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Coherent backscattering of light by nonlinear scatterers

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We theoretically study the propagation of light in a disordered medium with nonlinear scatterers. We especially focus on interference effects between reversed multiple scattering paths, which lead to weak localization and coherent backscattering. We show that, in the presence of weakly nonlinear scattering, constructive interferences exist in general between *three* different scattering amplitudes. This effect influences the nonlinear backscattering enhancement factor, which may thus exceed the linear barrier two.

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Light transport inside a nonlinear medium gives rise to a wide variety of phenomena, such as pattern formation, four waves mixing, self-focusing effects, dynamical instabilities, etc. [1]. These effects are well described and understood with the help of an intensity dependent susceptibility, e.g., $\chi^{(3)}$ nonlinearity. However, in these approaches, one usually discards the fact that interference phenomena in disordered systems may significantly alter wave transport properties. Indeed, considering the return probability to a given point, all scattering paths are now closed loops. Then the *two-wave* interference between amplitudes contra-propagating along these loops typically increases the return probability by at most a factor of 2, inducing a decrease of the diffusion constant (the weak localization effect). How nonlinear effects affect weak localization is basically unknown and the present paper is aimed at showing that this could be more important than naively expected. Coherent random lasers [2] are probably the most striking systems intrinsically combining both nonlinear effects and disorder. Even if in this case one would require an active (i.e., amplifying) medium, a key point is the understanding of the mutual effects between multiple interferences and nonlinear scattering.

An effect similar to weak localization is *coherent backscattering* (CBS) where an enhancement of the average intensity scattered around the direction opposite to the incident wave is observed [3]. In the linear scattering regime, CBS also arises from a two-wave interference between amplitudes entering and leaving the medium in opposite directions and contrapropagating along all possible scattering paths. Thus both the CBS and the weak localization are described by the so-called "maximally crossed diagrams" [4]. The CBS enhancement factor, defined as the signal detected in the exact backscattering direction divided by the diffuse background, never exceeds two. This maximum value is reached if each pair of interfering waves has the same amplitude, and if single scattering can be suppressed. Previous studies of the nonlinear regime have been restricted to the case of linear scatterers embedded in a uniform nonlinear medium [5–7]. Here, it has been shown that the maximum enhancement factor remains two.

As we will show in this paper, however, the situation drastically changes in the presence of nonlinear scattering (in contrast to nonlinear propagation). In particular, in the perturbative regime of at most one scattering event with $\chi^{(3)}$ nonlinearity, CBS arises from interference between three amplitudes. Depending on the sign of the nonlinearity, this leads to an increase or decrease of the nonlinear CBS enhancement factor compared to the linear value two. Since the same physics is at work for weak localization corrections to transport, a corresponding change of the diffusion constant is expected, too. Because, for photons, CBS is easier to observe than weak localization, we specifically concentrate on the former.

In this paper, we calculate CBS by a dilute gas of nonlinear scatterers. We assume that the cross section of a single scatterer situated at position $\mathbf{r}$ inside the disordered medium depends on the local intensity $I(\mathbf{r})$ as follows:

$$\sigma(\mathbf{r}) = \sigma_0[1 + \alpha I(\mathbf{r})], \tag{1}$$

where σ_0 denotes the linear cross section, and α quantifies the strength of the nonlinearity, which is proportional to the $\chi^{(3)}$ coefficient of the scattering material. The local intensity $I(\mathbf{r})$ is the intensity due to all external sources, i.e., the light radiated by all other scatterers and the incident light penetrating the medium until $\mathbf{r}$ without being scattered. For future convenience, we measure $I(\mathbf{r})$ in units of the incident intensity I_{in} (before entrance into the medium). Thus, α is dimensionless and also proportional to I_{in}. The general form (1) of the nonlinear cross section is obtained under the assumption of small scatterers, i.e., constant local intensity inside the scatterer, weak $\chi^{(3)}$ nonlinearity (i.e., higher-order terms like I^2 negligible), and isotropic scattering. The following treatment can also be generalized to the nonisotropic case, however. At the end, we will present numerical results where we take into account the polarization state of the light field.

Besides the scattering cross section, the second ingredient needed for the description of a multiple scattering process is the propagation between two scattering events. Under the condition that no other scattering event occurs in between, the disorder averaged intensity propagator is given by an exponentially damped spherical wave

$$\overline{P(\mathbf{r},\mathbf{r}')} = \frac{e^{-|\mathbf{r}-\mathbf{r}'|\langle 1/\ell\rangle}}{4\pi|\mathbf{r}-\mathbf{r}'|^2}. \tag{2}$$

Here, $\langle 1/\ell\rangle$ denotes the mean value of the inverse mean free path along a straight line connecting the two scattering events at $\mathbf{r}$ and $\mathbf{r}'$. In the linear case ($\alpha=0$), the mean-free-path ℓ_0 is constant, and is related via $1/\ell_0=\mathcal{N}\sigma_0$ to the linear cross section σ_0 and the scatterer density $\mathcal{N}$. This relation is a consequence of energy conservation, which ensures that the exponential attenuation of propagating field modes originates solely from scattering into other modes. Similarly, since we assume energy conservation for the nonlinear case, too (i.e., no absorbing or amplifying scatterers), we can also derive the nonlinear mean free path from the nonlinear scattering cross section, Eq. (1). Since the nonlinear contribution to the total intensity $\sigma(\mathbf{r})I(\mathbf{r})$ scattered from $\mathbf{r}$ is, according to Eq. (1), proportional to $\alpha I(\mathbf{r})^2$, we need to know the disorder-averaged *squared* intensity for this purpose. In a perturbative expansion up to first order in α, we can here replace $I(\mathbf{r})$ by its linear counterpart $I_0(\mathbf{r})$, whose fluctuation properties are well known [8]. By assuming uniformly distributed phases for the fields radiated by the other scatterers (which is valid in the case of a dilute medium), one obtains

$$\overline{I_0^2(\mathbf{r})} = 2\overline{I_0(\mathbf{r})}^2 - (e^{-z/\ell_0})^2. \qquad (3)$$

The second term, with z the distance from the boundary of the medium to $\mathbf{r}$ along the incident direction, represents the squared intensity of the incident, coherent mode. It accounts for the fact that—in contrast to the diffuse light—the (linear) coherent mode intensity does not fluctuate for different realizations of the disorder. By equating the intensity loss due to propagation with the scattered intensity (i.e., employing energy conservation), we therefore obtain from Eq. (3) different expressions for the mean free paths for diffuse and coherent light

$$\frac{1}{\ell(\mathbf{r})} = \frac{1}{\ell_0}(1 + 2\alpha\overline{I_0(\mathbf{r})}), \qquad (4)$$

$$\frac{1}{\ell_c(\mathbf{r})} = \frac{1}{\ell_0}(1 + 2\alpha\overline{I_0(\mathbf{r})} - \alpha e^{-z/\ell_0}). \qquad (5)$$

We can now write down a nonlinear radiative transfer equation for the average intensity $\overline{I(\mathbf{r})}$ inside the disordered medium. Radiative transport is obtained by representing $\overline{I(\mathbf{r})}$ as the incoherent sum of the coherent incident field mode plus the diffuse light radiated from all individual scatterers

$$\overline{I(\mathbf{r})} = e^{-z\langle 1/\ell_c\rangle} + \mathcal{N}\int_V d\mathbf{r}'\,\overline{P(\mathbf{r},\mathbf{r}')} \times \overline{\sigma(\mathbf{r}')I(\mathbf{r}')}, \qquad (6)$$

where $\langle 1/\ell_c\rangle$ denotes the mean value of the inverse coherent mean free path, Eq. (5), along the corresponding path of length z. In the second term, representing the diffuse light, the disorder average is decorrelated. This is justified by the fact that correlations between intensities at different positions (separated much further than the optical wavelength) can be neglected in the case of a dilute medium [4]. In the case $\alpha=0$, Eq. (6) reduces to the familiar linear radiative transfer equation [4], whose iterative solution yields $\overline{I_0(\mathbf{r})}$. To proceed, we expand Eq. (6) up to the first order in the nonlinearity parameter α. Introducing Eq. (3), we obtain a closed equation for the average intensity $\overline{I}$, which we solve by iteration. Finally, the average intensity of the backscattering signal follows via

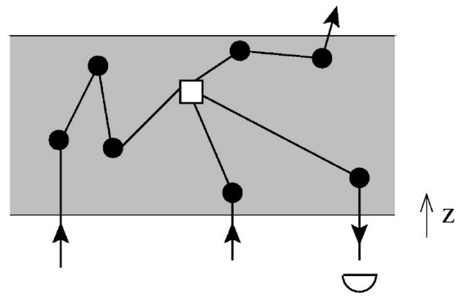

FIG. 1. In the perturbative approach, we assume a single nonlinear (□), but arbitrarily many linear scattering events (●). The nonlinear event symmetrically connects two linear propagators with each other. One of them finally reaches the detector placed in backscattering direction.

$$L = \mathcal{N}\int_V \frac{d\mathbf{r}}{A} e^{-z\langle 1/\ell\rangle} \times \overline{\sigma(\mathbf{r})I(\mathbf{r})}, \qquad (7)$$

with A the transverse area of the medium. Expanding again to the first order in α, we identify the linear and nonlinear part, $L=L_0+L_1$, respectively. According to whether α originates from the cross section σ or the mean-free-path ℓ (or ℓ_c), the latter splits into a nonlinear scattering and nonlinear propagation component, i.e., $L_1=L_1^{(sc)}+L_1^{(pr)}$. For a slab geometry of length L, i.e., (linear) optical thickness $b=L/\ell_0$, we obtain

$$L_0 = \int_0^b d\zeta\,\overline{I_0}(\zeta)e^{-\zeta}, \qquad (8)$$

$$L_1^{(sc)} = \alpha\int_0^b d\zeta\,\overline{I_0}(\zeta)[2\overline{I_0}^2(\zeta) - e^{-2\zeta}], \qquad (9)$$

$$L_1^{(pr)} = -\alpha\int_0^b d\zeta\,\overline{I_0}(\zeta)[2\overline{I_0}^2(\zeta) - 2\overline{I_0}^2(b) - e^{-\zeta} + e^{-2\zeta}], \qquad (10)$$

where we have introduced the (linear) optical depth $\zeta=z/\ell_0$. Note that the first terms in Eqs. (9) and (10) cancel each other. This is not surprising if one keeps in mind that energy conservation ensures the total outgoing flux to equal the incoming one. Thus, the nonlinear contribution vanishes even completely, if one considers the total detection signal, integrated over all directions in forward and backward direction. We have checked that Eqs. (8)–(10) are also obtained by using diagrammatic scattering theory [9], if only the so-called "ladder" diagrams are retained—thus neglecting recurrent scattering effects [10] and interferences between different scattering paths—and if, in addition, all diagrams with more than one nonlinear scattering event are discarded (see Fig. 1).

On top of the above background intensity, a narrow interference cone of height C is observed, originating from the interference between reversed scattering paths, which is de-

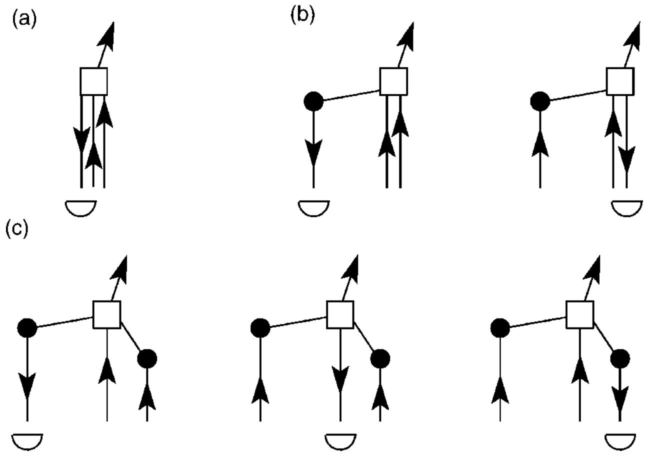

FIG. 2. In the presence of nonlinear scattering (□), there may be either (b) two, or (c) three interfering amplitudes contributing to enhanced backscattering, apart from single scattering (a), which only contributes to the background. In general, the case (c), which corresponds to the maximum enhancement factor three, is realized if either both incoming propagators, or one incoming and the outgoing detected propagator exhibit at least one linear scattering event (●) besides the nonlinear one.

scribed by the so-called maximally crossed diagrams. Due to time reversal symmetry, each maximally crossed diagram has the same value as the corresponding ladder diagram. In the linear case, there is exactly one reversed counterpart for each scattering path, except for those exhibiting only a single scattering event. Hence, the cone height equals the background, provided that single scattering is removed from the latter. In the presence of nonlinear scattering, however, there may be either two or three interfering amplitudes. As exemplified in Fig. 2, this is due to the fact that two linear propagators are symmetrically connected by the nonlinear event, which permits, in general, two different possibilities to reverse the propagator that finally reaches the detector. In the expression for the background component, Eq. (9), the three cases of Fig. 2 can be identified by writing the local intensity $\overline{I_0}=\exp(-\zeta)+\overline{I_d}$ as a sum of the coherent and diffuse part, respectively. Then, all terms of at least second power in $\overline{I_d}$ correspond to the case (c), those linear in $\overline{I_d}$ to case (b), and the remaining ones to case (a). From this decomposition, C is easily obtained, since the ratio of the cone height to the background depends solely on the number of interfering amplitudes. In particular, the three-amplitudes case (c) contributes to the interference cone twice as much as to the background. In the two-amplitudes case (b), a small complication arises, since the right-hand amplitude of Fig. 2(b) is twice as large as the left-hand one [11]. Only in the latter one, both propagators arriving at the nonlinear scattering event originate from the coherent mode, and hence the asymmetry is related to the different fluctuation properties of diffuse and coherent light, expressed by Eq. (3). Here, the ratio between cone height and background is obtained as $(1\times2+2\times1)/(1\times1+2\times2)=4/5$. Finally, the single scattering terms (a) do not contribute to the cone, and must be removed from Eq. (9). Thereby, we obtain

$$C_1^{(\mathrm{sc})}=4\alpha\int_0^L d\zeta[\overline{I_0}^3(\zeta)-2\overline{I_0}(\zeta)e^{-2\zeta}+e^{-3\zeta}]. \qquad (11)$$

Concerning nonlinear propagation, interference between the three amplitudes does not occur [5–7]. Formally, the reason is that in this case the two incoming propagators are not connected symmetrically by the nonlinear scattering event. Instead, they can physically be distinguished from each other, as one of them (the "probe") keeps the direction of propagation, whereas the other one ("pump") is scattered. Hence, there are only two interfering amplitudes, obtained by reversing the path of the probe. Just as for the linear component, it is sufficient to remove single scattering contributions from the background, Eq. (10).

The perturbative results derived above allow us to calculate the CBS enhancement factor $\eta=1+C/L$ up to the first order in the nonlinearity coefficient α. In particular, we obtain the first derivative of $\eta(\alpha)$ at $\alpha=0$, which quantifies the modification of CBS enhancement due to a small nonlinearity. In our case, the strength of the nonlinearity is limited by the perturbative assumption of at most one nonlinear scattering event. In order to estimate roughly its domain of validity, we have analyzed the statistical distribution of the number N of scattering events in linear backscattering paths, by numerical simulations with slab geometry. If we associate with each scattering event a constant probability proportional to α to be nonlinear (thereby neglecting the inhomogeneity of the local intensity), we find that the occurrence of two or more scattering events can be neglected provided that $\alpha b^2 \ll 1$. Let us note that a similar condition also ensures the stability of speckle fluctuations in a nonlinear medium [12].

We want to stress that the above treatment, valid for scalar point scatterers, can be extended to any kind of nonlinear scatterer with $\chi^{(3)}$ nonlinearity. Specifically, we have analyzed the vectorial case, where the polarization of the light field is taken into account. This case is especially interesting, since in the helicity preserving ($h\|h$) polarization channel single scattering contributions are filtered out, thus realizing the maximum linear enhancement factor two. Hence, any deviation of the enhancement factor from two can unambiguously be attributed to the nonlinear effect of interference between three amplitudes. Numerically, we have treated the vectorial case by using a Monte Carlo method, where the positions of the scattering events are randomly chosen.

The results for the scalar and vectorial ($h\|h$) case are shown in Fig. 3, as a function of the optical thickness b. Evidently, the slope $m=d\eta/d\alpha|_{\alpha=0}$ increases with b, since a nonlinear scattering event is more likely to occur at larger optical thickness. Thus, for large optical thickness, a significant change of η results already from a small nonlinearity α. In the vectorial case, the nonlinear influence on η is smaller. The main reason for this is the following: Due its explicit dependence on the polarization vectors attached to the two incoming and outgoing propagators, the nonlinear scattering amplitude does not remain invariant when exchanging a single incoming and outgoing propagator. (Only if *all* propagators are reversed, invariance is guaranteed by time-reversal symmetry.) This causes a polarization-induced loss of inter-

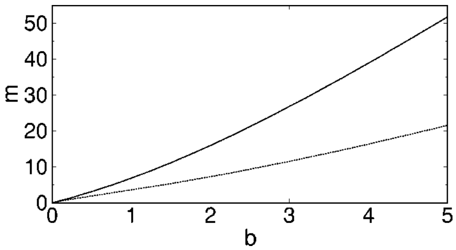

FIG. 3. Modification $m = d\eta/d\alpha|_{\alpha=0}$ of the CBS enhancement factor η induced by a small nonlinearity α, for backscattering from a slab of optical thickness b, in the scalar case (solid lines) and the $h\|h$ polarization channel (dotted lines). For a large optical thickness, already a small nonlinearity leads to a significant increase (or decrease, depending on the sign of α) of η. In the $h\|h$ channel, the nonlinear CBS modification is smaller than in the scalar case, as a consequence of decoherence due to polarization effects.

ference contrast, i.e., a reduction of the coherent nonlinear scattering component $C_1^{(sc)}$ (approximately by a factor 3/4). Nevertheless, the effect of the three-amplitudes interference still prevails, such that in total a positive slope is observed. In particular, the CBS enhancement factor is predicted to exceed the linear barrier two, if the sign of the nonlinearity α is positive. Due to the close relation between CBS and weak localization mentioned above, we thus expect that weak localization—and possibly also strong localization—are facilitated by positive nonlinearities.

An alternative method to observe enhancement factors larger than two is provided by using atomic scatterers. As a specifically quantum mechanical property of the atom-photon interaction, nonlinearity is here intrinsically related to *inelastic* scattering, where the frequency of scattered photons changes. On the one hand, inelastic scattering acts as a source of decoherence between reversed scattering paths, with ensuing decrease of the CBS enhancement factor [11]. On the other hand, however, it allows to distinguish linearly and nonlinearly scattered light in terms of its frequency. Thereby, the linear components L_0 and C_0 can be filtered out from the detection signal, so that the nonlinear effect of interference between three amplitudes can manifest itself especially clearly, unspoiled by linear contributions. To minimize decoherence, the frequency filter must be sufficiently narrow and be put as close to the initial frequency as possible, but far enough to filter out elastically scattered light. In this limit, the backscattering enhancement factor is exclusively given by the nonlinear scattering components derived above, i.e., $\eta = 1 + C_1^{(sc)}/L_1^{(sc)}$. For sufficiently large optical thickness, we thereby predict maximum values of the CBS enhancement factor up to 3 (scalar case) or 2.5 ($h\|h$ channel).

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[1] R. W. Boyd, *Nonlinear Optics* (Academic, San Diego, 1992).
[2] H. Cao, Waves Random Media **13**, R1 (2003).
[3] 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).
[4] M. C. W. van Rossum and T. M. Nieuwenhuizen, Rev. Mod. Phys. **71**, 313 (1999).
[5] V. M. Agranovich and V. E. Kravtsov, Phys. Rev. B **43**, 13691 (1991).
[6] A. Heiderich, R. Maynard, and B. A. van Tiggelen, Opt. Commun. **115**, 392 (1995).
[7] R. Bressoux and R. Maynard, in *Waves and Imaging through Complex Media*, edited by P. Sebbah (Kluwer Academic Publishers, Dordrecht, 2001).
[8] J. W. Goodman, in *Laser Speckle and Related Phenomena*, edited by J. C. Dainty (Springer, Berlin, 1984).
[9] A. J. van Wonderen, Phys. Rev. B **50**, 2921 (1994).
[10] D. Wiersma, M. P. van Albada, B. A. van Tiggelen, and A. Lagendijk, Phys. Rev. Lett. **74**, 4193 (1995).
[11] T. Wellens, B. Grémaud, D. Delande, and C. Miniatura, Phys. Rev. A **70**, 023817 (2004).
[12] S. E. Skipetrov and R. Maynard, Phys. Rev. Lett. **85**, 736 (2000).