Coherent backscattering of light from random media with inhomogeneous gain coefficient

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We study theoretically the coherent backscattering cone of light from an amplifying random medium using realistic gain profiles which are spatially inhomogeneous. By taking into account correctly the amplification of multiply scattered waves, we are able to obtain quantitative agreements with the measured data presented in Phys. Rev. Lett. **75**, 1739 (1995) by Wiersma *et al.* and resolve the discrepancy arising from earlier calculations based on a uniform gain profile. [S0163-1829(97)00125-2]

The propagation of light in random media has been a subject under extensive study in the past decade due to many interesting correlation and fluctuation phenomena arising from the interference of multiply scattered waves.[1] An important interference effect exists between a multiply scattered wave and its time-reversed counterpart. This gives rise to an enhanced intensity in the backscattering direction[2] and is also the main mechanism for the localization of waves.[1] Recently, random media containing laser active materials have shown to produce interesting phenomena due to the interplay between amplification and randomness.[3] Zyuzin studied theoretically coherent backscattering of light from a random amplifying medium and predicted a narrowing of the coherent backscattering peak.[4] The physics of this narrowing is easy to understand. In the presence of a constant gain coefficient k_g, the amplitude of a multiply scattered wave of path length S is amplified by a factor $\exp(k_g S/2)$. The central part of the coherent backscattering cone is dominated by the contribution from waves with long path lengths. Since waves propagating along long paths are amplified more than those propagating along short paths, this gives rise to a sharpening of the coherent backscattering cone. This sharpening has also been found by taking the saturation effect and the width of the probe beam into account.[5] Recently, coherent backscattering from an amplifying medium was studied experimentally by Wiersma, Albada, and Lagendijk in random media consisting of Ti_2O_3-doped sapphire powders.[6] They have also calculated the backscattering cone based on diffusion theory with a uniform gain coefficient in the medium. Although the theoretical results are in good agreement with the experimental data at low gain levels, the calculation always overestimates the narrowing of the cone, which becomes apparent at higher gain levels. As was pointed out in Ref. 6, the reason for this discrepancy is due to the use of a constant gain coefficient in the calculations.

In general, in any specific experimental situation, the gain coefficient will not be homogeneous but it will be position-dependent inside the sample. Therefore, a theory is needed for coherent backscattering with gain, that includes a position-dependent gain coefficient. Usually, samples are pumped from the front surface, which results in a gain coefficient that dies off as a function of distance away from the front surface.[6,7] Assuming a position-independent gain coefficient will overestimate the amplification for waves with long path lengths and give rise to a narrower cone than is observed experimentally.

In order to resolve this discrepancy, we have studied theoretically coherent backscattering from a medium with a position-dependent gain coefficient. Basically, we have solved numerically the diffusion equation with a depth-dependent gain coefficient, which allows us to calculate the shape of the backscattering cone for any given gain profile. Using realistic gain profiles, we have tested our theory by calculating explicitly various backscattering cones corresponding to previous experiments. The results of our calculations remove the discrepancy between theory and experiment. We find excellent agreement for the cases reported in Ref. 6. The inhomogeneity of a gain profile is taken into account in a quantitative way.

In the absence of gain, the angular distribution of the backscattering intensity can be described by the sum of three bistatic scattering coefficients, γ_i, γ_ℓ, and γ_c, which represent the contributions from single scattering, diffusive transport, and interference between counterpropagating waves, respectively.[8] Most of the angular dependence comes from the interference term $\gamma_c(\theta)$, which is peaked in the backscattering direction ($\theta=0$) and decays to zero at large θ. This leads to a narrow cone of coherent backscattering which has a width $\delta\theta\approx\lambda/\ell$, where λ is the wavelength and ℓ is the transport mean free path.[2] The diffusive term γ_ℓ has a much weaker angular dependence and can be well approximated by $\gamma_c(0)$. The term γ_i can also be considered as angular independent. Thus, the interference term gives rise to an enhancement factor

$$E\equiv\frac{\gamma_\ell+\gamma_i+\gamma_c(0)}{\gamma_\ell+\gamma_i} \tag{1}$$

at $\theta=0$, which is the ratio of the intensity at the peak of the backscattering cone to the intensity in its wings. Due to the presence of the single scattering term γ_i, the enhancement factor is smaller than 2.

In the presence of gain, the bistatic scattering coefficients, γ_i, γ_ℓ, and $\gamma_c(\theta)$, are amplified. It is convenient to define a normalized intensity according to the diffuse background at zero gain:

$$I(\theta)\equiv\frac{\gamma_\ell+\gamma_i+\gamma_c(\theta)}{\gamma_\ell^0+\gamma_i^0}, \tag{2}$$

where γ_ℓ^0 and γ_i^0 represent the bistatic coefficients γ_ℓ and γ_i at zero gain. It has been found experimentally that the enhancement factor E is essentially unchanged in the presence of gain.[6] Using this fact and Eq. (1), we are able to rewrite Eq. (2) as

$$I(\theta)=\frac{\gamma_c(0)}{\gamma_c^0(0)}+\frac{\gamma_c(\theta)}{\gamma_c^0(0)}(E-1). \tag{3}$$

The value of E can be extracted from the experimental data. The important quantity to calculate is the bistatic coefficient $\gamma_c(\theta)$ describing the shape of the backscattering cone.

In the following, we will discuss the calculations of $\gamma_c(\theta)$ in the presence of an inhomogeneous gain profile $k_g(z)$, where z measures the distance away from the front surface. The term γ_c^0 is obtained by setting $k_g=0$.

It has been shown that for a slab of thickness L, the interference term can be expressed in terms of a double integral of the following form:[8]

$$\gamma_c(\theta)=\frac{1}{2\ell}\int_0^L\int_0^L dz_1dz_2F(q_\perp,z_1,z_2)\times(e^{-(\eta+i\delta)z_1-(\eta-i\delta)z_2}+\text{c.c.}), \tag{4}$$

where $\delta\equiv2\pi(1-\cos\theta)/\lambda$, $\eta\equiv[1+(\cos\theta)^{-1}]/2\ell$, $q_\perp\equiv2\pi\sin\theta/\lambda$, and c.c. denotes the complex conjugate. In principle, due to the presence of gain, the factor $1/\ell$ in the definition of η should be replaced by a position-dependent extinction coefficient $(1/\ell)-k_g$. However, since k_g is orders of magnitude smaller than $1/\ell$, the effect due to gain on the coherent part can be ignored. The integrand $F(q_\perp,z_1,z_2)$ is a Fourier transformation of the intensity Green's function, and is the solution of the transformed stationary diffusion equation

$$\frac{1}{3}\ell^2\left(\frac{\partial^2}{\partial z_1^2}-q_\perp^2\right)F(q_\perp,z_1,z_2)+\ell k_g(z_1)F(q_\perp,z_1,z_2)+\delta(z_1-z_2)=0, \tag{5}$$

with the boundary condition $F(q_\perp,z_1,z_2)=0$ in ''trapping'' planes at $z_1=-z_0$ and $L+z_0$ with $z_0\approx0.71\ell$.[9] Here the gain coefficient $k_g(z_1)$ in the above equation is depth dependent.

The solution of Eq. (5) can be obtained from the solutions of the following eigenvalue problem:

$$-\left[\frac{\partial^2}{\partial z^2}+\frac{3k_g(z)}{\ell}\right]\varphi_n(z)=\varepsilon_n\varphi_n(z), \tag{6}$$

with the same boundary condition as that of the function F in Eq. (5). Equation (6) is a one-dimensional Schrödinger-like equation with an ''infinite potential well'' of width $L+2z_0$ and an ''effective potential'' $-3k_g(z)/\ell$ inside the ''well.'' It is easy to see that, with the use of the normalization condition $\Sigma_n\varphi_n(z_1)\varphi_n^*(z_2)=\delta(z_1-z_2)$, a formal solution of the Green's function F can be expressed in terms of the eigenvalues ε_n and eigenfunctions $\varphi_n(z)$ of Eq. (6), i.e.,

$$F(q_\perp,z_1,z_2)=\frac{3}{\ell^2}\sum_n\frac{\varphi_n(z_1)\varphi_n^*(z_2)}{\varepsilon_n+q_\perp^2}, \tag{7}$$

where the summation sums all eigenstates $n=1,2,\ldots$. Substituting Eq. (7) into Eq. (4), we obtain a simple expression for $\gamma_c(\theta)$:

$$\gamma_c(\theta)=\frac{3}{\ell^3}\sum_n\frac{1}{\varepsilon_n+q_\perp^2}\left|\int_0^L dz\varphi_n(z)e^{-(\eta-i\delta)z}\right|^2. \tag{8}$$

In the case of a small gain coefficient, the eigenvalues ε_n in Eq. (8) are all positive and the summation gives a well-defined function $\gamma_c(\theta)$ for all θ. However, $\gamma_c(\theta)$ can become unstable when the gain coefficient is such that the smallest eigenvalue ε_1 approaches zero. This is easy to see when the gain coefficient is a constant $k_g(z)=k_g$. In this case Eq. (8) recovers the result of Ref. 6, and Eq. (6) has the solutions $\varphi_n(z)=\sqrt{2/(L+2z_0)}\sin[n\pi(z+z_0)/(L+2z_0)]$ and $\varepsilon_n=[n\pi/(L+2z_0)]^2-\ell_{\text{amp}}^{-2}$, where $\ell_{\text{amp}}\equiv(\ell/3k_g)^{1/2}$ denotes the amplification length. The lowest eigenvalue ε_1 decreases with L and becomes zero when $L+2z_0$ reaches the critical thickness $L_{\text{cr}}\equiv\pi\ell_{\text{amp}}$, where the scattered intensity diverges.[10]

In order to check our theoretical results, we have calculated explicitly the backscattering cones for the experimental situation of Ref. 6. Here powdered Ti:sapphire crystals were optically excited with high-energy laser pulses that were incident from the front sample surface. The shape of the gain profile in these samples can be calculated by solving numerically the diffusion equation for the pump light that excites the laser material, together with the rate equations that determine the local excitation of the system. This calculation was described extensively in Ref. 7, and comes down to the following. The gain coefficient is calculated from the local excitation level of the laser material, using $k_g=\sigma_{\text{em}}N_1(\mathbf{r},t)$, where σ_{em} is the cross section for stimulated emission and $N_1(\mathbf{r},t)$ denotes the population of the upper lasing state. Ti:sapphire is a standard four-level laser system in which the lasing ground state is essentially unpopulated. The rate equation for the upper lasing state is given by[11]

$$\frac{\partial N_1(\mathbf{r},t)}{\partial t}=\sigma_{\text{abs}}v[N_t-N_1(\mathbf{r},t)]\left[W_{\text{G}}(\mathbf{r},t)+\frac{1}{c}I_{\text{G}}(\mathbf{r},t)\right]-\sigma_{\text{em}}vN_1(\mathbf{r},t)W_{\text{A}}(\mathbf{r},t)-\frac{1}{\tau_e}N_1(\mathbf{r},t), \tag{9}$$

where $W_{\text{G}}(\mathbf{r},t)$ denotes the energy density of the excitation light and $W_{\text{A}}(\mathbf{r},t)$ the energy density of (amplified) spontaneous emission. Furthermore, v is the transport velocity of the light inside the medium, σ_{abs} the absorption cross section, τ_e the lifetime of the excited state, $I_{\text{G}}(\mathbf{r},t)$ the intensity

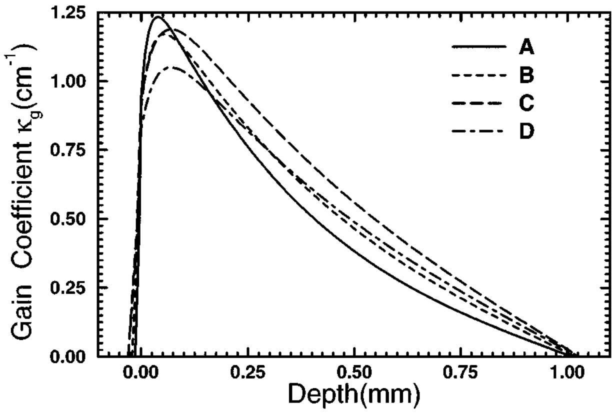

FIG. 1. The calculated gain coefficient $k_g(z)$ as a function of depth from the front surface for powdered Ti:sapphire samples at various transport mean free paths ℓ and pump energies E_p. Sample thickness 1 mm, particle diameter about 10 μm. Pump beam: diameter 5 mm, pulse width 14 ns, wavelength 532 nm. Curves A, B, C, and D represent ($\ell=18$ μm and $E_p=185$ mJ), ($\ell=28$ μm and $E_p=180$ mJ), ($\ell=40$ μm and $E_p=190$ mJ), and ($\ell=40$ μm and $E_p=165$ mJ), respectively.

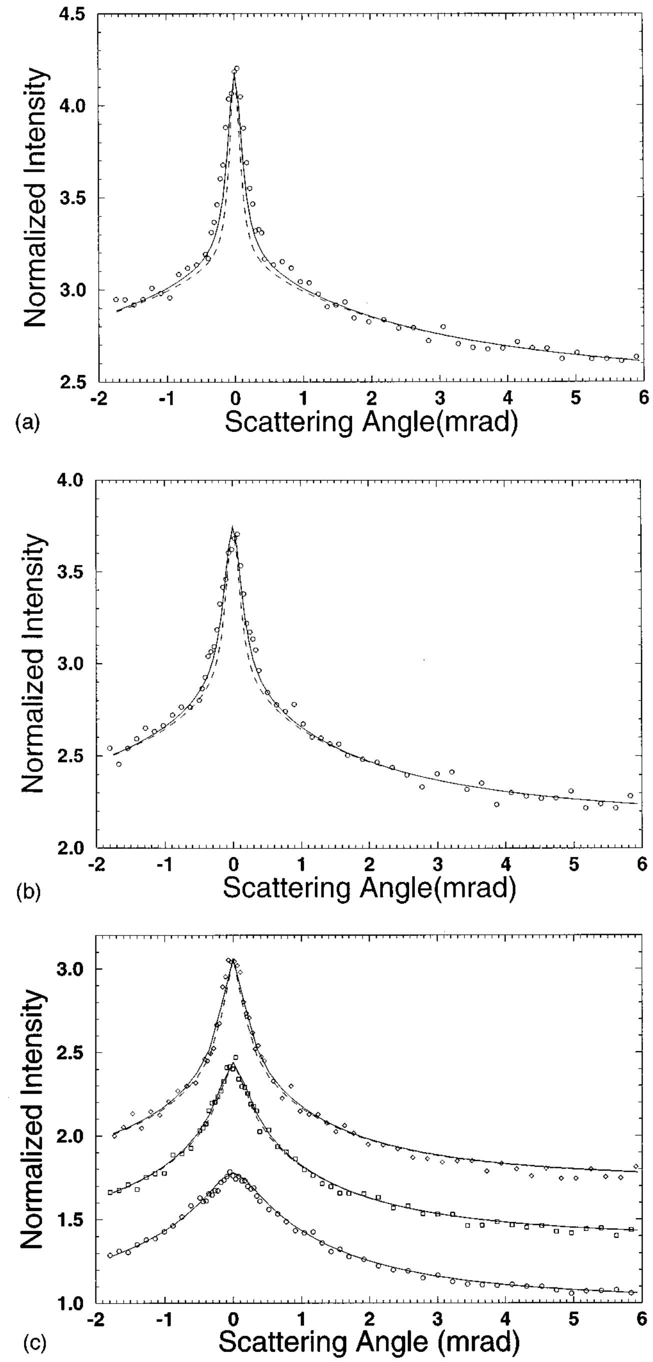

FIG. 2. Comparison between the calculated and measured backscattering cones (in the polarization conserving channel) from Ref. 6. The backscattered intensity is normalized to the diffusive background at zero gain. The backscattering cone in (a) is for the case of transport mean free path $\ell=18$ μm and pump energy $E_p=185$ mJ, which has the highest overall gain of 144%. The solid curve is the calculated result based on Eqs. (2) and (8). The open circles are the measured data. The dashed curve is the calculated result based on a uniform gain coefficient. (b) is for the case of $\ell=28$ μm and $E_p=180$ mJ. (c) is for the cases of $\ell=40$ μm and $E_p=190$ (upper curve) and 165 mJ (middle curve), which corresponds to the lowest gain level. The lower curve is the result of no gain. At high gain levels the inhomogeneity of the gain coefficient has to be taken into account to arrive at the right shape of the backscattering cone.

of the incoming pump light, and N_t is the total concentration of laser particles. For Ti:sapphire we have $\sigma_{\rm abs}=3.0\times10^{-24}$ m^2, $\sigma_{\rm em}=3.0\times10^{-23}$ m^2, and $\tau_e=3.2\times10^{-6}$ s. The samples consisted of 33% Ti:sapphire particles, corresponding (at a doping level of 0.15 wt % Ti_2O_3) to $N_t=1.6\times10^{25}$ m^{-3}. The diffusion equations that determine $W_{\rm G}(\mathbf{r},t)$ and $W_{\rm A}(\mathbf{r},t)$ are

$$\frac{\partial W_{\rm G}(\mathbf{r},t)}{\partial t}=D\nabla^2W_{\rm G}(\mathbf{r},t)-\sigma_{\rm abs}v[N_t-N_1(\mathbf{r},t)]W_{\rm G}(\mathbf{r},t)+\frac{1}{\ell}I_{\rm G}(\mathbf{r},t), \tag{10}$$

$$\frac{\partial W_{\rm A}(\mathbf{r},t)}{\partial t}=D\nabla^2W_{\rm A}(\mathbf{r},t)+\sigma_{\rm em}vN_1(\mathbf{r},t)W_{\rm A}(\mathbf{r},t)+\frac{1}{\tau_e}N_1(\mathbf{r},t), \tag{11}$$

where D is the diffusion coefficient inside the medium. Equations (9), (10), and (11) form a set of coupled differential equations that can easily be solved numerically.

The resulting depth dependence of the gain coefficient for the samples and pump energies used in the experiments of Ref. 6, is shown in Fig. 1. All curves show a similar behavior in the sense that the gain coefficient has a maximum close to the front sample interface, and than dies off with increasing depth. This shape is easy to understand if one realizes that the gain curve has roughly the shape of the energy distribution of the excitation light inside the slab. Without absorption, this energy distribution would decay linearly with thickness (regular diffusion in a slab geometry), but due to a small absorption, the curves become slightly bent in the middle.

Using the gain curves of Fig. 1, we can calculate explicitly $\gamma_c(\theta)$, as given by Eq. (8). The solution of Eq. (6) was found numerically by using the finite difference method. The number of obtained eigenstates depends on the spacing. We have systematically increased the number of eigenstates used in the evaluation of $\gamma_c(\theta)$ in Eq. (8) to ensure that a convergent result has been reached. A good convergence rate is guaranteed by the existence of a fast oscillating function $\varphi_n(z)$ in the integrand and the factor $(\varepsilon_n+q_\perp^2)^{-1}$ in Eq. (8). For each of the four cases shown in Fig. 1, more than 3000 eigenstates have been included. In order to check the accuracy of our calculations, we have also calculated $\gamma_c(\theta)$ for a case of constant gain. The result obtained this way from Eq. (8) agrees excellently with the analytic result given in Eq. (6) of Ref. 6. This provides a direct check on the reliability of our calculations. For the case of no gain, $\gamma_c^0(\theta)$ was calculated in a similar way.

For every sample, a measurement without gain was available, from which the mean free path ℓ and enhancement factor E could be determined. The enhancement factor lies between 1.70 and 1.78 in our case. Normally, the gain coefficient k_g is determined from the overall gain of the system, defined as the ratio between the total scattered intensity with amplification to the total scattered intensity without amplification. The overall gain can be measured at large angles, away from backscattering. We wish to include the depth dependence of the gain coefficient, as shown in Fig. 1. To match the overall gain of the system in our case, we multiplied the gain curves of Fig. 1 by a factor close to one (in practice between 1.12 and 1.36). The reason that the numerical calculation on the diffusion of pump light does not give immediately the overall value of k_g accurately is that the overall gain is very sensitive to, for instance, spatial inhomogeneities of the pump beam. In order to predict also the overall value of k_g accurately, one has to take into account the precise shape of the pump beam, which could be a subject of future work.

An example of a calculated backscattering cone is shown in Fig. 2(a), together with the corresponding experimental data. Here the transport mean free path is $\ell=18$ μm and the pulse energy of the pump beam $E_p=165$ mJ. The intensity at large θ gives an overall gain of 144%. The solid curve is our theoretical result including the explicit depth dependence of the gain. The agreement between the calculation and the measured data is excellent. Also shown in this figure is the calculated result obtained by using a constant gain coefficient (dashed curve). It is easy to see that a uniform gain profile always leads to a narrower cone compared to the measured one due to the overestimation of amplification for waves with long path lengths, and the narrowing is rather significant in this case. Since the amplification is correctly incorporated in Eqs. (5) and (6) by using a depth-dependent gain coefficient, we are able to obtain a much better agreement with the measured data.

Similar curves are shown in Fig. 2(b) for the case of $\ell=28$ μm and $E_p=180$ mJ, where the overall gain is 115%. The results for the case of $\ell=40$ μm are shown in Fig. 2(c), where the upper and middle curves are the data obtained for $E_p=190$ and 165 mJ, respectively. The corresponding overall gain is 71 and 36 %. The lower curve is the result of no gain. Excellent agreement between theory and experiment is also found in all three cases shown in Figs. 2(b) and 2(c). The dashed curves in Figs. 2(b) and 2(c) again represent the results of a constant gain coefficient. The narrowing due to a uniform gain profile diminishes with decreasing overall gain. For the case of $\ell=40$ μm and $E_p=165$ mJ, where the overall gain is 36%, the narrowing is insignificant and the middle dashed curve in Fig. 2(c) becomes indiscernible from the middle solid curve.

In conclusion, we have studied theoretically coherent backscattering from amplifying random media, considering explicitly the depth dependence of the gain coefficient. We find excellent agreement between theory and recent experiments on Ti:sapphire powders. Our study has resolved the discrepancy between the experimental data and theoretical calculations based on a constant gain coefficient. Although a calculation with a constant gain coefficient is capable of introducing good agreements with the measured data at low gain levels, it is necessary to consider a depth-dependent gain profile when the overall gain is above about 40%.

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