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Rayleigh scattering and weak localization: Geometric effects and fluctuations

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The coherently backscattered light from a disordered medium occupying a half space is discussed. For oblique incidence of the incident light it is shown that the coherently backscattered light is confined to a cone of roughly elliptical cross section (it is circular for normal incidence). The effects of lower dimensionality (two dimensions) and absorption on the coherent backscattering are discussed. The fluctuations in the intensity are discussed and in the diffusion approximation shown to obey a Rayleigh distribution. The light scattered at different angles is shown to be uncorrelated.

I. INTRODUCTION

There has been considerable experimental[1-4] and theoretical[5-8] interest recently in the coherent backscattering of light from a disordered medium. These experiments are interesting because the elastic scattering and interference effects associated with weak localization can be directly observed. The coherent backscattering arises because a wave elastically scattered through a certain multiple scattering path can interfere coherently with the wave which follows the time-reversed path. The interference is constructive in the backscattering direction. For scalar waves the angular line shape of the coherently backscattered light has been considered by Golubentsev[5] and by Akkermans, Wolf, and Maynard.[6] The experiments[1-3] (especially those of Ref. 3) show interesting polarization effects and these have been considered theoretically by Stephen and Cwilich (referred to as SC).

The purpose of this paper is to examine further the coherent backscattering. The above theoretical papers only deal with the case of normal incidence of the incident light, where the coherently backscattered light is confined to a circular cone. Experimentally it is advantageous to use an incident angle less than $\pi/2$ to avoid reflection from the surface. In this case we show that when polarization effects are included the coherently backscattered light is confined to a cone of roughly elliptic cross section. This effect has apparently been observed.[9] In some of the experiments[10] absorption effects are important. Absorption reduces the contribution of long multiple scattering paths to the coherent backscattering and we show here that it leads to a rounding of the backscattered peak. We also consider the effects of dimensionality D on the coherent backscattering and show that the interference effects in scattering of scalar waves from a half space $(D=3)$ and a half plane $(D=2)$ lead to identical line shapes. Finally we consider the angular correlation of the scattered light from a half space and show that in the diffusion approximation light scattered at different angles is uncorrelated. This is in agreement with the experiments of Etemad[3] who observed that the scattered light is uncorrelated over less than one mrad. We find that the localization effects increase the magnitude of the fluctuations in the backward direction but the intensity distribution still follows a Rayleigh distribution.

II. ANISOTROPY OF THE BACKSCATTERING PEAK

The disordered medium occupies the half space $z>0$ and light of frequency ω and wave vector $k=\omega/c$ is incident in the xz plane making an angle θ_0 with the normal (see Fig. 1). The incident light may be polarized along x' or y (perpendicular to xz plane) and the diffusely scattered light is observed in the direction of the unit vector $\mathbf{s}$ close to the backward direction, i.e., $(-z')$. We suppose that $\mathbf{s}$ has polar angles θ',ϕ' in the $x'yz'$ coordinate system. Then $\pi-\theta'$ is small, and $\phi'=0$ and $\phi'=\pi/2$ correspond to observing the scattered light in the xz and yz' planes,

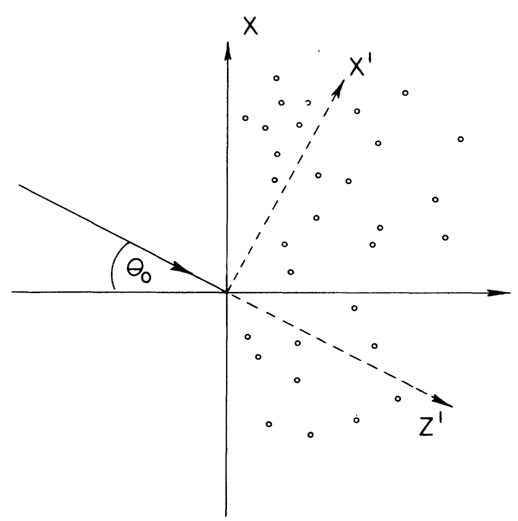

FIG. 1. Light is incident on the half space $z>0$ in the xz plane along z' making an angle θ_0 with the z axis. The light may be polarized along x' or y (perpendicular to the xz plane). The scattered light is observed in the backward direction close to the z' axis having polar and azimuthal angles θ',ϕ' in the $x'yz'$ coordinate system.

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respectively. We show below that the coherently backscattered light is confined to a cone with a roughly elliptical cross section if $\theta_0 \neq 0$ (it is circular for normal incidence).

In a previous paper[8] we gave an expression for the diffusely reflected light intensity from a half space [see Eq. (5.1) of SC]. For the geometry of Fig. 1 this expression becomes

$$J_{ij}^{(rs)}(\mathbf{s})=\frac{\pi\Delta}{c_0}\int_0^\infty dz_1 dz_2 \int d^2\rho e^{-(z_1+z_2)/l_0}[P_{ijrs}^{(L)}(\rho,z_1,z_2)+e^{-iQ_z(z_1-z_2)-i\mathbf{Q}_\perp\cdot\boldsymbol{\rho}}P_{ijrs}^{(C)}(\rho,z_1,z_2)]E_rE_s \ . \tag{2.1}$$

Here $J_{ij}^{(rs)}(\mathbf{s})$ is proportional to the intensity of the light scattered in direction $\mathbf{s}$ with polarization along i,j when the incident light of amplitude E_r has polarization along r $(i,j,r,s=x',y)$. We use the same notation as SC in which Δ is proportional to the mean-square fluctuation in the dielectric constant, $l_0=lc_0$ where l is the mean free path and $c_0=\cos\theta_0$. The wave-vector transfer when $\pi-\theta'$ is small is $(s_0=\sin\theta_0)$

$$\begin{aligned} Q_x &= kc_0\sin\theta'\cos\phi' \ , \\ Q_y &= k\sin\theta'\sin\phi' \ , \\ Q_z &= ks_0\sin\theta'\cos\phi' \ . \end{aligned} \tag{2.2}$$

The two "diffusion" propagators $P_{ijrs}^{(L,C)}$ give rise to the Rayleigh scattering (L) and interference effects (C), respectively. These are given in SC for the infinite space and are generalized to the half space problem by writing them in real space and using the method of images. There are three cases of interest: (i) the incident and reflected light are both polarized along $x', J_{x'x'}^{(x'x')}$; (ii) the incident light is polarized along x' and the reflected light along y or vice versa, $J_{yy}^{(x'x')}=J_{x'x'}^{(yy)}$; (iii) the incident and reflected light are both polarized along y, $J_{yy}^{(yy)}$. In each case the reflected light can be observed as a function of the angles θ' and ϕ'. For these three cases the Fourier transforms of the diffusion propagators in an infinite space are given by

$$\begin{aligned} P_{x'x'x'x'}^{(L,C)}(K)=\frac{1}{K^2l^2}&+2(1-3c_0^2s_0^2)f^{(L,C2)}(K) \\ &+4c_0^2s_0^2f_{xz}^{(L,C3)}(K) \ , \end{aligned} \tag{2.3}$$

$$P_{yyx'x'}^{(L)}(K)=\frac{1}{K^2l^2}-f^{(L2)}(K) \ , \tag{2.4}$$

$$\begin{aligned} P_{yyx'x'}^{(C)}(K)=c_0^2[&f_{xy}^{(C3)}(K)-f_{xy}^{(C4)}(K)] \\ &+s_0^2[f_{yz}^{(C3)}(K)-f_{yz}^{(C4)}(K)] \ , \end{aligned} \tag{2.5}$$

$$P_{yyyy}^{(L,C)}(K)=\frac{1}{K^2l^2}+2f^{(L,C2)}(K) \ , \tag{2.6}$$

where

$$f^{(L2)}(K)=10/(9+7K^2l^2) \ , \tag{2.7}$$

$$f_{ab}^{(L3)}(K)=\frac{35}{21+(23K^2-10K_a^2-10K_b^2)l^2} \ , \tag{2.8}$$

$$f_{ab}^{(L4)}(K)=\frac{5}{5+(3K^2-2K_a^2-2K_b^2)l^2} \ , \tag{2.9}$$

and $f^{(C2)}=\frac{7}{10}f^{(L2)}$, $f^{(C3)}=\frac{7}{10}f^{(L3)}$, and $f^{(C4)}=\frac{1}{2}f^{(L4)}$. We note that we no longer have a symmetry between x and y owing to the terms f_{xz} and f_{yz}. These propagators are easily calculated in real space giving $P(\rho,z_1-z_2)$ and the boundary conditions appropriate for the half space are satisfied by replacing $P(\rho,z_1-z_2)$ by

$$P(\rho,z_1,z_2)=P(\rho,z_1-z_2)-P(\rho,z_1+z_2) \ . \tag{2.10}$$

The resulting expressions are substituted in (2.1). In order to express the scattered intensities concisely we define

$$I_Q(b_1,b_2,b_3)=\frac{b_3^{-1}}{[1+(c_0/b_3^{1/2})(1+b_1Q_x^2l^2+b_2Q_y^2l^2)^{1/2}]^2+Q_z^2l_0^2} \ , \tag{2.11}$$

$$C^{-1}J_{x'x'}^{(x'x')}=1+\frac{1}{(1+Q_\perp l_0)^2+Q_z^2l_0^2}+\tfrac{20}{9}(1-3c_0^2s_0^2)[I_0(0,0,\tfrac{7}{9})+\tfrac{7}{10}I_Q(\tfrac{7}{9},\tfrac{7}{9},\tfrac{7}{9})]+\tfrac{20}{3}c_0^2s_0^2[I_0(0,0,\tfrac{13}{21})+\tfrac{7}{10}I_Q(\tfrac{13}{21},\tfrac{23}{21},\tfrac{13}{21})] \ , \tag{2.12}$$

$$C^{-1}J_{x'x'}^{(yy)}=1-\tfrac{10}{9}I_0(0,0,\tfrac{7}{9})+\tfrac{1}{2}c_0^2[\tfrac{7}{3}I_Q(\tfrac{13}{21},\tfrac{13}{21},\tfrac{23}{21})-I_Q(\tfrac{1}{5},\tfrac{1}{5},\tfrac{3}{5})]+\tfrac{1}{2}s_0^2[\tfrac{7}{3}I_Q(\tfrac{23}{21},\tfrac{13}{21},\tfrac{13}{21})-I_Q(\tfrac{3}{5},\tfrac{1}{5},\tfrac{1}{5})] \ , \tag{2.13}$$

$$C^{-1}J_{yy}^{(yy)}=1+\frac{1}{(1+Q_\perp l_0)^2+Q_z^2l_0^2}+\tfrac{20}{9}I_0(0,0,\tfrac{7}{9})+\tfrac{14}{9}I_Q(\tfrac{7}{9},\tfrac{7}{9},\tfrac{7}{9}) \ , \tag{2.14}$$

where $Q_\perp^2=Q_x^2+Q_y^2$, $C=\pi\Delta E_0^2lc_0^2/2$, and E_0 is the amplitude of the incident field.

For scalar waves only the first two terms of (2.12) and (2.14) would be present and for normal incidence $(Q_z=0)$ they reduce to the result of Ref. 6. More generally, these expressions reduce to those of SC for normal incidence. There are two effects leading to the angular anisotropy of the coherently backscattered light. (We define the anisotropy as the width of the peak in the xz plane divided by its width in the yz' plane, see Fig. 1.) Firstly there is the (trivial) geometric effect in the definitions of Q_x and Q_y in Eq. (2.2) which leads to an angular anisotropy of $1/c_0$, i.e., the backscattered cone is narrower in the yz' plane than in the xz plane. If we suppress this effect by using Q_x and Q_y as our variables the cone predicted by (2.14) is circular and the scalar wave part (second term) of (2.12) is circular. A further anisotropy arises from the polarization terms in (2.12) (last term) and in (2.13) (last two terms). These terms come from the unsymmetrical parts of the diffusion propagator (2.8) and (2.9). In (2.12) the anisotropic peak has $Q_x/Q_y \sim (\frac{23}{13})^{1/2}$ and is broadened in the xz plane. In (2.13) the two anisotropic peaks have $Q_x/Q_y \sim (\frac{13}{23})^{1/2}$ and $(\frac{1}{3})^{1/2}$, respectively and are narrower in the xz plane.

III. TWO DIMENSIONS

It is of interest to consider the effects of lower dimensions on the scattering. Suppose the disordered medium is contained in a cavity which is very thin in the y direction. (The scattering geometry is the same as in Fig. 1.) The modes of such a cavity can be divided into transverse electric (TE) or transverse magnetic (TM) modes. Such modes would not be expected to mix appreciably and polarization effects should be unimportant. Thus we only consider the case of scalar waves. The diffusion of the light will now occur in two dimensions, i.e., the xz plane. It is not difficult to see that the angular shape of the backscattering cone will be the same as in the $D=3$ case, i.e.,

$$J \sim 1+\frac{1}{(1+Q_x l_0)^2+Q_z^2 l_0^2} \tag{3.1}$$

which are the scalar terms in (2.12). A simple proof is to note that the diffusion propagator in the $D=3$ case for half space is proportional to

$$\frac{1}{[\rho_x^2+\rho_y^2+(z_1-z_2)^2]^{1/2}}-\frac{1}{[\rho_x^2+\rho_y^2+(z_1+z_2)^2]^{1/2}} . \tag{3.2}$$

If the scattering occurs in the xz plane this expression is integrated over $-\infty<\rho_y<\infty$ giving

$$\ln\left[\frac{\rho_x^2+(z_1+z_2)^2}{\rho_x^2+(z_1-z_2)^2}\right]$$

which is the diffusion propagator in the $D=2$ case for a half plane. Thus for the half-space reflection experiment scalar waves in $D=2$ and $D=3$ give an identical dependence on angle for the coherently backscattered light.

At first sight this result may appear strange but in $3D$ the light scattered from the uniform incident beam diffuses to the surface and in $2D$ diffuses to the boundary line. This is not the same as the frequently considered case of the return of a diffusing particle to the origin which are very different in two and three dimensions.

IV. EFFECTS OF ABSORPTION

The scattering medium may also absorb some of the light in addition to the elastic scattering. This will have the effect of reducing the contribution of long diffusion paths to the coherent backscattering. We add absorption to the model by including an imaginary part to the dielectric constant. The electric field satisfies

$$\{\nabla^2+k^2[1+i\alpha+\epsilon'(r)]\}E=0 , \tag{4.1}$$

where α is the constant absorptive part and ϵ' the real, random part of the dielectric constant. We only consider the case of scalar waves because only long diffusion paths are of interest. We define an absorption length by $l_i=1/\alpha k$. The angular distribution of the backscattered light for the case of normal incidence is

$$J=\frac{3\pi\Delta E_0^2 l}{2}\left[\frac{1}{(1+\bar{l}/l_e)^2}+\frac{\bar{l}/l}{[1+\bar{l}/l_e(1+Q^2l_e^2)^{1/2}]^2}\right]$$

when $\bar{l}^{-1}=l^{-1}+l_i^{-1}$ and $l_e^2=l_i\bar{l}^2/3l$, where l is the mean free path for elastic scattering. The effect of absorption is to reduce the intensity in the coherent backscattering peak by a factor $\bar{l}/l$ and also to smooth out the peak and give it a roughly Lorentzian rather than triangular shape.

V. FLUCTUATIONS

In this section we discuss intensity fluctuations and the angular correlation of these fluctuations in the light scattered from a disordered medium. There is considerable literature[11–13] on this subject which usually is under the heading of speckle patterns. Recently, such speckle patterns have been discussed for the present model of a disordered medium by Shapiro.[14] He evaluated the intensity-intensity correlation function for the light scattered from a point source at the origin in an infinite medium and for distances $R>l$ found the characteristic speckle pattern result $\langle I^2(R)\rangle_c=\langle I(R)\rangle^2$. (In addition, he determined the correlation of the intensity at different points and at different frequencies.)

We first generalize Shapiro's result to include the effects of polarization. We consider the case of a polarized point source at the origin of an infinite medium. We define correlation functions for the scattered field

$$\Gamma_{ij}(\mathbf{R})=\langle E_i(\mathbf{R})E_j^*(\mathbf{R})\rangle \tag{5.1}$$

which for $i=j$ is proportional to the intensity with polarization i, and

$$C_{ijkl}(\mathbf{R})=\langle (E_i(\mathbf{R})E_j^*(\mathbf{R}))(E_k(\mathbf{R})E_l^*(\mathbf{R}))\rangle_c . \tag{5.2}$$

C is defined as a cumulant so that the term $\Gamma_{ij}\Gamma_{kl}$ is omitted. In the diffusion approximation it is easily shown for $R>l$ that

$$\Gamma_{ij}(\mathbf{R})=\frac{\Delta j_0^2}{9\pi R}\delta_{ij}[1+\tfrac{7}{10}e^{-R/l_1}(3\delta_{ix}-1)] , \tag{5.3}$$

where $4\pi j_0$ is the amplitude of the point source which is polarized along x and $l_1=(\sqrt{7}/3)l$. Thus the scattered light for $R\gg l$ is depolarized and the effects of polarization decrease exponentially with R. In the diffusion approximation

$$C_{ijkl}(\mathbf{R})=\Gamma_{il}(\mathbf{R})\Gamma_{kj}(\mathbf{R}) \tag{5.4}$$

and because of this approximate factorization property of the correlation functions the intensity obeys a Rayleigh distribution, $P(I)\sim e^{-I/\langle I\rangle}$. The effects of weak localization will be to decrease the intensity reaching R because interference effects increase the intensity returning to the origin but the mean-square fluctuations will still obey (5.4). The intensity at different points in space will show power-law correlations.[15]

The angular correlation of the scattered light is also of interest. We only consider scalar waves because as shown above polarization effects are unimportant. We define a correlation function

$$C(\mathbf{R},\mathbf{q},\mathbf{q}')=\langle \Gamma(\mathbf{R},\mathbf{q})\Gamma(\mathbf{R},\mathbf{q}')\rangle_c \tag{5.5}$$

where $\Gamma(\mathbf{R},\mathbf{q})$ is the spectral intensity at $\mathbf{R}$ with wave vector $\mathbf{q}$. In terms of the scattered fields

$$C(\mathbf{R},\mathbf{q},\mathbf{q}')=\int d\mathbf{r}\,d\mathbf{r}'e^{-i\mathbf{q}\cdot\mathbf{r}-i\mathbf{q}'\cdot\mathbf{r}'}\left\langle \left[E\left[R+\frac{r}{2}\right]E^*\left[R-\frac{r}{2}\right]\right]\left[E\left[R+\frac{r'}{2}\right]E^*\left[R-\frac{r'}{2}\right]\right]\right\rangle_c . \tag{5.6}$$

In the diffusion approximation this cumulant can be factorized:

$$C(\mathbf{R},\mathbf{q},\mathbf{q}')=\int dr\,dr'e^{-i\mathbf{q}\cdot\mathbf{r}-i\mathbf{q}'\cdot\mathbf{r}'}\Gamma\left[R+\frac{r-r'}{4},\frac{r+r'}{2}\right]\Gamma\left[R+\frac{r'-r}{4},\frac{r+r'}{2}\right] \tag{5.7}$$

where

$$\Gamma(R,r)=\left\langle E\left[R+\frac{r}{2}\right]E^*\left[R-\frac{r}{2}\right]\right\rangle .$$

The averaged scattered intensity is slowly varying in space and

$$\Gamma\left[R\pm\frac{r-r'}{4},\frac{r+r'}{2}\right]$$

is almost independent of $r-r'$. Thus neglecting the dependence or $r-r'$ we find from (5.7)

$$C(\mathbf{R},\mathbf{q},\mathbf{q}')=\Gamma^2(\mathbf{R})(2\pi)^3\delta(\mathbf{q}-\mathbf{q}')F(q) , \tag{5.8}$$

where

$$F(q)=\frac{4\pi}{qk^2}[2\tan^{-1}2lq-\tan^{-1}2l(q+k)$$
$$-\tan^{-1}2l(q-k)] .$$

Thus the intensity of the scattered light at different angles is completely uncorrelated. A similar result for surface scattering is given by Goodman.[13]

These same arguments can be extended to light diffusely reflected from a half space. The reflected light intensity should exhibit the characteristic speckle and follow a Rayleigh distribution. In the backscattering direction interference effects increase the average intensity and correspondingly the fluctuations. The light scattered at different angles would be expected to be uncorrelated (for a finite system the δ function in (5.8) is replaced by a form factor depending on the shape and size of the scattering medium). These results are in accord with the observations of Etemad[3] who found that the scattered light at angles differing by 1 mrad was uncorrelated. It would be of interest to go beyond the diffusion approximation leading to (5.8) and estimate any remaining angular correlations.

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