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Temporal fluctuations in wave propagation in random media

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The effect of time-dependent fluctuations of a medium on the spectral intensity and intensity fluctuations of light scattered from the medium is discussed. In the case that the light is multiply scattered from the medium, it is possible in the diffusion approximation to relate the spectral intensity in the scattered wave to the angular-average structure factor of the medium. The relaxation time depends on the number of scattering events, i.e., on the lengths of the multiple-scattering paths, and the scattered intensity thus exhibits a broad range of relaxation times. The spectral intensity in coherent backscattering is also discussed.

I. INTRODUCTION

There has been considerable interest recently in the propagation, multiple scattering, and localization of waves in random media, with the observation of weak localization effects[1–3] and large intensity fluctuations[4] in light scattering. There has also been theoretical work on deriving the coherent backscattering intensity and fluctuations from the wave equation. Coherent backscattering has been discussed by Golubentsev,[5] Akkermans, Wolf, and Maynard,[6] and Stephen and Cwilich,[7] and intensity fluctuations have been discussed by Shapiro[8] and Stephen and Cwilich.[9] A large literature on the subject of multiple scattering exists and for reviews of some of this work we refer to Ishimaru[10] and Goodman.[11]

In this paper we investigate how the dynamical properties of the scattering medium determine the spectral properties of the scattered intensity and the intensity fluctuations. Often it is assumed that the scattering is weak and can be treated in the Born approximation. In this case the scattering is directly related to the dynamical structure factor of the medium. Here we are interested in the case in which the wave is multiply scattered in the medium, which will occur if the mean free path of the wave in the medium is much less than its dimensions. Owing to the multiple scattering the scattered intensity will not depend in an important way on the scattering angle but its spectral properties will be determined by the dynamics of the medium. The total scattered intensity is obtained by summing the contributions from all possible multiple-scattering paths. The relaxation time depends on the number of scattering events, i.e., on the length of the multiple-scattering path. The total intensity, therefore, contains a broad range of relaxation times. The sum over all diffusion paths also depends on the shape and size of the medium. The effect of particle dynamics on coherent backscattering has been discussed by Golubentsev.[5] Here we adopt the same model but we are concerned with the time dependence of the direct part of the scattering. A short discussion of backscattering is included for completeness. Recently Maret and Wolf[12] have measured the time autocorrelation function of the light intensity multiply scattered from an aqueous suspension of diffusing polystyrene spheres. They observed a fast decay of coherence which they interpreted as resulting from the short relaxation times associated with long diffusion paths.

II. COHERENCE FUNCTION

We consider a scalar field $E(\mathbf{r},t)$ propagating in a random medium which obeys a wave equation

$$\left[\nabla^2-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}[1+\epsilon(\mathbf{r},t)]\right]E(\mathbf{r},t)=0\ . \tag{2.1}$$

The random part of the dielectric constant ϵ has zero mean and a correlation function

$$\frac{\omega_0^4}{c^4}\langle\epsilon(\mathbf{r},t)\epsilon(\mathbf{r}',t')\rangle=C(\mathbf{r}-\mathbf{r}',t-t')\ , \tag{2.2}$$

where ω_0 is the frequency of the light. If it is assumed that the scattering is weak and can be treated in the Born approximation, the scattered intensity is related to the Fourier transform of the correlation function $C(\mathbf{q},\omega)$ where $\mathbf{q}$ and ω are the momentum and frequency transfer, respectively. $C(\mathbf{q},\omega)$ is often related to the density correlation function of the medium. In this paper we are interested in the case in which the wave is multiply scattered many times in the medium. The intensity of the diffusely scattered light will not depend in an important way on the scattering angle, but its time dependence will be determined by the properties of the medium. We investigate here how the spectral properties of the scattered intensity are related to the dynamic properties of the medium contained in $C(\mathbf{r},t)$.

It is useful to consider some simple examples. (a) The scatterers of mass m, polarizability α, and density n have a Maxwell-Boltzmann velocity distribution ($\beta=1/kT$)

$$C^{(a)}(\mathbf{q},t)=\Delta e^{-t^2q^2/2m\beta}\ , \tag{2.3}$$

where

$$\Delta=(4\pi\alpha)^2 n(\omega_0/c)^4\ .$$

(b) The scatterers diffuse with diffusion constant D_i,

$$C^{(b)}(\mathbf{q},t)=\Delta e^{-q^2D_i|t|}\ . \tag{2.4}$$

In general we assume that the scatterers have an average velocity v which is small compared with the wave velocity c. The frequency change on scattering is small ($\sim v/c\omega_0$) and we can consider the propagation of an almost-monochromatic wave with frequency close to the value $\omega_0=ck_0$. The mean free path of the wave l will not depend on the motion of the scatterers and is determined by the density of scatterers, $l=4\pi/\Delta$.

The quantity of interest is the correlation function of the scattered field

$$\Gamma(\mathbf{r}_1,\mathbf{r}_2,t)=\langle E(\mathbf{r}_1,t)E^*(\mathbf{r}_2,0)\rangle_c \tag{2.5}$$

averaged over the fluctuations in the medium. This is defined as a cumulant (subscript c) so that the coherent part of the incident field $\langle E(\mathbf{r},t)\rangle=E_c(\mathbf{r},t)$ is omitted. We introduce center of mass $\mathbf{R}=(\mathbf{r}_1+\mathbf{r}_2)/2$ and relative coordinates $\mathbf{r}=\mathbf{r}_1-\mathbf{r}_2$ in (2.5) giving $\Gamma(\mathbf{R},\mathbf{r},t)$. On taking the Fourier transform with respect to $\mathbf{r}$ and t we obtain the spectral density or coherence function[10] $\Gamma(\mathbf{R},\mathbf{q},\omega)$. This has the meaning of the energy density of the scattered field at $\mathbf{R}$ with wave vector $\mathbf{q}$ and frequency ω. The total scattered energy density at $\mathbf{R}$ is $\Gamma(\mathbf{R},r=0,t=0)$ and its spectral density is $\Gamma(\mathbf{R},r=0,\omega)$. We will mainly be concerned with the total scattered energy density $\Gamma(R,r=0,t)=\Gamma(R,t)$.

Experimentally it is often more convenient to measure the intensity correlation function

$$\Gamma_2(R,t)=\langle\, |E(R,t)|^2\,|E(R,0)|^2\rangle_c\ . \tag{2.6}$$

In the leading approximation $k_0l>1$ this correlation function factorizes[9] and is determined by $\Gamma(R,t)$:

$$\Gamma_2(R,t)=\Gamma(R,t)\Gamma^*(R,t)\ . \tag{2.7}$$

This function thus contains the same information as $\Gamma(R,t)$.

We now make the important assumption that the propagation of the light in the medium is diffusive. This is valid provided $\lambda\ll l\ll L$ where λ is the light wavelength, l is the light mean free path, and L is a characteristic dimension of the medium. It is also assumed that the Fourier-transformed structure factor $C(\mathbf{q},t)$ does not vary rapidly with q, i.e., the scatterers have dimensions smaller than λ. When $\lambda/l\ll 1$ the leading approximation to Γ is provided by the sum of the ladder diagrams with the result

$$\Gamma(1,1')=\int D(1-2)D^*(1'-2')L(22'33')\times E_c(3)E_c^*(3')d2\,d2'\,d3\,d3'\ , \tag{2.8}$$

where $1=(\mathbf{r}_1,t_1)$, etc., $D(\mathbf{r},t)$ is the average Green's function of (2.1),

$$D(\mathbf{r},\omega)=\frac{1}{4\pi r}e^{(ik_0-1/2l)r}\ , \tag{2.9}$$

and L is the ladder propagator. In this propagator we also introduce center of mass and relative coordinates

$$L(11'22')=L(\mathbf{R}T,\boldsymbol{\rho}_1\tau_1,\boldsymbol{\rho}_2\tau_2)\ , \tag{2.10}$$

where $\mathbf{R}=\frac{1}{2}(\mathbf{r}_1+\mathbf{r}_1'-\mathbf{r}_2-\mathbf{r}_2')$, $\boldsymbol{\rho}_1=\mathbf{r}_1-\mathbf{r}_1'$, $\boldsymbol{\rho}_2=\mathbf{r}_2-\mathbf{r}_2'$, and similarly for the time variables. The calculation of L is discussed in the Appendix. In the diffusion approximation the Fourier transform of L, i.e., $L(\boldsymbol{\kappa}\Omega,\mathbf{q}_1\omega_1,\mathbf{q}_2\omega_2)$ is independent of the directions of $\mathbf{q}_1$ and $\mathbf{q}_2$ and depends on $\omega_1-\omega_2$. Then setting $q_1=q_2=k_0$ and $\Omega=0$ we find

$$L(\kappa\Omega=0,k_0\omega_1,k_0\omega_2)=L(\kappa,\omega_1-\omega_2)\ , \tag{2.11}$$

with

$$L(\kappa,t)=\frac{12\pi/l}{F^2(t)+\kappa^2l^2}\ , \tag{2.12}$$

where $F^2(t)=3[1-f(t)]/f(t)$ and $f(t)$ is the angular average[5] of the correlation function (2.1),

$$f(t)=\frac{l}{(4\pi)^3}\int ds\,ds' C(k_0(\mathbf{s}-\mathbf{s}'),t)\ , \tag{2.13}$$

where $\mathbf{s}$ and $\mathbf{s}'$ are unit vectors. In the two examples (2.3) and (2.4) (Ref. 5)

$$f^{(a)}(t)=\frac{\tau_\lambda^2}{t^2}(1-e^{-t^2/\tau_\lambda^2})\ ,$$

$$f^{(b)}(t)=\frac{\tau_\lambda}{|t|}(1-e^{-|t|/\tau_\lambda})\ , \tag{2.14}$$

where τ_λ is the time for a scatterer to move a wavelength, i.e., $\tau_\lambda^{(a)}=(m\beta/2k_0^2)^{1/2}$, $\tau_\lambda^{(b)}=1/4k_0^2D_i$. We now consider the scattered intensity in two geometries.

A. Point source of waves

A point source of waves of unit strength is at the origin of an infinite medium. The coherent field is

$$E_c(\mathbf{r},t)=D(\mathbf{r},\omega_0)e^{-i\omega_0 t}\ .$$

When this is substituted in (2.8) together with (2.9) and (2.12) the spectral density at $R>l$ is

$$\Gamma(R,t)=I(R)e^{-(R/l)F(t)-i\omega_0 t}\ , \tag{2.15}$$

where $I(R)=3/(4\pi)^2 lR$ is the total scattered intensity. For static impurities $F=0$ and this reduces to Shapiro's[8] result. For $t<\tau_\lambda$, $F^2(t)=3t^2/2\tau_\lambda^{(a)2}$ or $3\,|\,t\,|\,/2\tau_\lambda^{(b)}$ in the two cases (2.14), and the spectral density has a frequency width $\sqrt{3}R/\sqrt{2}l\tau_\lambda^{(a)}$ or $3R^2/2l^2\tau_\lambda^{(b)}$, respectively. As the distance R from the source increases, the lengths of the diffusion paths increase leading to a larger spectral width. In case (a)

$$\Gamma_2(R,t)\sim\exp(-\sqrt{6}R\,|\,t\,|\,l\tau_\lambda^{(a)}) \tag{2.16}$$

and the spectral distribution is a Lorentzian of width $\sqrt{6}R/l\tau_\lambda^{(a)}$. In the single-scattering approximation it would be Gaussian (2.3). In case (b)

$$\Gamma_2(R,t)\sim\exp[-(R/l)(6\,|\,t\,|\,/\tau_\lambda^{(b)})^{1/2}] \tag{2.17}$$

and is a stretched exponential. This is a consequence of the distribution of relaxation times. In the single-scattering approximation it would be an exponential (2.4).

B. Reflection from a half-space

Radiation of unit amplitude is incident normally on a random medium occupying the half-space $z>0$. The coherent field in (2.8) is

$$E_c(r,t)=e^{(ik_0-1/2l)z-i\omega_0 t} .$$

The diffusely reflected light does not depend importantly on the scattering angle so we evaluate it in the backward direction. The boundary conditions at $z=0$ are satisfied by writing the diffusion propagation (2.12) in real space and using the method of images to impose the condition that it vanish on the plane $z=0$.[5,7] Using (2.9), we find the diffusely reflected intensity on the plane $z=0$ is

$$\Gamma(t)=\frac{6\pi^2}{[1+F(t)]^2}e^{-i\omega_0 t},\quad \Gamma_2(t)=\frac{36\pi^4}{[1+F(t)]^4} . \tag{2.18}$$

The time dependence is different from (2.15) because of the different geometry but is again determined by the average correlation function (2.13). In case (a) for $t<\tau_\lambda^{(a)}$ we have

$$\Gamma_2(t)\sim\frac{1}{(1+\sqrt{3}\,|\,t\,|\,/\sqrt{2}\tau_\lambda^{(a)})^4} , \tag{2.19}$$

and shows a power-law decay in time. In case (b) $\Gamma_2(t)$ also shows a power-law decay with

$$\Gamma_2(t)\sim\frac{1}{[1+(3\,|\,t\,|\,/2\tau_\lambda^{(b)})^{1/2}]^4} . \tag{2.20}$$

In each case the characteristic time is τ_λ. In reflection there are contributions from diffusion paths of all lengths so that no characteristic length enters (2.19) or (2.20) as in (2.15).

C. Transmission through a slab

Radiation of unit amplitude is incident normally on the plane $z=0$ of a slab of thickness L of the random medium lying between the planes $z=0,L$.

A similar calculation from (2.9) gives for the diffusely transmitted light on the plane $z=L$

$$\Gamma(L,t)=\frac{3}{2\pi}\frac{\sinh F(t)}{\sinh\left[\dfrac{LF(t)}{l}\right]} . \tag{2.21}$$

In this case a characteristic length L enters the problem and together with τ_λ determines the spectral width. Thus in case (a)

$$\Gamma_2(L,t)\sim\left[\frac{\sinh(\sqrt{3}t/\sqrt{2}\tau_\lambda^{(a)})}{\sinh(\sqrt{3}Lt/\sqrt{2}l\tau_\lambda^{(a)})}\right]^2 , \tag{2.22}$$

and in case (b)

$$\Gamma_2(L,t)\sim\left[\frac{\sinh(3\,|\,t\,|\,/2\tau_\lambda^{(b)})^{1/2}}{\sinh\left[\dfrac{L}{l}(3\,|\,t\,|\,/2\tau_\lambda^{(b)})^{1/2}\right]}\right]^2 , \tag{2.23}$$

and the decay of the correlation function is essentially exponential and similar to that for the point source (2.15).

III. COHERENT BACKSCATTERING

In diffuse reflection it is now well established that in the backward direction the constructive interference of time-reversed waves leads to an increase in the intensity by about a factor of 2. This effect also gives an increase in the intensity fluctuations in the backward direction.[9] The motion of the scatterers breaks the symmetry of the medium for time-reversed waves so that these waves no longer interfere constructively. In this section we discuss the spectral distribution of the scattered intensity $\Gamma^{(i)}$ resulting from the interference of time-reversed waves.

For the case of radiation of frequency ω_0 incident normally on a half-space the interference contribution to the scattered intensity close to the backward direction is[5]

$$\Gamma^{(i)}(t)=\frac{8\pi c}{l}e^{-i\omega_0 t}\int_0^\infty dt'\int_0^\infty du\, f(t+t')f(t-t')\frac{u^2}{(1+u^2)^2}e^{-D(q^2l^2+u^2)t'/l^2-\gamma(t,t')} , \tag{3.1}$$

where $D=cl/3$ is the diffusion constant, $q=k_c\sin\theta$, where θ is the scattering angle (θ is close to π for backscattering), and

$$\gamma(t,t')=\frac{c}{2l}\int_{t-t'}^{t+t'}dt''[1-f(t'')] . \tag{3.2}$$

In the purely elastic case $f=1$, and (3.1) gives

$$\Gamma^{(i)}=6\pi^2/(1+ql)^2 ,$$

which is equal to (2.18) (for $t=0$) in the backward direction $q=0$. In the two cases (2.14)

$$\gamma^{(a)}(t,t')=[(t+t')^3-(t-t')^3]/\tau_\phi^{(a)3} , \quad (3.3)$$

$$\gamma^{(b)}(t,t')=4\,|\,t\,|\,t'/\tau_\phi^{(b)^2}, \quad |\,t\,|>t'$$
$$=2(t^2+t'^2)/\tau_\phi^{(b)2}, \quad |\,t\,|<t' \quad (3.4)$$

where the phase-coherence time is

$$\tau_\phi^{(a)}=(6\tau\tau_\lambda^{(a)2})^{1/3}, \quad \tau_\phi^{(b)}=(4\tau\tau_\lambda^{(b)})^{1/2} , \quad (3.5)$$

and $\tau=l/c$ is the wave-scattering time. The phase-coherence time cuts off the long diffusion paths and reduces the intensity of the coherent backscattering by factors

$$\left[1-\left[\frac{72\tau}{\pi^2\tau_\lambda^{(a)}}\right]^{1/4}\Gamma(\tfrac{3}{4})\right]$$

and

$$\left[1-\frac{1}{\pi^{1/2}}\left[\frac{24\tau}{\tau_\lambda^{(b)}}\right]^{1/3}\Gamma(\tfrac{5}{6})\right]$$

in the two cases (3.3) and (3.4) when $\tau<\tau_\lambda$. The time dependence of the coherently backscattered intensity is determined by τ_ϕ, and not τ_λ, as found in Sec. II for the direct waves. As the contributions of long diffusion paths are cut off in (3.1) the spectrum resembles that obtained in the single-scattering approximation, i.e., Gaussian and Lorentzian in the two cases (3.3) and (3.4), respectively.

IV. DISCUSSION

We have shown that when a wave is multiply scattered in a medium the time dependence of the intensity and its fluctuations are determined by the angular average (2.13) of the dynamic correlation function. The important assumptions made are (a) the propagation of the light in the medium is diffusive [see discussion after Eq. (2.7)], and (b) the frequency change of the light is small [see discussion after Eq. (2.4)]. Both these situations are readily realized in suspensions of macromolecules or Latex spheres in solution. The scattered intensity is obtained by summing over all the multiple-scattering paths and then depends on the shape and size of the medium. The relaxation time depends on the number of scattering events and, hence, depends on the length of the path and gets shorter the longer the path. However, even in the multiple-scattering case it is possible to obtain information about the dynamics of the medium although this is not as direct as when the scattering is weak and is directly related to the correlation function (2.1). These results also allow us to obtain information on the dynamics of concentrated solutions when the single-scattering Born approximation breaks down. Some of the detailed predictions which can be experimentally tested are the length dependence of the spectrum (2.15) or (2.19), and the time dependence of the homodyne spectrum (2.16), (2.17), (2.20), or (2.21). The dynamics of concentrated solutions may not be represented by these simple cases and the more general results (2.15) and (2.19) can be used.

We considered two special cases in which the scatterers have a Maxwell-Boltzmann velocity distribution or in which their motion is diffusive. The above results should also apply for other forms of dynamics of the scatterers. For example, if the scattering is due to thermal phonons of speed s it is easily shown that

$$f(t)=\frac{1}{2(k_0st)^2}[\cos(2k_0st)-1+2k_0st\,\sin(2k_0st)] , \quad (4.1)$$

and $\tau_\lambda=(k_0s)^{-1}$. The phase-coherence time in (3.1) is $\tau_\phi=(3\tau/k_0^2s^2)^{1/3}$.

Another possible situation is where the scattering is due to impurities fixed in space but having some internal dynamics which modify the dielectric constant. In this case the correlation function (2.2) can be written in the form

$$C(r-r',t-t')=C(r-r')f_i(t-t') ,$$

where $f_i(0)=1$, and $f_i(t)$ is the function replacing (2.13) in (2.12) and subsequent formulas. The time decay of the intensity correlation functions is now determined by the internal dynamics of the scatterers contained in $f_i(t)$. For example, if $f_i(t)=e^{-|t|/\tau}$ the decay of the correlation function (2.15) is like (2.17).

The above results should also apply to the scattering of other types of waves, e.g., electrons. The important assumption made is that the frequency change of the wave on scattering is small, i.e., the scattering is almost elastic. This is the case for electrons scattering from atoms or from low-frequency phonons. We have considered only the case of scalar waves, but as the contributions from long diffusion paths are the most interesting, the case of transverse vector waves will give the same results apart from certain contributions from short diffusion paths.[7]

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APPENDIX

The Fourier transform of the sum of ladder diagrams (2.1) satisfies the integral equation

$$L(\boldsymbol{\kappa}\Omega,\mathbf{q}_1\omega_1,\mathbf{q}_2\omega_2)=C(\mathbf{q}_1-\mathbf{q}_2,\omega_1-\omega_2) + \frac{1}{(2\pi)^4}\int d\mathbf{p}\,d\omega\,C(\mathbf{q}_1-\mathbf{p},\omega_1-\omega)D(\mathbf{p}+\boldsymbol{\kappa}/2,\omega+\Omega/2)D^*(\mathbf{p}-\boldsymbol{\kappa}/2,\omega-\Omega/2)L(\boldsymbol{\kappa}\Omega,\mathbf{p}\omega,\mathbf{q}_2\omega_2) . \quad (A1)$$

In L and C we may set $\mathbf{q}_1=k_0\mathbf{s}_1$, $\mathbf{q}_2=k_0\mathbf{s}_2$, and $\mathbf{p}=k_0\mathbf{s}$ where the $\mathbf{s}$ are unit vectors. Also L will not depend importantly on the directions of $\mathbf{s}_1$ and $\mathbf{s}_2$. Then averaging over the directions of $\mathbf{s}_1$ and $\mathbf{s}_2$ and retaining only the spherical part, $L(\kappa\Omega,\omega_1-\omega_2)$, of L, Eq. (A1) becomes

$$L(\kappa\Omega,\omega_1-\omega_2)=\frac{4\pi}{l}f(\omega_1-\omega_2)+\frac{1}{2\pi}\int d\omega\, f(\omega_1-\omega)Q(\kappa\Omega)L(\kappa\Omega,\omega-\omega_2)\ , \tag{A2}$$

where

$$Q(\kappa\Omega)=\frac{1}{(2\pi)^3}\int d\mathbf{p}\, D(p+\kappa/2,\omega+\Omega/2)D^*(p-k/2,\omega-\Omega/2)\simeq 1-\frac{\kappa^2 l^2}{3}+i\Omega\tau\ . \tag{A3}$$

The approximation (A2) neglects a small contribution to the diffusion constant arising from higher harmonics of $C(k_0(\mathbf{s}_1-\mathbf{s}_2),\omega)$. Equation (A2) is easily solved by taking its Fourier transform and it gives (2.12).

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