Coherent Backscattering of Light from Amplifying Random Media

Diederik S. Wiersma,[1] Meint P. van Albada[1] and Ad Lagendijk[1,2]
[1]FOM-Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands
[2]Van der Waals-Zeeman Laboratorium, Valckenierstraat 65-67, 1018 XE Amsterdam, The Netherlands
(Received 23 February 1995)

We report on coherent backscattering measurements from amplifying random media using optically pumped Ti:sapphire powders. The top of the backscattering cone sharpens with increasing gain, reflecting a change in the relative contributions to the intensity from paths of different lengths. A calculation based on diffusion theory is performed on coherent backscattering from amplifying random media, which is in good agreement with the data.

PACS numbers: 42.25.Hz, 42.70.Hj, 78.45.+h

The recent discovery of various interesting interference effects in light that is scattered from disordered structures has led to a renewed interest in multiple light scattering [1]. It was found that the interference between counter-propagating waves in disordered structures gives rise to enhanced backscattering. The phenomenon is known as coherent backscattering or weak localization [2]. Later, more interference effects were recognized like the spatial correlations in the intensity transmitted through random media. So far, experimental studies on these phenomena have been restricted to passive random media.

It is challenging to extend the field of study to active media, such as laser materials. An amplifying disordered medium can be made by combining multiple scattering with stimulated emission. Recently, it was shown that powdered laser crystals can serve as incoherent quasi-monochromatic light sources [3]. The behavior of an amplifying random medium is very different from that of a passive one. Since the amplification along a light path depends on the path length, the overall scattering properties depend strongly on the sample size. There is, for instance, a critical sample size above which the intensity diverges and the system becomes unstable. In that sense, multiple light scattering with gain is very similar to neutron scattering in combination with nuclear fission [4].

Coherent backscattering is a consequence of reciprocity [5] in multiple scattering. Consider a disordered sample that is illuminated by a (spatially homogeneous) light beam. A partial wave that propagates over some distance through the sample, and then leaves the illuminated spot in the backscattering direction, will have a counterpropagating counterpart of equal amplitude and phase. The waves will interfere constructively. Moving away from the exact backscattering direction, a phase difference will develop between the counterpropagating waves that depend on the relative orientation of the points where the waves enter and leave the sample. For the ensemble of light paths, the relative phases will therefore gradually randomize. After averaging over all light paths, this leads to a cone of enhanced backscattering which has a width of the order of $2\pi\lambda/\ell$ with ℓ the (transport) mean free path for light in the medium and λ its wavelength [2]. The top of this backscattering cone is due to very long light paths (features that are due to $>10^4$ scattering events are experimentally accessible), and coherent backscattering can therefore be expected to be strongly influenced by gain [6].

In this Letter we report on the observation of coherent backscattering from amplifying random media. Our samples consist of (0.15 wt %) Ti_2O_3 doped Ti:sapphire powders. The samples are strongly polydisperse (average particle size 10 μm). Two types of sample are obtained by either using the dry powder or suspending it in water. The latter type exhibits a longer (transport) mean free path. The samples have a slab geometry, with thickness 1 mm and diameter 15 mm.

The Ti:sapphire is optically pumped through the front sample interface with a frequency doubled Nd:YAG laser (wavelength 532 nm, pulse duration 14 ns) with pulse energy varying between 0 and 190 mJ. The lifetime of the excited state of Ti:sapphire is 3.2 μs. After a time delay of 14 ns, the green pump pulse is followed by a low intensity (40 μJ) red probe pulse (wavelength 780 nm, pulse duration 14 ns). The beams overlap spatially on the sample surface, and both have a diameter of 5 mm. The angular distribution of the scattered probe light is recorded using a setup as described in Ref. [7]. The probe light is linearly polarized, and both the polarization conserving and reversing channels have been monitored. The repetition rate of the pump (5 Hz) is half that of the probe, so that the sample is pumped just before every second probe pulse only. The detected signals from even and odd probe pulses are averaged separately, so one experimental run yields two backscattering cones (one with gain and one without).

Figure 1 shows backscattering cones as recorded for both polarization components from one and the same sample at different pump power. The overall scattered intensity increases with pump power. Moreover, in the polarization conserving channel, the top of the backscattering cone is seen to sharpen upon increasing the gain.

This sharpening becomes even more apparent if the y axis is scaled such that the background levels coincide

0031-9007/95/75(9)/1739(4)$06.00

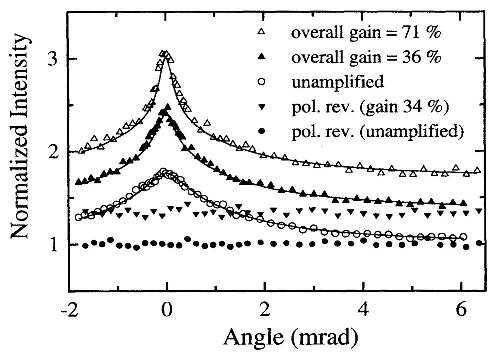

FIG. 1. Three backscattering cones in the polarization conserving channel and two backscattered intensities in the polarization reversing (pol. rev.) channel from the same sample at different pump power. Sample: 30% Ti:sapphire particles (10 μm) in water, transport mean free path: 40 μm. The overall gain of 0%, 36% (34% in the pol. rev. channel), and 71% corresponds to pump powers 0, 165, and 190 mJ. The intensity is normalized to the diffuse background at zero gain. Solid lines: calculated curves based on diffusion theory. Upon increasing the gain, the overall intensity increases and the top of the cone sharpens.

(see Fig. 2). It can be understood if one realizes that the shape of the cone reflects the path length distribution in the sample. The angular range of an interference contribution is inversely proportional to the distance between begin and end points of the paths that produce it. The wings of the cone are therefore mostly due to contributions from short paths. As we approach the top of the cone, interference contributions from successively longer paths add to the intensity. Because the amplification along a path will depend exponentially on its length, the introduction of gain will mainly affect the very central region of the backscattering cone, whereas the wings are hardly affected.

From Fig. 2 we also see that the enhancement factor is essentially independent of the gain. The enhancement

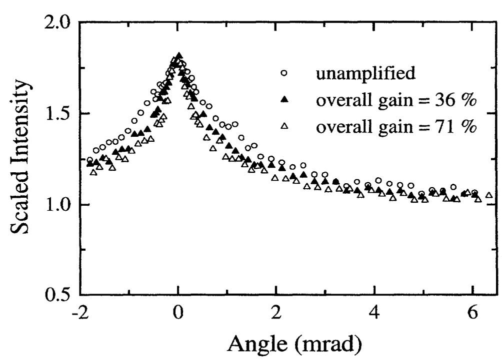

FIG. 2. Same backscattering cones as in Fig. 1 but y axis scaled such that background levels coincide. One can see that the top of the cone sharpens and that the enhancement factor is essentially independent of the gain.

is a consequence of reciprocity, which could be broken by the introduction of gain in the following way. After amplification of a pulse in part of the sample, the probed region is partly deexcited and the counterpropagating pulse will probe this region in a different state. For pulse durations comparable to the residence time of the light in the sample, this could affect reciprocity. In our system we do not expect this effect to occur, as the pulse duration by far exceeds the residence time and the pulse energy is low enough to leave the sample in almost the same state (there is no saturation of the gain at increasing probe power). This is consistent with our observation of a constant enhancement factor at increasing gain.

In the polarization reversed channel, correlation between the amplitudes and phases of counterpropagating waves diminishes with increasing order of scattering [8]. Coherent backscattering in this channel is due to low order scattering. Depending on the phase function of the scatterers, the enhancement factor ranges from 1.0 to about 1.2. We expect this enhancement factor to decrease with gain: Long path contributions are present in the background but not in the cone. In Fig. 1 it can be seen that, even without gain, the Ti:sapphire powders do not exhibit any significant enhancement factor in the polarization reversed channel and therefore do not allow us to measure this dependence of the enhancement factor on gain.

Coherent backscattering from passive random media can be calculated conveniently using diffusion theory [9]. Recently, Zyuzin [6] used diffusion theory to calculate the central cusp of the backscattering cone from an amplifying medium. In the following we will calculate the complete angular distribution of the backscattered intensity from an amplifying random medium. We start from the stationary diffusion equation with gain [4,10]:

$$\frac{1}{3}\ell^2\nabla^2_{\vec{r}_1}F(\vec{r}_1,\vec{r}_2) + \ell\kappa_g F(\vec{r}_1,\vec{r}_2) + \delta(\vec{r}_1 - \vec{r}_2) = 0\,. \tag{1}$$

Here $F(\vec{r}_1,\vec{r}_2)$ is the intensity Green function, κ_g is the (intensity) gain coefficient in the medium, and ℓ is the (transport) mean free path. We assume κ_g to be position independent. Because our system is translationally invariant over x and y, it is convenient to use the Fourier transform

$$F(\vec{q}_\perp, z_1, z_2) = \int F(\vec{r}_1,\vec{r}_2)e^{-i\vec{r}_\perp\cdot\vec{q}_\perp}d\vec{r}_\perp\,, \tag{2}$$

with $\vec{r}_\perp = \vec{r}_{1\perp} - \vec{r}_{2\perp}$ is perpendicular to z. The diffusion equation then reads

$$\frac{1}{3}\ell^2\left(\frac{\partial^2}{\partial z_1^2} - q_\perp^2\right)F(\vec{q}_\perp, z_1, z_2) + \ell\kappa_g F(\vec{q}_\perp, z_1, z_2) + \delta(z_1 - z_2) = 0\,. \tag{3}$$

The appropriate boundary condition for a slab with thickness L is $F(\vec{q}_\perp, z_1, z_2) = 0$ in "trapping" planes at distance z_0 on both sides of the slab, with $z_0 \approx 0.71\ell$

[11]. Solving Eq. (3) with this boundary condition we find

$$F(\vec{q}_\perp, z_1, z_2) = \frac{3\cos[\beta(L-z_s)] - 3\cos[\beta(L+2z_0-|z_d|)]}{2\ell^2\,\beta\,\sin[\beta(L+2z_0)]}, \tag{4}$$

where $z_s \equiv z_1 + z_2$, $z_d \equiv z_1 - z_2$, L is the slab thickness, and $\beta \equiv \sqrt{\ell_{\mathrm{amp}}^{-2} - q_\perp^2}$, with ℓ_{amp} the amplification length in the medium. We have to distinguish between the gain length $\ell_g \equiv \kappa_g^{-1}$, which is the length of a path along which the intensity is amplified by a factor e^{+1} and the amplification length $\ell_{\mathrm{amp}} \equiv \sqrt{\ell\,\ell_g/3}$, which is the (rms) average distance between begin and end points for paths of length ℓ_g. The amplification length ℓ_{amp} and gain length ℓ_g are the analogs of the absorption length ℓ_{abs} and the inelastic length ℓ_i in the case of absorption.

Note that the solution given in Eq. (4) is unstable for slabs thicker than a critical thickness $L_{\mathrm{cr}} \equiv \pi\ell_{\mathrm{amp}} - 2z_0$. In the limit $L \longrightarrow L_{\mathrm{cr}}$, the scattered intensity diverges. For a disordered laser material, the gain will saturate close to $L = L_{\mathrm{cr}}$, which provides an upper limit for the intensity.

To calculate the angular distribution of the backscattered intensity, the interference between counterpropagating waves must be incorporated. This interference yields a cosine modulation on the scattered intensity that depends on the phase difference $\vec{q}\,(\vec{r}_1 - \vec{r}_2)$ between a partial wave and its counterpropagating counterpart, with $\vec{r}_1$ and $\vec{r}_2$ the begin and end points of the path, and $\vec{q} \equiv \vec{k}_1 + \vec{k}_2$ the sum of incoming and outgoing wave vectors. In terms of a bistatic coefficient, the interference part of the backscattered intensity is given by [9]

$$\gamma_c(\theta) = \frac{1}{\ell}\int_0^L \int_{z_d}^{2L-z_d} F(\vec{q}_\perp, z_1, z_2) \times \cos(z_d\delta) e^{-\eta z_s} dz_s\, dz_d\,, \tag{5}$$

where $\delta \equiv k_0(1 - \cos\theta)$, $\eta \equiv (1 + \cos^{-1}\theta)/2\ell$, and θ is the angle of the outgoing wave vector $\vec{k}_2$ relative to the z axis. The incoming wave vector is taken along the z axis. The integrals in Eq. (5) can be performed explicitly using the intensity propagator from Eq. (4). The result is

$$\begin{aligned}\gamma_c(\theta) = {}& \frac{3e^{-\eta L}}{2\ell^3\beta\sin[\beta(L+2z_0)]}\,\frac{1}{(\beta^2+\delta^2+\eta^2)^2 - (2\beta\delta)^2} \\ & \times \Big[2(\beta^2+\delta^2+\eta^2)\cos(2\beta z_0)\cos(L\delta) - 4\beta\delta\sin(2\beta z_0)\sin(L\delta) \\ & + 2\frac{\beta}{\eta}(-\beta^2+\delta^2-\eta^2)\sin[\beta(L+2z_0)]\sinh(\eta L) \\ & + 2(\beta^2-\delta^2-\eta^2)\cos(L\delta) - 2(\beta^2+\delta^2+\eta^2)\cos[\beta(L+2z_0)]\cosh(\eta L) \\ & + 4\beta\eta\sin(\beta L)\sinh(\eta L) + 2(-\beta^2+\delta^2+\eta^2)\cos(\beta L)\cosh(\eta L)\Big].\end{aligned} \tag{6}$$

This solution agrees with the result of Zyuzin for small angles [6]. By taking the limit $\ell_{\mathrm{amp}} \longrightarrow \infty$, we obtain the solution for zero gain. In this limit, Eq. (6) is indeed equal to the bistatic coefficient from diffusion theory for absorbing media in the limit of zero absorption [9].

The backscattering cone is superimposed on an (essentially) angular independent background. This background consists of a diffusive term γ_ℓ, and contributions from paths that have no counterpropagating counterpart γ_{i} (e.g., single scattering and window artifacts), so the total backscattered intensity is

$$\gamma(\theta) = \gamma_\ell + \gamma_i + \gamma_c(\theta)\,. \tag{7}$$

We know (because of reciprocity) that γ_ℓ is equal to $\gamma_c(0)$. The value of γ_i can be found from the measured enhancement factor defined as

$$E \equiv \frac{\gamma_\ell + \gamma_i + \gamma_c(0)}{\gamma_\ell + \gamma_i}\,. \tag{8}$$

The solid lines in Fig. 1 are the backscattered intensities calculated using Eqs. (6) and (7). The mean free path ℓ for the sample and the enhancement factor E were inferred from the zero gain experiment (bottom curve). The gain coefficient κ_g was determined from the measured overall gain by comparing the measured and calculated intensities at large θ [12] and is the only parameter that varies between the three curves in Fig. 1. Values for the amplification length and gain coefficient at different pump energies are listed in Table I. The largest gain coefficient is about 0.9 cm^{-1}. The smallest amplification length is 0.336 mm and was measured in a sample with relatively large optical thickness.

Although general agreement between data and theory is very good in Fig. 1, we see that the upper calculated curve is slightly narrower than the measured one. The samples were pumped from the front side, so the gain decreases with increasing depth. Our assumption of a position independent κ_g therefore leads to a relative

TABLE I. Measured overall gain and calculated gain coefficient and amplification length at different pump powers and mean free paths. Also given is the calculated ratio of the sample thickness L to the critical thickness $L_{cr} \equiv \pi \ell_{amp} - 2z_0$. The actual sample thickness is always 1 mm.

ℓ (μm)	Pump energy (mJ)	Overall gain (%)	L/L_{cr}	κ_g (cm^{-1})	ℓ_{amp} (mm)
40	0	0	0	0	∞
40	165	36	0.74	0.720	0.428
40	190	71	0.83	0.896	0.384
28	180	115	0.91	0.746	0.351
18	180	134	0.94	0.537	0.338
18	190	155	0.95	0.542	0.336

overestimation of the amplification along the longer paths (that are more likely to probe the region where κ_g is below average). This effect is more apparent in Fig. 3, which shows a measurement on an optically thicker sample. We expect that a more homogeneous gain can be obtained by pumping the slab from both sides.

In conclusion, we have studied coherent backscattering from amplifying random media. In our system, reciprocity is preserved under optical amplification through stimulated emission. Upon increasing the gain we find a sharpening of the backscattering cone, reflecting an increased importance of long light paths inside the sample. A calculation based on diffusion theory is performed, which is in good agreement with our data. The possibility of making amplifying random media has opened the way to study the interesting combination of interference in multiple scattering with amplification through stimulated emission. Future experiments could involve speckle correlations from amplifying random media and studies on the effect of extremely strong scattering on the emission characteristics of laser materials. Also, amplification can be used to compensate for absorption which is present in all passive random media. Absorption has constituted a problem in the search for Anderson localization of light as it cuts off the longest light paths.

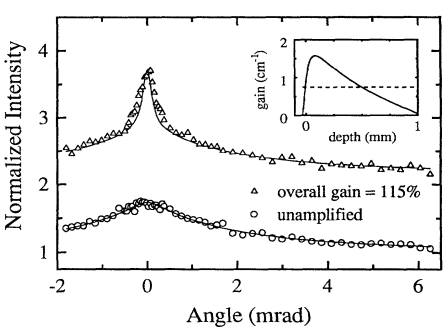

FIG. 3. Measured backscattering cones normalized to the angular independent background at zero gain compared with calculated curves. Upper curve: overall gain is equal to 115%, lower curve: unamplified. Sample: (dry) Ti:sapphire powder, transport mean free path: 28μm. The inset shows the calculated depth dependence of the gain coefficient $\kappa_g \equiv \ell_g^{-1}$ in cm^{-1} calculated by numerically solving the set of diffusion equations for pump and probe lights together with the rate equations of Ti:sapphire. The dashed line in the inset is the average κ_g, as determined from the data (see Table I) and used for the upper calculated curve. Because the calculation of the backscattering cone assumes constant gain, it overestimates the contributions from the longer (deeper penetrating) paths, resulting in a narrower cone.

We wish to thank Maarten Hagen for help with the experiment, Johannes de Boer and Mark van Rossum for discussions, Eric Jan Kossen for technical support, and Single Crystal Technology B.V. Enschede for providing us with the Ti:sapphire. The work in this Letter is part of the research program of the "Stichting voor Fundamenteel Onderzoek der Materie" (Foundation for Fundamental Research on Matter) and was made possible by financial support from the "Nederlandse Organisatie voor Wetenschappelijk Onderzoek" (Netherlands Organization for the Advancement of Research).

[1] For recent reviews, see *Classical Wave Localization,* edited by P. Sheng (World Scientific, Singapore, 1990); *Analogies in Optics and Micro Electronics,* edited by W. van Haeringen and D. Lenstra (Kluwer, Dordrecht, 1990).

[2] Y. Kuga and A. Ishimaru, J. Opt. Soc. Am. A **8**, 831 (1984); M.P. van Albada and A. Lagendijk, Phys. Rev. Lett. **55**, 2692 (1985); P.E. Wolf and G. Maret, Phys. Rev. Lett. **55**, 2696 (1985).

[3] C. Gouedard, D. Husson, C. Sauteret, F. Auzel, and A. Migus, J. Opt. Soc. Am. B **10**, 2358 (1993).

[4] B. Davison and J.B. Sykes, *Neutron Transport Theory* (Oxford University Press, New York, 1958).

[5] Optical measurements on linear physical systems obey the general principal of reciprocity, i.e., their results are invariant with respect to an interchange of source and detector (see, e.g., [11]). In the case of a conservative system, reciprocity is equivalent to time–reversal symmetry.

[6] A. Yu. Zyuzin, Europhys. Lett. **26**, 517 (1994).

[7] D.S. Wiersma, M.P. van Albada, and A. Lagendijk, Rev. Sci. Instrum. (to be published).

[8] M.P. van Albada and A. Lagendijk, Phys. Rev. B **36**, 2353 (1987).

[9] E. Akkermans, P.E. Wolf, and R. Maynard, Phys. Rev. Lett. **56**, 1471 (1986). Further developed in E. Akkermans, P.E. Wolf, R. Maynard, and G. Maret, J. Phys. (Paris) **49**, 77 (1988); M.B. van der Mark, M.P. van Albada, and A. Lagendijk, Phys. Rev. B **37**, 3575 (1988).

[10] V.S. Letokhov, Sov. Phys. JETP **26**, 835 (1968).

[11] H.C. van de Hulst, *Multiple Light Scattering* (Dover, New York, 1980).

[12] Here we assume that γ_i and γ_ℓ are amplified by the same factor. If part of the contributions to γ_i (e.g., single scattering) are not amplified, κ_g will be slightly underestimated.