Sharpening of enhanced backscattering peak in a disordered gain medium

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We study theoretically the phenomenon of enhanced backscattering (EBS) from a bulk disordered gain medium due to the presence of a dye and a pump beam. We show that the presence of the gain, in combination with the effect of saturation, can dramatically sharpen the EBS peak function. In the particular case of a point source incidence for the probe beam along with the presence of a plane-wave pump beam, the EBS approaches a δ function in angular space.

The phenomenon of enhanced backscattering (EBS) of light from a bulk disordered medium constitutes perhaps the clearest example of the importance of phase coherence in optical propagation in the multiple-scattering regime. Following its first experimental observation in the early 1980s,¹ this effect has received much attention in the optics community.^{2,3} The scientific importance of the EBS is further enhanced by its direct analogy to the quantum correction of conductance in disordered metals at low temperatures due to weak localization,⁴ as both effects originate from the perfect phase coherence of time-reversed interfering multiple-scattering paths in the backscattering direction. More recently, the phenomenon of EBS has been analyzed in the context of light scattering from random rough surfaces,^{5,6} and it is hoped that the EBS peak function can be used to extract useful information about the geometrical and scattering properties of the random surfaces under study.

The study of EBS has been mainly confined to various random systems in linear optics. Recently, several authors started to address the issue of what would happen in the nonlinear optical regime. In particular, Kravtsov, Agranovich, and Grigorishin⁷ showed that in the case of second-harmonic generation (due to χ_2 nonlinear susceptibility⁸) the EBS peak height for second-harmonic light is very much reduced from the usual factor of 2, due to the restriction that only second-harmonic light generated near the sample surface contributes to the EBS signal, whereas those generated anywhere in the bulk contribute to the diffuse background. Later Agranovich and Kravtsov⁹ predicted that when the χ_3 nonlinear susceptibility⁸ is taken into account, the EBS peak function of linear optics must be modified by the appearance of a narrow “dip” very close to the backscattering direction, although this result has recently been challenged by Heiderich, Maynard, and van Tiggelen.¹⁰

In this paper, we study how the EBS peak is modified in a disordered multiple-scattering medium by a different source of nonlinearity, namely when the probe light can gain intensity due to stimulated emission. Initial treatment of this problem was provided by Zyuzin recently,¹¹ who, however, did not take into account the important effect of saturation. We shall devise a simple model in the present paper to address this physical effect.

A concrete, experimentally relevant, system we have in mind is a dye-dissolved fluid containing a high concentration of scatterers (e.g., polystyrene spheres). For simplicity, we model the dye as a three-level atomic system with energies $E_1 < E_2 < E_3$ such that level E_2 is metastable. If a pump beam at frequency $\omega_p = (E_3 - E_1)/\hbar$ is applied to the system, population inversions between E_2 and E_1 can be achieved, such that a probe light at frequency $\omega = (E_2 - E_1)/\hbar$ passing through the sample will experience gain. Let us denote the gain coefficient as μ_g (in units of cm^{-1}). Another important feature of a disordered gain medium is the nonlinear effect of saturation. If the gain coefficient were to remain a constant, it is evident from physical ground that for a sufficiently large system, the probe light intensity will become exponentially amplified to infinity. This scenario is not permitted on physical ground, because when the probe light intensity increases, the population inversion decreases, which causes μ_g to decrease, such that in steady state, a saturation value for the probe light intensity is reached, which is a finite fraction of that of the pump light. Our main conclusion in this paper is that the presence of positive gain and the constraint of saturation in combination can induce a dramatic sharpening of the EBS peak, up to the limit of a *delta function* when the incident probe beam is of the form of a point source.

The geometry that we consider is the standard semi-infinite space where $z > 0$ defines the region of the multiple-scattering medium. We assume the incident pump beam is approximately a continuous (cw) plane wave, with a width W large compared to all the other length scales in the problem. The incoherent diffuse optical intensity of the pump beam can be obtained from the diffusion equation

$$-D_p \frac{d^2}{dz^2} \phi_p(z) = I_p [\delta(z - l_p^*) - \delta(z - l_p^* + a_p)], \quad (1)$$

where $\phi_p(z)$ is a function of z alone due to the assumption of a plane-wave incidence (in the normal direction), l_p^* is the transport mean free path of the pump beam, $D_p = cl_p^*/3$ the corresponding diffusion coefficient, I_p is the flux of the incident pump beam, and $a_p = 2(l_p^* + z_{0p})$ is the distance between the diffusive source and its image. We have followed

here the standard treatment of the boundary condition for the diffusion approximation for incoherent light transport in a multiple-scattering medium, and assumed $\phi_p = 0$ at the extrapolated boundary $z = -z_0$ with z_0 being a parameter dependent on the contrast in the index of refraction at the interface (e.g., $z_{0p} \approx 0.7l_p^*$ for the index-matched case). Solving Eq. (1) yields the result that the pump light intensity inside the medium for $z > l_p^*$ is approximately a constant, given by

$$\phi_p(z) = \frac{a_p I_p}{2D_p}, \quad \text{valid for } z > l_p^*. \quad (2)$$

Now we consider the incoherent light intensity for the probe beam at $\omega = (E_2 - E_1)/\hbar$. Let us first discuss the case where the incident probe beam is also a cw plane wave, with optical flux I , and beam width $W \gg l^*, l_p^*$, where l^* is the transport mean free path for the probe beam. For simplicity, we approximate the gain process at ω phenomenologically by a constant gain coefficient μ_g , together with a cutoff-type saturation procedure, as we now illustrate.

If the incident source were not a cw but an impulse, i.e., $S(z, t) = I[\delta(z - l^*) - \delta(z - l^* + a)]\delta(t)$ where $a = 2(l^* + z_0)$, then the light intensity at time t should be described by the time-dependent diffusion equation

$$\frac{\partial}{\partial t} \phi(z, t) - D \frac{\partial^2}{\partial z^2} \phi(z, t) - c\mu_g \phi(z, t) = S(z, t), \quad (3)$$

where $D = cl^*/3$. The solution is simply given by the time-domain one-dimensional (1D) diffusion propagator

$$\begin{aligned} \phi(z, t) = \frac{I}{\sqrt{4\pi Dt}} & \left\{ \exp\left[-\frac{(z - l^*)^2}{4Dt}\right] \right. \\ & \left. - \exp\left[-\frac{(z - l^* + a)^2}{4Dt}\right] \right\} e^{c\mu_g t}. \end{aligned} \quad (4)$$

We see that with a constant gain coefficient, the light intensity response to an impulse at $t = 0$ for the probe beam diverges as $t \rightarrow \infty$. This is clearly unphysical; so we impose a saturation condition by introducing a phenomenological saturation time parameter T , such that the intensity response to a plane wave cw source $S(z, t) = I[\delta(z - l^*) - \delta(z - l^* + a)]$ is given by

$$\begin{aligned} \phi(z) = I \int_0^\infty \frac{dt}{\sqrt{4\pi Dt}} & \left\{ \exp\left[-\frac{(z - l^*)^2}{4Dt}\right] \right. \\ & \left. - \exp\left[-\frac{(z - l^* + a)^2}{4Dt}\right] \right\} e^{f(t)}, \end{aligned} \quad (5)$$

where $f(t) = c\mu_g t$ for $t \leq T$ and $f(t) = c\mu_g T$ for $t > T$. It is clear that T models physically, albeit crudely, the nonlinear saturation effect which cuts off gain when the probe light intensity reaches a certain threshold value, corresponding to a finite fraction of the pump light density in the medium.

Equation (5) can be rewritten in the form

$$\phi(z) = I e^{c\mu_g T} \left[\int_0^\infty P_0(z, t) dt - \int_0^T P_0(z, t) g(t) dt \right], \quad (6)$$

where $g(t) = 1 - e^{-c\mu_g(T-t)}$, and $P_0(z, t)$ is the 1D diffusion propagator in the absence of gain:

$$\begin{aligned} P_0(z, t) = \frac{1}{\sqrt{4\pi Dt}} & \left[\exp\left(-\frac{(z - l^*)^2}{4Dt}\right) \right. \\ & \left. - \exp\left(-\frac{(z - l^* + a)^2}{4Dt}\right) \right]. \end{aligned} \quad (7)$$

We shall see later that the condition $c\mu_g T \gg 1$ usually holds. In this case, for t within the range of integration $0 < t < T$ in the second term of Eq. (6), the function $g(t)$ can be well approximated by $g(t) \approx \theta(T' - t)$ (step function) with $T' = T - 1/c\mu_g \approx T$, since $g(t = T) = 0$ exactly. Thus we arrive at the approximate expression

$$\phi(z) \approx I e^{c\mu_g T} \int_{T'}^\infty P_0(z, t) dt. \quad (8)$$

After some simple manipulations, we arrive at a form for $\phi(z)$:

$$\phi(z) \approx \frac{aI}{2D} e^{c\mu_g T} E\left(\frac{z + z_0}{\sqrt{4DT'}}\right), \quad (9)$$

where $E(x) \equiv 2/\pi \int_0^x e^{-y^2} dy$ is an error function, and $z_0 \approx 0.7l$ for index-matched interface. The behavior of $\phi(z)$ is quite simple, it rises linearly for $z \ll z_T$, and saturates at $\phi_\infty = (aI/2D) e^{c\mu_g T}$ for $z \gg z_T$, where $z_T \equiv \sqrt{4DT'}$ is a diffusive depth associated with the saturation time T .

The saturation time parameter can be determined experimentally by making the physical saturation requirement $\phi(z \gg z_T) = \gamma \phi_p$ where the constant percentage factor $\gamma < 1$ is a property of the dye material. Thus we can write

$$c\mu_g T = \ln \frac{\gamma I_p}{I} + \ln \frac{a_p D}{a D_p} \approx \ln \frac{\gamma I_p}{I}, \quad (10)$$

i.e., T is mainly determined by the ratio of incident pump light intensity and that of the probe light. So if I is chosen to be much smaller than γI_p , the condition $T \gg 1/c\mu_g$ is easily fulfilled.

In order to calculate the EBS peak function in the presence of gain, we need to calculate the following cw diffusion propagator [$\vec{r} \equiv (\vec{\rho}, z)$]

$$\begin{aligned} Q(\vec{\rho}, z) &= \int_0^T e^{c\mu_g t} P_0(\vec{\rho}, z; t) dt + \int_T^\infty e^{c\mu_g T} P_0(\vec{\rho}, z; t) dt \\ &\approx e^{c\mu_g T} \int_{T'}^\infty P_0(\vec{\rho}, z; t) dt, \end{aligned} \quad (11)$$

where $P_0(\vec{\rho}, z; t)$ is the 3D time-dependent diffusion propagator without gain, given by

$$\begin{aligned} P_0(\vec{\rho}, z; t) &= \frac{1}{(4\pi Dt)^{3/2}} \left[\exp\left(-\frac{\rho^2 + (z - l^*)^2}{4Dt}\right) \right. \\ & \left. - \exp\left(-\frac{\rho^2 + (z - l^* + a)^2}{4Dt}\right) \right]. \end{aligned} \quad (12)$$

Setting $y^2 = 1/4DT$, $Q(\vec{\rho}, z)$ can be rewritten as

$$Q(\vec{\rho}, z) = \frac{e^{c\mu_g T}}{4\pi D} \frac{2}{\sqrt{\pi}} \int_0^{1/z_T} dy [e^{-y^2[\rho^2 + (z-l^*)^2]} - e^{-y^2[\rho^2 + (z-l^*+a)^2]}] \quad (13)$$

where $z_T = \sqrt{2D(T - 1/\mu_g)} \approx \sqrt{2DT}$. In the linear optics case without gain, we let $T \rightarrow 0$ and $z_T \rightarrow 0$ in the above expression, and we recover the familiar expression²

$$Q(\vec{\rho}, z) = \frac{1}{4\pi D[\rho^2 + (z-l)^2]^{1/2}} - \frac{1}{4\pi D[\rho^2 + (z-l+a)^2]^{1/2}}. \quad (14)$$

The z -dependent incoherent light intensity $\phi(z)$ is related to the propagator $Q(\vec{r})$ simply by $\phi(z) = \int d^2\rho Q(\vec{r})$. Following Akkermans, Wolf, and Maynard,² the albedo function $\alpha(\hat{k}_i, \hat{k}_f)$ (defined as the ratio of detected optical flux per unit area per solid angle in the detection direction $\hat{k}_f$ to the incident flux of the probe light) contains a contribution from the incoherent re-radiated light as well as that due to the coherent interference between the time-reversed multiple-scattering paths, which has the form

$$\alpha(\hat{k}_i, \hat{k}_f) = \frac{c}{4\pi} \int d^2\rho [1 + \cos(\vec{q} \cdot \vec{\rho})] Q(\vec{\rho}, z \approx l^*), \quad (15)$$

where $\vec{q}$ is the projection of $\vec{k}_i + \vec{k}_f$ in the x - y plane, which measures the angular deviation from the exact backscattering direction where the phase coherence of the time-reversed multiple-scattering paths is perfect. Near normal incidence, $q \approx |\vec{q}| \approx k\theta$ where θ is the detector angle measured from the backscattering direction. After some algebra, we find

$$\alpha(\theta) = \frac{ce^{c\mu_g T}}{(4\pi)^2 D} \frac{2}{\sqrt{\pi}} [I(0,0) - I(0,a) + I(q,0) - I(q,a)], \quad (16)$$

where

$$I(q,z) = \int d^2\rho \cos(\vec{q} \cdot \vec{\rho}) \int_0^{1/z_T} dy \exp[-y^2(\rho^2 + z^2)]. \quad (17)$$

In the absence of gain, it has been shown² that

$$\alpha(\theta) \approx \alpha_{\text{inc}} \left(1 + \frac{1 - e^{-qa}}{qa} \right),$$

where α_{inc} is the incoherent background. This can be obtained from our formalism by taking the limit $T \rightarrow 0$. Thus the angular width of the EBS peak in linear optics is $\Delta\theta \approx 1/ka$ and the height of the EBS peak relative to the incoherent background is a factor of 2. In the presence of gain, it follows from Eq. (16) by setting $q=0$ ($\theta=0$) that the relative EBS height is still a factor of 2.

For $q \neq 0$, using the following integral involving the Bessel function J_0 ,¹²

FIG. 1. Comparison of normalized EBS peak function $\alpha(\theta)/\alpha_{\text{inc}}$ in the absence (dotted line) to that in the presence of gain (solid line). We choose a saturation time parameter $T \approx 15$ ps. Other parameters chosen are $c \approx 3 \times 10^8$ m/s, $l^* = 19$ μm , $\lambda = 0.515$ μm , corresponding to realistic experimental values. This gives a saturation depth $z_T = 107$ $\mu\text{m} \gg l^*$.

$$\int_0^\infty x e^{-\alpha x^2} J_0(\beta x) dx = \frac{1}{2\alpha} e^{-\beta^2/4\alpha}.$$

$I(q,z)$ may be rewritten as

$$I(q,z) = \pi \int_0^{1/z_T} dy \frac{\exp(-z^2 y^2 - q^2/4y^2)}{y^2}. \quad (18)$$

For $q \neq 0$, setting $x = q/y$, $I(q,z)$ may be written in the following form:

$$I(q,z) = \frac{\pi}{q} \int_{qz_T}^\infty e^{f(x)} dx, \quad (19)$$

where

$$f(x) = -\frac{(qz)^2}{x^2} - \frac{x^2}{4}.$$

For $qz_T \gg 1$, $I(q,z)$ can be approximately evaluated using the method of steepest descent, with the result

$$I(q,z) \approx \frac{\pi \sqrt{\pi}}{2q} e^{-qz} e^{-(qz_T)^2} \quad (20)$$

leading to the following expression for the albedo function:

$$\alpha(\theta) \approx \alpha_{\text{inc}} \left[1 + \frac{1}{2} \left(\frac{1 - e^{-qa}}{qa} \right) e^{-(qz_T)^2} \right] \quad (21)$$

which is valid for $\theta \gg 1/kz_T$. Since $z_T \gg a$, the angular width of the EBS in the presence of gain is reduced to $\Delta\theta \approx 1/kz_T$. As z_T depends on the ratio of the incident pump beam intensity and that of the probe beam, it should be interesting to observe experimentally how the EBS beam width gets reduced when the pump beam intensity is increased while keeping that of the probe beam fixed.

In Fig. 1, we illustrate the sharpening effect due to gain by plotting the normalized albedo $[\alpha(\theta)/\alpha_{\text{inc}}]$ in the absence (dotted line) as well as in the presence of gain (solid line). We choose a saturation time parameter $T \approx 15$ ps, and $c \approx 3 \times 10^8$ m/s, $l^* = 19$ μm , $\lambda = 0.515$ μm , corresponding to

realistic experimental values. This gives a saturation depth $z_T = 107 \mu\text{m} \gg l^*$. Thus the angular width in the absence of gain (dotted line) is $\Delta\theta \approx \pi/ka \approx 0.22^\circ$ and the angular width in the presence of gain (solid line) is $\Delta\theta \approx \pi/kz_T \approx 0.03^\circ$. The effect of the narrowing and sharpening of the EBS peak is quite obvious.

We also remark that in the usual linear optics regime without gain, the diffusion approximation usually used is not really valid for $\Delta\theta \gg 1/ka$ as at these large angles only scattering paths of order a few mean free paths contribute to $\alpha(\theta)$. But in the presence of gain, the albedo is dominated by long scattering paths, thus the diffusion approach becomes a very good approximation for the problem.

The above discussion is based on the assumption of a plane-wave probe beam incidence. In the case where the incident probe beam is focused to an area A of order l^2 (since $l \gg \lambda = 2\pi/k$, we can still use the notion of a well-defined incident wave vector $\vec{k}_i$), the sharpening effect of the EBS peak due to gain is even more dramatic. In this case, the diffusive probe-light intensity is simply

$$\phi(\vec{\rho}, z) = IAQ(\vec{\rho}, z), \quad (22)$$

assuming the incident light is focused at the origin $\vec{\rho} = 0$. Since in the saturation regime, we must have

$$\phi(\vec{\rho}, z) = \gamma\phi_p(z) \quad (23)$$

and since ϕ_p is ρ independent, we must require a ρ -dependent saturation time $T(\rho)$ such that ϕ is independent of ρ . From Eqs. (22), we see that this implies a ρ -independent 3D propagator $Q(\rho, z \approx l^*)$! Using Eqs. (15), (2), (22), and (23), we find that in this case the EBS peak function is sharply peaked, i.e., we may write

$$\alpha(\theta) = \frac{cW^2\gamma I_p a}{8\pi D_p IA} (1 + e^{-k^2 W^2 \theta^2}), \quad (24)$$

where $W \gg l^*$ is simply the width of the pump beam which can be made arbitrarily large! This effect of extreme sharpening of the EBS peak in the presence of gain with a point source incidence probe beam deserves immediate experimental verification.

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⁸ χ_2 and χ_3 are defined by the following expansion of the relation between the displacement vector $\vec{D}$ and the electrical field $\vec{E}$:

$$\vec{D} = \vec{E} + 4\pi\chi_1\vec{E} + \chi_2 \cdot \vec{E}\vec{E} + \chi_3 \cdot \vec{E}\vec{E}\vec{E},$$

where χ_1 is the usual linear optical susceptibility with $\epsilon = 1 + 4\pi\chi_1$. χ_2 is a second-rank nonlinear optical tensor which is responsible for the well-known phenomenon of second-harmonic generation. Similarly, χ_3 is a third-rank next-order nonlinear optical tensor.

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