Coherent Backscattering of Light by Disordered Media: Analysis of the Peak Line Shape

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Recent experiments have confirmed that coherent effects in the multiple scattering of light affect the angular dependence of the intensity reflected by disordered media. By considering the constructive interferences between time-reversed paths of light in a semi-infinite medium, we analyze the experimental line shape of the albedo within the diffusion approximation and explain the observed effects of polarization.

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The propagation of a wave in a dense distribution of elastic scatterers is a difficult problem to handle in the framework of the multiple-scattering theory. The classical approach, which assumes that phases are uncorrelated on scales larger than the elastic mean free path l, leads to an intensity transport equation of the Boltzmann type in which any interference effects are neglected. In the simple regime where the length or time scales are larger than l or τ ($\tau = l/c$, where c is the wave velocity), this equation reduces to a diffusion equation with a diffusion constant $D = lc/3$. The classical approach is generally well justified for study of the intensity scattered from a bounded, weakly disordered medium. However, it must be corrected when the wave emerges from the medium around the backscattering direction (i.e., the direction opposite to the incident one). In this case, constructive interferences arise and must be taken into account in order to explain the enhancement of the backscattered intensity with respect to the classical prediction. This phenomenon has been recognized almost independently in two different fields. In condensed-matter physics, it is the basis of the weak-localization regime for electrons in impure metals,[1,2] where the quantum correction to the diffusion constant[3] is obtained from the pioneering work of Langer and Neal.[4] In optics, it was first considered by de Wolf[5] for electromagnetic waves propagating in turbulent atmosphere. More recently, this effect has been directly demonstrated by three experiments[6-8] which show that the intensity of light scattered from a concentrated aqueous suspension of latex microspheres presents a sharp peak centered at the backscattering direction. The sharpness of this peak as well as the effects of light polarization are typical enough to call for a detailed analysis of this coherent backscattering effect, which is the purpose of this Letter. Two previous works must be mentioned here: one[9] about the contribution of double scattering to the backscattering intensity enhancement and another one by Golubentsev[10] who discusses the reduction of this peak due to the motion of impurities and the gyrotropy of the medium. We shall follow here the features of his analysis.

The basis of the interference effect in multiple scattering is very general. We assume that the waves are of scalar nature (the important effect of light polarization will be discussed later). Consider a sequence of n scattering events characterized by the wave vectors $\mathbf{k}_i, \mathbf{k}_1, \ldots, \mathbf{k}_n = \mathbf{k}_f$, where $\mathbf{k}_j$ is the wave vector after the jth scattering event and $\mathbf{k}_i$ and $\mathbf{k}_f$ stand for the initial and final wave vectors. In classical transport theory all n-order sequences are assumed to be uncorrelated as a result of the random nature of the distribution of scatterers. However, any given sequence and its time reverse $\mathbf{k}_i, -\mathbf{k}_{n-1}, -\mathbf{k}_{n-2}, \ldots, -\mathbf{k}_1, \mathbf{k}_f$, where the light is scattered by the same centers but in opposite order, can interfere constructively for a special choice of $\mathbf{k}_f$ relative to $\mathbf{k}_i$. The total phase shift between the two corresponding partial waves is simply $\mathbf{q}\cdot(\mathbf{r}_1 - \mathbf{r}_n)$, where $\mathbf{q}$ is the transfer wave vector $\mathbf{k}_i + \mathbf{k}_f$ and $\mathbf{r}_1$ and $\mathbf{r}_n$ are the positions of the first and last scattering centers. For the backscattering situation ($\mathbf{k}_f = -\mathbf{k}_i$) these two partial waves have the same amplitude and phase and add coherently. If θ is the relative angle between $\mathbf{k}_i$ and $\mathbf{k}_f$, the coherence is lost for angles θ larger than $\lambda/|\mathbf{r}_1 - \mathbf{r}_n|$, where λ is the

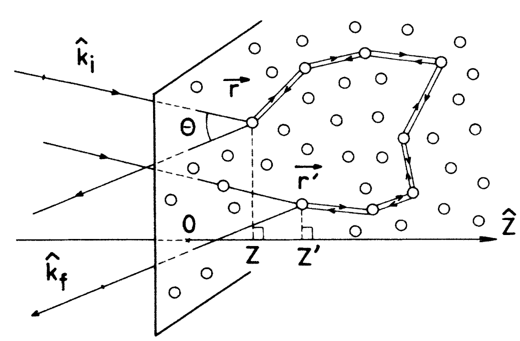

FIG. 1. Geometry used for the calculation of the coherent albedo, showing two interfering light paths.

wavelength. Since the average value of $|\mathbf{r}_1-\mathbf{r}_n|$ for the shortest sequence $(n=2)$ is the mean distance between two scattering events, i.e., the elastic mean free path l, one expects[11] the reflected intensity to increase by up to a factor 2 inside a cone of angular width of order λ/l. More quantitatively, the interference correction to the multiple-scattering contribution of all paths of n steps from $\mathbf{r}_1$ to $\mathbf{r}_n$ will be obtained by a weighting of the corresponding incoherent intensity by a factor $\cos[\mathbf{q}\cdot(\mathbf{r}_1-\mathbf{r}_n)]$. It is now well known that these interferences reduce the bulk transport coefficients,[3,12] like the diffusion constant D. Here, we consider their consequence on the albedo problem.

We study the light reflected by a semi-infinite scattering medium occupying the half space $z>0$ (Fig. 1). We define the albedo $\alpha(\hat{\mathbf{k}}_i,\hat{\mathbf{k}}_f)$ as the ratio of the emergent flux per unit solid angle $d\Omega$ and unit interface area around the direction $\hat{\mathbf{k}}_f$ to the incident energy flux. α is given by

$$\alpha(\hat{\mathbf{k}}_i,\hat{\mathbf{k}}_f)=(c/4\pi l^2)\int dz\,dz'\,d^2\rho\exp(-z/\mu_0 l)\{1+\cos[\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}')]\}Q(\mathbf{r},\mathbf{r}')\exp(-z'/\mu l), \tag{1}$$

where z and z' are the projections of $\mathbf{r}$ and $\mathbf{r}'$ on the z axis, $\boldsymbol{\rho}$ is the projection of $\mathbf{r}-\mathbf{r}'$ on the interface plane, while μ_0 and μ are respectively the projections of $\mathbf{k}_i$ and $\mathbf{k}_f$ on the z axis. The physical meaning of the different terms appearing in (1) is the following: $l^{-1}\exp(-z/\mu_0 l)$ is the ratio to the incident flux of the energy scattered per unit time and unit volume at point $\mathbf{r}$. $Q(\mathbf{r},\mathbf{r}')$ is the Green's function which describes the light transport from a point source located at $\mathbf{r}$ to $\mathbf{r}'$. It is given by the ratio of the energy density at $\mathbf{r}'$ to the source production rate of energy. $(c/4\pi)dz'/l$ is the fraction of the energy density at $\mathbf{r}'$ which contributes to the flux scattered per solid angle $d\Omega$ around $\hat{\mathbf{k}}_f$, while $e^{-z'/\mu l}$ is the part of this flux which emerges without being again scattered. Finally, the factor $1+\cos[\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}')]$ accounts for the interference effect. Note that since this interference is irrelevant for single scattering, Eq. (1) is only valid for the multiple-scattering part of the albedo.

The problem of the calculation of $Q(\mathbf{r},\mathbf{r}')$ has been studied in classical transport theory.[13] From the intensity transport equation, it can be shown that far enough from the interface, $Q(\mathbf{r}-\mathbf{r}')$ obeys a diffusion equation with the boundary condition $Q(-z_0)=0$, i.e., with a trapping plane located at $-z_0$ (for pointlike scatterers, z_0 is about $0.7l$). $Q(\mathbf{r}-\mathbf{r}')$ is then given by

$$Q(\mathbf{r}-\mathbf{r}')=Q_0(\mathbf{r}-\mathbf{r}')-Q_0(\mathbf{r}-\mathbf{r}'^*). \tag{2}$$

Here, Q_0 is the homogeneous solution of the stationary diffusion equation, $Q_0(\mathbf{r}-\mathbf{r}')=[4\pi D|\mathbf{r}-\mathbf{r}'|]^{-1}$ (i.e., the "3D Coulomb potential"), where $D=lc/3$ is the diffusion constant[14] of the light, while $\mathbf{r}'^*$ is the image of $\mathbf{r}'$ in the mirror plane in $-z_0$. Physically, the image term in Eq. (2) ensures that only paths not crossing the trapping plane are selected in the evaluation of Q since the emergent light is lost for the medium.

Let us now simplify the expression for α by assuming that in $Q(\mathbf{r},\mathbf{r}')$, $\mathbf{r}$ and $\mathbf{r}'$ are located on the same plane $z=l$. This is justified by the exponential terms $\exp(-z/\mu_0 l)$ and $\exp(-z'/\mu l)$. For quasinormal incidence and emergence $(\mu\simeq\mu_0\simeq 1)$, the albedo (1) becomes

$$\alpha(\theta)=(3/4\pi^2 l)\int d^2\rho[1+\cos(\mathbf{q}_\perp\cdot\boldsymbol{\rho})][\rho^{-1}-(\rho^2+a^2)^{-1/2}], \tag{3}$$

where $a=2(l+z_0)$ and $\mathbf{q}_\perp$ is the component of $\mathbf{q}$ normal to the z axis, with $q_\perp\simeq 2\pi\theta/\lambda$. The integration in Eq. (3) leads to

$$\alpha(\theta)=(3a/4\pi l)\{1+[1-\exp(-q_\perp a)]/q_\perp a\}. \tag{4}$$

This expression exhibits the following features: (i) For $\theta=q_\perp=0$, i.e., right in the backscattering direction, the albedo is exactly twice the incoherent value $\alpha_{\rm inc}$ obtained for "large" angles. (ii) The angular width in which the coherent effect is observable is of order $\lambda/2\pi l$. (iii) Close to the exact backscattering direction, the albedo varies linearly, $\alpha(\theta)=\alpha_{\rm inc}[2-2\pi l+z_0/\lambda|\theta|]$, so that, near $\theta=0$, the line shape is triangular. The exact expression of $\alpha(\theta)$ derived by going beyond the previous approximation is given by

$$\alpha(\theta)=\frac{3}{8\pi}\left[1+\frac{2z_0}{l}+\frac{1}{(1+q_\perp l)^2}\left(1+\frac{1-\exp(-2q_\perp z_0)}{q_\perp l}\right)\right] \tag{5}$$

which preserves the previous features. This expression is in principle only valid for isotropic scattering. An approximate extension to the experimental case of anisotropic scattering is obtained[15] by replacing l everywhere by the transport mean free path l^*. In Fig. 2, we thus compare an experimental line shape with the predicted one, obtained by a convolution of Eq. (5) with the relevant instrumental profile.[8] No adjustable parameter is used, since l^* was determined by a different experiment.[16] If we consider the approximate nature of the diffusive behavior, the agreement between the two curves is remarkable.

The small-angle linear behavior of $\alpha(\theta)$ is the signature of the large-distance asymptotic diffusive behavior of $Q(\mathbf{r}-\mathbf{r}')$ in the presence of the interface. Because of the image term in Eq. (2), the ρ^{-1} law valid for Q in the bulk is modified to ρ^{-3}. The surface element in the integration over the separation plane provides an additional factor ρ which leads to a ρ^{-2} behavior whose Fourier transform yields the linear dependence at small angles.. It should be noted that a similar analysis in other dimensions yields the same triangular line shape.[17]

Expression (3) shows that the coherent contribution to the albedo is nothing but the small-angle "structure factor" of the correlation function $Q(\mathbf{r}-\mathbf{r}')$ given by Eq. (2). Therefore, the smaller the angle θ is, the larger the maximal size of the probed diffusion paths is. We can be more explicit by considering the contribution to the albedo of paths of a given length L (this contribution is also of interest since it can be measured experimentally, e.g., by a pulse experiment). Within the previous approximation, $z=z'=l$, justified for large L, the contribution of these loops to $Q(\rho)$ is

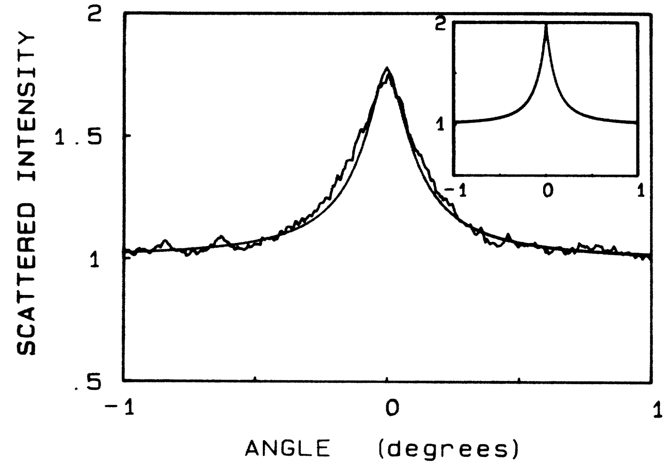

FIG. 2. Comparison between experimental and theoretical line shapes for the coherent backscattering. The experimental recording is taken from Ref. 8 [Fig. 3(a)]. The inset represents the bare theoretical curve taken from Eq. (5) with $\lambda=0.515\ \mu$m and $l^*=19\ \mu$m. The full line is the theoretical prediction obtained by the convolution of this previous curve and the instrumental profile.

$$Q(\rho,L)=\left(\frac{3}{4\pi l}\right)^{3/2}\frac{1-\exp[-3(l+z_0)^2/Ll]}{L^{1/2}}\frac{\exp(-3\rho^2/4Ll)}{L}. \quad (6)$$

The ρ Fourier transform of $Q(\rho)$ is a Gaussian function. Its width, of order $\lambda/(Ll)^{1/2}$, results from the diffusion process parallel to the interface, while the $1/L^{3/2}$ dependence of its amplitude is the signature of a one-dimensional random walk with a trap.[18] Thus, at a given small angle θ, only the paths of length L smaller than $\lambda^2/l\theta^2$ contribute significantly to the coherent albedo. In this respect, $\lambda^2/D\theta^2$ is analogous to the phase coherence time τ_ϕ introduced by Khmelnitskii (see Ref. 1).

The previous results have been obtained within the diffusion approximation. Nevertheless, it must be noted that an expression of the coherent part of the albedo can be carried out from a direct nth-order multiple-scattering theory by means of an expansion in maximally crossed diagrams.[15] As expected, we find that the angular width and amplitude of the coherent nth-order contribution vary respectively as $1/n^{1/2}$ and $1/n^{3/2}$. It is worth noting that in recent experiments (see Fig. 2), the coherent albedo still varies for angles about 10 times smaller than the total measured cone aperture. This shows that in such experiments, the light explores the medium along paths of length larger than about one hundred mean free paths.

So far, we have considered the case of scalar waves. However, the importance of light polarization has been demonstrated in recent experiments.[6-8] When the analyzed polarization is parallel to the incident one (parallel configuration), the backscattered intensity for $\theta=0$ is about 1.7 times larger than the incoherent intensity (see Fig. 2). On the other hand, the enhancement factor in the crossed configuration is about 1.3. This difference can be understood from the form of the amplitude Rayleigh scattered by an individual center. It is proportional to $(\mathbf{k}\times\mathbf{P}_0)\times\mathbf{k}\equiv\mathbf{M}(\mathbf{k})\cdot\mathbf{P}_0$, where $\mathbf{P}_0$ is the incident polarization, $\mathbf{k}$ the emergent wave vector, and $\mathbf{M}(\mathbf{k})$ a 3×3 symmetric matrix. Thus, for the n-scattering sequence $(\mathbf{k}_i,\mathbf{k}_1,\ldots,\mathbf{k}_{n-1},\mathbf{k}_f)$ the emergent polarization state is $\mathbf{P}_n=[\mathbf{k}_f\times(\mathbf{M}_n\cdot\mathbf{P}_i)\times\mathbf{k}_f$, where $\mathbf{P}_i$ is the incident polarization and $\mathbf{M}_n$ is the ordered product $\prod_{j=1}^{n-1}\mathbf{M}(k_{n-j})$, while its time-reversed counterpart is $\mathbf{P}_n'=[\mathbf{k}_f\times(\mathbf{M}_n'\cdot\mathbf{P}_i)]\times\mathbf{k}_f$. Since the $M(k)$'s are symmetric, $\mathbf{M}_n'$ is the transpose of $\mathbf{M}_n$. Let us denote by $P_{n\parallel}$ and $P_{n\perp}$ the components of P_n respectively parallel and perpendicular to $\mathbf{P}_i$. We have $P_\parallel=P_\parallel'$, so that, in the parallel configuration, the coherence is totally maintained for any loop and the expected enhancement is, as previously noticed,[7,8] a factor 2. Furthermore, because we expect the large-distance behavior of the intensity in any polarization state to be diffusive, the line shape at small angles should be similar to that given by (5). The case of perpendicular polarization is different. Except for $n=2$, $P_{n\perp}$ differs from $P_{n\perp}'$ and one must determine the coherence ratio,

$C(n)=\langle P_{n\perp}P'_{n\perp}\rangle/\langle P^2_{n\perp}\rangle$, averaged over all the n-scattering sequences, in order to estimate the enhancement factor at $\theta=0$. Within the assumption of uniform distribution for the successive $\mathbf{k}_j$'s, which for isotropic scattering should be true in the limit of large n, we can derive linear recurrence equations involving the different averages $\langle M_{nij}M_{nkl}\rangle$. Their solution leads to an exponential decay

$$C(n)=\tfrac{3}{2}[(0.7)^{n-1}-(0.5)^{n-1}]/[1-(0.7)^{n-1}].$$

Finally, we estimate the desired enhancement factor for crossed polarization to be about 1.5 by summing over n the scalar n-scattering contribution weighted by $C(n)$. We thus understand the existence of a partial coherent effect in the crossed configuration (the difference between 1.3 and 1.5 is likely due to the various approximations in our calculation).

In the present analysis, we have presented physical arguments to explain the observed coherent backscattering peak in the albedo of disordered media. This peak is the last coherence effect which survives in the presence of complete disorder. It is built up by constructive interferences in the random walk of light, the same phenomenon responsible for the weak localization of electrons in bulk impure metals. On the other hand, the particular peak line shape, valid for two and three dimensions, is specific to the presence of an interface and could not be obtained from bulk considerations. It results from the addition of all contributions of nth-order multiple scattering, and the triangular singularity at $\theta=0$ can be reached only in the limit of n going to infinity. Any process which introduces a limitation in the order n of the multiple scattering or in other words in the total length of the random paths will round the peak. Finite-size confinement, absorption of light, or modulation of the light intensity are good candidates for these rounding processes.[15] Furthermore, since the peak line shape gives direct information on the transport of light in a random medium, an extension of this analysis towards non-Euclidean random walks is of great interest and will be reported in a forthcoming publication.[15] These considerations show the interest in this kind of experiment and analysis for the characterization of random media.

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[14]We neglect the weak-localization correction to the diffusion constant, which is allowed for λ/l small enough.

[15]E. Akkermans, G. Maret, R. Maynard, and P. E. Wolf, to be published.

[16]l^* is determined to be 19 μm by division of the experimental scattering mean free path 2.8 μm by the calculated average of $1-\cos\omega$, where ω is the scattering angle for a single latex sphere. $\lambda=0.515$ μm is the value of the wavelength in air, which accounts for the refraction at the water-air interface.

[17]However, in two dimensions, the bulk low critical dimensionality, one should also renormalize the diffusion constant, taking into account the presence of the interface.

[18]This dependence must not be confused with that of the recurrence probability in a three-dimensional bulk.