving the latter equation can be justified only if the parameter $L\mathbf{q}\cdot\mathbf{k}/k$ is large. This condition is obviously violated for directed (forward-scattered) waves, where vector $\mathbf{q}$ is not only small, but is also perpendicular to $\mathbf{k}$. Here, however, we explore another situation: the radiation wavelength is comparable with the correlation scale of the disorder, and the wave undergoes large angle scattering. In this case, not too much energy of the spectrum $\Psi_\varepsilon(\mathbf{q},\Omega)$ is concentrated near the origin of the $\mathbf{q}$-space, and therefore the fraction of ballistic photons (phonons) contributing to the registered field is vanishingly small.

It is worth noting that, as for the single-scattering approximation, Eq. (13) is easily mapped onto the Ewald construction (see Fig. 2), but is *not local* here. Indeed, within the Born approximation, only one Bragg lattice of the spectral density, $\Psi_\varepsilon(\mathbf{k}_s-\mathbf{k}_i,\Omega)$,

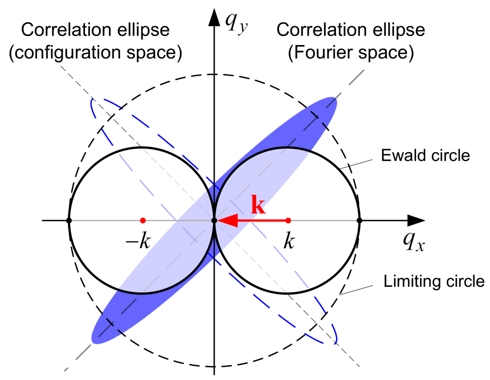

Fig. 2. Spectral density $\Psi_\varepsilon(\mathbf{q},\Omega)$ of a random medium for fixed Ω is shown schematically as an ellipse in both configuration and Fourier space. The major axis of the ellipse in configuration space corresponds to the direction of higher correlation.

contributes to the scattered field. In contrast, in order to evaluate the decrement $\widehat{\chi}(\mathbf{k},\Omega)$ in our solution for fixed values of $\mathbf{k}$ and Ω, we should integrate the spectral density $\Psi_\varepsilon(\mathbf{q},\Omega)$ outside the eight curve formed by the two Ewald spheres, with a weighting factor q^{-2}.

Let us illustrate the behavior of the decrement for three-dimensional (3D), statistically isotropic media, where spectral density $\Psi_\varepsilon(\mathbf{q},\Omega)$ depends only on the absolute value of vector $\mathbf{q}$. After integrating over the angular variables, Eq. (13) becomes

$$\widehat{\chi}(k,\Omega) = \frac{\pi^2}{4}k^2 L\int_0^\infty \mathrm{d}q\,\min(q,2k)\,\Psi_\varepsilon(q,\Omega). \tag{14}$$

There are many systems the dynamics of which is described by different models, as for instance polydisperse colloids, interacting scatterers, particles dispersed in viscoelastic complex fluids, and convective motions, including turbulence in fluids, etc. If $\Psi_\varepsilon(\mathbf{q},\Omega)$ is a non-separable function, the temporal dynamics depends on the details of the spectrum behavior in the $\mathbf{q}$-space. Indeed, it is rather natural, for example, that small inhomogeneities (with high spatial frequencies q) are less correlated in time, i.e., they change their form, orientation, or position more rapidly than the large-scale perturbations. Apart from the intrinsic evolution of the structure (as it is observed, e.g., in a turbulent medium), other reasons (diffusion or flow) also lead to strong space-time coupling.

Instead of going into the analysis of a variety of possible physical models, we will consider a simple example of the temporal evolution satisfying some general conditions. First of all, we assume that the static correlation function $B_\varepsilon(r)$ is of a Gaussian form,

$$B_\varepsilon(r) = \sigma_\varepsilon^2\exp\left(-\frac{r^2}{\ell_\varepsilon^2}\right), \tag{15}$$

with correlation length ℓ_ε and variance σ_ε^2. In the 3D case, this corresponds to the power spectrum

$$\Phi_\varepsilon(q) = \left(2\sqrt{\pi}\right)^{-3}\sigma_\varepsilon^2\,\ell_\varepsilon^3\exp\left(-\frac{\ell_\varepsilon^2 q^2}{4}\right). \tag{16}$$

Although the Gaussian function is not related directly to a specific physical mechanism responsible for the formation of a heterogeneous medium, and further cannot even correspond to any two-phase random medium, it is an effective mathematical model widely used to characterize wave propagation in a broad class of random media, when the exact form of $B_\varepsilon(r)$ is not known, or to perform qualitative analysis (Ishimaru, 1978).

Second, the most attractive model for the normalized spectral density $\psi_\varepsilon(q,\Omega)$ that conforms to these requirements mentioned above is

$$\psi_\varepsilon(q,\Omega) = \frac{1}{\sqrt{2\pi}\,\Omega_c(q)}\exp\left[-\frac{\Omega^2}{2\,\Omega_c^2(q)}\right], \tag{17}$$

where $\Omega_c(q)$ is the characteristic evolution frequency of inhomogeneities with linear scale q^{-1}. For example, in a random flow with isotropic Gaussian velocity fluctuations $\Omega_c(q) \sim q^2$, while in turbulent media the dynamics is characterized by a fractal-type time dependence $\Omega_c(q) \sim q^{2/3}$, at least within the inertial interval.

For simplicity, we adopt here the square law model, which results in

$$\chi(\kappa,\tau) = \frac{\sqrt{\pi}}{2}\sigma_\varepsilon^2\left(\frac{L}{\ell_\varepsilon}\right)\left[A(\kappa,1) - A\left(\kappa,\sqrt{1+\frac{\tau^2}{\tau_c^2}}\right)\right]. \tag{18}$$

Here, $\kappa = k\ell_\varepsilon$ is the normalized wave number, τ_c is the characteristic time, the auxiliary function $A(\kappa,x)$ is given by

$$A(\kappa,x) = \left(\frac{\kappa}{x}\right)^2\left[1-\exp\left(-\kappa^2x^2\right)+\sqrt{\pi}\,\kappa x\,\mathrm{erfc}(\kappa x)\right], \tag{19}$$

and $\mathrm{erfc}(x)$ denotes the complementary error function. The behavior of $\chi(\kappa,\tau)$ is illustrated in Fig. 3. It is seen that the dynamics of the medium depends essentially on the wave number.

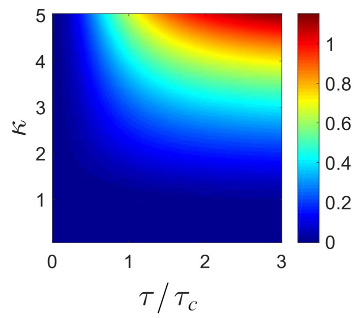

Fig. 3. The normalized decrement $\chi(\kappa,\tau)/\chi(1,\tau_c)$ shown as a function of delay time τ and normalized wave number κ.

Further analysis of the solution obtained for the temporal ACF leads to two main conclusions. First, for fixed $\mathbf{k}$, the information content of cumulant $\chi(\mathbf{k},\tau)$ is, in a sense, similar to the corresponding ACF decrement in the classical DWS theory. In fact, $\chi(\mathbf{k},\tau)$ may be interpreted as a (half of the) *structure function* of the phase fluctuations in the measured field. Moreover, there is another fact that makes our approach very similar to the DWS technique: both provide us with only a $\mathbf{q}$-weighted average, in striking contrast to the weak scattering experiments, where the registered quantity is $\mathbf{q}$-selective. Although much more sensitive to small displacements, the multiple scattering and associated $\mathbf{q}$-averaging effect make it impossible to directly invert the measured decrement for information on the dynamics of the structure at a specific value of $\mathbf{q}$.

The second important conclusion is that the temporal ACF $\gamma(\mathbf{k},\tau)$ depends essentially on the wave number k, or more precisely, on wave vector $\mathbf{k}$, when the random medium is statistically anisotropic. Hence, considering wave vector $\mathbf{k}$ as a free parameter that can vary at will, we provide an additional dimension to the data, which may result in a *tomographic-type reconstruction* of the space-time dynamics, rather than a plain spectroscopic technique. We will explore this idea in Sec. 4.

4. Inversion

As was conjectured in Sec. 3, in order to make the inversion, we must measure the ACF, or simply its decrement $\chi(\mathbf{k},\tau)$, for waves of different frequencies and a variety of angular orientations of the structure with respect to the straight line connecting the source with the observation point. Indeed, to recover the spectral density $\Psi_\varepsilon(\mathbf{q},\Omega)$, we should perform the two-step procedure set forth below.

First, we differentiate both sides of Eq. (7) with respect to τ and invert the resulting Fourier transform, yielding

$$\widehat{\chi}(\mathbf{k},\Omega) = \frac{1}{2\pi\Omega}\int_0^\infty \mathrm{d}\tau\ \sin(\Omega\tau)\,\chi'(\mathbf{k},\tau). \tag{20}$$

The integral in Eq. (20) is convergent, if the derivative $\chi'(\mathbf{k},\tau)$ approaches zero for $\tau\to\infty$, and also satisfies the condition $\lim_{\tau\to 0}\tau^2\chi'(\mathbf{k},\tau)=0$. This is compatible, for instance, with a fractal τ^α time dependence, when $\alpha<1$. For $1\le\alpha<2$ we should differentiate twice, which leads to another inversion algorithm, see (Rytov *et al.*, 1989). In principle, this step may be unnecessary, if our goal is to directly recover a structure function of the disorder; see Eqs. (29), (34), and the related discussions below.

At the second step, having at hand $\widehat{\chi}(\mathbf{k},\Omega)$, which can now be considered as a function of $\mathbf{k}$ for each fixed value of Ω, we have to invert the integral transform (13). At this point, it is worthwhile to recall that in classical X-ray tomography, the integration is performed over planes (or along straight lines in the 2D case), the operation that constitutes the well-known Radon transform (Natterer, Wübbeling, 2001). In this case, the submanifold that represents the integration domain has dimensionality of one less than that of the ambient manifold, over which the unknown function is defined. For the tomographic modalities based on the so-called soft field (say, diffuse optical tomography), where the influence domain constitutes a kind of blurred banana-like region connecting the source and receiver in the configuration space (Gibson *et al.*, 2005), analytical inversion based on the integral geometry transforms is hardly to be expected (see, however, the recent studies by Samelsohn (2016; 2017; 2018), where an efficient Radon-to-Helmholtz mapping has been proposed). Since the multiple scattering effects are taken into account, our situation is not exceptional in that sense, but surprisingly, integral transform (13) can be reduced to an operator that is invertible. In fact, performing the integration by parts (with respect to the absolute value of the vector $\mathbf{q}$) in Eq. (13), we arrive at

$$\widehat{\chi}(\mathbf{k},\Omega) = \frac{\pi}{8}k^3L\int \mathrm{d}\mathbf{q}\,\delta\left(q-\left|2\mathbf{k}\cdot\frac{\mathbf{q}}{q}\right|\right)F_\varepsilon(\mathbf{q},\Omega), \tag{21}$$

where we have introduced an auxiliary function

$$F_\varepsilon(\mathbf{q},\Omega) = q^{1-m}\int_q^\infty \mathrm{d}q\,q^{m-3}\Psi_\varepsilon(\mathbf{q},\Omega) \tag{22}$$

(here m is the dimensionality of the problem). In order to find the spectral density $\Psi_\varepsilon(\mathbf{q},\Omega)$, provided the auxiliary function is known, we multiply Eq. (22) by q^{m-1} and then differentiate, which leads to

$$\Psi_\varepsilon(\mathbf{q},\Omega) = -q^{3-m}\frac{\mathrm{d}}{\mathrm{d}q}\left[q^{m-1}F_\varepsilon(\mathbf{q},\Omega)\right]. \tag{23}$$

The spectral density $\Psi_\varepsilon(\mathbf{q},\Omega)$ and then $F_\varepsilon(\mathbf{q},\Omega)$ are both even functions, so that we can keep the integration over only one Ewald sphere and double the result. Therefore, Eq. (21) is nothing other than a spherical mean of the function $F_\varepsilon(\mathbf{q},\Omega)$ for a family of spheres containing the origin (Cormack, Quinto, 1980). These are just the Ewald spheres constructed for different frequencies and propagation directions (the information contained actually in the wave vector $\mathbf{k}$). The spherical mean operator is known to be invertible (Cormack, Quinto, 1980). In particular, it may be converted into the classical Radon transform by using a geometric inversion of the $\mathbf{q}$-space with respect to a reference sphere centered at the origin (Yagle, 1992; Samelsohn, 2009).

Although in the general case the inversion procedure is rather involved, for statistically isotropic 3D media, the final results are unexpectedly simple. Indeed, after integrating over the angular variables, integral transform (21) is reduced to

$$\widehat{\chi}(k,\Omega) = \frac{\pi^2}{4}k^2L\int_0^{2k}\mathrm{d}q\,q^2F_\varepsilon(q,\Omega), \tag{24}$$

so that the inversion becomes straightforward,

$$F_\varepsilon(2k,\Omega) = \frac{1}{2\pi^2k^2L}\partial_k\left[k^{-2}\widehat{\chi}(k,\Omega)\right], \tag{25}$$

where ∂_k means the derivative with respect to k. Then, as follows from Eq. (23),

$$\Psi_\varepsilon(2k,\Omega) = -\frac{1}{\pi^2L}\partial_k^2\left[k^{-2}\widehat{\chi}(k,\Omega)\right]. \tag{26}$$

Note that for isotropic media, we can avoid calculating the auxiliary function and directly arrive at Eq. (26). Indeed, if we divide Eq. (14) by k^2 and then differentiate, we have

$$\partial_k\left[k^{-2}\widehat{\chi}(k,\Omega)\right] = \frac{\pi^2}{2}L\int_{2k}^\infty \mathrm{d}q\,\Psi_\varepsilon(q,\Omega). \tag{27}$$

The second differentiation leads to Eq. (26).

Finally, converting Eq. (26) into the time domain,

$$D_\varepsilon(2k,\tau) = 2\int_{-\infty}^{\infty} \mathrm{d}\Omega\,[1-\exp(i\Omega\tau)]\,\Psi_\varepsilon(2k,\Omega), \quad (28)$$

we obtain the temporal structure function taken at a specific value of the spatial wave number (namely, at $q = 2k$):

$$D_\varepsilon(2k,\tau) = -\frac{1}{\pi^2 L}\partial_k^2\left[k^{-2}\chi(k,\tau)\right]. \quad (29)$$

By definition, $D_\varepsilon(\mathbf{q},\tau)$ characterizes how quickly the contribution of spatial harmonic $\mathbf{q}$ is changed in time. It is reminiscent of the "image structure function" used within the framework of DDM (Cerbino, Trappe, 2008). However, the DDM approach implies that the $\mathbf{q}$-information is captured directly (via Fourier transformation of the two-dimensional differential images). This setup is easily realized in optics, but is hardly possible in acoustics. Our solution permits probing the structure by recording the signal in one point only but sweeping the wavenumber (frequency) of the radiation used.

As follows from Eq. (29), to recover the spectral density, we need to differentiate the noisy sampled data twice. As is known, calculating the derivative is a numerically unstable operation. In principle, we can avoid the evaluation of one derivative by converting the spectral density $\Psi_\varepsilon(\mathbf{q},\Omega)$ into a true structure function $D_\varepsilon(\boldsymbol{\rho},\tau)$ at the outset, and then applying an integration by parts.

Let us illustrate this approach for a statistically isotropic 3D structures. In this situation, the spectral expansion (38) of the structure function, after integration over angular variables, becomes

$$D_\varepsilon(\boldsymbol{\rho},\tau) = 8\pi\int_{-\infty}^{\infty}\mathrm{d}\Omega\int_0^{\infty}\mathrm{d}q\,q^2 \cdot [1-\exp(-i\Omega\tau)\,\mathrm{sinc}(q\rho)]\,\Psi_\varepsilon(q,\Omega), \quad (30)$$

where $\mathrm{sinc}(x) = \sin(x)/x$. Changing the integration variable $q = 2k$ in the latter equation, we substitute the solution (26) for the spectral density $\Psi_\varepsilon(2k,\Omega)$, and arrive at

$$D_\varepsilon(\boldsymbol{\rho},\tau) = -\frac{64}{\pi L}\int_{-\infty}^{\infty}\mathrm{d}\Omega\int_0^{\infty}\mathrm{d}k\,k^2 \cdot [1-2\exp(-i\Omega\tau)\,\mathrm{sinc}(2k\rho)] \cdot \partial_k^2\left[k^{-2}\widetilde{\chi}(k,\Omega)\right]. \quad (31)$$

As follows from Eq. (27), for any reasonable spectral density $\Psi_\varepsilon(q,\Omega)$, the first derivative of $k^{-2}\chi$ approaches zero faster than k^{-2} as k tends to infinity, such that the integration by parts gives

$$D_\varepsilon(\boldsymbol{\rho},\tau) = \frac{128}{\pi L}\int_{-\infty}^{\infty}\mathrm{d}\Omega\int_0^{\infty}\mathrm{d}k\,k \cdot [1-\exp(-i\Omega\tau)\,g(\rho,k)] \cdot \partial_k\left[k^{-2}\widetilde{\chi}(k,\Omega)\right], \quad (32)$$

where

$$g(\boldsymbol{\rho},k) = \cos(2k\rho) + \mathrm{sinc}(2k\rho). \quad (33)$$

The integration over Ω may also be performed, and the unknown structure function is then expressed directly via the measured quantity $\chi(k,\tau)$:

$$D_\varepsilon(\boldsymbol{\rho},\tau) = \frac{64}{\pi L}\int_0^{\infty}\mathrm{d}k\,k\left\{[1-g(\rho,k)]\,\partial_k\left[k^{-2}\chi(k,\infty)\right] + g(\rho,k)\,\partial_k\left[k^{-2}\chi(k,\tau)\right]\right\}. \quad (34)$$

Unfortunately, the value of $k^{-2}\chi(k,\tau)$ is not zero at infinity, and we cannot use the same trick as has been applied to Eq. (31). Anyway, the kernel of the integral operator that arises here is an oscillatory function, and therefore appropriate interpolation and smoothing of raw data should be a critically important ingredient of the CWT inversion, irrespective of the domain (configuration or Fourier) in which the inversion is performed.

In conclusion, one important remark is in order. Our analysis is based on the simplest model, Eq. (2), which describes propagation in a medium with variable wave velocity. Density fluctuations have been neglected. In principle, the latter can be easily included in consideration, if we use an appropriate substitution for the acoustic pressure (Bergmann, 1946). The problem is then reduced again to Eq. (2), but with a new, generalized, potential $\widetilde{\varepsilon}(\mathbf{r},t)$ that depends now on both sound velocity and the density of the medium. Thus, the solution of the direct problem, i.e., finding the coherence function for a given ACF of the medium, remains intact. However, a new situation arises if we try to make the inversion, since the generalized potential will be frequency-dependent. Likewise, the spectral dependence of the decrement $\chi(k,\tau)$ can be altered by the effects of frequency dispersion of materials constituting the random medium. Overall, these phenomena may lead to artifacts in the reconstruction of space-time dynamics, and the subject deserves further research.

5. Summary

In this paper, we have developed a new concept of probing the space-time dynamics of time-varying random structures. The solution of the direct problem has a clear physical interpretation. In contrast to the DWS technique, our results have been derived from first principles, without using any phenomenological parameters, such as a diffusion constant. Our theory of temporal correlation is valid for situations where the size of the scattering system is comparable to the transport mean-free-path, i.e., it is suitable for filling the gap between a ballistic regime, on the one hand, and a diffusion regime, on the other. The better accuracy is predicted to occur for long and intermediate time scales (relatively short paths), where diffusion approximation fails to describe the wave transport correctly.

Concerning the inversion, the CWT concept extends the conventional diffraction tomography technique to the mesoscopic scattering regime. Indeed, we have arrived at the tomographic-type reconstruction based, after all, on a classical Radon transform. In principle, the CWT technique permits recovery of the full space-time dynamics of the structure under study. The disordered systems are not required to be composed of identical particles; statistically anisotropic and fractal density fluctuations can be probed equally well, with no *a priori* information needed. The CWT technique may find applications in various areas of soft condensed matter science, structure analysis, and biomedical research, to name a few.

Appendix

For a statistically homogeneous random field $\widetilde{\varepsilon}(\mathbf{r},t)$, the correlation function is defined as

$$B_\varepsilon(\boldsymbol{\rho},\tau) = \langle \widetilde{\varepsilon}(\mathbf{r},t)\,\widetilde{\varepsilon}(\mathbf{r}+\boldsymbol{\rho},t+\tau)\rangle . \qquad (35)$$

Its spectral expansion is of the form

$$B_\varepsilon(\boldsymbol{\rho},\tau) = \int \mathrm{d}\mathbf{q}\int \mathrm{d}\Omega \exp\left[i\left(\mathbf{q}\cdot\boldsymbol{\rho}-\Omega\tau\right)\right]\Psi_\varepsilon(\mathbf{q},\Omega). \qquad (36)$$

Note that hereafter we follow the Fourier transform convention adopted in the book by Rytov *et al.* (1989).

The locally homogeneous random field can be characterized by a structure function

$$D_\varepsilon(\boldsymbol{\rho},\tau) = \left\langle\left[\widetilde{\varepsilon}(\mathbf{r}+\boldsymbol{\rho},t+\tau)-\widetilde{\varepsilon}(\mathbf{r},t)\right]^2\right\rangle . \qquad (37)$$

The spectral expansion of the structure function is given by

$$D_\varepsilon(\boldsymbol{\rho},\tau) = 2\int \mathrm{d}\mathbf{q}\int \mathrm{d}\Omega \left\{1-\exp\left[i\left(\mathbf{q}\cdot\boldsymbol{\rho}-\Omega\tau\right)\right]\right\}\Psi_\varepsilon(\mathbf{q},\Omega). \qquad (38)$$

For a homogeneous medium, the structure function is related to the correlation function by the obvious relation

$$D_\varepsilon(\boldsymbol{\rho},\tau) = 2\left[B_\varepsilon(0,0)-B_\varepsilon(\boldsymbol{\rho},\tau)\right]. \qquad (39)$$

Moreover, if $B_\varepsilon(\infty,\infty) = 0$ then $D_\varepsilon(\infty,\infty) = 2B_\varepsilon(0,0)$, and we arrive at the inverse relation

$$B_\varepsilon(\boldsymbol{\rho},\tau) = \frac{1}{2}\left[D_\varepsilon(\infty,\infty)-D_\varepsilon(\boldsymbol{\rho},\tau)\right]. \qquad (40)$$

Integration of $\Psi_\varepsilon(\mathbf{q},\Omega)$ over all frequencies Ω recovers a static spatial spectrum $\Phi_\varepsilon(\mathbf{q})$ of the random medium, while the integration over all $\mathbf{q}$-space reduces $\Psi_\varepsilon(\mathbf{q},\Omega)$ to a temporal spectrum $S_\varepsilon(\Omega)$, measured at a fixed point in the configuration space. Note that only for separable spectra,

$$\Psi_\varepsilon(\mathbf{q},\Omega) = \Phi_\varepsilon(\mathbf{q})\,S_\varepsilon(\Omega), \qquad (41)$$

the temporal and spatial statistics are decoupled.

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